

OPTIMIZATION OF THE PELLETIZATION PROCESS OF AGRICULTURAL WASTES ORIGINATING FROM OLIVE FARMS FOR THEIR APPLICATION IN DOMESTIC BOILERS

OPTIMIZACIÓN DEL PROCESO DE PELLETIZACIÓN DE RESIDUOS AGRÍCOLAS PROCEDENTES DEL OLIVAR PARA SU APLICACIÓN EN CALDERAS DOMÉSTICAS

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MANIFIESTAN:

DE Maraver para optar al grado de Doctor con Mención Internacional por la Universidad de SU APLICACION EN CALDERAS DOMESTICAS', presentada por Dña. Ángela García Granada, ha sido realizada bajo nuestra dirección en el Departamento de Ingeniería Civil de la Universidad de Granada y, por ello, autorizamos la presentación de la PELLTIZACION DE RESIDUOS AGRICOLAS PROCEDENTES DEL OLIVAR PARA PROCESO presente Memoria titulada "OPTIMIZACION DEL Que la misma.

Granada, a 14 de enero de 2013

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Fdo. Angel Fermín Ramos Ridao

Memoria presentada por Dña. Ángela García Maraver para optar al Grado de Doctor

con Mención Internacional por la Universidad de Granada.

Fdo. Ángela García Maraver

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TÍTULO DE DOCTOR CON MENCIÓN INTERNACIONAL

Con el fin de obtener el Título de Doctor por la Universidad de Granada con Mención Internacional, que el Real Decreto 99/2011 establece en su artículo 15, se han cumplido los siguientes requisitos:

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(ii) Parte de la tesis doctoral se ha redactado y presentado en una de las lenguas habituales para la comunicación científica en su campo de conocimiento, distinta a cualquiera de las lenguas oficiales en España.

(iii) La tesis ha sido informada por un mínimo de dos expertos doctores pertenecientes a alguna institución de educación superior o instituto de investigación no española.

(iv) Un experto perteneciente a alguna institución de educación superior o centro de investigación no española, con el título de doctor, y distinto del responsable de la estancia mencionada en el apartado (i), forma parte del tribunal evaluador de la tesis.

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PUBLICACIONES INTERNACIONALES INCLUIDAS EN EL JCR

A. García-Maraver, V. Popov, M. Zamorano, 2011. A review of European standards for pellet quality. Renewable Energy; 36 (12): 3537-3540. Impact Factor: 2.972 (Q2 Energy and Fuels).

Zamorano M, Popov V, Rodríguez ML, García-Maraver A, 2011. A comparative study of quality properties of pelletized agricultural and forestry lopping residues. Renewable Energy; 36: 3133-3140. Impact Factor: 2.972 (Q2 Energy and Fuels).

A. García-Maraver, M. Zamorano, A. Ramos-Ridao, L.F. Díaz, 2012. Analysis of olive grove residual biomass potential for electric and thermal energy generation in Andalusia (Spain). Renewable and Sustainable Energy Reviews; 16 (1): 745–751. Impact Factor: 6.018 (Q1 Energy and Fuels).

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RESUMEN

Los problemas ambientales y de dependencia energética, derivados del elevado consumo de combustibles fósiles, han hecho necesario el desarrollo de medidas dirigidas a impulsar nuevos modelos energéticos renovables, sostenibles, eficientes, económicamente viables, prácticos y seguros, que den respuesta a las necesidades de abastecimiento de energía en el contexto de un desarrollo sostenible.

Entre las energías renovables, la biomasa residual está destinada a jugar un papel importante en el nuevo marco energético, ya que los residuos agrícolas se producen en cantidades relativamente grandes en todo el mundo, por lo que suponen una forma renovable y medioambientalmente segura de proporcionar energía.

En las regiones mediterráneas del suroeste europeo, entre ellas Andalucía, el cultivo del olivar presenta un elevado valor ambiental gracias a su contribución en el desarrollo de sistemas sostenibles, como sumidero de gases de efecto invernadero y por su potencial en la generación de energías renovables, ya que parte de la biomasa que se produce como residuo agrícola e industrial de este sector presenta un alto contenido energético.

El uso de la biomasa residual del olivar destinado a la producción de energía térmica a nivel doméstico (calefacción y agua caliente) se presenta por tanto, como una alternativa que precisa sin embargo de un impulso en la investigación asociada a impulsar canales de comercialización de un biocombustible competitivo. En este sentido su densificación haría posible la conversión de un residuo difícil de gestionar en una fuente de energía especialmente indicada para su uso en las zonas donde es generada.

Dada la falta de información en lo relativo al uso de la biomasa residual del olivar con los fines indicados, el presente trabajo de investigación ha evaluado el potencial de la biomasa residual del olivar como fuente de energía renovable en zonas mediterráneas, más concretamente en el caso de Andalucía y ha estudiado el proceso de pelletización de éstos para su aplicación en calderas domésticas, de acuerdo al marco



regulador que establece los parámetros de calidad físicos, químicos y mecánicos de los pellets a nivel europeo, y las emisiones generadas en el proceso de combustión.

Los resultados obtenidos ponen de manifiesto el importante potencial de este tipo de residuos en las áreas en las que se extiende el cultivo del olivar. Este tipo de biomasa, al igual que otras de naturaleza residual agrícola, ha mostrado algunas limitaciones en la fabricación de pellets de aplicación en sistemas doméstico; éstas han sido debidas fundamentalmente a un mayor contenido en cenizas y cloruros, especialmente consecuencia de la presencia de la hoja, que también han afectado a las emisiones en caldera doméstica, sobre todo en una mayor emisión de partículas. No obstante, la reducción de la presencia de hoja en el proceso de fabricación, un diseño de calderas adaptado a las características de pellets de este origen, así como el desarrollo de un marco regulador acorde a la naturaleza de este tipo de biomasa, supondrá el impulso de una industria estratégica desde el punto de vista de la seguridad energética y del desarrollo económico, social y medioambiental sostenible, en las regiones del área mediterránea, entre ellas Andalucía.

ABSTRACT

Environmental and energy dependency problems derived from the high fossil fuels consumption have made necessary the development of new energy models that have to be renewable and sustainable, efficient and economically effective, practical and save, to meet the demand of a sustainable energy supply.

Among renewable resources, residual biomass is destined to play an important role in the new energy model since agricultural residues are a local energy resource which is produced in relatively large amounts throughout the world and it is regarded as a renewable and environmentally safe way of providing energy.

In the Mediterranean areas of European Southwest, such as Andalusia, olive tree pruning residues have a high environmental value since they contribute to the development of sustainable systems and reduce the greenhouse effect. Moreover, agricultural and industrial residues from the olive sector have a great energy potential.

The use of residual biomass from olive trees in thermal energy production at a domestic scale is thus an alternative that needs more research on the improvement of the quality of solid biofuels. In this regard, the densification of these residues would allow the conversion of a residue difficult to manage into an energy resource especially indicated to be used in the areas where the residues are generated.

Given the lack of sufficient information concerning residual biomass from olive trees as well as the combustion and emission characteristics of these residues, the aim of the current research was to analyze the energy potential of this residual biomass as a renewable energy source in Andalusia, to analyze the pelletization process of this biomass that would allow it to be used in domestic boilers according to the regulatory framework that establishes the chemical, physical and mechanical quality parameters of pellets at a European level, as well as to better understand the thermal behavior and the gaseous and particulate emissions.

The results obtained state the important energy potential of these residues in the areas where they are generated. This type of biomass, as well as other agricultural



residues, has shown some limitations regarding the production of pellets for nonindustrial use. These limitations are due to high contents of ash and chlorine, specially as a consequence of the presence of leaves, which also affects to the gaseous and particulate emissions. Nevertheless, the reduction of the use of leaves in the production process, the design of combustion systems fully adapted to the biomass of each region, and the development of a regulatory framework in accordance with the origin of the biomass, will lead to the impulse of a strategic industry in the Mediterranean regions and especially in Andalusia.

INTRODUCCIÓN, MOTIVACIÓN Y OBJETIVOS

El desarrollo de la sociedad ha venido acompañado de un gran consumo de energía (IDAE, 2005). Para satisfacer la creciente necesidad energética, el ser humano ha recurrido al uso de fuentes de energía fósiles como son el carbón, el petróleo o el gas natural, lo cual ha hecho que países que carecen de estos combustibles, como es el caso de España, dependan en más de un 75% de un suministro exterior de energía (Dinica, 2009).

Esta situación hace que el bienestar social, el crecimiento económico y la competitividad empresarial de las regiones se vean amenanzados por la elevada vulnerabilidad del modelo energético actual ante futuros problemas de abastecimiento energético. Además, en los últimos años se ha introducido una nueva varible que está siendo sin duda la fuerza impulsora hacia un cambio de la percepción mundial de la cuestión energética: el cambio climático derivado de las emisiones de gases de efecto invernadero (Omer, 2008). Debido a la gravedad de estos problemas, de índole mundial, se ha hecho necesario el desarrollo de medidas desde el punto de vista energético y medioambiental por parte de todos los países con el fin de reducir los efectos negativos derivados del uso de combustibles fósiles sobre el planeta.

Entre estas medidas se destaca la necesidad de utilización de fuentes de energía renovables como alternativa al uso de los combustibles fósiles como eje de un nuevo modelo energético que de respuesta a las necesidades de abastecimiento de energía sin generar desequilibrios ambientales, económicos y sociales, en el contexto de un desarrollo sostenible. Las fuentes de energías renovables se producen de forma continua, siendo inagotables a escala humana. Hoy en día, estas energías son una de las soluciones más eficientes y efectivas para la consecución de un desarrollo energético sostenible y de la prevención de la contaminación ambiental.

En este sentido, la política energética y de protección del clima en Europa, se ha plasmado en tres objetivos estratégicos: seguridad en el suministro, eficiencia y



compatibilidad ambiental. Para alcanzarlos hay que actuar en las áreas prioritarias de generación y consumo energético, así como en la de emisiones, asumiendo los compromisos de adaptación recogidos en la Directiva 2009/28/CE del Parlamento Europeo y del Consejo, de 23 de abril de 2009, relativa al fomento del uso de energía procedente de fuentes renovables, que establece objetivos mínimos vinculantes para el conjunto de la Unión Europea y para cada uno de los Estados miembros. Esta Directiva establece como objetivo conseguir una cuota mínima del 20% de energía procedente de fuentes en el consumo final bruto de energía de la Unión Europea y una cuota mínima del 10% de energía procedente de fuentes renovables en el consumo final bruto de genergía en el consumo de energía en el sector del transporte en cada Estado miembro para el año 2020. Además, la Directiva requiere que cada Estado miembro elabore y notifique a la Comisión Europea (CE), un Plan de Acción Nacional de Energías Renovables (PANER) para el periodo 2011-2020, con vistas al cumplimiento de los objetivos vinculantes que fija la Directiva.

España, en su paso hacia el establecimiento un nuevo modelo energético, ha desarrollado una serie de herramientas jurídicas, normativas y de planificación que permitan abanderar el proceso de transformación necesario para hacer efectivos esos compromisos. Para ello, una vez agotado el período de vigencia del Plan de Energías Renovables (PER) 2005-2010 y atendiendo al mandato establecido en la legislación vigente¹, el Consejo de Ministros aprobó el pasado 11 de noviembre de 2011 el PER 2011-2020. Este Plan incluye el diseño de nuevos escenarios energéticos así como la incorporación de objetivos acordes con la Directiva 2009/28/CE y los mandatos del Real Decreto 661/2007, por el que se regula la actividad de producción de energía eléctrica en régimen especial y de la Ley 2/2011, de 4 de marzo, de Economía Sostenible². Tras el anterior PER (2000-2010), que estableció una contribución de las fuentes de energías renovables del 12% con respecto al consumo total de energía en España, la aprobación del PER 2011-2020 pretende mantener el compromiso de cubrir con fuentes renovables al menos el 20% del consumo de energía final en el año 2020,

¹ Real Decreto 661/2007, por el que se regula la actividad de producción de energía eléctrica en régimen especial y, posteriormente, Ley 2/2011, de 4 de marzo, de Economía Sostenible

² La política energética estará orientada a garantizar la seguridad del suministro, la eficiencia económica y la sostenibilidad medioambiental. En especial, el modelo de consumo y de generación y distribución de energía debe ser compatible con la normativa y objetivos comunitarios y con los esfuerzos internacionales en la lucha contra el cambio climático. A dichos efectos, en esta Ley se fijan objetivos nacionales de ahorro energético y participación de las energías renovables.

mismo objetivo que para la media de la UE, junto a una contribución mínima del 10% de fuentes de energía renovables en el transporte para ese año; estos objetivos, a su vez, han quedado recogidos en la Ley 2/2011, de Economía Sostenible.Además la aprobación de este plan eleva el alcance de las finalidades de la política energética, aspirando a un nuevo modelo energético que propicie cambios estructurales en el sistema y la consolidación de una cultura energética impregnada de una conciencia colectiva que considere la energía como un bien valioso y escaso. En esta reorientación estructural del sistema energético español se debe buscar un reposicionamiento basado en el mejor aprovechamiento de los recursos energéticos del país y en la definición global de un modelo energético plenamente adaptado a las condiciones climáticas, culturales y económicas de las diferentes regiones.

España, aunque deficitaria en recursos energéticos fósiles, es muy rica en recursos renovables cuyo aprovechamiento permitirá dotar al país de una energía autóctona y segura que minimice el futuro impacto de inestabilidades del mercado energético internacional. En el nuevo modelo energético, las energías renovables ocupan por derecho propio un puesto predominante por su carácter sostenible, distribuido y en armonía con el medio ambiente. Las fuentes de energía renovables a las que se refieren las herramientas de planificación actuales son: biocombustible sólidos y líquidos, biogás, biomasa, energías del mar, eólica, geotermia y otras energías del ambiente, hidroeléctrica, residuos (municipales, industriales y lodos de EDAR) y solar (fotovoltaica, térmica y termoeléctrica). Como resultado de la política de apoyo a las energías renovables en España, en el marco del PER 2005-2010, el crecimiento de éstas durante los últimos años ha sido notable, y así, en términos de consumo de energía primaria, han pasado de cubrir una cuota del 6,3% en 2004 a alcanzar el 11,3% en 2010. Este porcentaje correspondiente al año 2010 se eleva al 13,2% si se calcula la contribución de las energías renovables sobre el consumo final bruto de energía, de acuerdo con la metodología establecida en la Directiva 2009/28/CE.

También a nivel regional, como en el caso de Andalucía, se está trabajando en la búsqueda de la implementación de nuevos modelos energéticos. Así, la Reforma del



Estatuto de Autonomía fijó el marco competencial de la Comunidad Autónoma, en el que reconoce explíticamente estas políticas a partir de la definición de competencias compartidas en energía en instalaciones de producción, distribución y transporte, en fomento y gestión de las energías renovables y en eficiencia energética. Para ello, una vez agotado el período de vigencia del Plan Energético de Andalucía 2003-2006, se puso en marcha el Plan Andaluz de Sostenibilidad Energética 2007-2013 (PASENER), que establece un nuevo modelo energético para esta región basado en sus condiciones climáticas, culturales y económicas. Por otro lado, la aprobación del Código Técnico de la Edificación, mediante el Real Decreto 314/2006, el nuevo marco retributivo a las energías renovables establecido en el Real Decreto 661/2007, la Ley 2/2007 de Fomento de las Energías Renovables y del Ahorro y la Eficiencia Energética de Andalucía y la consolidación de la Agencia Andaluza de la Energía como instrumento para alcanzar el objetivo de optimizar el abastecimiento energético de Andalucía, conforman un marco robusto en el que cimentar los pilares de la política energética de la Comunidad Andaluza en los próximos años.

En los planes de fomento de energías renovables, tanto en el marco europeo como español y andaluz, se reconoce la importancia de la biomasa residual agrícola como fuente de energía renovable, y medioambientalmente segura de proporcionar energía: (i) es una abundante fuente energética cuyo uso conlleva beneficios socioeconómicos y ambientales significativos a partir de la generación de residuos; (ii) es una fuente de biomasa barata, obtenida a partir de los productos residuales procedentes del procesado de la madera o de la agricultura y que permite por tanto la valorización energética de residuos orgánicos forestales y agrícolas; (iii) su uso mejora la gestión de residuos y mitiga el impacto ambiental de la disposición de residuos, de acuerdo con la Directiva 98/2008 de residuos.

En el caso de España, el potencial de biomasa disponible, bajo hipótesis conservadoras, se sitúa en torno a 88 millones de toneladas de biomasa primaria en verde, incluyendo restos de masas forestales existentes, restos agrícolas, masas existentes sin explotar y cultivos energéticos a implantar (PER 2011-2020). A este potencial se suman más de 12 millones de toneladas de biomasa secundaria seca obtenida de residuos de industrias agroforestales. En el año 2010 el conjunto de biomasa, biogás y residuos representaba el 4,2% del consumo final bruto de energía,

con un 1,4 % en el caso de biomasa sólida, estando previsto en el PER para el año 2020 un 2,2% del consumo final bruto de energía para la biomasa sólida. En la actualidad la mayor parte de los 3.655 ktep de consumo térmico final de biomasa en España proviene del sector forestal, utilizándose en sector doméstico y en industrias forestales para consumo térmico o cogeneración. Existe una potencia instalada de 533 MW abastecida con residuos de industrias agroforestales y restos de cultivos agrícolas principalmente. Para alcanzar los objetivos fijados en el área de biomasa el PER 2011-2020 ha definido una serie de propuestas dirigidas a cada fase del aprovechamiento de la misma, tanto en el desarrollo de aplicaciones térmicas, especialmente en edificios, como en el crecimiento de la producción eléctrica con biomasa mediante la generación distribuida a través de pequeñas cogeneraciones y centrales eléctricas en el entorno de los 15 MW.

En el caso de Andalucía, la elevada disponibilidad de biomasa, la gran capacidad para la producción de cultivos energéticos y, la abundancia de recurso solar, hacen que esta comunidad destaque respecto del resto de las Comunidades Autónomas y parte de las regiones de la Unión Europea. El potencial de biomasa disponible en Andalucía se puede cifrar en 3.327 ktep/año (PASENER 2007-2013), distribuidos entre los residuos agrícolas (43%), cultivos energéticos (17%), residuos forestales (4%) y los residuos industriales (18%), de los que se aprovechan aproximadamente el 30%. De entre las diferentes fuentes de energía renovable, la biomasa proporciona el 6,3% del consumo total de energía primaria en la región y supone el 78,7% del consumo de energía procedente de fuentes renovables (García-Maraver et al., 2012). Este potencial permite que la región pueda optar a un liderazgo científico e industrial y convertirse así, en un referente en el desarrollo de tecnologías relacionadas con la generación y el aprovechamiento de las energías renovables.

La subida de precios de los combustibles convencionales, las necesidades de autosuficiencia energética y los objetivos medioambientales de nuestra sociedad han impulsado el desarrollo de nuevos proyectos tanto de generación eléctrica como de producción de energía térmica. Este hecho, unido al desarrollo tecnológico de la biomasa, presenta unas expectativas de crecimiento importantes para el sector de la biomasa en Andalucía. Debido a que la reorientación estructural del sistema energético requiere de una reposición en materia de mejora del uso de las fuentes de energía


locales, se hace necesario el análisis de las materias primas propias de cada región, ya que pueden convertirse en la fuente de energía de la zona donde esta biomasa residual se genera. En las zonas mediterráneas del suroeste europeo, las actividades agrícolas producen grandes cantidades de biomasa residual. Este es el caso de los residuos de la poda del olivar, que han sido tradicionalmente usados para calefacción doméstica en las zonas rurales. Una hectárea de olivar genera aproximadamente tres toneladas de diferentes tipos de residuos de poda cada año (AAE, 2008), la mayoría de las cuales son actualmente quemadas de forma ilegal o dejadas sobre el suelo. Es por tanto evidente que los residuos procedentes de industrias olivareras y del mantenimiento de los cultivos del olivar, pueden convertirse en una importante fuente de energía en estas zonas donde el aprovechamiento de esta biomasa incrementaría la autonomía, la diversificación energética y el desarrollo.

En este marco, el sector del olivar representa una importancia clara para Andalucía, en los ámbitos económico, social, medio-ambiental y cultural. El cultivo del olivar representa en Andalucía aproximadamente el 26% de la producción agraria (Junta de Andalucía, 2009a). Además, la industria asociada al olivar vertebra y cohesiona el medio rural donde se asienta, apoyándose en un fuerte movimiento asociativo de base. Este cultivo posee además un alto valor ambiental, aparte de las meramente comerciales que puede realizar, por ejemplo la provisión de bienes públicos, el mantenimiento de la población rural, la vigilancia del territorio, y puede llegar a ser un referente dentro del sector agrario, en la lucha contra el cambio climático. Junto a su contribución por el desarrollo de sistemas sostenibles cabe citar también su papel como sumidero de gases de efecto invernadero y su potencialidad para la generación de energías renovables. Así gran parte de la biomasa andaluza se produce en forma de residuos en este sector, tanto en el sector agrícola como a nivel industrial, por ejemplo el orujillo y los huesos de aceituna (García-Maraver, et al., 2012). Esta biomasa residual procedente del olivar puede ser utilizada para generar electricidad, energía térmica y biocombustibles, dependiendo su destino final de las características de la materia prima. El daño al medio ambiente que estos residuos producen puede evitarse mediante la implantación de técnicas medioambientalmente sensibles y gracias a una gestión integrada que abarque programas de minimización, compostaje, reciclado, reutilización y valorización energética (Read et al., 1997; Wilson et al., 2006); esta gestión permite un tratamiento medioambiental y económicamente sostenible dentro de la Europa del Reciclado, en el marco de la Directiva 2008/98/CE de residuos, que avanza en los principios de la recuperación y máximo aprovechamiento de los materiales y recursos contenidos en los residuos. La legislación española, representada en España por la Ley 22/2011 de residuos y suelos contaminados, y por el Plan Director Territorial de Gestión de Residuos no Peligrosos de Andalucía (2010-2019), avanza en la misma dirección, favoreciendo la valorización y la utilización de materiales valorizados a fin de preservar los recursos naturales. En este sentido, y bajo el marco de la gestión integrada de residuos, la valorización energética de los residuos está adquiriendo una creciente atención (Sabbas et al., 2003; Ghani et al., 2009).

Las aplicaciones térmicas para producción de calefacción y agua caliente sanitaria son las más comunes dentro del sector de la biomasa. Estos usos térmicos pueden ser: (i) a pequeña escala, en calderas o estufas individuales utilizadas tradicionalmente en los hogares; (ii) a mediana escala en calderas para edificios públicos, bloques de viviendas u otros usos; o (iii) a gran escala en redes de climatización. En este tipo de equipos es necesario llevar a cabo una densificación previa de masa y volumen que permita el uso de una biomasa originalmente heterogénea y de baja densidad aparente en pequeños sistemas de combustión. Esta densificación se lleva a cabo normalmente mediante un proceso de pelletización. Para llevar a cabo un correcto proceso de pelletización, la calidad de los productos densificados debe alcanzar los requisitos del consumidor, así como los estándares del mercado. La calidad final del pellet es función de las propiedades de la materia prima y del proceso de fabricación (Carone et al., 2011).

Mientras los pellets de madera procedentes de residuos forestales han establecido ya tecnologías y mercados con éxito en algunos países de Europa, en otros países como España, esta técnica no está aún muy extendida por no ser los residuos forestales la principal fuente de biomasa, sino la biomasa residual agrícola. En el caso de la biomasa residual procedente del olivar, se han observado características propias de la materia prima tales como su elevado contenido en cenizas (Ollero et al., 2003) si se compara con otras biomasas europeas (Vassilev et al., 2010), aunque se ha observado un vacío bibliográfico importante en esta materia. Por ello es necesario ampliar el conocimiento que permita optimizar la eficiencia de la producción y mejorar la calidad



del producto final, ya que tan sólo el uso de pellets estandarizados procedentes del olivar permitiría el uso de estos residuos en procesos de combustión eficientes.

Dada la falta de investigación en residuos procedentes del olivar como fuente de energía térmica, el estudio descrito en este documento pretende alcanzar un conocimiento en profundidad de estos residuos que ayude en el aprovechamiento de los mismos como combustible renovable. Por tanto, el objetivo principal de este trabajo es la optimización del proceso de pelletización de residuos agrícolas procedentes del olivar para su aplicación en calderas domésticas. Para alcanzarlo, se han definido los siguientes objetivos secundarios:

• Evaluar el potencial de la biomasa residual del olivar como fuente de energía renovable en zonas mediterráneas, más concretamente en el caso de Andalucía.

• Analizar el marco regulador que establece los parámetros utilizados para la determinación de la calidad y el ámbito de aplicación de los pellets a nivel europeo.

• Caracterizar física y químicamente la biomasa residual del olivar, de acuerdo a los requisitos establecidos para el uso de biocombustibles sólidos a escala no industrial.

• Determinar las variables óptimas para la fabricación de pellets a partir de biomasa residual del olivar.

• Determinar el comportamiento en el proceso de combustión de los pellets fabricados a partir de biomasa residual del olivar, bajo condiciones reales en caldera doméstica.

INTRODUCTION, MOTIVATION AND OBJECTIVES

Society development has been accompanied by a large energy consumption (IDAE, 2005). To satisfy the increasing energy demand, the human being has used fossil energy resources, such as coal, oil, or natural gas. This fact has made that countries that do not have this type of fuels, which is the case of Spain, depend more than 75 % on an external energy supply (Dinica, 2009).

This situation threatens social comfort, economical development and business competitiveness of the regions due to the vulnerability of the current energy model to the future energy supply. Moreover, a new concept that leads to a change in the worldwide perception of energy has been introduced during the last years: climate change caused by emissions of greenhouse gasses (Omer, 2008). Due to the importance of these problems, the development of energetic but environmentally friendly measures in countries all over the world has become a necessity in order to reduce the negative effects derived from the use of fossil fuels.

Among these measures it is noteworthy the promotion of the use of renewable energies, as the axis of a new energy model that reacts to the needs of an energy supply without generating environmental, economic or social disequilibrium in the context of a sustainable development. Renewable energy sources are produced in a continuous basis, being inexhaustible on a human scale. Currently, these sources of energy are some of the most efficient and effective solutions to the achievement of a sustainable energetic development and the prevention of environmental pollution.

In this sense, energy and climate policies in Europe have been expressed in three strategic objectives: safe supply, efficiency and environmental compatibility. To achieve these objectives it is necessary to act in the generation and consumption areas, as well as in the emissions, assuming the agreements in Directive 2009/28/CE on the promotion of renewable energies, which establishes the minimum objectives to be achived by the European countries and the European Union in general. This Directive



establishes that a minimum of 20% of the gross energy consumed in Europe has to be produced by renewable sources in 2020, as well as 10% of the energy consumed in the transport sector. Moreover, the Directive demands that National Action Plans for Renewable Energy for the period 2011-2020 have to be implemented in every member state.

Spain, in its way towards a new energy model, has developed some legal, normative and planning instruments that allow the transformation process. For that purpose, once the valid period of the Renewable Energy Plan (2005-2010) finished, and according to the current legislation³, a new Renewable Energy Plan 2011-2020 was approved. This Plan includes the design of new energetic scenaries as well as objectives according to the Directive 2009/28/CE, the Royal Decree 661/2007 that regulates the production of electricity in the special regime, and the Royal Decree Law 2/2011, dated 4 March, on Sustainable Economy⁴. After the previous Renewable Energy Plan (2000-2010), which established 12% contribution from renewable energies in the global energy consumption in Spain, the approval of the Renewable Energy Plan (2011-2020) aims to meet the agreements established in Directive 2009/28/CE, also presented in the Law 2/2011. The approval of this Renewable Energy Plan envisages a new energy model that consolidates an energy culture that considers energy as a scarce and valuable resource. With this structural reorientation of the Spanish energy system it is expected to go towards a repositioning based on the best exploitation of the energy resources of the country and on the global definition of an energy model fully adapted to climatic, cultural and economic conditions in the different regions.

Spain, despite not having fossil energy resources, is rich in renewable resources which explitation will give autonomous and safe energy to the country, minimizing the future impact of the imbalances from the international energy market. In the new energy model, renewable energies are destined to play an important role because of their

³ Royal Decree 661/2007, regulating the production of electricity in the special regime, and Law 2/2011 of March 4th on Sustainable Economy.

⁴ Energy policies will be orientated to guarantee the energy supply, the economic efficiency and the environmental sustainability. Energy consumption, generation and distribution models have to be compatible to the regulatory framework and the communitarian objectives, as well as with the international efforts to reduce the climate change.

sustainability and environmentally friendly character. Renewable energies considered by the current planning instruments are: solid and liquid biofuels, biogas, wave energy, wind energy, geothermal energy and another energies from the ambient, hydropower, energy from waste (municipal, industrial, sludge) and solar energy (photovoltaic, thermal and thermoelectric).

Due to the promotion of renewable energies by the Renewable Energy Plan (2005-2010), their increase has been notable, and have covered, in terms of primary energy consumption, from 6.3 % in 2004 to 11.3% in 2010, percentage corresponding to 13.2% in terms of gross final consumption according to the Directive 2009/28/CE.

At a regional level, as in the case of Andalusia, new energy models are being implemented as well. The Reform of the Statute of Autonomy established the responsibilities of the Autonomous Community and explicitly recognizes shared responsibilities in energy production, distribution and transport, as well as in renewable energy promotion and management, and energy efficiency. For that purpose, once the validity period of the Andalusian Energy Plan (2003-2006) finished, the new Andalusian Sustainable Energy Plan 2007–2013 established a new energy model for the region based on its climatic, cultural and economical conditions. On the other hand, the approval of the Royal Decrees 314/2006 and 661/2007, the Law 2/2007 and the consolidation of the Andalusian Energy Agency as an instrument to optimize the energy supply of Andalusia, define a robust framework to base the regional energy policy on.

In the European, Spanish and Andalusian renewable energies promotion plans, the importance of residual biomass from agricultural activities is widely recognized, because: (i) it is an abundant energy source which use entails significant socioeconomic and environmental benefits from waste generation; (ii) it is a cheap biomass source, since it allows the energy valorization of organic residues; (iii) its use improves waste management and mitigates the environmental impact derived from waste disposal, according to the Directive 98/2008 on waste.



In the case of Spain, the available biomass potential, under conservative hypothesis is 88 million tonnes of green primary biomass, including forestry and agricultural residues and energy crops to be implemented (PER 2011-2020). More than 12 million tonnes of dry seconday biomass from agroforestry industries can be added to this biomass potential. In 2010, biomass consumption represented 4.2% of final gross energy consumption, being 1.4% the contribution of solid biomass. It is expected a 2.2% in 2020 (PER 2011-2020). Currently, the major part of the 3,655 ktoe of final thermal energy consumption from biomass in Spain comes from the forestry sector and is used in the residential sector and in forestry industries. There is an installed power of 533 MW mainly supplied by agroforestry industries and agricultural residues. To achieve the goals for biomass established in the Renewable Energy Plan 2011-2020 a series of proposals for thermal and electric installations have been defined.

In Andalusia, the large biomass availability and energy crop capacity, as well as the abundant solar catchment, make this region to be in a privileged position with respect to the rest of the Autonomous Communities and other European regions. Available biomass potential in Andalusia can be valued in 3,327 ktoe/year (PASENER 2007-2013), distributed among agricultural residues (43%), energy crops (17%), forestry residues (4%) and industrial residues (18%), but only 30% of them are exploited. Of the various renewable energies, biomass comes to 6.3% of the total primary energy consumption and 78.7% of the renewable energy consumption (García-Maraver et al., 2012). This potential allows the region to opt for a scientific and industrial leadership and become a model for the development of technologies for renewable energies.

The rise in the price of conventional fuels, the necessity of energy selfsufficiency and the environmental objectives of the society have motivated the development of new projects on electricity and thermal energy generation. This fact, toguether with the technological development of biomass, mean important growth espectations for the biomass sector in Andalusia.

Given the fact that the structural reorientation of the energy system requires a repositioning in terms of a better exploitation of the local energy source, it is necessary

to conduct an analysis of the raw materials typical of each region, since they can become the energy source of the areas where they are generated.

In the Mediterranean areas of southwest Europe, agricultural activities are very important, but they produce large quantities of residue. This is the case of olive tree pruning residues, which have traditionally been used for domestic heating in rural areas and which are an important source of residual biomass. One hectare of olive trees generates approximately three tons of different types of pruning per year (AAE, 2008), most of which are now illegally burnt or left on the ground. The energy exploitation of this biomass would allow the sustainable replacement of fossil fuels. Furthermore, it would increase self-sufficiency and energy diversification, besides contributing to the development of rural areas.

Olive farms represent 26% of agricultural production in Andalusia (Junta de Andalucía, 2009a). Moreover, the olive sector has an important environmental added value because it maintains the rural development of the areas where this grove exists and can be regarded as a sink of greenhouse gasses and as a renewable energy source. 25% of residual biomass in Andalusia is produced by the olive sector (García-Maraver, et al., 2012) and its final use of biomass depends mainly on the characteristics of the raw material, but it can be used for electricity or thermal energy generation and biofuels for transportation.

The environmental damage that these residues currently produce, can be avoided through environmentally sensitive techniques and an integrated waste management that includes minimization, composting, recycling, reusing and energetic valorization programs (Read et al., 1997; Wilson et al., 2006). This management allows a sustainable and economic treatment within the *Recycling Europe* and the Directive 2008/98/CE on waste. Spanish legislation, represented by the Law 22/2011 and the Director Plan on Non Hazardous Residues Management in Andalusia (2010-2019) move forward in the same direction, favoring the valorization and utilization of valorized materials to preserve the natural resources. In this sense, and within the framework of integrated waste management, the energetic valorization of residues is gaining increasing attention (Sabbas et al., 2003; Ghani et al., 2009).



Thermal applications for domestic heating and sanitary hot water are the most common in the biomass sector. These thermal uses can be: (i) at small scale, in individual boilers or stoves; (ii) at medium scale, for building boilers; or (iii) at large scale, in heating and cooling distribution systems. In this type of applications it is necessary a previous mass and volume densification that allows the originally heterogeneous and low bulk density biomass to be used in combustion systems. This densification is usually done through a pelletization process. For success of the densification process, the quality of the densified products must meet the consumer requirements and market standards. The final quality of pellets is a function of the properties of the raw material and of the manufacturing process (Carone et al., 2011).

Whereas wood pellets from forestry residues already has successfully established technologies and markets for production and consumption in some European countries, in countries such as Spain this technology is not widely used since the main source of residual biomass is agricultural biomass. In the case of residual biomass from olive trees, certain characteristics, such as the higher ash content (Ollero et al., 2003) in comparison with other European pellets (Vassilev et al., 2010), have raised doubts concerning their use in thermal applications, especially in domestic heating systems. Nevertheless, a lack of references on this matter has been observed. Due to this fact, it is necessary to increase the knowledge that leads to the optimization of the production efficiency and improve the quality of the final product, since only the use of standardized pellets would allow their use in efficient combustion processes. Given the lack of research on olive tree residues as a source of thermal energy, the study described in this document aims to achieve an in-depth knowledge of these residues to help in the energetic exploitation of them to be used as a renewable fuel.

The main objective of this work is therefore to optimize the pelletization process of agricultural wastes originating from olive farms for their application in domestic boilers. In order to achieve this objective, the following secondary objectives were defined:

• Evaluate the availability of residual biomass from olive trees as a renewable energy source in the Mediterranean regions, more concretely in the case of Andalusia

• Analyze the framework that establishes the requirements that solid biofuels have to achieve to be properly used at non-industrial scale, as well as its application at a European level

• Characterize the residual biomass from olive trees from a physical and chemical point of view according to the requirements for non-industrial pellets

• Analyze the optimum variables for the residual biomass from olive tree pellet production

• Analyze the combustion behavior of pellets from olive residual biomass under real conditions in a domestic boiler.



PART 1

BACKGROUND



Optimization of the pelletization process of agricultural wastes originating from olive farms for their application in domestic boilers

Ángela García Maraver

CHAPTER 1

STATE OF THE ENERGY SUPPLY WORLDWIDE

1. Past, present and future of energy supply in the world

The importance of energy in the generation of wealth and in economic development has been universally recognized. Historic data demonstrate a strong relationship between energy availability and economic activity (Spalding-Fecher et al., 2005). However, economic development, industrialization and the implantation of economic models that base growth on the sustained increase of consumption have resulted in a society that is highly dependent on energy. Due to this fact, the need for this resource is increasing (Marcos, 2001), as a direct result of the increase in the world's population and energy consumption that accompany development and industrialization.

The data in Figure 1 demonstrate the evolution of fuel consumption all over the world between 1971 and 2010. As the data in the figure show, the use of electricity, gas and renewable fuels has increased with time, while the increase of the use of coal has maintained nearly constant. This growth tendency will continue, since current



predictions estimate that the energy demand for 2030 will be 50 to 80% higher in comparison with that required in 1990 (IEA, 2012).



Figure 1. World total final energy consumption from 1971 to 2010 by fuel (Mtoe) (IEA, 2012)

Energy is essential for economic and social development, as well as for improving the quality of life, but depending upon the way it is produced, transported and used, it can contribute or not to environmental degradation (Farinelli, 1999; Davidson and Sokona, 2002; Johansson and Goldemberg, 2002). As a consequence of energy consumption, millions metric tons of greenhouse gases, such as carbon dioxide, methane or nitrous oxide among others, are released to the atmosphere, 80% of which come from the combustion of fossil fuels (IPCC, 2001). These greenhouse gases are precursors to global warming, air pollution, acid rain, ozone layer decrease, destruction of forests and radioactive emissions (Omer, 2008) that negatively affect the meteorology and cultivation and increase disease proliferation and sea level (Spalding-Fecher et al., 2005).

The combustion of fossil fuels such as petroleum or coal during the last 100 years has produced an increase of CO_2 levels in the atmosphere (Dobelmann et al., 2004), although it has been since the 80s when the risk and reality of this environmental

degradation have been more evident due to the environmental impact of human activities, that have grown with world population and industrial activity.

This tendency can be observed in Figure 2, which shows the progressive increase of CO_2 emitted from the combustion of fossil fuels. It is predicted that in 2015 carbon emissions will increase by 54% with respect to those in 1990, prompting global warming (Omer, 2008). In fact, the greenhouse effect has already increased the average temperature in the Earth by 0.7°C, and it is estimated that this temperature will increase by 4 to 6°C in 2050 (IEA, 2012). The main responsible for CO_2 emissions is industry, but 40% of global energy consumption is due to the energy consumption in heating systems and lighting (Omer, 2008).



Figure 2. World CO₂ emissions from 1971 to 2010by fuel (Mt of CO₂) (IEA, 2012).

Current methods to produce and consume energy have impacts on the environment and health; moreover, there are problems regarding the supply of fossil fuels, since they are connected to economic and politic instabilities (Romm and Lovins, 1993; Goldemberg, 1996; Holdren and Smith, 2000). The data in Figure 3 show the world's petroleum reserves in 2008, making evident that the use of this fossil fuel would not be able to cover the increasing energy demand. The exhaustion of fossil fuels reserves has been estimated to be between 40 (BP, 2009) and 60 (Santamarta, 2004)



years; in consequence it will result in a continuing increase in the cost of them, affecting global economic development.



Figure 3. Demonstrated petroleum reserves in 2008 (thousand million barrels) (BP, 2009)

Nowadays, the majority of the energy is produced and consumed in a way that could not be maintained if the technology remains constant and the energy consumption keeps rising (Kaygusuz and Sari, 2003). Consequently, the new energy models have to be renewable and sustainable, efficient and economically effective, practical and save (Chum and Overend, 2001; Kaygusuz and Sari, 2003). Therefore, energy planning faces up to a historical change marked by the necessity to guarantee a qualified, safe and sufficient energy supply with no environmental imbalances. Until now, energy planning studies had focused on forecasting energy demand in order to meet this demand under the approach that considers energy as a finite resource. Nevertheless, the demonstrated impact that this approach and the current energy planning have on the environment and its important contribution on global warming, make necessary a new energy model.

2. Energy framework

Climate change is the environmental impact that has received the most attention since the United Nations Conference on Environment and Development took place in 1992 in Rio de Janeiro; this problem cannot be dealt without important changes in the energy sector (IPCC, 2001). In this regard, the challenge for the global energy sector is

double: (i) increase dramatically the access to affordable and modern energetic services in countries that lack them and (ii) find the combination of energy sources, technologies, policies and behavioral changes that will reduce adverse environmental impacts (Spalding-Fecher et al., 2005). A considerably large number of measurements have tried to be implemented as a response to the necessary fight against climate change; some of them are analyzed in the section below.

2.1. Kyoto Protocol

The Kyoto Protocol is an international agreement linked to the United Nations Framework Convention on Climate Change. It was adopted in Kyoto, Japan, on 11 December 1997 and entered into force on 16 February 2005.

The major distinction between the Protocol and the Convention is that while the Convention encouraged industrialized countries to stabilize greenhouse gas emissions (GHG), the Protocol commits them to do so. The detailed rules for the implementation of the Protocol were adopted at COP 7 in Marrakesh in 2001, and are called the "Marrakesh Accords".

The Kyoto Protocol sets binding targets for 37 industrialized countries and the European community for reducing GHG emissions to an average of 5% against 1990 levels over the five-year period 2008-2012, varying among the different developed countries. Moreover, this protocol identifies for first time the promotion of renewable energies as a key strategy to reduce greenhouse gasses emissions (Flavin and Dunn, 1998).

The targets cover emissions of the following six main greenhouse gases: carbon dioxide (CO₂); methane (CH₄); Nitrous oxide (N₂O); hydrofluorocarbons (HFCs); perfluorocarbons (PFCs); and sulphur hexafluoride (SF₆). Under the Protocol, countries' actual emissions have to be monitored and precise records have to be kept of the trades carried out.

The 15 States which were EU members when the Kyoto Protocol was adopted, took on that 8% target that will be redistributed among themselves, but which combined make an overall target for that group of countries.

The Protocol places a heavier burden on developed nations under the principle of "common but differentiated responsibilities" because it recognizes that developed countries are principally responsible for the current high levels of GHG emissions in the atmosphere as a result of more than 150 years of industrial activity.

By the end of the first commitment period of the Kyoto Protocol in 2012, a new extension for the period 2013-2020 was negotiated and ratified in order to deliver the stringent emission reductions the Intergovernmental Panel on Climate Change (IPCC) had clearly indicated were needed.

2.2. European Union climate and energy package (2008-2020)

After the Kyoto Protocol, a new international agreement to reduce emissions of greenhouse gases was to be negotiated at Poznan (Poland) and in Copenhagen in 2009. To play a leading role in these negotiations, the European Union wanted to develop as quickly as possible a common position in the fight against climate change, and thus implemented its own measures to deal with climate change.

The European plan on climate change consists of a range of measures adopted by the members of the European Union to fight against climate change. The plan was launched in March 2007, and it was adopted by the European Parliament on December 2008. The package focuses on emissions cuts, the use of renewable sources of energy and energy efficiency.

The plan included the so-called "20-20-20 targets", but in reality it consisted in four proposals. These aims were:

- To reduce emissions of greenhouse gases by 20% by 2020.
- To increase energy efficiency to save 20% of EU energy consumption by 2020.
- To reach 20% of renewable energy in the total energy consumption in the EU by 2020.
- To reach 10% of biofuels in the total consumption of vehicles by 2020.

The 20-20-20 targets represent an integrated approach to climate and energy policy that aims to combat climate change, increase the EU's energy security and strengthen its competitiveness. They are also headline targets of the Europe 2020 strategy for smart, sustainable and inclusive growth. This reflects the recognition that tackling the climate and energy challenge contributes to the creation of jobs, the generation of "green" growth and a strengthening of Europe's competitiveness.

The climate and energy package comprises four pieces of complementary legislation which are intended to deliver on the 20-20-20 targets:

• Reform of the EU Emissions Trading System (EU ETS)

The EU ETS is the key tool for reducing industrial greenhouse gas emissions most cost-effectively. The climate and energy package includes a comprehensive revision and strengthening of the legislation which underpins the EU ETS, the Emissions Trading Directive.

• National targets for non-EU ETS emissions

Under the so-called Effort Sharing Decision, Member States have taken on binding annual targets for reducing their greenhouse gas emissions from the sectors not covered by the EU ETS, such as housing, agriculture, waste and transport (excluding aviation). Around 60% of the EU's total emissions come from sectors outside the EU ETS.

The national targets, covering the period 2013-2020, are differentiated according to Member States' relative wealth. They range from a 20% emissions reduction (compared to 2005) by the richest Member States to a 20% increase by the least wealthy (though this will still require a limitation effort by all countries). Member States must report on their emissions annually under the EU monitoring mechanism.

• National renewable energy targets

Under the Renewable Energy Directive, Member States have taken on binding national targets for raising the share of renewable energy in their energy



consumption by 2020. These targets, which reflect Member States' different starting points and potential for increasing renewables production, range from 10% in Malta to 49% in Sweden.

The national targets will enable the EU as a whole to reach its 20% renewable energy target for 2020 - more than double the 2010 level of 9.8% - as well as a 10% share of renewable energy in the transport sector. The targets will also help to cut greenhouse gas emissions and reduce the EU's dependence on imported energy.

• Carbon capture and storage

The fourth element of the climate and energy package is a directive creating a legal framework for the environmentally safe use of carbon capture and storage technologies. Carbon capture and storage involves capturing the carbon dioxide emitted by industrial processes and storing it in underground geological formations where it does not contribute to global warming.

The directive covers all CO_2 storage in geological formations in the EU and lays down requirements which apply to the entire lifetime of storage sites.

Regarding energy efficiency, the climate and energy package does not address the energy efficiency target directly. This is being done through the 2011 Energy Efficiency Plan and the Energy Efficiency Directive.

2.3. Spanish National Renewable Energy Plan (2011-2020)

On 11th November the new Renewable Energy Plan (REP 2011-2020) was approved by the Spanish Government for the years 2011 to 2020, establishing the development framework for the renewable energy sector during the next 10 years.

This plan aims to be the main tool for fulfilling and going beyond the objectives set up by the European Union of reaching 20% of the total energy consumption covered by renewable sources by 2020. The REP 2011-2020 establishes the Spanish objectives and suggests the measures to be implemented in order to reach the 20% goal in 2020 and includes the Spanish vision on the evolution of the renewable energy sector regarding each of the types of renewable energy available in the coming years. The public entity in charge of the implementation of the Renewable Energy Plan 2011-2020 is the Institute for Energy Diversification and Saving (IDAE).

The REP 2011-2020 specifically takes into account the different renewable sources such as wind power, biofuels and bioliquids, hydro energy, biogas, biomass, wave and tidal energy, geothermal energy, waste energy and solar technologies, including photovoltaic, thermal and concentrated solar power.

According to the REP 2011-2020, the increase in the use of renewable energy in the Spanish energy mix will contribute to improve supply guarantee, reduce energy dependency, improve trade balance, reduce greenhouse gas emissions and, in general, will have a positive effect on the creation of qualified jobs, stimulation of the economy and reduction of environmental impacts caused by the energy system. All of these factors explain the fact that renewable energy has become a strategic sector in Spain, which is a key issue in the Spanish economic, political and social development strategy.

Final energy consumption will hardly decrease in the present decade in Spain, going from 96.4 Mtoe in 2010 to 89.7 Mtoe in 2020. By that time, renewable energy will cover 20.8% of the energy consumption, with a contribution of the renewable energy sources of 11.3% to the energy consumption in the transport sector.

Nevertheless, 61% of the objectives set up by the REP for 2020 will come from the generation of electricity from renewable energy sources. This way, the importance of renewable sources for electric uses in the energy mix will increase by 30%.

The main objective of the plan is to reach at least 20% of final energy consumption by 2020, with a contribution of renewable sources to the transport sector of at least 10% by the same year.

By the end of the Renewable Energy Plan 2011-2020, it is estimated that wind energy will continue to be the renewable energy with the highest contribution, followed by solar energy. Meanwhile, biomass, biogas and waste will increase their participation in the electricity supply. Geothermal energy and wave energy will gain importance and will be mature in the next period 2020-2030. The importance of thermal biomass will equally be divided between the domestic and industrial sector, while solar thermal energy will increase in terms of installed surface. Geothermal energy will continue with its development and biodiesel will see an increase in its consumption moderately, while bioethanol consumption will be doubled until 2020.

2.4. Andalusian Sustainable Energy Plan (2007-2013)

In the Spanish Autonomous Community of Andalusia, energy policy has been developed according to European and Spanish policies and betting on the diversification of renewable energy sources with low carbon emissions, the decentralization of the production and the increase in energy efficiency.

The structural reorientation of the Spanish energy system evidently requires a repositioning in regards to the best possible use of the country's energy resources and the overall definition of a model for energy production and use. Such a model should be fully adapted to the climatic, cultural, and economic conditions of the different regions, where energy policies are generally implemented through regional development plans, such as the Andalusian Energy Plan 2003–2006 (PLEAN) in Andalusia, which was followed by the Andalusian Sustainable Energy Plan (PASENER) 2007–2013.

In this regard, the PASENER is a new energy plan based on the commitments in the Kyoto Protocol. It incorporates changes in energy policies and envisages a new energy model that consolidates an energy culture that considers energy as a scarce and valuable resource. It introduces a new energy culture into society, so that a collective conscience can emerge that evaluates the capacity to access different energy sources with high levels of security and quality, and assesses the effects that this has on the environment, taking decisions consistent with the situation.

Its strategic objectives are the following:

- To prioritize the use of renewable sources to increase Andalusia's selfsufficiency in terms of energy, to protect the environment, and to establish a distributed energy system.
- To involve the community in the rudiments of an energy culture in order to raise awareness that energy is a basic and scarce resource and to promote energy saving and efficiency in all sectors.

- To contribute to balanced town planning and to economic growth through an energy infrastructures system that guarantees a secure, efficient and highquality supply.
- To promote a competitive entrepreneurial network through innovation in the field of energy technologies.

There are 11 objectives to fulfill in 2013:

- To ensure that 18.3% of the primary energy structure comes from renewable energy sources, purely for energy-providing purposes.
- To provide 39.1% of the total electricity supply from electricity produced using renewable technologies.
- To set the gross electricity production produced from renewable sources at 32.2% of the net target consumption of electricity in Andalusia.
- To reach a saving equivalent to 8% of primary energy consumed purely for energy-providing purposes in 2006.
- To reduce primary energy intensity by 1% compared with 2006.
- To set biofuels consumption at 8.5% of the total gasoline and gasoil fuel consumption in the transport sector.
- To set a quality-of-service index (TIEPI) at 0.86 hours in urban areas, 1.37 hours in semi-urban areas, 2.89 hours in concentrated rural areas, and 3.81 hours in dispersed rural areas, obtaining a TIEPI of 1.56 hours for Andalusia.
- To provide access to a natural gas supply for 80% of residents in towns of between 10,000 and 20,000 inhabitants.
- To reduce by 20% CO₂ emissions per unit of electricity generated.
- To prevent the emission of 11 million tons of CO₂.



• To provide a total of 27.7% of the final energy consumed in Andalusia from renewable energy sources.

With this structural reorientation of the Andalusian energy system it is expected to go towards a new energy model that adapts economic growth to social cohesion throughout Andalusia and that protects Andalusia's natural and cultural heritage, without generating imbalance in the global ecosystem, especially that associated with the great challenge posed by climate change. An energy model fully adapted to climatic, cultural and economic conditions in the region.

3. Renewable energies in the new energy framework

Renewable energies are produced in a continuous basis, being inexhaustible on human scale. Renewable sources of energy include wind, solar, hydro-electric and tidal power as well as geothermal energy and biomass. Excepting geothermal energy, the rest of the renewables derive direct or indirectly from the solar energy. Directly in the case of the light and heat produced by solar radiation and indirectly in the case of wind, hydro-electric, tidal and biomass energy.

Renewable energy sources have been important for human beings since the beginning of civilization (Sattar, 1996). Along the history, and until 19th century, they covered society's energetic needs. However, because of the massive use of coal and petroleum and the later use of natural gas in the last years, renewables were replaced by mostly fossil fuels (Santamarta, 2004).

The growing consumption of fossil fuels together with the environmental degradation have resulted in a generalized interest in renewable energy as a response to the concern for the climatic change (Akella et al., 2007; Cowie and Gardner, 2007) and the energy efficiency, its transmission, distribution and consumption (Kaygusuz and Sari, 2003). Nowadays, renewable energy resources are one of the most efficient and effective solutions to sustainable energy development and the prevention of environmental pollution.

Consequently, within the new energy framework, renewable energies are intended to cover part of the future fossil fuels demand (Marcos, 2001) and therefore enable the world to reduce greenhouse emissions and make it less dependent on imported energy. Moreover, boosting the renewables industry will encourage technological innovation and employment.

Among renewable resources, biomass can play an important role since it is regarded as a renewable and environmentally safe way of providing energy for electricity, heating, and cooling (Fernandez-Llorente and Carrasco-Garcia, 2005). Biomass is an abundant renewable energetic resource that entails significant socioeconomic and environmental benefits from the generation of residues. Consequently, converting this residue into energy can revalorize waste materials, mitigate the environmental impacts of waste disposal, and reduce the mass and volume of the waste.



CHAPTER 2

ENERGY FROM BIOMASS

Biomass is the progenitor of fossil fuels in that natural gas, oil and coal are biomass that has been converted into concentrated energy forms by geological processes akin to pyrolysis (heating in the absence of oxygen) under pressure. This means that with appropriate industrial processing, newly harvested biomass can be converted into homologs of current gas, liquid and solid fossil fuel resources.

Though until the mid-19th century biomass dominated global energy consumption, the steep increase in fossil-fuel usage has resulted in a decline of biomass consumption for energy purposes over the last 50 years. Nevertheless, biomass still provides about 14% of the world's annual energy consumption (Werther et al., 2000; Purohit et al., 2006; Zeng et al., 2007), standing as the fourth-largest energy resource in the world behind only coal, oil and natural gas (Ladanay and Vinterbäck, 2009).

Moreover, the use of biomass as a traditional energy source for economically developing countries can play a pivotal role in helping the industrialized world to reduce the environmental impact of burning fossil fuels to produce energy (McKendry, 2002).

Wood fuels, agricultural wastes, and grasses are the most prominent biomass energy sources. Biomass, if properly managed, offers many advantages because of



being a renewable and sustainable energy feedstock. It can significantly reduce net carbon emissions when compared with fossil fuels, making it a clean development mechanism for reducing greenhouse gas emissions (Junfeng and Runqing, 2003). The cheapest biomass sources are the waste products from wood or agro-processing operations, but their supply is limited. To overcome this limitation, countries around the world are planting biomass crops for energy purposes.

Most developed and industrialized nations have begun developing technologies to use biomass more efficiently. In the United States and most of Europe, biomass has already penetrated the energy market. Therefore the potential of biomass for energy production is promising.

4. Biomass definition and classification

4.1. Biomass definition

The European Technical Specification EN 14588 (2010) defines biomass as "all biological materials excluding those that were included in geological formations suffering a mineralization process". This is the case of coal, petroleum and natural gas, which formation and composition millions of years ago is not comparable with the "carbon neutral balance" of biomass.

The biomass carbon cycle and carbon neutrality differentiate the carbon in biomass from the carbon in fossil fuels. Fossil fuels contain carbon that has been out of the atmosphere for millions of years. When fossil fuels are burned, they put carbon in the atmosphere that is in addition to what has been cycling between the atmosphere and the earth, causing the amounts of CO_2 in the atmosphere to increase and contributing to the greenhouse effect. Indeed, the primary source of increased CO_2 in the atmosphere since pre-industrial times is the combustion of fossil fuels (Denman, 2007).

In the context of biomass derived fuels, the term "carbon neutrality" is best understood as an attribute of biomass carbon that reflects the fact that this carbon was only recently removed from the atmosphere and is part of a natural cycle. Through photosynthesis, carbon in atmospheric CO_2 becomes carbon in plant tissue, also called biomass. When biomass is burned, decays or is otherwise oxidized, the chemical energy is released and the CO_2 is placed back into the atmosphere, completing a natural carbon cycle (Figure 4).



Figure 4. Biomass carbon neutrality (IDAE, 2007)

When this cycle is in balance, it has a net zero impact on atmospheric carbon, which is why biomass carbon is often called "carbon neutral balance" (Varela et al, 1999; McKendry, 2002; Cowie and Gardner, 2007; Miner, 2010). This is an important distinction between biomass carbon and the carbon in fossil fuels.

The cycle, however, is never in exact balance. If plants are removing carbon from the atmosphere faster than it is being returned to the atmosphere, the cycle is accomplishing net removal of carbon from the atmosphere, and stocks of stored carbon (primarily in forests) are increasing. On the other hand, if biomass carbon is being returned to the atmosphere faster than it is being removed by plants, the cycle is adding carbon to the atmosphere and stocks of stored carbon are decreasing (Miner, 2010). Therefore, only a sustainable management can supply renewable fuels indefinitely.

4.2. Biomass classification

Biomass resources include natural and derived materials mainly classified according to their origin (Vinterbäck, 2004; VIEWLS, 2005; AAE, 2008;



Rofiqul et al, 2008; Van Loo and Koppejan, 2008; Ladanai and Vinterbäck, 2009).

Biomass sources can be divided into 3 principal groups: (i) Natural biomass, directly available in natural ecosistems; (ii) Residual biomass, coming from the development of different activities; and (iii) Energy crops, with the only objective to produce biomass with energy purposes. Residual biomass and energy crops are those that are most used to produce energy. In Table 1, these groups are classified by different types of feedstock.

The product derived from a particular feedstock is dependent on which part of the plant and the conversion process used. The possible products that can be derived from biomass include solid biofuels, biodiesel, ethanol, butanol, methane, hydrocarbons, and natural oils, which can be further processed into any number of desirable fuels (Tumuluru et al., 2011).

Regarding solid biofuels, the European Committee for Standardization, CEN under committee TC335 has published 27 technical specifications (pre-standards) for solid biofuels during 2003 – 2006. Now these technical specifications have been upgraded to full European standards (EN).

The two most important technical specifications being developed deal with classification and specification (EN 14961-1, 2010) and quality assurance for solid biofuels (EN 15234-1, 2011). The classification of solid biofuels is based on their origin and source. The fuel production chain of fuels shall be unambiguously traceable back over the whole chain. The solid biofuels are divided to the following sub-categories for classification in EN 14961-1 (2010):

- Woody biomass: from trees, bushes and shrubs (Table 1 EN 14961-1).
- Herbaceous biomass: from plants that have a non-woody stem and which die back at the end of the growing season. It includes grains and their byproducts such as cereals (Table 2 EN 14961-1).
- Fruit biomass: from the parts of a plant which are from hold seeds. If appropriate, also the actual species (e.g. spruce, wheat) of biomass should be stated.

TYPE OF BIOMASS	SUPPLY SECTOR	TYPE OF FEEDSTOCK	EXAMPLES
RESIDUAL BIOMASS	Agricultural residues	Dry lignocellulosic agricultural residues	Straw (maize, cereal, rice) Sugar beet leaves Residue flows from bulb sector
		Livestock waste	Solid manure (chicken manure) Liquid manure (cattle, pigs, sheep manure)
	Forestry	Forestry by- products	Bark; Wood blocks; Wood chips from thinning; Logs from thinning
	Industry	Wood industry Residues	Industrial waste wood from sawmills and timber mills (bark, sawdust, wood chips, slabs, off-cuts)
			Fibrous vegetable waste from virgin pulp production of paper from pulp, including black liquor
		Food industry Residues	Wet cellulosic material (beet root tails); Fats (used cooking oils); Tallow, yellow grease; Proteins (slaughter house waste)
		Industrial products	Pellets and briquettes from sawdust and shavings; Bio-oil (pyrolisis oil); Ethanol; Biodiesel
	Parks and	Herbaceous	Grass
	gardens	Woody	Pruning
	Solid Waste	Contaminated waste	Demolition wood; Biodegradable; Municipal waste; Sewage sludge; Landfill gas; Sewage gas
	Others	Roadside hay	Grass/Hay
		Husks/Shells	Almond; Olive; Walnut; Palm pit; Cacao
ENERGY CROPS		Dry lignocellulosic woody energy crops	SRW-willow SRC-poplar Eucalyptus
		Dry lignocellulosic herbaceous energy crops	Miscanthus; Switch grass; common reed; Reed canary grass; Giant reed; Cyanara cardu; Indian shrub
		Oil energy crops	Sugar beet; Cane beet; Sweet sorghum; Jerusalem Artichoke; Sugar millet
		Starch energy	Wheat; Potatoes; ;aize; Barley; Triticae;
		crops	Corn (cob); Amaranth
		Others	Flax (Linum); Hemp (Cannabis); Tobacco stems; Aquatic plants (lipids from algae); Cotton stalks; Kenaf

Table 1. Type of biomass feedstock available for energy purposes (VIE	WLS, 2005; Van
Loo and Koppejan, 2008; Tumuluru et al., 2011)	

• Blends and mixtures: blends are intentionally mixed biofuels, whereas mixtures are unintentionally mixed biofuels (Table 3 EN 14961-1).



For the avoidance of doubt, demolition wood is not included in the scope of the EN 14961-1. Demolition wood is "used wood arising from demolition of buildings or civil engineering installations" (EN 14588, 2010).

5. Biomass advantages and barriers

5.1. Biomass advantages

Increases in the price of fossil fuels, growing environmental concerns regarding their use and impact (including climate change) and considerations regarding the security of energy supply have motivated the rising use of renewable energy sources worldwide (Luque et al., 2008).

Biomass-to-energy facilities present many benefits. Biomass is a renewable resource, and generating electricity and other energy products from biomass offsets consumption of fossil fuels. Biomass is typically waste material from another industry, such as logging or dairy operations, and converting it to energy not only reduces disposal, but also mitigates environmental impacts that these wastes can have on air, groundwater, and surface water quality. These advantages, together with those described below, turn biomass into a very important element for the territorial balance, especially in rural areas.

• Reduction of pollutant emissions such as CO, HC and NO_x

Use of biomass for energy production incorporates the benefits such as reduced CO_2 emissions from combustion of fossil fuels (Bram et al., 2009), reduced SO_2 formation through a decrease in fuel bound sulphur and reduced NO_x formation through a reduction in fuel bound nitrogen (Lehtikangas, 2001; Li et al., 2009; Qian et al., 2011). The low levels of particulates, CO and C_xH_y , contribute to avoid impacts on the photochemistry of the atmosphere (Garcia et al., 2012).

• Biomass carbon neutrality with no contribution to the greenhouse effect

One of the most important advantages related to the use of biomass is the carbon neutrality cycle (Wahlund et al, 2004; Munir et al., 2009; Gil et al., 2010a,b)

previously described (Figure 5). An increase of 1695 MW in electric power generation from biomass and 582.5 ktep in primary energy from thermal biomass energy would avoid the emission of 9 million tones of CO_2 to the atmosphere (IDAE, 2007)

• Reduction of risks from forest fires and insects plagues

The exploitation of agricultural biomass as a fuel for biomass boilers is one of the solutions to clean up the forests.

• Exploitation of agricultural wastes

Agricultural cuttings and prunings, traditionally burnt in the country, can find a new market as residual biomass to produce energy. Therefore the use of residual biomass improves waste management and mitigates the environmental impacts of waste disposal, according to the Directive 98/2008 on waste.

• Use of fallow lands for energy crops

The reforestation with energy crops can increase water retention as well as reduce the degradation and erosion of the soil. The right management of this energy resource stops the erosion and contamination of the water because the vegetation cover reduces the superficial run-off and stabilizes the particles from the soil, preserving animal and vegetal biodiversity.

• The use of biomass can reduce dependence on foreign oil because biofuels are the only renewable liquid fuels currently available

Biomass and biological wastes are an autonomous fuel resource, partially averting dependence on foreign energy supplies, improving a country's trade balance and economical sustainability (Wolf et al, 2006; Demirbas, 2008).

• Socioeconomic improvement of rural areas

From a social point of view, promoting biomass production for energetic purposes can develop a new activity in rural areas, which can be energetically self-sufficient because of the use of local biomass. The development of biomass and energy crops stimulates job creation, avoiding the depopulation of rural



areas and supports local agriculture. In this way the impact on food production can be minimized (Bahng et al., 2009).

5.2. Biomass barriers

In contrast to the benefits, there are significant barriers to biomass-to-energy facilities.

Compared to conventional heating systems, such as oil or gas boilers, biomass technology still has disadvantages in terms of space demand, efficiency, particulate emissions and maintenance. Therefore, due to the low energy densities exhibited by biomass, collection and transportation of the biomass can be cost prohibitive. On the other hand, biomass fuels tend to have a high moisture content, which adds weight and increases the cost of transportation. The moisture content also decreases combustion performance. Nevertheless, an intelligent design and sophisticated technology can minimize these disadvantages (Fiedler, 2004).

But there are not only logistical barriers regarding the use of biomass. It is very important to use it in a sustainable way, which requires comprehensive management of natural resources such as land and water. Unsustainable biomass production would erode the climate-related environmental advantage of bioenergy. It is more important than ever to reliably demonstrate that the advantages of biofuels exceed the cost of the potential environmental damage caused by their production. Therefore, sustainable development of biomass and biofuels is the major challenge in increasing the production of biomass and biofuels. Criteria to ensure the sustainable production of biomass are urgently needed (Ladanai and Vinterbäck, 2009).

6. Biomass-to-energy technologies

All other renewable or low carbon energy technologies (for example solar, wind) can only produce heat and power; whereas biomass is able to supply a range of carbonbased products with material qualities, such as liquid fuels, metallurgical reductants, lubricants and a wide range of petrochemical substitutes. This could contribute relatively seamlessly to a low emission future with respect to existing downstream technologies and infrastructure assets, and in general it will wise be concentrate the use of biomass to applications where material qualities can be brought to bear. Where opportunities for biomass initiatives exist around the world, these are primarily the result of tightening environmental regulations and increasing waste disposal costs. Increased landfill costs, restrictions on land-application of raw manures, and curtailment of open burning of forest residues are all circumstances that would make biomass-to-energy a more attractive solution. Parts of the world, such as Europe, where environmental restrictions are more stringent and energy prices are higher are looking to biomass-to-energy facilities as an opportunity to solve both issues.

Biomass utilization is as old as the discovery and use of fire for heating and cooking systems using wood as fuel. Nevertheless, biomass is a complex resource that can be processed in many ways leading to a variety of products.

There are six generic technologies for energy conversion based on: direct combustion for power; anaerobic digestion for methane rich gas; fermentation of sugars for alcohols; oil exaction for biodiesel; pyrolysis for biochar, gas and oils; and gasification for syngas (Barber and Warknen, 2008). These can then be followed by an array of secondary processing options (stabilisation, dewatering, upgrading, refining) depending on specific final products.

Anaerobic digestion, fermentation and oil extraction are suited to specific biomasses that have easily extractable oils and sugars or high water contents. Such biomass resources are mainly associated with the food chain and are strategically constrained (limited availability and/or competition for land). Digestion, oil extraction and fermentation of sugars are all well established commercially.

Thermal technologies such as direct combustion, pyrolysis and gasification, can effectively process all forms of biomass, including complete utilisation of lignocellulosic materials. Lignocellulosic material (woody biomass) is overwhelmingly the most common form of biomass in agricultural, industrial, municipal, forest and natural environments. Tree based lignocellulosic biomass can be essentially harvested on demand therefore offers the most stable and scalable primary resource for biomass


technologies. The abundance of lignocellulosic biomass provides strategic advantages to thermal technologies and to emerging technologies for cellulosic fermentation. Combustion and gasification of biomass for heat and power is commercially proven. Of all of the thermal processing options currently available, pyrolysis provides the most flexible product platform, being able to generate solid, liquid and gas outputs roughly analogous to their coal, oil and gas fossilised counterparts. However, there are still relatively few commercial pyrolysis operations using biomass feedstocks. Pyrolysis outputs are mainly used in basic heat and power applications, with developments underway to produce higher quality transport fuels through the further refining of biocrude. Pyrolysis char (biochar) can be used for energy, metallurgical reduction, activated carbon, or for carbon sequestration in soils.

Technology for providing bioheat to households, commerce and industry is available, reliable and efficient but has to compete against well established systems based on fossil fuels. Bioenergy can provide both low-temperature heat and steam, and high temperature heat suitable for industrial processes. Small-scale heating systems fired with wood logs, chips or pellets offer good ease of use, low operating costs and are replacing oil heating in many European regions. Biomass district heating is of growing importance in countries where demand for heat by the residential/service sector is high (RHC, 2011).

Each biomass resource has different characteristics in terms of calorific value, moisture and ash content, as well as distinct molecular structures that require appropriate conversion technologies for bioenergy production. These conversion routes use chemical, thermal and/or biological processes. The selection of processing technologies needs to be aligned to the nature and structure of the biomass feedstocks and the desired project outputs. Therefore, a particular biomass technology must be tailored to the biomass type to achieve optimum outcomes.

7. Biomass potential and consumption

Biomass is the fourth largest energy source after coal, oil and natural gas (Ladanay and Vinterbäck, 2009) and in consequence, it is the largest and most important renewable energy option at present.

It is, together with the other renewable energy options, capable of providing all the energy services required in a modern society, both locally and in most parts of the world. Renewability and versatility are, among many other aspects, important advantages of biomass as an energy source. Moreover, compared to other renewables, biomass resources are common and widespread across the globe (Santamarta, 2004; WEC, 2004). Therefore it can be concluded that the potential of biomass for energy production is promising.

7.1. Global biomass potential and consumption

Biomass is the primary source of energy for nearly 50% of the world's population (Karekezi and Kithyoma, 2006) and wood biomass is a major renewable energy source in the developing world, representing a significant proportion of the rural energy supply (Hashiramoto, 2007). In the past decade, the number of countries exploiting biomass opportunities for the provision of energy has increased rapidly, and has helped to make biomass an attractive and promising option in comparison to other renewable energy sources. The global use of biomass for energy production increases continuously and has doubled in the last 40 years (Figure 5).



Figure 5. World use of combustible renewables and waste 1971 – 2006 (World Bank, 2009)

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The annual global primary production of biomass is equivalent to the 4,500 EJ of solar energy captured each year (Sims, 2004). The potential of global biomass as a sustainable energy source is widely recognized. Thus, at present, a bioenergy supply of 270 EJ, possible to be achieved on a sustainable basis by only 6% of the annual global primary production of biomass, can cover almost 50% of the world's total primary energy demand (Ladanay and Vinterbäck, 2009). However, contribution of biomass to the global energy demand of 532 EJ in 2010 was only 10% (IEA, 2012)

The future potential for energy from biomass depends in part on land availability. Currently, the amount of land devoted to growing energy crops for biomass fuels is only 0.19% of the world's total land area (0.025 billion hectares) and only 0.5-1.7% of global agricultural land (Figure 6). A mere 10% increase in the efficiency of biomass production through irrigation, manuring, fertilizing and/or improved management through the cultivation of idle land, would create energy equivalent to the total current global energy demand (Ladanay and Vinterbäck, 2009). The current forest standing stock is a large reservoir of bioenergy and in line with the theoretical potential of biomass energy.



Figure 6. Distribution of land use types in world's total land area (Smeets et al., 2004; Faaij, 2008).

There are many scenarios that predict a future potential in biomass. There are also many studies performed during the past decades which attempt to estimate the future demand and supply of bioenergy (Lashof and Tirpak, 1990; Hall et al., 1993; WEC, 1994; Fujino et al., 1999; IPCC, 2000; Rogner, 2000; Fischer and Schrattenholzer, 2001; Hoogwijk, 2004). However, published estimates of the total global bioenergy production potential in 2050 ranged from 33 to 1,135 EJ annualy (Hoogwijk et al., 2003), from which 0 to 358 EJ annually came from woody biomass (Sørensen, 1999; Hoogwijk et al., 2003). Energy crops from surplus agricultural land have the largest potential contribution of 0-988 EJ/year (Hoogwijk et al., 2003).

This large range of estimates was the result of: differences in the type of biomass included; differences in the theoretical, technical, economic, or ecologic limitations related to the supply of woody biomass for energy use; differences in data on key parameters, such as the consumption of wood fuel, the annual growth of forests, and the efficiency of conversion; differences in scope whereby most of the existing bioenergy potential assessments focused on either the demand (WEC, 1994) or the supply (Yamamoto et al., 1999) of bioenergy and consequently ignored demand-supply interactions (Smeets and Faaij, 2007). Moreover, as Smeets and Faaij (2007) pointed out, most of the studies they reviewed ignored existing studies on the demand and supply of wood (Lazarus et al., 1993; Sørensen, 2001), despite the extensive literature and data on the subject (Solberg et al., 1996; FAO, 1998; Sedjo and Lyon, 1998). Overall, differences between the various scenarios are due to large differences in demand and energy mix, as a result of variations in population dynamics, and economic and technological development.

The global primary energy demand was 302 EJ in 1980, 420 EJ in 2000, 470 EJ in 2007 (IEA, 2009a) and 532 EJ in 2010 (IEA, 2012), but was expected to increase to 591 and 712 EJ by 2015 and 2030 respectively (IEA, 2009b), and to 826 EJ (as averaged for different scenarios) by 2050 (Smeets et al., 2004). However, even in the case featuring the highest consumption scenarios of 1,041 EJ in 2050 (Smeets et al., 2004), the 1,342 EJ which is the average of the cited bioenergy production potentials in 2050 is enough to meet this demand (Ladanai and Vinterbäck, 2009). The current



theoretical potential of biomass energy is 2,900 EJ (WEA, 2000). The current stock of standing forest, with a total energy content corresponding to 2,011 EJ, is a large reservoir of bioenergy (Figure 6). Overall, the world's bioenergy potential seems to be large enough to meet global energy demand in 2050.

7.2. European biomass potential and consumption

Europe is in the midst of a dramatic energy transition, away from fossil fuels, and in some cases also from nuclear power, to renewable energy. Bioenergy is the major renewable energy source, accounting for almost 70 percent of European renewables (EEA, 2005a; AEBIOM, 2011), and showing steady growth. This development did not start with the 20-20-20-goals formulated in 2007, or the Renewable Energy Directive adopted in 2009. Already before these goals were in place renewable energy sources had almost doubled their production from the early nineties (AEBIOM, 2011).

The national action plans presented by the EU member states will further stimulate this development, particularly for the "slow starters". A requirement for success is, however, that the governments come forward with strong incentives, not only for power production and for the transport sector, but in particular for the heating sector. A price on carbon dioxide emissions, a common European carbon tax, is necessary to give the extra push for the conversion of heating systems from fossil fuels to biomass (EC, 2004; EREC, 2004; EEA, 2005b; Ragwitz et al, 2005).

Year	Renewables	Biomass and Waste	Solar	Geothermal	Hydro Power	Wind Energy	Biomass Share
2000	98.2	59.5	0.4	3.4	30.4	1.9	60.6%
2001	101.4	60.4	0.5	3.6	32.0	2.3	59.6%
2002	99.8	62.1	0.5	3.9	27.1	3.0	62.2%
2003	107.8	67.8	0.6	5.3	26.3	3.8	62.9%
2004	116.2	72.5	0.7	5.3	27.8	5.0	62.4%
2005	120.9	77.5	0.8	5.3	26.4	6.0	64.1%
2006	129.1	83.5	1.0	5.5	26.5	7.0	64.7%
2007	143.1	91.8	1.2	5.7	26.6	8.9	64.2%
2008	144.2	98.2	1.7	5.7	28.1	10.2	68.1%
2009	152.6	104.7	2.4	5.8	28.1	11.4	68.6%

Table 2. Gross inland consumption of renewable 1995-2009 in EU27 (stacked, Mtoe)(AEBIOM, 2011)

The consumption of renewable energies has significantly increased in recent years. While hydro power stagnates, solar and wind energy show impressive growth rates, but starting from a relatively small market share, biomass is by far the most important source of renewable energy in Europe. Biomass represented 68.6% of the consumption of renewable energies in the EU in 2009, remaining stable compared to the previous year (Table 2).

At the EU level, in absolute terms, total gross final energy consumption fell from 1193.3 Mtoe in 2006 to 1113.6 Mtoe in 2009, while consumption of renewable energy rose from 129.1 Mtoe in 2006 to 152.6 Mtoe in 2009 with an average biomass share of 65% (AEBIOM, 2011).

Figure 7 depicts the bioenergy balance for 2008. Over 1/3 of the biomass consumed in EU is fed to electricity, combined heat and power (CHP) and district heating plants, while the rest is consumed in the private, commercial and industrial sectors for heating purposes. About 10% is used as biofuels in the transport sector, as well as 60% of biomass from CHP plants is used as recovered heat.



Figure 7. European bioenergy balance in 2008 and 2020 targets (AEBIOM, 2011; RHC, 2011)



The Renewable Heating and Cooling (RHC)-Platform expects biomass use to more than double by 2020 and to reach around 370 Mtoe of primary energy in 2050 (RHC, 2011), mostly to meet heat demand (231 Mtoe total contribution to heat demand in 2050). By 2020 the biomass supply should be increased significantly to meet the demand of all sectors of heat, electricity and transport biofuels. Such developments in the biomass supply should be realised taking into account the need for the other sectors like food as the priority for agriculture and materials production. The greatest contribution to increase the biomass supply is due to energy crops, by-products from agriculture and the use of forest logging residues.

In order to understand the future role of bioenergy in Europe, it is important to analyze the potential of biomass. There are three relevant sectors: agriculture, forestry and waste; under these main sectors there are categories of dedicated biomass production such as biofuel crops, agricultural byproducts or primary and secondary forestry residues. The forest (41%) and waste (38%) sectors can contribute the lion share of the potential. The remaining 21% may come from the agricultural sector and is scattered over many different small categories (AEBIOM, 2011); within the agricultural group the largest contribution may come from straw, dedicated cropping and prunings.

	2003	2010	2020	2030	-
TOTAL	69	187	228	284	-
Wood direct from forest	-	43	43	55	
Wastes and residues	67	100	100	102	
Energy crops from agriculture	2	44	85	122	

Table 3. Environmentally-compatible bioenergy potential (Mtoe) in the EU25 (EEA, 2005a)

The environmentally-compatible potential (Table 3) is the quantity of biomass that is technically available for energy generation based on the assumption that this places no additional pressures on biodiversity, soil and water resources compared to a development without increased bioenergy production. Moreover, it should be in line with other current and future environmental policies and objectives. This concept was realized by taking into account a number of environmental constraints when calculating the available technical potential. It was also assumed that greenhouse gas emissions are further reduced by 20 % in 2020 and 40 % in 2030 by the EU-25 (EEA, 2005a). The data in Table 3 show the production potential for biomass in the EU 25 as calculated by the European Environmental Agency (EEA, 2005a). The figures for EU 27 can be estimated as 10% higher. The main growth is seen in the sector of "waste and residues" and "energy crops from agriculture", due to the Utilised Agricultural Area (UAA), which represents 38 % of the whole EU-27 territorial area (AEBIOM, 2011).

The agricultural biomass resource for bioenergy mainly depends on the available land area and the yield of bioenergy crops grown. Priority was given to domestic food production needed to satisfy food demand. In consequence the share of UAA in the total area varies greatly from country to country, from only 2% in Latvia or 7% in Finland and Sweden to 65% in the United Kingdom. As part of UAA, arable land represents almost one quarter of the whole EU-27 territory and Denmark has the highest share of arable land (57%). Permanent grassland represents 14% of EU-27 territory; while more than 45% of the land in Ireland and the United Kingdom is used for permanent grassland, extreme northern and southern countries (Finland and Cyprus) have less than 1% of their land under permanent grassland. Finally, land under permanent crops represents less than 3% in the EU-27; however, several southern European countries have a higher share of land under permanent crops: 10% in Spain, 9% in Greece and Italy and 8% in Portugal) (AEBIOM, 2011).

7.3. Biomass potential and consumption in Spain

Spain has been one of the countries with the highest dependency on imported energy resources in the European Union. In 1973, domestic energy resources covered only 28.6% of the total energy demand so Spain was severely hit by the oil crises of mid 1970s. In consequence the Spanish governments have developed policies aiming at energy saving and the diversification of primary energy resources since the 1970s (Dinica, 2009). In this framework, the use of all types of domestic energy resources has become top priorities.

Renewable energy in Spain has developed into a growing share of the energy system, which is evidenced by the coverage of demand, expressed in terms of primary



energy as final. This has seen a significant rise from 2005 and in 2009 renewables have accounted for 9.4% of primary energy supply, and exceeded 12% in terms of gross final energy. Electricity production from renewable sources, oscillating due to climatic variations that affect water resources, shows a more stable and upward trend since 2005. In the past ten years, electricity from renewable sources has experienced an increase of over 40%, reaching in 2009 24.7% of gross electricity production in Spain (AVEBIOM, 2011).

The relevant objectives for the promotion of renewable energy in Spain are included in the Renewable Energy Plan, for the periods of 2005-2010 and 2011-2020. The Renewable Energy Plan (PER) 2005-2010 provided that, in 2010, biomass would account for 57% within a global contribution of renewable energy sources of 12.6% on energy consumption primary. The fact is that biomass has only about 500 MW, compared with 1,317 MW expected in the period 2005 to 2010 (APPA, 2009).

Despite of having 23.10% of utilised agricultural area and a total biomass potential of 17,286 ktoe/year (IDAE, 2011), the share of biomass in the final energy consumption of the country was only 4,630 ktoe (5.21%) in 2009, a value lower than the average of the UE-27 (7.51%) (AEBIOM, 2011). This situation was far from covering the goal of 12,257 ktoe share of biomass planned for 2010 (Fernandez, 2009).

According to objectives settled in the Spanish Resource and Energy Analysis Programme, Spain should be consuming 9 to 11 million tons per year of biomass for heating purposes by 2020. An affordable goal, taking into account all Spanish biomass that every year remains in the forests (it is being used only 29% of the annual available biomass stock for all industrial uses of wood).

Biomass consumption in the industrial sector from 2000 to 2008 stabilized its upward trend in key subsectors, but the changes from 2005 in some companies in the pulp and paper sector, the new cogeneration projects in pellet plants and in agrialimentary sector, coupled with measures to boost renewable thermal energy (such as incentives or credit lines) allows to establish a significant increase of biomass consumption in the industrial sector. Currently, the consumption in the industrial sector was 1,478 ktoe in 2008. Concerning the evolution of consumption in the domestic sector, from 2003 to 2008, the market has begun to take off for modern thermal biomass for domestic purpose, agricultural applications, developments in public administration and services sector. Biomass consumption for the household sector and miscellaneous uses was a total of 2,138 ktoe in 2008. The use of biomass in the domestic sector is the most competitive application and in which you get the biggest savings of greenhouse gases (GHG) emissions and energy efficiencies (90%), but its development has been slowed down by the lack of incentives to cover the initial risks of implementation related to high investment costs of equipment, uncertainties in the supply and maintenance of facilities.

7.4. Biomass potential and consumption in Andalusia. The importance of biomass from olive groves⁵

Andalusia is in a privileged position because of its capacity for wind power production, as well as its abundant solar catchment. However, biomass is the renewable source which most contributes to Andalusian energy infrastructure (AAE, 2009) because of the availability of residual biomass. This is hardly surprising since farming is the main economic activity in Andalusia. In fact, there are 4.7 million agricultural hectares (57% of the territory). Of these hectares, 1.4 million are olive groves, which are the main source of residual biomass in the Autonomous Community (INE, 2003; Junta de Andalucía, 2007).

Of the various economic sectors, the primary sector (agricultural activities, hunting, silviculture and fishing) is more important in Andalusia than in the rest of Spain, and in 2003 represented 6% of the gross added value of Andalusian economy. This result is an excellent indicator of the importance of this sector in Andalusia, which generates a high percentage of employment (27% of all agricultural jobs in Spain) (Junta de Andalucía, 2007).

⁵ The results shown in this section were presented in: A. García-Maraver, M. Zamorano, A. Ramos-Ridao, L.F. Díaz. Analysis of olive grove residual biomass potential for electric and thermal energy generation in Andalusia (Spain). Renewable and Sustainable Energy Reviews 16 (2012) 745–751.



Andalusia estimates its primary energy consumption at 20.5 Mtoe, based on the use of fossil fuels imported as crude oil (50.5%), natural gas (31.7%) and coal (8.5%). Fossil fuels are not produced in Andalusia because there is no crude oil. Since there are only five deposits of natural gas and four deposits of coal in the region (PASENER, 2007), its self-sufficient energy supply is estimated at 9.1%.

Unit: ktoe	Coal and derivative products	Crude oil and derivative products	Natural gas	RENEWABLE ENERGIES	Electric energy	Derivative energies (heat for sale)	TOTAL
Production	23.4	0.0	15.4	1,668.3	0.0	0.0	1,957.1
Recoveries	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Exchange balance (imp-exp)	1,823.0	10,135.8	6,438.8	-58.2	286.1	0.0	18,625.5
Stock variation (initial-final)	-355.9	45.6	70.7	0.0	0.0	0.0	-239.6
Bunkers (maritime transport)	0.0	199.2	0.0	0.0	0.0	0.0	199.2
GROSS INTERNAL CONSUMPTION	1,740.5	10,380.6	6524.9	1,610.0	286.1	0.0	20,542.2
Available for final consumption	31.6	8,871.3	2,391.6	750.6	3,134.9	33.9	15,213.9
Non-energetic final consumption	0.0	1,312.7	276.7	0.0	0.0	0.0	1,589.4
ENERGETIC FINAL CONSUMPTION	31.6	7,558.6	2,115.1	750.6	3,134.9	33.9	13,624.7

Table 4. Energy balance of Andalusia in 2008 (AAE, 2009)

Conversely, in this same region, 1668.3 ktoe of energy comes from renewable sources. Of this energy, 58.2 ktoe are exported. The gross internal consumption of renewable energies is thus 1610 ktoe, with 750.6 ktoe available for final energy consumption. This is 5.5% of the 13.6 Mtoe final energy consumption in the region (Table 4), which is mainly used by the industrial and domestic sectors (Table 5).

Of the various renewable energies, biomass comes to 6.3% of the total primary energy consumption and 78.7% of the renewable energy consumption (Figure 8). The progressive increase in renewable energy sources is mainly due to the use of biomass

for energy. The rain calendar as well as the agricultural crops and harvests have a bearing on this contribution. In 2008, the contribution of primary energy from biomass was 40.5% (365.1 ktoe), which was higher than in 2007 (Figure 8).

Unit: ktoe	Coal and derivative products	Crude oil and derivative products	Natural gas	RENEWABLE ENERGIES	Electric energy	Derivative energies (heat)*	TOTAL
Industry	31.6	682.4	1,786.1	466.5	789.6	33.9	3,790.1
Transport	0.0	5,405.3	6.3	98.0	33.0	0.0	5,542.6
Primary (agriculture and fishing)	0.0	896.7	88.2	7.5	126.1	0.0	1,118.6
Service sector	0.0	33.9	122.5	56.3	1,065.6	0.0	1,278.3
Residential	0.0	540.3	112.0	122.3	1,120.6	0.0	1,895.2
ENERGETIC FINAL CONSUMPTION	31.6	7,558.6	2,115.1	750.6	3,134.9	33.9	13,624.7

Table 5. Energetic consumption distribution by sectors in Andalusia (AAE, 2009)



Figure 8. Evolution of total renewable primary energy consumption in 2008 in Andalusia (AAE, 2009)

The information in Table 6 shows the installed power capacity for each renewable technology in Andalusia. It also specifies the attainment level of the objectives established in the PASENER 2007–2013. The goal is for renewable energies to cover 12% of Andalusian energy consumption in 2010 and 25.4% of this consumption in 2013. In the case of biomass, it is necessary to distinguish between

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biomass for the generation of electricity and biomass for thermal use. Since the enactment of the Andalusian Energy Plan (PLEAN) 2003–2006, the required levels for thermal biomass have not as yet been attained, though in the case of electricity biomass, the goal has been reached (Table 6). The reason for this was the transfer of the little fuel available in 2006 to electricity generation plants and the exportation of biomass during this same period to European countries (PASENER, 2007). When the PLEAN 2003–2006 was in force, biomass was exported to other European Union countries, mainly because of a compensation system in Spain, which could not compete with the prices of this biofuel in other countries. This situation was remedied with the passing of Royal Decree RD 661/2007, which was an important advance since it subsidized installations that used biomass to generate electricity.

		PLEAN	Degree of	PASENER	PASENER	PASENER
	2006	OBJECTIVES	Compliance	OBJECTIVES	OBJECTIVES	OBJECTIVES
		2006	2006 (%)	2007	2010	2013
Hydraulic (special regime), MW	129.8	107.2	121.1	129.8	137.8	148.0
Hydraulic (ordinary regime), MW	464.2	-	-	464.2	476	476
Wind, MW	607.9	2,700	22.5	1,284	4,000	4,800
Solar photovoltaic (insulated), MWp	5.8	6.1	95.1	36.2	55.4	100
Solar photovoltaic (connected), MWp	15.4	4.4	348.7	30.2	55.4	100
Solar thermal, m^2	347,182	411.5	84.4	407,000	765,228	1,341,554
Solar thermoelectric, MW	11	100	11	60	250	575
Biomass thermal, ktoe	367.5	643	57.2	583.5	615.6	649.0
Biomass electric, MW	164.6	164	100.4	169.9	209.9	256.0
Biofuels consumption, ktoe	36.1	90	40.1	263.7	2,000	2,300

Table 6. PLEAN and PASENER objectives (PASENER, 2007)

At the end of 2009, the total power capacity available in installations in the Autonomous Community of Andalusia came to 14051 MW. Renewable sources provided 4419 MW (equivalent to 877.2 ktoe/year, which is the energy consumption of over two million homes). It is significant that 62% of this amount was produced with biomass (Europapress, 2010).

Biomass is the renewable energy that most contributes to the energy system in Andalusia (Figure 8). This region has a cultivated land surface of 8,759,531.18 ha. Approximately 40% of this land is forest and 60% is farmland, of which certain crops cover a total surface of 1,937,107.76 ha (EC, 2002).

The biomass potential in Andalusia is about 3327 ktoe/year (PASENER, 2007). Of this annual amount of biomass, 1434 ktoe is obtained from agricultural residues, of which approximately 50% is olive tree residues (803 ktoe/year) whereas the other half is mainly obtained from sunflowers, cotton and fruit trees (Table 7). This was the capacity necessary to generate 5% of the primary energy consumption of Andalusia in 2007 (AAE, 2008; AAE, 2010). On the other hand, since olive oil industries generate 77.25% of the industrial residual biomass (Table 7), the olive sector can be regarded as the greatest producer of residual biomass in Andalusia.

The energy exploitation of biomass allows the sustainable replacement of fossil fuels, increases self-sufficiency and energy diversification, and contributes to the development of rural areas. Furthermore, biomass is a renewable energy that can be used as an energy source for electricity, heating, and vehicle transport (AAE, 2008), and reduces 890 g CO₂/kWh if it is used instead of oil. Biomass is a fuel of biological origin, such as woody biomass. If properly cultivated and managed, biomass does not contribute to climate change through emissions of CO₂ to the atmosphere because it absorbs the same amount of carbon in growing as it releases when consumed as fuel. In the future, CO₂ emissions from power and heat generation will have to be greatly reduced to meet environmental constraints and commitments (Wahlund et al., 2004).



	ktoe/year
TOTAL BIOMASS POTENTIAL IN	2277
ANDALUSIA	5521
AGRICULTURAL RESIDUES	1434
Olive grove	803
Fruit trees	86
Corn	72
Sunflowers	186
Greenhouse residues	100
Rice	43
Cotton	143
INDUSTRIAL RESIDUES	589
Olive oil industry	455
Bark	35
Rice husk	19
Cotton	15
Nuts	17
Wood	41
Olive stones	2
Sugar refinery	4
Cork	1
FORESTRY RESIDUES	136
Quercus	59
Eucalyptus	53
Pine	18
Poplar	7
ENERGY CROPS	559
Cynara	559
BIODEGRADABLE	609
MSW	54
Mud	43
Animal waste	15

Table 7. Biomass potential in Andalusia (PASENER, 2007; AAE, 2008)

The final use of biomass depends mainly on the characteristics of the raw material. Most of the biomass in Andalusia generates electricity (45%) and thermal energy (40%), which is mainly used in domestic heating systems (13%) and olive oil industries (14%) due to the abundance of olive groves and their proximity to this type of industry (Figure 9).

Olive groves are particularly relevant in Andalusia because of their importance in the region and more concretely, for the environmental management and energy use of the residues from olive oil industries and olive grove maintenance. In fact, Andalusia has specific legislation concerning olive groves, which enhances the efficient use of water and energy, and promotes the use of renewable sources of energy, in particular of biomass (Junta de Andalucía, 2009a). These regulations were passed in order to conserve energy and generally improve energy efficiency in the olive sector. The goal is to foment the energy exploitation of agricultural and industrial residues, the production of energy from biomass, and the use of renewable energies.

There are 1.4 million hectares of olive crops in Andalusia (INE, 2003), producing an average of 4 million tons of olives per year (AAE, 2008). Of this quantity, 3.7 million tons per year are used to make olive oil (800,000 tons/year), and the rest to obtain olives (300,000 tons/year), to be consumed as table olives. Furthermore, olive crops generate a variety of energy residues, such as olive pruning residues (4.2 million tons/year of branches and leaves that can be pelletized for domestic heating), olive stones (suitable for domestic and industrial heating or electricity production), olive-pomace and orujillo (Arjona et al., 1999; Ollero et al., 2003), residues that have been used traditionally for domestic heating in rural areas.



Figure 9. Biomass energy uses in Andalusia (AAE, 2009)

One hectare of olive grove generates three tons of pruning residues (Figure 10). This means that about four million tons are generated per year in Andalusia, most of which are now illegally burnt or left on the ground to generate greenhouse effect gases (Junta de Andalucía, 2009b).





Figure 10. Biomass potential from olive grove (AAE, 2008)

In Andalusia, there are 19 plants that use biomass to generate a total of 205.3 MW of electrical energy (AAE, 2008). This amounts to 1.46% of the total power capacity of 14,051 MW available in existing installations in the Autonomous Community. This quantity is very near to the objectives established in the PASENER for the year 2010 (Table 6), signifies a reduction of approximately 8% of the CO_2 emissions (PASENER, 2007). The majority of these installations use olive tree residues as fuel though there are plants that consume greenhouse residues, wood, and sugar cane bagasse (AAE, 2008). Olive-pomace, orujillo, and olive stones are the olive tree residues that are best for producing electrical energy because their lower bulk density, and in the case of olive-pomace, their higher moisture content (EC, 2002), make them less effective for domestic heating systems.

The demand for heating accounts for a significant portion of the total energy demand in the world today. The building sector consumes 35.3% of the final energy demand of which 75% is for space and domestic water heating (IEA, 2006). Renewable energy sources used for heating and cooling purposes have received relatively little attention compared with those used to generate electricity and to produce transport fuels. This is surprising because the demand for heat consumes the largest share of the primary energy supply. Renewable energy sources can thus offer a practical alternative

to fossil fuels under many circumstances (IEA, 2007). The potential to increase the use of solar, geothermal, and biomass resources for renewable energy heating is therefore large (Jurczak, 2006).

Biomass consumption for thermal applications during the year 2008 was approximately 613.55 ktoe. This biomass was mainly obtained from olive-pomace and olive stones. This resulted in a reduction of carbon dioxide emissions of 1895.87 metric tons (IPCC, 2006) in Andalusia.

Moreover, the use of standardized pellets from olive pruning residues would permit the use of these residues in efficient combustion processes. Nevertheless, certain characteristics, such as the higher ash content of olive tree pellets (Ollero et al., 2003) in comparison with other European pellets, have raised doubts concerning their use in thermal applications, especially in domestic heating systems.

Pelletized biomass made from agricultural and forestry products and residues are rapidly becoming an important renewable energy source for industrial and domestic purposes (Larsson et al., 2007). The pelletization of biomass is a mass and energy densification process for materials that possess low bulk densities. The typical bulk density of biomass chips is less than 150 kg/m³ while that of wood pellets is typically over 600 kg/m³ (Gilbert et al., 2009). In the pelletizing process, raw material is dried, ground, homogenized, and densified, which reduces transportation costs, improves storage, and results in better handling characteristics with less dust formation, besides providing potential storage for off-season utilization (Gilbert et al., 2009).

Andalusia is in the process of improving its infrastructure, and has become the first Spanish region in densified biofuel production, with 13 industrial pellet production plants. Six of these plants are in operation, and are producing 44 ktoe/year. All of them are strategically located near the olive groves to be able to use olive pruning residues (AAE, 2010). There are currently three more plants under construction in Jaen that will produce 34.6 ktoe. This will be in addition to the energy generated by the other plants, which is steadily increasing.



CHAPTER 3

DENSIFICATION OF BIOMASS

Because of its high moisture content, irregular shape and size, and low bulk density, biomass is very difficult to handle, transport, store, and utilize in its original form (Li and Liu, 2000; Kaliyan and Morey, 2009). In order to make the biomass available for a variety of applications, the challenges with the use of biomass materials must be resolved.

Densification of biomass materials could reduce the costs of transportation, handling, and storage. Because of uniform shape and sizes, densified products can be easily handled using the standard handling and storage equipment, and can be easily adopted in direct-combustion or co-firing with coal, gasification, pyrolysis, and in other biomass-based conversions (Vinterbank, 2004; Kaliyan and Morey, 2009).

Conventional processes for pressure-assisted densification can be classified into three types: extruding, pelleting, and briquetting (Li and Liu, 2000; Pietsch, 2002):

• In an extruder, the raw material is compressed by a screw or a piston through a die to form compacted particles, often of cylindrical shape. The diameter of extrusion products may range from 20 to 100 mm. Extrusion



technologies are commonly used for biomass densification in Asian countries such as India, Japan, and Thailand (Bhattacharya et al., 1985; Grover and Mishra, 1996; Varadharaju and Gothandapani, 1998; Klass, 1998).

- In pelleting equipment, the feed material is pressed through open-ended cylindrical holes (dies) made in the shape of a ring. One to three small rotating rolls push the feed material into the holes of the die from inside of the ring towards the outside of the ring. The skin friction between the feed particles and the wall of the die resists the free flow of feed and thus the particles are compressed against each other inside the die to form pellets. One or two adjustable knives placed outside the ring cut the pellets into desired lengths. The diameter of the pellets may range from 4.8 to 19.0 mm, and the length of the pellets may range from 12.7 to 25.4 mm (Li and Liu, 2000). Pelleting technology is commonly used in the U.S. and in Europe for producing animal feed.
- In a roll press briquetter/compactor, material is densified by compression between two counter-rotating rolls. Initial densification of the material may occur through compressing the material with a tapered auger in the feed mechanism. The primary purpose of this initial densification phase is to remove air from low bulk density material. The final compaction occurs because of high pressures created as the material flows between the two rollers. The roll surfaces have pockets to form briquettes of the desired size and shape when the material passes between the rolls. The briquettes are easily separated and handled after leaving the machine. The densified products are mostly of pillow-shape with a size of 10 to 40 mm or larger (Li and Liu, 2000). The roll press is generally used for densifying coal, minerals, and metals in the U.S. and in Europe (Pietsch, 2002).

Pelletization is the most common densification process used for solid fuel applications. Biomass pellets represent a promising alternative to fossil fuels for heat and power production, in particular in the residential sector, where automated heating systems can be implemented. The growing domestic and industrial demand of biomass for heat and power production in many European countries has resulted in a strong growing global pellet marked during the last decades, and continuous growth of the market is predicted for the next years (Junginger et al., 2009; Samuelsson et al, 2009).

Wood pellets from forestry residues already has successfully established technologies and markets for production and consumption in some European countries. However, in other countries such as Spain, this technique is still not widely used. For success of the densification process, the quality of the densified products must meet the consumer requirements and market standards. The final quality of pellets may vary depending on the raw material properties and the manufacturing process (Carone et al., 2011). These variables can be controlled to optimize production efficiency and improve the quality of finished product.

8. Pelletization advantages

There are several advantages of densified fuel pellets. The higher bulk and energy densities result, respectively, in lower transport costs and higher energy conversion efficiency, while the reduced moisture content increases the long term storage capability (Holm et al., 2006). These advantages, detailed below, are likely to stimulate its use in small stoves and boilers, where its enhanced properties will outweigh its extra cost (Carroll and Finnan, 2012):

- Increase the bulk density of biomass from an initial bulk density (including baled density) of 40–200 kg/m³ to a final bulk density of 600–800 kg/m³ (Obernberger and Thek, 2004).
- The uniform size and shape make pelleted biomass easier to handle using existing handling and storage equipment and increase the durability, avoiding the damage derived from the utilization of this material. Therefore,



this densification process reduces transportation costs and provides better handling and feeding of the biomass with less dust formation (Gilbert et al, 2009; Kaliyan and Morey, 2009), reducing cleaning and maintenance activities and making pellets an optimum fuel for domestic heating systems.

- High energetic density, dependent on the raw material, due to the compaction and the low moisture content (6-8%). In the majority of the cases the high calorific value ranges from 4,000 to 4,500 kcal/kg.
- Versatility due to the possibility of producing pellets from a wide range of raw materials. The main raw materials used are wood residues, such as saw dust, wood shavings and wood chips, but also agro-residues, such as straw, waste products of the food industry and fuel crops, are used for the production of fuel pellets (Sultana et al., 2010; Verma et al., 2012).

9. Pelletization process

Before entering into any densification equipment, the feed material is ground to a desired particle size, subjected to pretreatments such as mixing with additives or fats, and conditioned with steam or expander to increase the temperature and/or moisture level. During the pelletization process, the raw material is dried, ground, homogenized and densified (Figure 11), improving its storage, transport and handling characteristics (Larsson et al, 2007).

The principal stages of the process are described below:

a) Size reduction

Whether the raw material is wood, grass, straw or any other type of biomass, it must be reduced to a sufficient uniform size for the pellet mill. The general rule is that the milled material going into the pellet mill must be smaller than the die holes in the pellet mill to avoid blocking the holes (Arshadi et al, 2008). For example to produce 6mm pellets the milled material must be smaller than 6mm.

There are various types of equipment to conduct size reduction (chippers/shredders, hammer mills); each has different abilities and strengths (PelHeat,

2010). In some cases one piece of equipment is required, but the most common process two different stages of grinding are needed: a chipper before the dryer and a hammer mill after the dryer, capable of reduce the particle size to powder.



Figure 11. Diagram of the pelletization process (Zamorano et al., 2011)

b) Drying

The moisture content of the raw material is an important parameter for the pelletizing (compaction) process because moisture facilitates the heat transfer and friction. If the moisture content is too low the friction between the raw material and the die will become too high resulting in increasing power consumption and eventually the die holes might become clogged. The moisture content is also important for the thermal softening and self-bonding of individual particles in the pellet (Rhén et al., 2005).

In consequence, to produce quality pellets the moisture percentage of the raw material has to be not only homogeneous but also adequate to allow the pelletization process. Most materials produce the highest quality pellets with a moisture content of around 10-15% (raw weight); however, the moisture content to produce high-quality pellets is specific to each raw material (PelHeat, 2010).



Since it is not possible to reach such low moisture contents by natural drying, the material has to be artificially dried. The rotary/drum dryers are the most commonly used in the drying process.

c) Pelletization

The friction between the woody material and the die during the forced passage results in a rise in temperature. This, in combination with the moisture and lignin contents of the raw material, softens the wood components and is assumed to have a crucial influence on the self-bonding processes (Rhén et al., 2005; Gilbert et al., 2009). So for most of the biomass it is not necessary to add binders, since the composition of lignocellulosic materials works as a binder once the temperature inside the pellet press has increased.

There are two types of pellet mills: flat die pellet mill and round die pellet mill. Generally flat die pellet mills are used for small to medium scale pellet production and round die pellet mills are used for medium to large-scale pellet production. The flat die pellet mill works on the principle of material falling from above on to the rollers, which are rotating over the pellet mill die, while the round die pellet mill comprises of a vertical ring die with rollers on the inside, applying pressure against ring die (PelHeat, 2010).

d) Cooling, storing and packaging

The temperature of the densified products after leaving the densification equipment is generally higher than the conditioned feed due to frictional heat developed in the pressing systems and, therefore, the densified products are cooled using ambient air before storing/packaging. This last process step also results in a reduction of the moisture content and the pellet is hardened (Lehtikangas, 2001).

Once the pellets are produced and cooled, they are stored in silos before being transported by tankers to the consumer, or packed in bags of 15 kg.

10. Factors affecting pellet quality

The quality of pellets is determined by the end-user's requirements on the heating system and the handling properties (Lehtikangas, 2001). In general, the pellet quality depends on the properties of the feedstock – in terms of biomass type, moisture content and particle/fiber size; and quality management of the manufacturing process – in terms of operating conditions, pelletizer type and binding agent (Li and Liu, 2000; Lehtikangas, 2001; Rhén et al, 2005; Temmerman et al, 2006; Arshadi et al, 2008; Gilbert et al, 2009). These parameters involve a high sensitivity when the feedstock changes. As a consequence, a better knowledge of mechanical, physical and chemical properties of the biomass used is necessary to determine the thermal behavior (Lehtikangas, 2001; Gilbert et al, 2009; Kaliyan and Morey, 2009). These properties are described below.

10.1. Chemical properties

Chemical properties have large differences which are dependent on the raw material. These properties vary from species to species and within the same species it varies with the habitat, age and location in the tree (trunk, branches, top, leaves and roots) (Orfao et al, 1999).

a) Chemical composition

Biomass types are composed of a number of constituents and they have complex structures. With respect to chemical and physical properties, biomass species have large differences. These varying amounts of organic and inorganic elements have to be considered.

• Because of its organic nature, carbon, oxygen, nitrogen and hydrogen are the main components of biomass (Byrne and Nagle, 1997). These components are related to the calorific value of the raw material (Telmo et al, 2010).



- Hemicellulose, cellulose, lignin and extractive matter are the major constituents of biomass which are present in different ratios and structures in different biomass species. The mass loss of biomass during its devolatilization is controlled by these main components, which decompose at different temperature ranges (Prins et al., 2006).
- Inorganic elements can negatively affect to the combustion process. Sulfur and chlorine can reduce the melting point of the ash causing operational problems in combustion, such as formation of deposits (slagging) in the furnace (Öhman and Nordin, 1998). Sulfur and chlorine can reduce the protective oxide layer in the furnace because of its corrosive character. Chlorine can induce, e.g., formation of extremely toxic dioxins. Potassium content influences the ash melting behavior and aerosol formation. An increased potassium content leads to a decreased ash melting point, which can cause slag and hard deposit formation in the furnace and boiler. Moreover, increased potassium concentrations rise the amount of aerosols formed during combustion and thus fouling in boilers and fine particulate emissions (Obernberger and Thek, 2004).

b) Calorific value

The calorific value of biomass is an indication of the energy chemically bound in it and in the combustion process it is converted into heat energy. Calorific value is one of the critical properties of interest of a fuel because it determines the energy value of it. The design and control of a biomass combustor depend strongly on the calorific value of the biomass fuel (Erol et al., 2010).

c) Particle size distribution

The mechanical properties of binderless particleboards depend much on the size and shape of the raw material particles and it is considered beneficial with a mixture of particle sizes because this increases the durability of the board (Okuda and Sato, 2004). Particle size and its distribution has also for the same reason been considered important in biomass densification. For pelletizing, it is generally assumed that small particles with a large surface area will increase density and result in stronger pellets (Lehtikangas, 2001; Mani et al., 2006; Kaliyan and Morey, 2009).

A narrow particle size distribution of the raw material also facilitates even moisture distribution during the drying stage. In a mixture of different sizes, small particles might become over dried in the process and make self-bonding more difficult later on in the densification stage (Bergström et al., 2008).

d) Moisture content

Water has a crucial role in the pelletizing process (Mani et al., 2003; Kaliyan and Morey, 2009; Nielsen et al., 2009). It is found to be the most important factor influencing pellet quality and it can act as both a binding agent that affect mechanical durability and fines, and as a lubricant that lower the friction in the die resulting in low bulk density and low energy consumption (Kaliyan and Morey, 2009).

e) Ash content

One of the problems associated with biomass combustion is related to the ash content of the material. The quantity and quality of ash produced during the biomass combustion process are strongly influenced by the properties of the biomass and the combustion technology, including the characteristics of the furnace, temperature of the process, and ash extraction systems. Inorganic species in biomass fuels, such as alkali oxides and salts, can intensify agglomeration, deposition, and corrosion problems on the heat transfer surfaces of boilers (Tortosa Masiá, 2007). Therefore, despite the obvious benefits of using biomass as a fuel source, technical difficulties related to the inorganic part of biomass fuels can lead to the reduced accessibility of biomass combustion systems. A higher ash content lowers the heating value of the pellets and increases the risk of sintering. Since this also negatively affects milling and pelleting equipment (Lehtikangas, 2001), the calculation and subsequent reduction of the ash content in



biofuels is crucial to enhance their quality. It goes without saying that this is also bad publicity for the residential pellet market (Öhman et al., 2004a; Rhén et al., 2005).

f) Volatile content

Compared to coal, biomass has a high volatile content. Despite being considered as one of the most important renewable energy sources, biomass generates a number of pollutants, including particulate matter, unburned heavy hydrocarbons, and nitrogen oxides (Koziński and Saade, 1998). These emissions increase with the use of biomass with high ash and nitrogen contents (Johansson et al., 2004), but differences are observed depending on the type of burner, boiler, or stove in use (Olsson et al., 2002).

Like coal volatiles, biomass volatiles result from pyrolysis and typically are composed of H_2 , H_2O , CO, CO_2 , hydrocarbon gases, hydrocarbon liquids, and PAHs. If biomass is to be more widely used as an energy source, a greater understanding of the detailed processes is necessary. According to Williams et al. (2012), prediction for gas phase flow and volatile combustion including the detailed chemical mechanism model for calculating pollutants of PAHs and soot particles is one of the main tasks in biomass combustion modeling. Biomass conversion mechanism at high temperatures is still uncertain, specifically how the nature of the biomass affects the formation of PAHs and soot during combustion.

g) Thermal behavior

The thermal decomposition of lignocellulosic materials involves a number of chemical reactions as well as physical stages such as heat and mass transfer (Bilbao et al, 1997). Wood is a complex material, mainly composed of cellulose (40-60 wt%), hemicelluloses (20-40 wt%) and lignin (10-25 wt%), in addition to extractives (terpens, tannins, fatty acids, resins, etc), water and mineral matter (McKendry, 2002).

The differences between the main chemical components of biomass (i.e. cellulose, hemicelluloses, and lignin) have a direct impact on their chemical reactivity. Wood composition varies from species to species and within the same species it varies with the habitat, age and location in the tree (trunk, branches, top, leaves and roots)

(Barton, 1981; Pereira et al., 1984; Pereira, 1988). Therefore, given the heterogeneous nature of biomass, it is not easy to make generalizations about thermochemical treatment systems and their equipment.

Moreover, when thermal decomposition takes place in an air atmosphere, the presence of oxygen causes additional complexities. It enhances the degradation of the material at low temperatures (Shafizadeh and Bradbury, 1979), causes the appearance of gas-phase reactions between the volatile compounds released and oxygen, and when the temperature is high enough, oxygen is likely to promote the combustion of the char residue generated in the early stages of the solid degradation (Shafizadeh and Bradbury, 1979), thereby combustion and pyrolysis may coexist (Liu et al., 2002). The kinetics of this thermal decomposition is more related to the combustion process.

The nature of a thermogravimetric analysis (TGA) curve in combination with its corresponding derivative DTG peaks indicates the number of stages of thermal degradation, related to chemical composition. DTG is used to determine the characteristic temperatures such as the temperature of maximum mass loss rate, the initial decomposition temperature and the final decomposition temperature. The nature of a TGA curve in combination with the corresponding DTG peaks indicates the number of stages of the thermal degradation, which are directly related to the decomposition of the main components (Liu et al, 2002; Grioui et al, 2006; Marcilla et al, 2009).

Knowledge of the combustion characteristics is the basis and thus essentially important for a better understanding of thermal-chemical conversion of biomass.

10.2. Mechanical and physical quality properties of the pellets

Mechanical and physical properties are related to the manufacturing process. Changes in the operational conditions of the pelletization process affect these final parameters.



10.2.1. Mechanical properties

a) Durability

Mechanical durability (or abrasive resistance) is a quality parameter that is defined as the ability of densified biofuels to remain intact when handled (EN 14588, 2010). It is measured by the resistance of densified fuels towards shock or/and friction. Since this fuel type is susceptible to mechanical wear, which leads to the production of fine particles or dust during transport, transhipment and storage (Temmerman et al, 2006), this test will indicate the dusting potential of the pellets during handling, transportation, and storage. Dust emissions are not only an inconvenience for the consumer; they are also a health hazard (Vinterbäck, 2004). Additionally, fine particles and dust can disturb feeding systems of boilers and may lead to inhomogeneous combustion processes. Finally, dust may contribute to fire and explosion risks during handling, storage and transhipment (Lehtikangas, 2000).

b) Hardness

Hardness (or compressive or crushing resistance) is the maximum crushing load a pellet can withstand before cracking or breaking (Kaliyan and Morey, 2009). The tensile strength is related to the adhesion forces between particles at all contact points in the agglomerate (Pietsch, 2002; Gilbert et al, 2009).

Compressive resistance test simulates the compressive stress due to weight of the top pellets on the lower pellets during storage in bins or silos, and crushing of pellets in a screw conveyor. The compressive resistance test provides a quick measure of the quality of pellets as soon as the pellets are produced from the pellet mill and aids in adjusting the pelleting process to improve the pellet quality.

10.2.2. Physical properties

a) Bulk density

Bulk density of pellets is a parameter used for piles of wood fuels that create voids among the woody pieces which may be bigger or smaller depending on the size and shape of the pellets (EN 15103, 2009).

The bulky characteristic of non densified biomass makes it difficult to transport over long distances and demands space for storage. Therefore, densification of biomass by pelleting can significantly increase the bulk density of biomass from 40–250 kg/m³ up to 600–800 kg/m³ (Mani et al., 2003; Kaliyan and Morey, 2009) and thus improve storability and reduce transportation costs (Holm et al., 2006; Kaliyan and Morey, 2009). Moreover, bulk density is important to be considered since the fuel is fed by volume, not weight, so variations in bulk density can cause large variations in combustion efficiency.

b) Particle density

Particle density is defined as ratio of the mass and the volume of a sample including pore volume. Its value is affected by the particle size, the compression strength, and protein and moisture content (Mani et al, 2006).

This property has an influence on bulk density and on combustion properties of pellets (heat conductivity, burning time and rate of degasification) (Böhm and Hartmann, 2004; Obernberger and Thek, 2004).

c) Size of the pellets

Wood and wood-based materials interact with a humid environment by adsorbing and desorbing moisture. One factor that may limit the hygroscopicity was the hard circumference surface around the pellet. This surface is a result of the manufacturing through the high pressures needed to produce the pellet. However, the ends of the pellets are exposed to the humid environment and any moisture uptake by the pellet would be from the ends (Lehtikangas, 2000; Hartley and Wood, 2008), so an important parameter is the ratio length/diameter.

In this context the ratio length/diameter is also in particular of great relevance if pneumatic feeding systems are used, due to the fact that even a single long pellet is able to block the transport pipe of such systems (Obernberger and Thek, 2004).



d) Moisture content of the pellets

Moisture resistance is an important pellet property. The pelletizing process implies high temperatures, and attention has to be paid to proper cooling and heat removal before the pellets leave a production plant, especially with regard to the storage stage (Rhén et al, 2005; Mani et al, 2006). If moisture content exceeds 20% of d.b., bacterial growth might occur (Lehtikangas, 2000) causing material degradation and self-heating, which in worst case can result in self-ignition (Rhén et al, 2005).

11. Pellet quality standards⁶

Apart from the energy content of the pellets, end-consumers are mainly concerned with two problem areas: (i) emissions generated by pellet combustion; (ii) the use of pellets in boilers and stoves. This is understandable since emissions and low-quality biofuels can damage combustion equipment and produce undesired effects such as slagging, corrosion, and interference with process control (Lehtikangas, 2001). Furthermore, the content of fine particles in the fuel, which is extremely dependent on mechanical durability as well as logistics and storage, can disturb the regulation of highly automated heating systems or interrupt automated fuel feeding (Li and Liu, 2000). In addition, fine particles burn more quickly, and these higher temperatures can favor ash melting.

Consequently, many European countries have developed standards for the quality, storage, transport, and combustion of densified biomass fuels. These include parameters and guidelines pertaining to the following: particle and bulk density, moisture content, crushing resistance or hardness, particle number, particle size (length and diameter), chemical composition, ash content, and heating value. As trade between countries becomes more widespread, it is also necessary to create international standards to facilitate the purchase and sale of biomass fuels. Accordingly, the European Committee for Standardization, CEN, received a mandate from the European Commission to develop standards for solid biofuels within the Technical Committee (TC) 335 Solid Biofuels.

⁶ The results shown in this section were presented in: A. García-Maraver, V. Popov, M. Zamorano, 2011. A review of European standards for pellet quality. Renewable Energy; 36 (12): 3537–3540

Nevertheless, current national standards regarding pellet quality are very heterogeneous. This is reflected in the differences detected in their quality control parameters and guidelines. For this reason, the parameters and the limit values for densified biomass fuels were compared, as defined in different European standards.

11.1. Pellet quality: standards and certification

11.1.1. Austrian standard: ÖNORM M 7135

The Austrian standard for energy pellets is the ÖNORM M 7135, which establishes requirements and test specifications for compressed wood or compressed bark in its natural state – pellets and briquettes (ÖNORM M 7135, 2003). Furthermore, it also includes requirements for non-wood biomass pellets (i.e. M7139 for energy grains, C 4000 for Miscanthus pellets, and C 4002 for straw pellets). The main parameters and guidelines of the ÖNORM M 7135 are summarized in Table 8. Compliance with this standard is certified by the ÖNORM tested label for high-quality pellets. Other pellet related standards in Austria are those pertaining to pellet logistics (M 7136) and pellet storage tanks (M 7137).

	PARAMETER	LIMITS
	Diameter (mm)	4-10
Physical	Length (mm)	<5D
	Particle density (kg/dm ³)	<1.12
Mechanical	Durability* (%)	<2.3
	Moisture content (%)	<10
	Ash content (%)	<0.5
	Heating value (kcal/kg)	>4302
Chemical	N (%)	< 0.3
	S (%)	< 0.04
	Cl (%)	< 0.02
	Additives (%)	<2

Table 8. Parameters and guidelines included in Austrian standards for energetic pelletsÖNORM M 7135 (2003)

^{*} Durability has been defined in terms of percentage of fines after testing



11.1.2. Swedish standards: SS 187120

Sweden was one of the first countries to establish pellet quality standards in its SS 187120 (1998). This standard includes three categories of pellet quality, depending on the parameter limit values considered. The physical, mechanical, and chemical parameters, as well as the guidelines in this standard are listed in Table 9.

T		PELLETS CATEGORY			
PARAMETER		Group 1	Group 2	Group 3	
	Diameter (mm)	<25	<25	<25	
Dhysical	Length (mm)	<4D	<5D	<5D	
Fliysical	Bulk density (kg/dm ³)	>600	>500	>500	
	Moisture content (%)	<10	<10	<10	
Mechanical	Durability (%)*	< 0.8	<1.5	<1.5	
	Ash content (%)	< 0.7	<1.5	<1.5	
Chamical	Heating value (kcal/kg)	>4039	>4039	>3609	
Chemical	S (%)	< 0.08	$<\!\!0.08$	-	
	Cl (%)	< 0.03	< 0.03	-	

Table 9. Parameters and guidelines included in Swedish standards for biofuels and peatSS 187120 (1998)

[•] Durability has been defined in terms of percentage of fines after testing

11.1.3. German standards: DIN 51731 and DIN EN 15270

Germany has two standards: DIN 51731 (1996) for pellets and briquettes, and DIN EN 15270 (2007) for high-quality pellets. The first standard for compressed wood was introduced in Germany in 1996. The use of this standard in the small-scale pellet heating sector is limited because of its high maximum ash content (<1.5%) and the absence of an abrasion limit (Table 10).

In consequence, the DINplus certification, based on the DIN EN 15270 (2007) standard for high-quality wood pellets, was subsequently developed. This standard has contributed significantly to the success of the residential pellet market in Germany. In fact, today it is the most important quality certification for wood pellets worldwide. Its parameters and limit values are listed in Table 10.

• A.1. Deciduous and conifer tree trunk without bark; untreated wood from wood-yielding industry; untreated wood and wood without bark after being used; a mixture of these materials.

- A.2. Raw materials not included in category A.1; untreated herbaceous biomass; a mixture of these materials.
- A.3. Raw materials not included in category A.2. Table 11 summarizes the parameters, values, and guidelines for each pellet category.

-	-	· · · ·	• •
	PARAMETER	DIN 51731	DIN EN 15270
	Diameter (mm)	4-10	-
Physical	Length (mm)	<5D	<5D
	Particle density (kg/dm ³)	<1.2	<1.2
Mechanical	Durability (%)*	-	<2.3
	Moisture content (%)	<12	<10
	Ash content (%)	<1.5	< 0.5
	Heating value (kcal/kg)	3705-4661	>4302
	N (%)	< 0.3	< 0.3
	S (%)	< 0.08	< 0.04
	Cl (%)	< 0.03	< 0.02
Chaminal	Additives (%)	-	<2
Chemical	As (mg/kg)	< 0.8	< 0.8
	Cd (mg/kg)	< 0.5	< 0.5
	Cr (mg/kg)	<8	<8
	Cu (mg/kg)	<5	<5

Table 10 Parameters and guidelines included in German standard DIN 51731 (1996), for pellets and briquettes, and DIN EN 15270 (2007), for high quality pellets

Durability has been defined in terms of percentage of fines after testing

Hg (mg/kg)

Pb (mg/kg)

Zn (mg/kg)

Table 11. Parameters and guidelines	included in	Italian recom	mendation C	TI - R	04/5
	(2004)				

	PARAMETER		CATEGORIES OF PELLET				
Diamatan (mm)		A.1	A.2	A.3			
	Diameter (mm)	$6\pm0.5-8\pm0.5$	$6\pm 0.5-8\pm 0.5$	$10\pm0.5-25\pm1.0$			
Physical	Length (mm)	-	<5D	-			
	Bulk density (kg/dm ³)	620-720	600-720	≥550			
Mechanical	Dust emission (%)	≤1	≤1	-			
	Moisture content (%)	≤10	<10	≤15			
	Ash content (%)	≤0.7	<1.5	-			
	Heating value (kcal/kg)	>4039	>3870	-			
Chemical	N (%)	≤0.3	≤0.3	-			
	S (%)	≤0.5	≤0.5	-			
	Cl (%)	≤0.03	-	-			
	Additives (%)	banned	-	-			



Optimization of the pelletization process of agricultural wastes originating from olive farms for their application in domestic boilers

< 0.05

<10

<100

< 0.05

<10

<100
11.1.4. French recommendation: ITEBE

Even though France has no official standard for wood pellets, the government has established certain quality controls (ITEBE, 2009), which classify pellets according to their usage context (stove, boiler, large boiler, and incinerator). Table 12 summarizes the control and parameter values in these recommendations.

Doromotor		Type of c	ombustion	installation	
r ai ailietei		Stove	Boiler	Big boiler	Incinerator
	Diameter (mm)	6±1	8-10±1	>16	>16
Dhysical	Length (mm)	10-30	10-15	>16	>16
Filysical	Bulk density (kg/dm ³)	>650	>650	>580	>580
	Particle density (kg/dm ³)	1.2-1.4	1.2-1.4	-	-
	Moisture content (%)	<10	<10	-	-
	Ash content (%)	<10	<10	-	-
	Heating value (kcal/kg)	>4052	>4052	-	-
Chemical	N (%)	< 0.3	< 0.3	-	-
	S (%)	< 0.08	< 0.08	-	-
	Cl (%)	< 0.3	-	-	-
	Na (ppm)	<300	-	-	-

Table 12. Parameters and guidelines included in French recommendation ITEBE (2009)

11.1.5. European standard committee CEN/TC 335

Finally, the European Standard Committee CEN/TC 335 is the technical committee that has developed the draft standard to describe all forms of solid biofuel in Europe, including wood chips, wood pellets and briquettes, logs, sawdust, and straw bales.

The result is the experimental technical specification EN 14588 (2010) for solid biofuels, which includes and specifies the relevant terminology, definitions, and descriptions.

The guidelines and pellet classification in the European standard are different from those in the national standards. More specifically, it classifies pellets in three general categories, depending on the origin of the raw materials that pellets are made of: (1) woody biomass; (2) herbaceous biomass; (3) biomass from fruit and a mixture of the previous types of biomass.

Each category includes various subcategories that are specified to up to four levels. For example, the category of woody biomass includes the following subcategories: (1.1) forest, plantation and other virgin wood; (1.2) by-products and residues from wood processing industry; (1.3) used wood; (1.4) blends and mixtures. Subcategory 1.1 has seven subcategories: (1.1.1) whole trees without roots; (1.1.2) stemwood; (1.1.3) logging residues; (1.1.4) stumps/roots; (1.1.5) bark (from forestry operations;(1.1.6) segregated wood from gardens, parks, roadside maintenance, vineyards and fruit orchards;(1.1.7) blends and mixtures. Finally each of these subcategories also contains various subdivisions. For instance, category 1.1.1 is subdivided as follows: (1.1.1.1) deciduous; (1.1.1.2) coniferous; (1.1.1.3) bushes; (1.1.1.4) blends and mixtures. Each parameter also has various subdivisions, depending on their limit values (Table 13). Labels should provide information about the characteristics of each type of pellet. Furthermore, the annexes of this draft standard also include recommendations for the potential uses of pellets. For example, Table 14 shows the recommended parameter values for pellets used in domestic heating. In this case, guidelines are stricter because (i) domestic equipment does not employ advanced cleaning and control mechanisms; (ii) the equipment is not handled by professional staff; (iii) the combustion of pellets takes place in inhabited areas.

11.2. Comparative analysis of European pellet quality standards

The standards for pellet quality developed by various European countries differ not only in their basic requirements, but also in their limit values and their mode of application to the pellets. Pellet size, moisture content, heating value, and ash content are the pellet quality parameters that appear most frequently in standards and recommendations. However, pellet size (diameter, length and/or length/diameter ratio) is the only parameter that is included in all the norms.

Most of the standards specify moisture content limit values except in the case of large boilers and incinerators in the French recommendations (ITEBE, 2009). Bulk density is also included in practically all the norms with the exception of the German standards (DIN 51731, 1996; Din EN 15270, 2007).

Nitrogen and sulfur limit values are provided in the Austrian norm (ÖNORM M 7135, 2003), the German standards (DIN 51731, 1996; Din EN 15270, 2007), the Italian

standard (for A1 and A2 pellets) (CTI-R 04/5, 2004), and the French recommendations (for the use of pellets in stove and heating) (ITEBE, 2009).

Heating limit values appear in all the norms except in the Italian standard for pellet type A3 (CTI-R 04/5, 2004).

PARAMETER	CLASSIFICATION
	D06: D≤6±0.5 and L≤5D
	D08: D≤8±0.5 and L≤4D
Size (diameter and length) (mm)	D10: D≤10±0.5 and L≤4D
	D12: D≤12±1.0 and L≤4D
	D25: D≤25±1.0 and L≤4D
	M10: ≤10%
Moisture content (%)	M15: ≤15%
	M20: ≤20%
	A0.7: ≤0.7%
	A1.5: ≤1.5%
Ash content (%)	A3.0: ≤3%
	A6.0: ≤6%
	A6.0+: >6%
	N0.3: ≤0.3%
	N0.5: ≤0.5%
N (%)	N1.0: ≤1%
	N3.0: ≤3%
	N3.0+: >3%
	N0.05: ≤0.05%
$\mathbf{S}(0/)$	N0.08: ≤0.08%
S (%)	N0.1: ≤0.1%
	N0.2+: >0.2%
	CL0.03: ≤0.03
C1(0/2)	CL0.07: ≤0.07
	CL0.1: ≤0.1
	CL0.1+: >0.1
	DU97.5: ≥97.5
Durability*	DU95.0: ≥95
	DU90: ≥90
	F1.0: ≤1%
Fines content (%<3.15 mm)	F2.0: ≤2%
	F2.0 +: >2%
Bulk density (ka/m^3)	Recommended value should be included by
Burk density (kg/m/)	manufacturer
Heating value (kcal/kg)	Recommended value should be included by
	manufacturer
Additives	Binding materials and ash inhibitory should be
	included in the label

Table 13. Classification of parameters included in EN 14961-1 (2011) including specifications for solid biofuels

* Durability has been defined in terms of percentage of whole pellets after testing.

Most standards also include ash content limit values except in the case of A3 pellets in the Italian standard (CTI-R 04/5, 2004) and pellets for large boilers and incinerators in the French recommendations (ITEBE, 2009).

Finally, there are other parameters that appear in only a few of the standards. For example, the German standards include limit values for the As, Cd, Cr, Cu, Hg, Pb and Zn content of pellets. This is true even in standard DIN 51731 for lower quality pellets (DIN 51731, 1996; Din EN 15270, 2007).

Particle density is specified in the German standards and the French recommendations (i.e. for the use of pellets in stove and boilers) (DIN 51731, 1996; Din EN 15270, 2007; ITEBE, 2009). Nevertheless, the German standard DIN 15270 (2007) is the only one that includes limit values for additives, though this parameter is restricted to higher quality pellets.

In reference to the limit values of the various parameters, no important differences were observed, except in the case of ash content, durability, and heating value. In these cases, the German standard DIN EN 15270 (2007) and the Austrian norm (ÖNORM M 7135, 2003) were found to be the most restrictive.

Finally, there are norms that establish quality parameters, according to a preestablished classification of pellets. For example, the Swedish standard specifies three categories of pellet quality depending on the limit values set. The set of values for group 1 defines pellets of the highest quality; the set for group 2 defines pellets of medium quality; and the set for group 3, pellets of lesser quality (SS 187120, 1998). The classification of pellets in the Italian recommendation CTI-04/5 (2004) is based on the origin of the raw material, and the limit values are more restrictive for pellets made with higher quality raw materials, such as deciduous and conifer tree trunks without bark, which belong to group A.1. For pellets made of untreated herbaceous biomass (group A.2) a lower heating value is specified, and the ash content can be higher. Group A.3 has even fewer restrictions, and, the guidelines only specify values for the diameter, bulk density, and moisture content of the pellets. French quality controls establish four groups of pellets depending on their final use. Accordingly, pellets of higher quality should be used in stoves or boilers, while less exacting requirements are applicable to pellets used in large boilers and incinerators (ITEBE, 2009).



PARAMETER	(CLASSIFICATION		
	A1	A2	В	
Size (diameter and	D06:	D≤6±1 and 3.15≤L≤40		
length) (mm)	D08:	$D \leq 8\pm 1$ and $3.15 \leq L \leq 40$		
Moisture content (%)		M10: ≤10		
Ash content (%)	$A0.7 \le 0.7$	A1.5 ≤ 1.5	$A3.0 \le 3.0$	
N (%)	$N0.3 \le 0.3$	$N0.5 \le 0.5$	$N1.0 \leq 1.0$	
S (%)	$S0.03 \le 0.03$	$\mathrm{S0.03} \leq 0.03$	$S0.04 \le 0.04$	
Cl (%)	$C10.02 \leq 0.02$	$C10.02 \le 0.02$	$C10.03 \le 0.03$	
Durability	$DU97.5 \geq 97.5$	$DU97.5 \geq 97.5$	$DU96.5 \geq 96.5$	
Fines content ($\% < 3.15$	F10<10	F1.0 < 1.0	F1.0 < 1.0	
mm)	$11.0 \le 1.0$	$1.0 \le 1.0$	$\Gamma 1.0 \ge 1.0$	
Bulk density (kg/m ³)		$BD600 \ge 600$		
Lower Heating Value	016 5. 16 5<0<10	0163.163<0<10	$0.16.0 \cdot 16.0 < 0 < 10$	
(MJ/kg)	Q10.3. 10.3 <u>></u> Q <u>></u> 19	Q10.3. 10.3 <u>></u> Q <u>></u> 19	Q10.0. 10.0 <u>></u> Q <u>></u> 19	
Additives	≤2% (type	and quantity to be speci	ified)	

Table 14. Classification of parameters included in EN 14961-2 (2011) in relation to guidelines specifications for pellets applied for domestic heating use

^{*} Durability has been defined in terms of percentage of whole pellets after testing.

Given the heterogeneous nature of national pellet quality standards, the experimental technical specification EN 14961-1 (2011) for solid biofuels created a set of guidelines for pellet quality and use. It also elaborated a very specific categorization of pellets, based on the classifications in national norms. As previously mentioned, like the Italian standard, the EN 14961-1 (2011) also classifies pellets depending on the origin of the raw materials.

However, each quality parameter considered is subdivided into different categories depending on its value and therefore on its final quality (Table 13). Moreover, the annexes of this norm include recommendations concerning the final use of pellets. The most restrictive limit values are for A1 pellets used in domestic heating devices (Table 14) (EN 14961-2, 2011).

11.3. Conclusions

In the vast majority of the European countries, there are very few regulations for the manufacture and quality of pellets. All too often, pellets only come under the jurisdiction of very general biomass laws. The only European countries with official standards for compacted biomass fuels are Austria, Sweden, and Germany. A few other countries, such as France and Italy have published recommendations or guidelines. The comparative analysis of these standards and recommendations for pellets highlights significant differences in their quality parameters as well as in their guidelines. National standards that specify pellet quality requirements at an individual level have not been accepted by all stakeholders in Europe, and this has contributed to the development of national pellet types, whose characteristics depend on the country where they are manufactured. The situation will doubtlessly change when the application of the European standard for solid biofuels becomes official. A common standardization will be beneficial for Europe since it will remove an important barrier to the further development of European pellet and combustion technology market in general and will increase international pellet trade in particular.



PART 2

PRELIMINARY ANALYSIS OF QUALITY PROPERTIES AND THERMAL BEHAVIOR OF AGRICULTURAL AND FORESTRY RESIDUES⁷

⁷ The results shown in this part were presented in: (i) Zamorano M, Popov V, Rodríguez ML, García-Maraver A, 2011. A comparative study of quality properties of pelletized agricultural and forestry lopping residues. Renewable Energy; 36: 3133-3140; (ii) García-Maraver A, Terron LC, Zamorano M, Ramos-Ridao A. Thermal events during the combustion of pelletised agricultural and forestry lopping residues. Submitted to Journal of Material Cycles and Waste Management



12. Introduction

The progress of civilization and world population growth has significantly contributed to an important increase in the quantity and types of waste. The intensive consumption of resources leads to the production of large amounts of solid wastes from industrial, domestic and other activities. This poses an important threat to human health (Frosch, 1996). Moreover, mounting quantities of waste along with inappropriate treatment processes and final disposal can result in negative impacts not only on public health and the environment (Leone et al., 1979; El-Fadel et al., 1997; Sarkar et al., 2003; Diaz et al., 2005), but also on society in general because of the social alarm generated by waste accumulation, treatment, and disposal (Garrod and Willis, 1998; Joos et al., 1999; Al-Yaquot et al., 2002; Paolini et al., 2008).

Nevertheless, damage to the environment due to poor waste management can be avoided by implementing environmentally sensitive waste management techniques and an integrated management, involving minimization, composting, recycling, reuse, and waste-to-energy programs (Read et al., 1997; Wilson et al., 2006).

In the framework of integrated waste management, thermal treatment is gaining attention as landfill costs and environmental concerns soar in many countries (Sabbas et al., 2003; Tsai and Chou, 2006; Ghani et al., 2009). In recent years, pressures on global environment and energy safety have led to an increasing demand for renewable energy and the diversification of energy sources. Among these resources, biomass could play an important role since it is regarded as a renewable and environmentally safe way of providing energy for electricity, heating, and cooling (Fernandez-Llorente and Carrasco-Garcia, 2005).

Forestry and agricultural residues are produced in large amounts in several countries throughout the world (Ioannidou and Zabaniotou, 2007), and most of these residues are composed of biomass that can be used for energy generation (Matteson and Jenkins, 2007; Skoulou and Zabaniotou, 2007; Liu et al., 2008). However, because of its relatively high moisture content, irregular shape and size, and low bulk density, biomass, in most cases, is very difficult to handle, transport, store, and utilize in its original form. In consequence, densification has prompted significant interest in

developing countries in recent years as a technique for utilization of residues as an energy source (Vinterbäck, 2004; Kaliyan and Morey, 2009) and the pellets' market has increased for different raw materials (Li and Liu, 2000; Lehtikangas, 2001; Obernberger and Thek, 2004; Gilbert et al., 2009).

Pelletization of biomass is a mass and energy densification applied to materials that possess low bulk densities. This kind of densification increases the bulk density of biomass from an initial bulk density of 40-200 kg/m³ to a final bulk density of 600-800 kg/m³ (Obernberger and Thek, 2004). The process improves pellets' transportation and storage, and provides better handling and feeding of the biomass with less dust formation (Gilbert et al., 2009; Kaliyan and Morey, 2009).

The quality of the pellets is determined by the end-user's requirements considering the heating system and the handling properties (Lehtikangas, 2001). It depends on the properties of the feedstock, in terms of biomass type, moisture content and particle/fibre size, and also on the quality management of the manufacturing process, in terms of operational conditions, pelletizer type and binding agents (Li and Liu, 2000; Lehtikangas, 2001; Rhén et al., 2005; Temmerman et al., 2006; Arshadi et al., 2008; Gilbert et al., 2009). Consequently, an understanding of the physical and chemical properties of biomass residues is very important in order to determine their behavior in a combustion process (Lehtikangas, 2001; Gilbert et al., 2009; Kaliyan and Morey, 2009). For example the physical properties, such as density, determine the particle heating rate and thermal efficiency while the chemical properties, such as ultimate analyses (moisture content, volatile content, the free carbon remaining and the ash content) and proximate analysis (the composition of the biomass in wt% of carbon, hydrogen, oxygen, sulfur and nitrogen), functional groups and calorific value, govern the combustion process.

The purpose of this study was to analyze physical and chemical characteristics of pellets produced in a pelletizing plant, to identify the quality of the products generated and to increase the current knowledge of the effects of raw materials on



quality properties of pellets as well as on the thermal events occurring during combustion of the solid residue.

The relationships between various parameters and a comparison with guidelines established by some European norms have been also investigated in order to determine possible restrictions on the use of the pellets.

13. Materials and methods

13.1. The manufacturing process

All of the samples of the pellets studied in this research were produced at a privately owned industrial plant under the same manufacturing process conditions. Figure 11 summarizes the manufacturing process.

The raw material was received in a reception area where it was weighed and its moisture content measured. The throughput of the plant was 5000 kg/h. In a first phase, the size of the material was reduced to 20-40 mm in a hammer mill. The moisture content of the material was reduced until it reached a value lower than 15% in a rotary drier operated at an entrance temperature of 250 °C and an exit temperature of 85 °C. Magnets and screens were used to remove undesired particles before the dried material reached a hammer mill that reduced the particle size of the material to the range of 6-8 mm. Pellets with a diameter of 6 mm and without any additives were produced with a Mabrik (PVR 220 2 N) press operating between 1500 and 1600 kg/cm² and around 200 °C. Since the pelletizing process implied high temperatures, a cooling process was necessary to reduce the pellets' temperature to 20-25 °C before storage.

Finally, pellets were packaged in plastic bags of 15 kg each or they were stored in two pits (400,000 kg), equipped with systems to keep the pellets from temperature and moisture changes.

13.2. Materials

13.2.1. Raw materials for pellets production

Lopping residues from almond trees (AT), black poplars (BP), branches of different sizes and leaves from olive trees (OT), holm oaks (HO) and only leaves from

olive tree (OTL) were used to make up pellets tested in this research. All the raw materials were delivered from agricultural activities in Granada (South of Spain).

Fresh materials were collected and chipped at the point of origin with special equipment and transported in trucks to the pelletizing plant to be processed into pellets. Figure 12 shows the different types of pellets tested.

13.2.2. Sample preparation for thermal analysis

About 100 mg of different types of pellet manufactured at the industrial plant previously described (Figure 12) were selected and milled in order to obtain a homogenous, representative sample of each type.

13.3. European standards related to quality of pellets

Parameters that determine the quality of pellets and limiting values defined in Austrian (Ö NORMM 7135, 2003), Swedish (SS 187120, 1998) and German norms (DIN 51731, 1996; DIN 15270; 2007), Italian (CIT-R04/05, 2004) and French (ITEBE, 2009) recommendations and guidelines of European Standard Committee (EN 14961, 2011) for densified biomass fuels have been considered in this research. They have been summarized by García-Maraver et al. (2011).

13.4. Laboratory procedures

13.4.1. Particle density

The particle density of pellets was determined according to CEN/TS 15150 (2005). The calculated particle density represents the average value of these measurements.

13.4.2. Bulk density

The volume and the weight of a pellet sample have to be measured in order to be able to determine the bulk density according to EN 15103 (2009). The average bulk density was calculated from three measurement series per sample.



Figure 12. Pellets tested in the study

13.4.3. Moisture content

Three samples from each raw material were milled to determine the moisture content in an oven-drier at 105 ± 2 °C to a constant weight and according to EN 14774-1 (2007).

13.4.4. Crushing resistance or hardness

Three randomly chosen pellets of each material were chosen for the calculation of crushing resistance or hardness (compressive resistance) defined as the maximum crushing load a pellet can withstand before cracking or breaking.

Compressive resistance and tensile strength of the densified products were determined by diametrical compression test using a KAHL tester working between 0 and 100 kg.

13.4.5. Number of particles of pellets

Three random samples of 100 g of each material were used to determine the number of pieces per 100 g of pellets.

13.4.6. Size

Three randomly selected pellets from each raw material were used to determine the length and the diameter of the pellets (mm) using a digital vernier caliper CLD-150.

The size distribution of the pellets (in percentage) was studied by screening the three random samples of 100 g of each type of pellets through 16, 10, 8, 4 and 2.5 mm circular openings.

13.4.7. Chemical composition

Three samples from each raw material were milled and mixed to obtain a homogenous sample of about 7 mg to determine percentage of carbon, nitrogen, sulfur and hydrogen according to EN 15104 (2011). A FISONS CARLO ERBA EA 1108 CHNSO detector type was used.

13.4.8. Ash content

Three samples of pellets were chosen and the ash content was measured by measuring loss of ignition at 550 °C according to EN 14775 (2009).

13.4.9. Heating value

Three samples from each raw material were milled to determine their heating value by using a bomb calorimeter IKA C 2000 and according to EN 14918 (2009).

13.4.10. Thermogravimetric analysis (TGA/DTG)

A thermogravimetric (TGA) analysis of solid fuel was performed using a SHIMADZU TGA-50H thermo analyzer in an oxidative atmosphere. The size of the particles was reduced to less than 0.5mm (500µm) to ensure the heat transfer rate within the kinetic regime of decomposition (Colomba, 1998; Varhegyi et al., 1997; Rath et al., 2002), and the initial mass of the sample was kept between 6-8 mg to avoid any possible effect on the mass and heat transfer during the process of biomass decomposition (Blasi et al., 2001). The samples were heated from 30 to 950°C at a heating rate of 20 K/min of air with a constant airflow rate of 100 ml/min through the sample chamber to maintain an oxidative atmosphere for the thermal decomposition of the particles.

13.4.11. Differential Scanning Calorimetry (DSC)

A SHIMADZU DSC-50 tester was used to perform Differential Scanning Calorimetric (DSC) measurements. Samples of about 4 mg were heated up to 470°C at a constant heating rate of 10°C/min (DSC) with a constant airflow rate of 100 ml/min through the sample chamber.

14. Results and discussions

14.1. Quality properties of pellets

14.1.1. Characteristics of pellets

The results of the analysis of densified biomass tested have been summarized in Table 15, Table 17 and Table 18, and analyzed and discussed in the subsections below.

a) Particle density

The variation in the particle density of pellets was between 1083 and 1259 kg/m³ for OTL and OT respectively (Table 15). Values obtained were similar to other wood pellets given in the literature (Li and Liu, 2000; Lehtikangas, 2001; Rhén et al., 2005; Kaliyan and Morey, 2009), except for the samples produced from OTL, which had shown lower values. The energy density of the pellets defines the efficiency of energy storage, transport and energy production (Vinterbäck, 2004; Gilbert et al., 2009; Kaliyan and Morey, 2009) as well as the combustion behavior because dense particles show a longer burnout time (Obernberger and Thek, 2004). Austrian and German standards, as well as French recommendations for pellets used in stoves and boilers have included guideline values for these parameters (Garcia-Maraver et al., 2011).

Sample	Bulk d (kg/	ensity m ³)	Dens (kg/1	sity m ³)	Moi (9	sture %)	Crus resist (k	hing tance g)	Compr resist (k	ression tance g)
	Avg	SD	Avg	SD	Avg	SD	Avg	SD	Avg	SD
AT	575.28	3.23	1187.18	57.48	4.78	1.56	23.33	0.58	21.33	0.76
BP	523.38	12.26	1151.06	43.62	5.78	0.68	22.83	0.29	17.50	3.77
НО	558.52	8.23	1124.48	56.77	6.50	2.01	23.33	0.29	12.50	1.50
OT	582.53	12.43	1259.22	20.43	5.37	0.53	22.50	0.00	21.33	1.04
OTL	481.56	1.72	1083.47	49.95	6.57	0.47	4.00	0.87	8.33	2.36

Table 15. Physical parameters of samples of pellets used in the study

Some samples of pellets from AT fulfilled the limit established by the German standard and by the French recommendations for pellets used in stoves and boilers; no sample of AT pellets fulfilled guidelines established by the Austrian norm (Table 16). All samples of BP and HO pellets showed values that fulfilled the limit established by both German standards (Table 16); furthermore, all samples would not have density value following guidelines of Austrian standard and French recommendations. Density

values of pellets from OT only fulfilled guidelines of French recommendations for pellets used in stoves and boilers (Table 16). Finally samples of pellets from OTL fulfilled limits of both German standards (Table 16) and only some samples fulfilled the limit established by the Austrian norm; French recommendations were not fulfilled.

b) Bulk density

This parameter varied in a range between 482 and 583 kg/m³ (Table 15) corresponding to pellets made from waste from OTL and OT respectively. Values obtained in this research were always lower than bulk density of pellets from sawdust, bark and logging residues chipped and stored for several months; but they were similar to bulk density of pellets from fresh logging residues (Vinterbäck, 2004), other wood pellets (Obernberger and Thek, 2004) or pellets from some herbaceous crops (Gilbert et al., 2009). The bulk density is of importance with regard to pellet storage and transport (Vinterbäck, 2004; Obernberger and Thek, 2004; Kaliyan and Morey, 2009); thus some norms have included guideline values for this parameter to increase the efficiency and decrease the costs of storage and transport.

Swedish and Italian standards, French recommendations and EN 14961 (2011) include guidelines related to the bulk density of pellets. The comparison between bulk density of pellets analyzed and guiding value of these standards (Table 16) showed different grades of fulfillment. Pellets produced from OTL did not fulfill any of the established limits for this parameter because values were always lower than the limit; pellets from BP lopping waste only fulfilled guidelines established by Swedish norm in groups 2 and 3 and EN 14961 (2011); OT samples satisfied several guidelines, including Swedish (G.2 and G.3) and Italian (A.3) standards. The average value of bulk density also fulfilled limits established in French recommendations for large heaters and incinerators; samples from AT, HO and OT fulfilled several limits established by Swedish and Italian standards (Garcia-Maraver et al., 2011), depending on type of pellets; French recommendations have been only fulfilled by some samples from OT and large heaters and incinerators.



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	OTL	~	•			•	~	•	•	•	•	•	•	•	*
tn	AT	x	x	x	x	x	х	x	x		x	x	•	•	х
ətu	BP	x	x	x	x	х	x	х	x	•	x	x	•	•	х
00	ЮН	x	x	x	x	x	х	х	х	•	x	x	•	•	х
ųs	oT	x	x	x	x	x	x	x	x	•	x	x	•	•	х
V	OTL	x	x	x	x	х	х	х	x	•	x	x	•	•	х
ուղ	AT	~	7	~	~	~	*	٨	*	•	×	*	*	*	*
BA	BP	~	7	7	7	~	~	2	2	•	۲	*	~	*	*
9ui	HO	7	7	7	7	7	7	7	7		7	~	2	7	~
teat I	5	7	7	7	7	~	7	~	~		7	7	7	7	7
Η	OT,	7	7	7	7	7	7	7	7	,	7	7	7	7	7

Table 16. Guidelines fulfillment corresponding to norms considered in the research (x: guideline not fulfilled; $\sqrt{}$ guideline fulfilled; \pm guideline fulfilled only by some samples; - guideline not included in the norm)

c) Moisture content

The water content of a pellet has an influence on the net calorific value, the combustion efficiency, the temperature of combustion and the equilibrium moisture content with the ambient moisture content, affecting storage conditions (Lehtikangas, 2000). Table 15 shows the average value and standard deviation of moisture content of samples tested which were between 4.78 and 6.57% for AT and OTL respectively. These values were similar to those of other wood pellets (Obernberger and Thek, 2004) although lower than the moisture content of pellets from fresh sawdust, logging and bark, with values between 7.9 and 21.3% (Lehtikangas, 2001).

All samples tested had moisture contents according to the guideline values of standards considered in this study (Table 16).

d) Crushing resistance or hardness

This parameter has not been included in the standards considered in the study, however some literature includes a compressive resistance test to simulate the compressive stress due to the weight of the pellets on top on the lower pellets during storage in bins or silos as well as crushing of pellets in a screw conveyor (Li and Liu, 2000; Gilbert et al., 2009). The compressive resistance test provides a quick measure of the quality of pellets as soon as the pellets are produced from the pellet mill and aids in adjusting the pelleting process to improve the pellet quality.

Pellets tested showed similar crushing resistance values except for the pellets from OTL, which showed significantly lower values. Values of tensile strength showed important differences between different pellets tested (Table 15). It was detected that higher moisture content, lower densities and lower length values could reduce the tensile strength of the pellets (Figure 13 and Figure 14). This could be explained by the fact that the tensile strength is related to the adhesion forces between particles at all contact points in the agglomerate (Pietsch, 2002).





Figure 13. Relationship between tensile strength, particle density and moisture content



Figure 14. Relationship between tensile strength and particle length

e) Pellet number

Table 17 shows the average value and standard deviation of the number of pellets in a 100 g sample of pellets. Samples of pellets from OTL and HO had higher values while pellets from OT showed lower pellet number density.

Sample	Len (mi	gth m)	Diar (m	neter m)	Ratio	o L/D	Nu per	mber 100 g	Particle size distribution (%) > 16/16-10/10-7/7-4/4-2.5/<2.5 mm
	Avg	SD	Avg	SD	Avg	SD	Avg	SD	
AT	21.27	2.40	5.87	0.06	3.62	0.39	229	10.02	0.00/7.41/58.41/27.29/3.17/3.72
BP	20.99	7.63	5.83	0.23	3.64	1.47	273	18.23	0.00/4.25/67.38/26.88/1.27/0.21
HO	13.22	1.11	6.08	0.08	2.18	0.19	327	12.14	0.00/0.53/85.10/10.04/3.15/1.18
OT	20.36	2.01	5.94	0.02	3.43	0.35	176	9.64	0.22/1.81/93.69/3.71/ 0.04/0.01
OTL	10.65	2.09	6.04	0.11	1.76	0.31	322	36.06	0.00/1.76/76.22/13.23/5.39/3.39

Table 17. Physical parameters of samples of pellets in the study



Figure 15. Relationship between number of particles per 100 g, particle density and tensile strength

These results could be related to the tensile strength of samples and density of particles (Figure 15). Pellets with lower values of tensile strength and density of particles were broken down easily during handling, transportation, and storage increasing the number of particles (Lehtikangas, 2000; Lehtikangas, 2001; Kaliyan and Morey, 2009).

f) Size

Visual observations of the pellets tested showed differences in size (Figure 12). These observations were confirmed by the results of the study of dimensions of pellets



tested (Table 17). The main part of the pellet samples obtained had a diameter of about 6 mm because all the samples were produced in the same industrial plant and under the same conditions; however, some differences had been detected in relation to their length. Pellets from OTL and HO showed lower length values than the rest of samples.

Some relationship between length, density of particle and moisture content had been detected. Pellets more densified and with lower moisture content broke down less than pellets with lower particle density values (Figure 16), confirming the conclusions explained in the subsection about the number of pieces, and they should be considered in relation to pellet storage and transport (Lehtikangas, 2000).



Figure 16. Relationship between length, particle density and moisture content

The dimensions of the pellets, both diameter and length, are important factors with respect to combustion (Lehtikangas, 2001; Obernberger and Thek, 2004). As a consequence all the standards considered in this study include a guideline value about pellets' sizes or the relationship length/diameter (Garcia-Maraver et al., 2011).

In relation to the diameter of the pellets, all the samples fulfilled requirements except for the limit established by the Italian norm for group A.3 and French recommendations for pellets used in boilers, large boilers and incinerators (Table 16).

In relation to the length of pellets only French recommendation includes a limit value not related to the value of the diameter of pieces of pellets. The value of these parameters of pellets made from AT, BP and OT waste fulfilled the requirements established by French recommendation except for the application in boilers because the length of the pellets was higher than that in the guidelines (Table 16). Samples of HO only fulfilled requirements for stove and boiler; OTL pellets did not fulfill them.



Figure 17. Particle size distribution of pellets

The analysis of values of the ratio length/diameter of pellets tested (Table 17) showed that samples of OTL pellets had the lowest ratio and samples from BP had the highest ratio. However, all samples fulfilled guidance values established in the standards.

Finally, the particle size distribution of pellets' length was also analyzed (Figure 17). Results showed that the higher percentage of particles were between 4 and 10 mm for all the pellets although percentages depended on different materials. Samples from OTL had the higher percentage of particles lower than 2.5 mm and OT pellets the lowest one. These results are related to the conclusions obtained regarding the number of particles, density, length and crushing resistance or hardness.

g) Chemical composition

The percentage of sulfur, nitrogen, carbon and hydrogen content in different types of pellets was determined (Table 18). Values obtained were similar to the values



reported in the literature (Obernberger and Thek, 2004) except for OT and OTL pellets which showed a higher percentage of nitrogen than that reported in the literature.

Sample	Ch	emical c (%	ompositi 6)	ion	Ash co (%	ontent	Higher h value (M	eating IJ/kg)
	С	Ν	S	Η	Avg	SD	Avg	SD
AT	44.29	0.78	0.00	8	3.35	0.26	18.01	0.07
BP	46.15	0.29	0.00	7.19	2.37	0.69	18.79	0.02
HO	47.02	0.34	0.00	7.62	3.32	0.25	18.82	0.05
OT	44.84	0.95	0.00	7.64	4.79	0.11	18.47	0.03
OTL	42.80	1.09	0.00	7.05	12.34	0.55	18.68	0.06

Table 18. Chemical parameters of samples of pellets used in the study

Norms considered in this research include different chemical parameters. All of them include guidelines for nitrogen and sulfur contents except for the Swedish norm. Requirements established for nitrogen were fulfilled only by BP pellets because in the rest of samples the values obtained in this study were higher than the limits established (<0.3%). Limits established for sulfur content were fulfilled by all of the samples (Table 16).

The manufacturing process of the pellets did not use additives so requirements established in relation to this parameter were fulfilled in all cases.

h) Ash content

Table 18 shows some differences in relation to the ash content of different pellets tested. BP pellets were the ones that showed lower value of ash upon completion of the combustion process; samples from OTL and OT generated the highest percentage of ash. It was noticeable that all values exceeded the usual ash content of softwood (0.4-0.8%), hardwood (1-1.3%) (Obernberger and Thek, 2004) and sawdust (lower than 0.5%) (Lehtikangas, 2001); however, values were similar to the ash content from fresh bark (2.65%), fresh logging residues (2.36%), switchgrass (3.49%), bark (3%) and logging residues stored for months (3.81%) (Lehtikangas, 2001; Öhman et al., 2004a) for AT, BP and HO samples and to values of pellets from torrefied switchgrass (4.8%), wheat straw (5.28%) (Obernberger and Thek, 2004), and olive husk (between 4.6 and 5.8%) (Arvelakis and Koukios, 2002; Ollero et al., 2003) for OT pellets. Finally, the ash content of OTL pellets was higher than the rest of the materials tested including values given in the literature.

Increasing the ash content lowers the heating value and implies the risk of sintering and dust emissions (Ollero et al., 2003; Obernberger and Thek, 2004; Öhman et al., 2004b). In order to provide operation comfort for end users in the residential heating sector, all of the European standards and recommendations included in this study provide guiding limits related to ash content in the case of the residential heating sector (Garcia-Maraver et al., 2011). If the pellets are used in medium and large-scale applications (A.3 group of the Italian norm and French recommendations for big installations and incinerators), due to the higher robustness as well as to the more sophisticated combustion and process control technology, pellets' ash content could have higher values (Obernberger and Thek, 2004) and guidelines for this parameter have not been included.

The analysis of results obtained in the research showed that no samples fulfilled guidelines established in the norms (Table 16). The higher values of ash contents of samples could possibly be an indication that the producers had not exclusively used chemically untreated raw material or had problems with mineral contamination of the raw material used during storage and handling (Lehtikangas, 2001; Gilbert et al., 2009). Consequently, both the processes of collecting the raw material and of drying the fuels should be conducter very carefully in order to avoid mineral contamination and therefore minimize the ash content.

i) Heating value

Table 18 shows the average and standard deviation of upper heating values of different types of pellets tested and no differences between them were detected. The gross calorific values for the different samples of pellets were within the typical ranges found in the literature (Lehtikangas, 2001; Öhman et al., 2004a; Celma et al., 2007; Gilbert et al., 2009).

This parameter is included in Austrian, Swedish, German and Italian standards, and even in the experimental European Standard specifications for pellets applied for domestic uses. Results obtained for all the samples tested were within the specified limit (Table 16).



14.1.2. Comparison of the characteristics of pellets and fulfillment of the European standards

Table 19 summarizes the percentage of parameters that each type of pellet has fulfilled for each norm, considering only requirements included in this study as well as average value of parameters.

		PEF	RCENT	AGE O	F SAM	PLES	AVEDACE
NC	ORM			(%)			AVERAUE
		AT	BP	HO	OT	OTL	(%)
Ö NORM		66.7	72.2	77.8	66.7	72.2	71.1
	G.1	66.7	66.7	66.7	66.7	66.7	66.7
SS 187120	G.2	83.3	83.3	83.3	83.3	66.7	80.0
	G.3	83.3	83.3	83.3	83.3	66.7	80.0
DIN 51731		68.8	75.0	87.5	62.5	75.0	73.8
DIN PLUS		72.2	77.8	88.9	66.7	77.8	76.7
CTI R04/05	A.1	62.5	62.5	75.0	62.5	62.5	65.0
	A.2	62.5	62.5	75.0	62.5	62.5	65.0
	A.3	75.0	50.0	75.0	75.0	50.0	65.0
ITEBE	Stove	61.1	55.6	55.6	66.7	50.0	57.8
	Boiler	38.9	33.3	55.6	44.4	38.9	42.2
	Large boiler	50.0	50.0	25.0	50.0	25.0	40.0
	Incinerator	50.0	50.0	25.0	50.0	25.0	40.0
EN 14961-2 (A1)		85.7	85.7	85.7	85.7	85.7	85.7
Average (%)		66.2	64.9	68.5	66.1	58.9	

Table 19. Percentage of samples fulfilling guidelines established by norms considered in the study

It is possible to conclude that samples of pellets made from lopping waste from BP fulfilled the higher percentage of parameters (70.3%) followed by samples of pellets from OT (66.1%); finally OTL pellets fulfilled the lowest percentage of parameters.

According to the norms considered in this study, Table 19 shows that EN 14961-2 (2011) published by the European Standard Committee CEN/TC 335, containing the specifications for pellets applied for domestic use, is the norm with the highest percentage of parameters satisfied (85.7%) by different pellets. Otherwise French recommendations (ITEBE, 2009), for large boilers and incinerators, have shown the lower percentage of guidelines fulfilled (40%).

Table 20 shows that some requirements have been fulfilled by all the samples, moisture, S content, additives content and heating value. The requirements for diameter of pellets have been fulfilled by 71.4% and length by 88.6% of the samples. The limits

established on ash content have not been fulfilled by any of the samples analyzed. In addition requirements about particle density (46%), bulk density (24%) and N content (20%) have been fulfilled by less than half of the samples.

DARAMETER	PERCENTAGE OF
	SAMPLES (%)
Particle density	46.0
Bulk density	24.0
Moisture	100.0
Diameter	71.4
Length or ratio L/D	88.6
N content	20.0
S content	100.0
Additives content	100.0
Ash content	0
Heating value	100

Table 20. Per	centage of samples fulfilling	ng guidelines	established for	r parameters
	included in	n the study		

14.2. Thermal behavior

The thermal decomposition of lignocellulosic materials involves a number of chemical reactions as well as physical stages such as heat and mass transfer (Bilbao et al., 1997; Zheng and Kozinkski, 2000).

Wood is a complex material, composed mainly of cellulose (40-60 wt%), hemicelluloses (20-40 wt%), and lignin (10-25 wt%), in addition to extractives (terpens, tannins, fatty acids, resins, etc), water, and mineral matter (McKendry, 2002). Biomass composition varies from species to species, and within the same species, it varies with habitat, age, and location in the tree (trunk, branches, top, leaves and roots) (Barton, 1981; Pereira and Sardinha, 1984; Pereira, 1988). The mass loss of biomass is controlled by its main components (i.e. hemicelluloses, cellulose and lignin), which decompose at temperature ranges of 225-325°C, 305-375°C, and 250-500°C, respectively (Byrne and Nagle, 1991; Bilbao et al., 1997; Orfao, 1999; Liu, 2002; Safi et al., 2004; Fang et al, 2006; Prins et al., 2006; Shen et al., 2009). In general, mass loss kinetics may change as the temperature increases (Liu et al., 2002).



When thermal decomposition takes place in an air atmosphere, the presence of oxygen is a source of added complication. More specifically, it enhances the degradation of the material at low temperatures (Shafizadeh and Bradbury, 1979) and causes the appearance of gas-phase reactions between the volatile compounds released and oxygen. In fact, when the temperature is sufficiently high, oxygen is likely to promote the combustion of the char residue generated in the early stages of the degradation of the solid (Shafizadeh and Bradbury, 1979), which means that combustion and pyrolysis may coexist (Liu et al., 2002). Since the kinetics of this thermal decomposition is more closely related to the combustion process, this research contributes to the development of efficient biomass combustion applications. Knowledge of combustion characteristics is the basis for this and is thus crucial for a better understanding of the thermal chemical conversion of biomass.

Figure 18 and Figure 19 show the typical thermograms for the different types of pellets tested under the experimental conditions described in the previous section. They represent weight loss (TGA curve) and the derivative of the TGA curves (DTG), respectively. Figure 20 shows the result of the DSC experiment, which is a heat flux curve in contrast to the temperature for each pellet tested.



Figure 18. TG curves of several types of pellets heated up to 950°C under a heating rate of 20 °C/min and a constant airflow rate of 100 ml/min through the sample chamber



Figure 19. The DTG curves of several types of pellets heated up to 950 °C under a heating rate of 20 °C/min and a constant airflow rate of 100 ml/min through the sample chamber

The results of this study show the presence of three decomposition phases in all cases. These phases were similar to those described in other wood combustion studies (Byrne and Nagle, 1991; Tsujiyama and Miyamori, 2000; Grioui et al., 2006; Shen et al., 2009). They were the following: (i) drying phase and elimination of certain volatile compounds; (ii) roasting phase; (iii) solid residue decomposition phase.

The nature of a TGA curve in combination with the corresponding DTG peaks indicates the number of stages of the thermal degradation. During thermal decomposition in air, weight loss occurs continuously until the weight becomes almost constant.

14.2.1. Drying phase and elimination of certain volatile compounds

The first decomposition phase corresponded to a slow initial weight loss because of moisture content and the elimination of certain volatile compounds (Marcilla et al., 2009). Weight loss (Figure 18) showed a slight decrease in the curve, whereas DSC curves (Figure 20) corresponded in this phase to a negative peak because drying and



elimination of volatile compounds is an endothermic reaction, and in consequence, energy was consumed.



Figure 20. The DSC curves of several types of pellets heated up to 470 °C under a heating rate of 10 °C/min and a constant airflow rate of 100 ml/min through the sample chamber

This phase lasted until the temperature reached 200 °C. The results obtained showed that weight loss varied slightly, depending on the type of pellet (Table 18). For samples from olive trees (OT), olive tree leaves (OTL) and holm oak trees (HO), the weight loss rate was greater than 3.5%. In the case of pellets from black poplar (BP) and almond trees (AT), the values were lower than 3%. Since there were no significant differences in the chemical composition of the samples (Table 18), this result could be explained by the fact that the moisture content of the samples was higher in the first cases.

14.2.2. Roasting phase

The second phase showed a faster weight loss when the samples were roasted. The results showed that the decomposition occurred in the 200 - 500°C range. In this phase, a maximum weight loss occurred for all the samples tested. AT and BP samples showed the highest percentage of weight loss, followed by HO and OT samples. Samples from OTL showed the lowest value (Table 21). These results could be related to the hemicellulose, cellulose, and lignin contents of the raw material tested (Yang et al., 2007; Shen et al., 2009).

Dallata		Weight loss (%)	- Ach content $(0/)$
reliets	1 st phase	2 nd phase	3 rd phase	Asir content (%)
AT	2.9	92.6	1	3.5
BP	1.6	94.5	0	3.9
НО	3.6	89.4	2.5	4.5
OT	4.3	82.5	7.3	5.9
OTL	37	78.4	46	13.3

Table 21. Percentage of weight loss per phase and final ash content

The roasting phase is a complex process, involving various steps that are reflected in the overlapping peaks in the DTG curve (Figure 19). The thermograms obtained for each type of pellet were analyzed to determine the relevant combustion parameters, such as peak temperatures (PT) and maximum weight loss rate (Rmax, %min-1), regarded as the reactivity index. This was determined on the basis of the DTG plot, and corresponded to the maximum rate at the DTG-peak temperature. The TGA–DTG results are shown in Table 22. The combustion profile of pellets in this phase showed two sharp double peaks in the DTG curve at a similar temperature range. Such double peaks (DTG) corresponded to mass loss phenomena.

Sampla	DTG-	peak tempe	erature (PT	Г), °С	R _{max1}	R _{max 2}	R _{max 3}	R _{max 4}
Sample -	1	2	3	4	%/min	%/min	%/min	%/min
AT	371.6	471.6	-	-	14.17	17.85	-	-
BP	351.4	451.4	-	-	16.77	9.26	-	-
HP	371.2	451.3	-	-	13.98	9.24	-	-
OT	352	492	711.7	-	9.3	9.35	0.70	-
OTL	351.8	491.8	671.5	711.5	11.19	7.54	0.81	1.10

Table 22. Combustion parameters (TG-DTG)

Weight loss at 200°C is indicative of polymer decomposition when the hemicellulose component begins to break down (Prins et al., 2006). Consequently, the first peak corresponds to the mass loss phenomena caused by the combination of the total decomposition of hemicellulose and cellulose, and the partial decomposition of lignin (Bilbao et al., 1997; Orfao, 1999; Liu, 2002; Safi et al., 2004).





Figure 21. The DTG curves of holm oak and leaves of olive tree samples with detail of a "shoulder peak" in the first step of roasting phase

The first stages of the DTG curves of the samples showed certain differences (Figure 19), and reactivity index values varied from 9.3 to 16.77 %/min at a temperature range of 351.4 – 371.6°C (Table 22). This could be caused by the different constituent fractions of the five samples (Shen et al., 2009). Figure 21 shows a detailed representation of the DTG curves for samples of HO and OTL. As can be observed, a "shoulder peak" occurs in the first stage of the DTG curve at around 271°C for HO and 311°C for OTL, which could be attributed to the total decomposition of hemicellulose (Bilbao et al., 1997; Orfao, 1999; Liu, 2002; Safi et al., 2004; Fang et al., 2006). However, there is no "shoulder peak" in the DTG curve of the other samples. This could possibly be due to their lower content of hemicellulose, which caused them to react slower than the other samples (Shen et al., 2009).

Another explanation for the "non-shoulder peak" DTG curve of these samples could be that their hemicellulose contained components which postponed the thermal decomposition of the hemicellulose. In that case, the hemicellulose decomposition peak was concealed by that of the cellulose, whereas the temperature demarcation for the hemicellulose decomposition overlapped with that of the cellulose. The cellulose dominated the first stage of the thermal decomposition of the biomass, and as a result, the "shoulder peak" of the DTG curve disappeared.

When temperatures exceeded 400°C, the rate of change in weight loss again increased, as the cellulose had largely decomposed while the lignin components continued to break down (Byrne and Nagle, 1991). It reached a maximum at 450–490°C, depending on the pellet type. This corresponded to the second peak in the DTG curves. The second stage of the DTG curves was noticeably different for the five samples (Figure 19), and the reactivity index values varied from 7.54 to 17.85%/min at a temperature range of $451.3 - 491.8^{\circ}$ C (Table 22). This could be the result of the different constituent fractions of the five samples, especially their lignin content.

Lignin is the major contributor to the production of char residue, and leads to different processes of char formation, which directly affect the thermal decomposition of the biomass in the second stage in an oxidative atmosphere (Shen et al., 2009). It is probable that samples from OT and OTL have a higher percentage of lignin than the other samples.

In this phase, DSC curves (Figure 20) corresponded to positive peaks because hemicelluose, cellulose, and lignin decomposition are exothermic reactions, and in consequence, energy was generated. The curve obtained for each pellet type was analyzed to determine the relevant combustion parameters, such as DSC-peak temperatures (PT), onset temperature for combustion, and burnout temperature (BOT). In this case, the DSC experiment was performed by using a tester that heated samples up to 470°C. At this temperature, samples were not completely roasted. Consequently, the curves are incomplete, and thus it was not possible to determine all the parameters of DSC combustion.

Despite the fact that DSC curves were not complete, it was possible to identify two DSC-peak temperatures (PT), corresponding to peak energy production because of the maximum hemicellulose, cellulose, and lignin decomposition rate (Tsujiyama and



Miyamori, 2000). The curves indicated that the decomposition of olive tree leave samples was slower than the rest of samples (Figure 20).

The DSC curve can be used to determine enthalpies of transitions. This is done by integrating the peak corresponding to a given transition. The enthalpy of transition can thus be expressed in an equation that includes the calorimetric constant, which varies from instrument to instrument, and also in consonance with the area under the curve. Enthalpy of transition is related to the heating value of the samples.

Since the tester used for the DSC experiment heated samples up to 470°C, they were not completely roasted, and in consequence, the curves were incomplete. It was thus impossible to determine the area under the curve. As a result, the heating value had to be determined calorimetrically, as described in the previous section. The values summarized in Table 18 show that there were no significant differences among the heating values obtained for all the samples tested.

14.2.3. Solid residue decomposition phase

After reaching the second temperature peak, which corresponded to the roasting phase, the weight loss rate decreased to very low values. However, samples from OT and OTL showed a higher percentage of weight loss than the rest of the samples. Both samples showed peak temperatures in this phase although reactivity indexes were very low (near 1%). The possibly higher content of lignin in these samples decomposed at a much wider temperature range and produced more char residue during the second stage of combustion (Shen et al., 2009). Further heat treatment to 950°C yielded ash content. The last part of the TGA and DTG curves showed no differences in weight loss, which seems to indicate that all chemical reactions were completed. This indicates that an exclusively ash residue was obtained that was free of any carbon content. Consequently, the TGA curve also provided information about residual mass or ash content in the pellets tested.

Figure 19 and Table 21 reflect the differences in the final residual mass of samples in regards to ash content. AT pellets showed the lowest residual mass value at the end of the burning process, whereas pellets from OTL generated the highest ash values. These values clearly exceed the usual ash content of wood, the values of which

are 0.4–0.8% for softwood and 1.0–1.3% for hardwood (Obernberger and Thek, 2004). Sawdust has a value lower than 0.5%; fresh bark has a value of 2.65%; and fresh logging residue, a value of 2.36% (Lehtikangas, 2001; Öhman et al., 2004a). The high values obtained in this study are probably due to the fact that forestry and agricultural lopping residues also include leaves and small tree branches, not only woody portions. These percentages would be higher than the limit established for some applications of biomass, for example, domestic heating.

However, the ash content of AT, BP and HO pellets was similar to that of pellets made from other raw materials, such as raw switchgrass (3.49%), torrefied switchgrass (4.8%), bark (3%), and logging residues that had been stored for months (3.81%) (Lehtikangas, 2001; Öhman et al., 2004a). The ash content of OT pellets had higher values than other types of wood pellet, but was similar to that of pellets made from wheat straw (5.28%) (Gilbert et al., 2009), olive husks (4.6–5.8%) (Arvelakis and Koukios, 2002; Ollero et al., 2003), and grape waste (Celma et al., 2001). Finally the ash content of pellets from OTL was clearly higher than that of the rest of the materials tested or the values obtained in other studies.

The very high ash content of certain samples could be explained by the high lignin content of OT and OTL samples, as explained in the previous subsection. However, another possibility is that the manufacturers may not have only used chemically untreated raw material. There may also have been problems with the mineral contamination of the raw materials. For example, sand or soil might also have been dragged in during the storage and manipulation of the raw material at the collection point by the equipment used for this purpose (Öhman et al., 2004a). Lehtikangas (2001) concluded that the ash content of pellets was consistently higher in pellets made from chips stored for six months. Furthermore, stored chips were usually found to contain a greater amount of relatively large mineral particles. The fact that pellets had fewer mineral particles than those found in raw material may be due to pellet production, which includes a screening process.

High ash content lowers the heating value and means a higher risk of sintering. It also negatively affects milling and pelletizing equipment (Lehtikangas, 2001).



Consequently, both raw materials and drying fuels/processes should be carefully treated to avoid mineral contamination in order to minimize ash content, and thus meet current quality standards for pellets. Standards for pellets used in domestic heating are stricter than standards for pellets used in industrial processes. The collection of raw material should be optimized to reduce contamination in pellets.

15. Conclusions

15.1. Quality properties of pellets

Physical and chemical characteristics of samples of pellets, produced in a pelletizing plant with different raw materials, have been analyzed and compared to guidelines established by seven European norms.

The use of different type of raw materials from agricultural and forestry lopping residues results in different physical and chemical properties of pellets imposing restrictions on application of the pellets. It was concluded that samples of pellets made from lopping waste from holm oak have higher quality, followed by samples of pellets from black poplar, almond tree and olive tree; while pellets made from leaves of olive tree have shown the lowest quality.

Some relationships have been observed between some of the parameters analyzed. These relationships could be explained by the fact that tensile strength is related to the adhesion forces between particles at all contact points in the agglomerate and in consequence to density of particles. Density of particles could be also affected by other parameters, for example moisture content. Pellets with lower density and tensile strength were broken down easily during handling, transportation, and storage, reducing length of pellets and producing higher dust emissions.

No pellets tested fulfilled all parameters from guidelines established in the norms considered in the research. The higher ash content is a common problem in the samples analyzed, although it is higher in pellets made from olive tree waste.

More research is needed on the effects of raw material characteristics, seasonal variations, collection and storage of raw material as well as manufacturing process in order to design a production line which would produce pellets with the required quality established by norms while considering the specific application of pellets.

15.2. Thermal behavior

Thermoanalytical techniques, such as thermogravimetry (TGA) and differential scanning calorimetry (DSC), were used to analyze combustion differences in pellet samples made from agricultural and forestry lopping residues. The results were analyzed both experimentally and theoretically, and the following conclusions were derived:

The results of the TGA experiment confirmed those obtained in previous studies, which stated that the process of thermal decomposition of wood in an oxidative atmosphere takes place in three stages: (i) the drying phase and elimination of certain volatile compounds; (ii) the roasting phase; (iii) the solid residue decomposition phase.

The roasting phase in this study generally showed a faster weight loss rate though certain differences were detected in the samples tested. These differences could be related to the hemicellulose, cellulose, and lignin content of the raw material used to manufacture the pellets.

The very high ash content of olive tree and olive tree leaf samples could be explained because of their higher lignin content. However, it could also be an indication of the mineral contamination of this material as a consequence of the collection or storage systems.

Although no differences in heating value were detected in the samples tested, it is probable that pellets made from olive trees and olive tree leaves will lead to more maintenance problems in domestic heating than pellets made from the other materials tested. Thus, it would be necessary to improve the collection, storage, and production process to optimize the quality of pellets.

Evidently, the presence of olive tree leaves could reduce pellet quality because of the higher ash production in the combustion process. Higher ash content could lead to problems in combustion processes, such as domestic heating, which require pellets of higher quality than those used in industrial processes.

Finally, all samples tested, with the exception of olive tree leaves, were found to be suitable for use in combustion processes, such as domestic heating. However, further


studies are necessary to ascertain whether these pellets meet the high quality standards for pellets currently in force in other countries.

PART 3

CHARACTERIZATION OF RESIDUAL BIOMASS FROM OLIVE TREES



Optimization of the pelletization process of agricultural wastes originating from olive farms for their application in domestic boilers

Ángela García Maraver

As it was described in the previous section, the generation of agricultural residues from the olive sector in the Mediterranean region is an important source of residual biomass highly suitable for thermal energy generation. This biomass comes from olive groves and olive oil production plants that generate byproducts with high energy content.

Since the properties of biomass are dependent on a wide range of factors, the focus of this part of the current research was to analyze all of its forms (leaves, prunings and wood) separately in order to better understand their thermal behavior and assure the quality of the final product.

The determination of the ash content for each type of residue from olive tree pruning indicated that the leaves from olive trees were responsible for the high ash content of this biomass. Since ash content could be explained by the presence of inorganic species from the soil (Öhman et al., 2004a), various cleaning methods were used to study the effect of the mineral contamination from leaves on the final ash content. The ashes were also analyzed with a microscope to ascertain their composition.

On the other hand and due to the fact that chemical differences in the structural components of the biomass (cellulose, hemicellulose, and lignin) have a direct impact on its chemical reactivity, the influence of the components of the raw material from olive trees (leaves, pruning residues, and wood) in the combustion behavior of this biomass was analyzed. Accordingly, the study used a thermogravimetric analyzer to monitor the different states and complex transitions that occurred in the biomass as the temperature varied. The decomposition rates of the different samples were analyzed in order to establish a link between each combustion phase and the composition of the raw materials. Two methods were used to determine the hemicellulose and cellulose contents of biomass from olive trees. Given the high cost of gas chromatography (GC), the results obtained with GC were compared with those obtained with a colorimetric method in order to evaluate the feasibility of using this less expensive alternative in future studies.

CHAPTER 1

ORIGIN AND EFFECTS OF MINERAL CONTAMINATION IN OLIVE TREE PRUNINGS⁸

16. Introduction

One of the problems associated with biomass combustion is related to the ash content of the material. The quantity and quality of ash produced during the biomass combustion process are strongly influenced by the properties of the biomass and the combustion technology, including the characteristics of the furnace, temperature of the process, and ash extraction systems (Samuelsson et al., 2009; Verma et al., 2009).

Inorganic species in biomass fuels, such as alkali oxides and salts, can intensify agglomeration, deposition, and corrosion problems on the heat transfer surfaces of boilers (Tortosa Masia et al., 2007; Vamvuka and Kakaras, 2011). Therefore, despite the obvious benefits of using biomass as a fuel source, technical difficulties related to

⁸ The results shown in this part were presented in: García-Maraver A, Terron LC, Ramos-Ridao A, Zamorano M. Origin and effects of mineral contamination in olive tree prunings. Submitted to Biomass and Bioenergy



the inorganic part of biomass fuels can lead to the reduced accessibility of biomass combustion systems. It goes without saying that this is also bad publicity for the residential pellet market (Öhman et al., 2004a; Rhén et al., 2005). The quality of pellets is determined by the end-user's requirements for the heating system and for handling properties. Higher ash content lowers the heating value of the pellets and increases the risk of sintering. Since this also negatively affects milling and pelletizing equipment (Lehtikangas, 2001), the calculation and subsequent reduction of the ash content in biofuels is crucial to enhance their quality.

In the Mediterranean areas of southwest Europe, agricultural activities are very important, but they produce large quantities of residue. This is the case of olive tree pruning residues, which have traditionally been used for domestic heating in rural areas and which are an important source of residual biomass. One hectare of olive trees generates approximately three tons of different types of pruning residue (i.e. 0.7 tons of wood, 1.5 tons of branches, and 0.8 tons of leaves) per year (AAE, 2008), most of which are now illegally burnt or left on the ground (Junta de Andalucía, 2009c). The energy exploitation of this biomass would allow the sustainable replacement of fossil fuels. Furthermore, it would increase self-sufficiency and energy diversification, besides contributing to the development of rural areas. Nevertheless, certain characteristics, such as the higher ash content of pellets from olive trees (Ollero et al., 2003; Zamorano et al., 2011) in comparison to other European pellets (Vassilev et al., 2010) have raised doubts concerning the viability of their use in thermal applications, especially in domestic heating systems.

During the harvesting of olive tree pruning residues, mobile and mechanical equipment adapted to the conditions of the grove are used to improve their processing and transport (Figure 22). According to previous research, the mechanized harvesting of this pruning residue requires that the dimensions of the residue between the tree rows be 1.5 m in width and at least 1 m in height (IDAE, 2008). However, during this collection, soil is also picked up along with the biomass, which impoverishes its final quality and increases the ash content, primarily because of the sand contamination of the raw material during storage and handling (Öhman et al., 2004a).



Figure 22. Collection equipment of olive tree pruning residues (IDAE, 2008)

In this research, fresh and stored materials from different olive tree pruning residues were separated to determine their ash content (% dry wt.) and to analyze its relation to the presence of inorganic compounds from the soil. Given that olive leaves were the pruning residues with the highest ash content, they were subjected to different cleaning processes and analyzed with an environmental scanning electron microscope (ESEM) and R-Ray Fluorescence (XRF). The objective was to determine the composition of the ash in order to obtain a higher-quality biomass and thus improve the pelletization process and combustion performance.

17. Experimental setup

17.1. Sampling program

This study analyzed the following types of olive tree pruning residue: (i) leaves; (ii) branches; (iii) bark; (iv) wood (Figure 23). Branches and leaves were obtained after the pruning that takes place at the beginning of each year to obtain the olives that will be used in olive oil production. Since roughly similar quantities of branches and leaves are picked up from the ground, this research studied them together as well as each separately in order to better ascertain the behavior of each component.



Wood and bark from olive trees are obtained every year after the olive harvest, and they are used as firewood. Because of the relatively low percentage of bark in a piece of wood, wood and bark were studied separately. They were collected while the pruning was still fresh so that the bark could be easily separated from the wood .

17.2. Soil characteristics

The olive tree pruning residues in this study were taken from an olive grove located in the province of Granada in the south of Spain. The soil in the grove was Calcareous Orthents (Icona, 1992), which is rich in calcium, magnesium, and silica.



Figure 23. Olive tree pruning residues

17.3. Laboratory procedures

17.3.1. Analysis of biomass from olive tree pruning

a) Ash content

The ash content of different samples was determined at 550°C and 900°C with a conventional laboratory furnace. The char was maintained for 5 h at 550 °C to reduce the organic carbon content to below 1 wt% and for 2 h at 900°C.

b) X-Ray fluorescence (XRF)

The 550°C ashes of the olive tree pruning residues as well as a soil sample from the grove were analyzed with a sequential spectrometer with dispersive wavelength and an X-Ray generator of 4 KW power, model PHILIPS Magix Pro (PW-2440).

17.3.2. Analysis of olive leaf samples

a) Cleaning procedures

Olive leaves were subjected to different cleaning methods in order to eliminate the particles adhered to them. The purpose was to analyze the impact of sand contamination on the final ash content. The following laboratory cleaning methods were applied:

- *Forced air:* A Ligno tester to determine the durability of pellets was used to make air rapidly circulate 50 g of the sample around a perforated chamber for 60 s. The chamber, which is an inverted square pyramid with perforated sides, was set at 70 mbar pressure. Forced air was the destructive force. The test was repeated three times for each sample.
- *Sieve shaker:* A sieve for particle size analysis was used with a sample of less than 30 g to avoid the blocking of the sieve. The sieve was placed on a sieve shaker for 1 minute. After sieving, particles larger than 4 mm were retained and their mass was weighed. This analysis was repeated three times for each sample.
- *Hand-washing:* Samples were hand-washed with a brush and water so that they could act as a point of reference for the other samples.

b) Storage conditions of the samples

Each of the previously described methods was applied to the samples of olive tree pruning residue with a high ash content at different stages of their handling and storage: (i) from an olive tree; (ii) from a pile in an olive oil factory; (iii) after three weeks of storage in a pile; (iv) after six weeks of storage, spread on the ground; (v) after 6 weeks of storage in a pile. In each case, samples were analyzed without being cleaned, as well as after having been cleaned by the different methods previously described.



c) Chemical composition of the ash: Environmental Scanning Electron Microscopy (ESEM)

Hand-washed samples (as a point of reference) and originally dirty samples from olive leaves were burnt at 550°C to be subsequently analyzed by ESEM. An Environmental Scanning Electron Microscope FEI model Quanta 400 was used to obtain the image, size of particles, and composition of the ash analyzed.

18. Results and discussions

18.1. Analysis of biomass from olive tree pruning

18.1.1. Ash content

Table 23 shows the ash contents of the different olive tree pruning residues. Olive leaves were found to have the highest ash content, which came to nearly 9% at 550°C, followed by bark (4.79%) and branches (3.78%). The results obtained were consistent with those of other authors, who obtained higher ash contents for agricultural residues when they were burnt with bark (Lehtikangas, 2001), as well as when leaves were burnt (Liu et al., 2002). On the other hand, olive wood without bark showed values (0.98%) similar to the normally lower ash content of hardwoods (1-1.3%) (Obernberger and Thek, 2004).

	ASH CONTENT (%)		
	550°C	900°C	
BARK	4.79	3.14	
WOOD (without bark)	0.98	0.70	
BRANCHES (without leaves)	3.78	2.77	
BRANCHES (with leaves)	6.34	4.32	
LEAVES	8.91	6.26	

Table 23. Ash content of the different residues obtained from olive tree pruning

However, at 550°C, the combustion is not complete. On the contrary, it continues until approximately 650°C, when a residue consisting only of ashes is obtained and the mass remains constant (Liu et al., 2002). Despite being calcined at 900°C, olive leaves presented an ash content of 6.26% (Table 23), a percentage higher than that of other species studied (Vassilev et al., 2010).

The high ash content of olive leaves, similar to the results obtained for fir leaves, wheat straw, and corn stover (Liu et al., 2002; Mani et al., 2006), could be the result of contamination by sand and chemicals (Lehtikangas, 2001; Öhman et al., 2004a). During the pruning season, since branches and leaves are picked up off the ground together, the ash content of both is increased because of the presence of olive leaves (Table 23). The leaves are collected from the ground; soil is swept up with them during their collection; and they are more exposed to mineral contamination during storage because of their larger surface. Consequently, it is clear that olive leaves are responsible for ash-related problems in biomass from olive pruning. For this reason, it was necessary to perform a detailed analysis of this biomass and thus obtain a deeper understanding of its characteristics.

18.1.2. X-Ray fluorescence (XRF)

Studies of the chemical composition of wood ash in the past have been limited to its basic composition. The focus of this research was largely on the agricultural use of wood ash because of its content in elements such as calcium, potassium, and phosphorus, and its capacity to modify soil health and crop performance. However, little has been said about the formation reactions of the ash during combustion, and the problem of ash deposition on heat transfer surfaces in boilers.

According to other studies (Vassilev et al., 2010), relatively large concentrations of certain elements, such as Ca and K, were found in olive wood and prunings because of the woody nature of these samples. On the other hand, as expected, more Si, Mg, Al, and Fe were found in leaf samples because they contained more clay from the soil (Table 24) in comparison to pruning and wood samples, which were not in such close contact to the ground. The differences in the mineral composition of the leaves made it necessary to focus on these samples because they made up a high percentage of the prunings that were used as a biomass source.

18.2. Analysis of olive leaf samples

Because of the differences in the ash content and composition of the olive leaves, the leaves were analyzed to ascertain whether or not the presence of inorganic



components was due to a superficial mineral contamination or to the composition of the leaves themselves.

Composition			Concentration (% dry wt.)					
Com			Olive leaves Olive pruning		Soil			
	Na ₂ O	0.2050	0.1350	0.0920	0.5170			
	MgO	3.6870	3.8490	2.1420	1.1300			
	Al ₂ O ₃	5.3700	0.5580	0.2680	17.0500			
	SiO ₂	15.1400	1.7200	0.8130	41.8600			
	P_2O_5	2.6000	3.4600	3.0000	0.1480			
	SO ₃	1.4200	1.6900	1.2000	0.0730			
	Cl	0.1670	0.2570	0.3550	-			
	K ₂ O	5.1240	8.5390	18.4400	2.3510			
	CaO	38.5700	59.2300	52.1500	10.2000			
Compounds	TiO ₂	0.4490	0.0590	0.0210	0.8020			
	Cr_2O_3	-	-	-	0.0140			
	MnO	0.0715	0.0769	0.0280	0.0713			
	Fe ₂ O ₃	3.5330	0.6890	0.7720	6.5560			
	NiO	-	-	0.0069	0.0124			
	CuO	0.0673	0.0628	0.0669	0.0067			
	ZnO	0.0337	0.0284	0.0181	0.0123			
	Rb ₂ O	0.0087	-	0.0071	0.0099			
	SrO	0.0999	0.0677	0.3577	0.0137			
-	ZrO ₂	0.0202	0.0032	0.0057	0.0300			
	0	27.5800	24.2900	22.1000	36.6700			
	Na	0.1520	0.0999	0.0682	0.3840			
	Mg	2.2230	2.3210	1.2920	0.6840			
	Al	2.8400	0.2950	0.1420	9.0260			
	Si	7.0770	0.8060	0.3800	19.5700			
	Р	1.1400	1.5100	1.3100	0.0645			
	S	0.5690	0.6790	0.1410	0.0292			
	K	4.2530	7.0890	15.3100	1.9510			
	Ca	27.5600	42.3300	37.2700	7.2890			
Elements	Ti	0.2690	0.0350	0.0130	0.4810			
	Cr	-	-	-	0.0095			
	Mn	0.0554	0.0595	0.0220	0.0552			
	Fe	2.4710	0.4820	0.5400	4.5860			
	Ni	-	-	0.0054	0.0098			
	Cu	0.0537	0.0502	0.0535	0.0053			
	Zn	0.0271	0.0228	0.0145	0.0099			
	Rb	0.0079	-	0.0065	0.0091			
	Sr	0.0845	0.0572	0.3025	0.0116			
	Zr	0.0150	0.0024	0.0042	0.0222			

Table 24. XRF results for dirty samples and soil

18.2.1. Cleaning of the samples

With a view to discovering whether or not the mineral contamination from the soil was responsible for the higher ash content of olive leaves (Table 23), we subjected them to different cleaning methods so as to eliminate the particles on them. Nevertheless, even though they were washed very thoroughly, the ash content of olive leaves at 550°C was always higher than 7.5% (Figure 24). Therefore, the composition of the olive leaf itself could be responsible for the high ash content of this type of biomass, rather than any type of external contamination.

During their growth on the tree, olive leaves are almost completely clean. The ash content for an unwashed sample was thus very similar to the ash content of a hand-washed one (Figure 24 and Figure 25).

18.2.2. Storage conditions of the samples

According to other studies (Afzal et al., 2010), the storage conditions of biomass can change its fuel properties and affect its final quality. In order to analyze the influence of storage and handling on the ash content, olive leaves were studied at different stages throughout their handling and storage (Figure 24 and Figure 25): (i) when they had just been picked from the tree in order to avoid any contamination during their handling and storage; (ii) in an olive oil factory when the olives had just been removed from the branches; (iii) during storage under different conditions.

As can be observed in Figure 24 and Figure 25, an increase of about 2% takes place when the leaves are taken from the tree and start to be handled and stored. This increase is higher for the unwashed samples, which are constantly accumulating dust and particles during the whole storage process, especially if leaves are stored in piles.

Stored samples that had been left spread on the ground for a period of six weeks had a similar ash content at 550°C to those stored in a pile for three weeks. However, the ash content of those stored in a pile for six weeks increased 2% in the last three weeks (Figure 24). Therefore the storage condition of the samples was found to be an important factor in the reduction of the mineral contamination of the leaves, which affected their ash content and possible slagging tendencies.





Figure 24. Ash content of olive leaves at 550°C under different storage and cleaning conditions



Figure 25. Ash content of olive leaves at 900°C under different storage and cleaning conditions

As shown in this study, the handling and storage of olive leaves increased their final ash content up to 4% after six weeks of storage (Figure 24 and Figure 25) in comparison to the hand-washed samples. It was thus necessary to analyze the chemical composition of dirty and hand-washed samples in order to discover the origin of the inorganic compounds of the ashes.

18.2.3. Environmental Scanning Electron Microscopy (ESEM)

The source of the ashes from olive leaves was not only the mineral contamination from the soil (Table 24), but also the chemical composition of this biomass. Accordingly, it was necessary to take the XRF results for the inorganic components of the unwashed leaf samples and compare them with the ESEM results for the components of hand-washed leaf samples.



Figure 26. ESEM of the dirty samples

Whereas Si and Al were only present in the unwashed leaf samples because of the soil on them (Figure 26), other elements, such as Mg, P, Ca, and K, were found in both the unwashed and hand-washed samples (Figure 26 and Figure 27) because these are components of the biomass as well as of the soil (Table 24) in which the olive trees were planted (Icona, 1992). Since the absorption of these components by the leaves made it impossible to remove them by physical methods such as those used in this

study, they were found to be responsible for the high ash content of the samples even in the hand-washed leaf samples.



Figure 27. ESEM of the hand-washed samples

19. Conclusions

Despite the low ash content of the woody fractions of olive tree pruning residues, which makes them suitable for domestic heating systems, the largest part of this biomass is composed of small branches and leaves with a high ash content at 550 °C and 900°C, mainly due to the presence of olive leaves.

These leaves became progressively dirtier during their handling and storage since they were contaminated by mineral compounds from the soil. In order to reduce the ash content of olive leaves, two cleaning methods were assayed with hand-washed samples used as a point of reference. The degree of cleanliness obtained was always higher with a Ligno test, which achieved a 2% reduction in ash content with respect to the unwashed samples throughout each of the storage stages. This approximately doubled the reduction achieved with the sieving method.

Nevertheless, it was impossible for the ash production to be lower than 7.5% at 550°C in the case of leaves. This production was even as high as 5% when the leaves were incinerated at 900°C.

Since physical cleaning methods were not sufficient to reduce the ash content of the leaves, the origin of the high ash content of this type of biomass was the inherent composition of olive leaves. Though the absorption of inorganic components from the soil, such as Mg, Ca, and K, is responsible for part of the ash content of the leaves, the next chapter describes how the organic components were also analyzed to better understand if there was any relationship between the organic composition of the raw materials and the final ash content.



CHAPTER 2

ORIGIN AND EFFECTS OF MINERAL CONTAMINATION IN OLIVE TREE PRUNINGS⁹

20. Introduction

Organic matter such as wood or plants (lignocellulosic biomass) is a potential source of energy that can be used to generate heat and electricity as well as to foment green oil production for automobiles (Carrier et al., 2011). In the environment, lignocellulose is found in wood, grass, agricultural residues, forestry wastes, and municipal solid wastes (Perez et al., 2002). In fact, it is the main component of this type of biomass.

Lignocellulose is composed of three types of polymers: cellulose, hemicellulose, and lignin. Cellulose is the most abundant component in lignocellulosic structure,

⁹ The results shown in this part were presented in: García-Maraver A, Salvachúa D, Martínez MJ, Zamorano M. Analysis of the relationship between cellulose, hemicelluloses and lignin contents and the thermal behavior of residual biomass from olive trees. Submitted to Biomass and Bioenergy



consisting of a linear polymer of β -1,4 linked glucose units (Stelte et al., 2011). Cellulose is closely associated with hemicellulose, which is a branched polymer composed of sugars such as pentoses and hexoses. Lignin occurs throughout the plant cell-wall and is mainly concentrated in the middle lamella and the primary cell-wall. This polymer is mostly formed by phenylpropane units, which makes it a highly complex and recalcitrant compound (Stelte et al., 2011). The composition and percentages of cellulose, hemicellulose, and lignin vary depending on the plant species (Perez et al., 2002), and their differences in chemical structure give them different chemical reactivities.

Given the wide range of biomasses that can be used for thermal treatment as well as their heterogeneous nature, the consideration and analysis of their composition is a crucial factor in any type of process design (Carrier et al., 2011). Since the thermal degradation process is primarily influenced by the chemical nature and components of the biomasses (Faravelli et al., 2010), an in-depth knowledge of the composition of each type of biomass is necessary to ascertain its thermal behavior and properly design the combustion process. Consequently, each component must be studied in order to be able to predict the efficiency of the biomass conversion process. This means that if lignocellulosic biomass is to become a renewable fuel and a viable replacement for fossil fuels, its exact composition should be analyzed and specified.

Modern biomass energy production recovers organic leftovers from forestry and agriculture, such as corn stover, rice husks, wood waste, and bagasse, or uses special, fast-growing energy crops like willow and switchgrass, as fuel (Carrier et al., 2011). Olive groves can be found throughout Mediterranean regions, where they are one of the most important agricultural crops. It is thus hardly surprising that the residues from olive oil industries and olive grove maintenance have become an important energy resource in this area (Garcia-Maraver et al., 2012). Accordingly, the use of olive tree residues permits their exploitation in efficient combustion processes.

Nevertheless, certain characteristics, such as the higher ash content of olive tree pellets (Ollero et al., 2003; Zamorano et al., 2011), have raised doubts concerning their use in thermal applications, especially in domestic heating systems. As the main producer of char residue, lignin has naturally an important role in char formation. This directly affects thermal biomass decomposition in the second stage of an oxidative atmosphere (Demirbas, 2001; Shen et al., 2009; Stelte et al., 2011). Consequently, the high ash content obtained from olive tree pruning residues is related to the high percentage of lignin where agricultural lopping residues also include leaves or small tree branches and not only woody portions (Ollero et al., 2003; Gilbert et al., 2009).

Numerous studies have estimated the chemical composition of different types of biomasses, based on the analysis of thermal curves (Liu et al., 2002; Grioui et al., 2006; Marcilla et al., 2009). However, up until now, this chemical composition has not been compared to the real composition of the samples. Given the lack of research on olive tree residues as a source of thermal energy, the study described in this chapter analyzed the chemical composition of the different olive tree pruning residues (leaves, prunings, and wood) in order to gain a better understanding of their thermal behavior under combustion conditions. Moreover, an in-depth knowledge of the composition of these residues was found to be crucial in the design of the most efficient combustion process, which would permit these residues to be used as a renewable fuel.

21. Materials and methods

21.1. Samples and preparation for the analysis procedure

Olive pruning residues (leaves, pruning residues, and wood) were obtained from an olive grove near Granada, Spain. The granulometric values of the raw materials had to be reduced since a particle size smaller than 0.5 mm was necessary for the chemical analysis and for the thermogravimetric analysis to ensure the heat transfer rate within the kinetic regime of decomposition (Rath et al., 2002). Therefore, a Retsch MM301 hammer mill was applied for 5 min at 25 Hz, as well as a sieve of the corresponding size.



21.2. Raw material characterization

21.2.1. Lignin content

Raw materials (300 mg, dry weight) were subjected to acid hydrolysis as specified in standard Tappi methods (Tappi, 1974; Tappi, 1975) for acid-insoluble lignin and polyssacharide content analysis. After hydrolysis, soluble lignin (g/L) was measured by UV-Spectrophometry (ϵ 205= 110 L/g cm). The analyses were carried out in triplicate.

21.2.2. Cellulose and hemicelluloses content

Tappi methods (Tappi, 1975) were followed to analyze the sugar composition by gas chromatography (GC) (Prieto et al., 2008). However, other colorimetric methods were also tested for faster polysaccharide analysis and both methodologies were subsequently compared. The glucose was quantified with the Glucose-TR kit (Spinreact) and the Total Reducing Sugars (TRS) were determined with the Somogyi-Nelson method (Somogyi, 1945). The difference between TRS and glucose corresponds to the sugars from hemicellulose, in which pentoses are usually the main carbohydrates. The cellulose content was calculated from the glucose and hemicellulose in the other sugars, using an anhydro correction of 0.90 and 0.88 for both hexoses and pentoses, respectively.

21.2.3. Ash content

The ash contents of the various raw materials was determined as specified in the standard EN 14775 (2009).

21.3. Thermogravimetric analysis (TGA)

The TGA of the solid biofuels was performed with a Mettler Toledo TGA/DSC 1 thermogravimetric analyzer under air atmosphere. The initial mass of the samples was kept between 6-8 mg to avoid any possible effect on mass and heat transfer during the biomass decomposition process (Blasi et al., 2001). The samples were heated from 30 to 1000°C at a heating rate of 10 K/min and 60 ml/min of air to maintain an oxidative atmosphere for the thermal decomposition of the particles. This heating rate and air flow

ensured that there was not a temperature gap between the sample and its surroundings (Bilbao et al., 1997).

22. Results and discussion

22.1. Composition of the samples

Table 25 shows the composition of the different lignocellulosic samples analyzed by the methods previously described. As shown by the results of the gas chromatography (GC), glucose (mainly from cellulose) was the most abundant sugar in all samples, especially in the woody samples. Regarding the hemicellulose components, arabinose, mannose, and xylose were the main sugars in leaves, olive prunings, and wood, respectively (Table 25).

Cellulose and hemicellulose were also analyzed and estimated with colorimetric methods. For the olive prunings and olive wood, the result of the glucose analysis with the Glucose-TR kit was similar to that obtained with GC. However, for the leaves from olive trees, there was a significant difference between the results obtained with each method. In the case of hemicellulose, the Somogyi-Nelson method was evidently not the only viable procedure for estimating its content in the samples, especially in the olive wood. The results obtained showed that, as described earlier (Salvachua et al., 2011), these colorimetric methods were an effective way to estimate the cellulose and hemicellulose content in wheat straw, but regarding the current results, they can also be used in olive leaves and pruning samples when the identifications of specific sugars is not required.

The percentage of insoluble lignin was found to be considerably higher in olive leaves than in the rest of the samples, and was similar to that obtained for fresh bark (37.4%) (Lehtikangas, 2001). In contrast, olive prunings and wood showed similar values to those obtained for other types of biomass such as pine (27.45%) (Shen et al., 2009).

The raw materials analyzed were primarily composed of cellulose, hemicellulose, and lignin as well as a lesser quantity of minerals (e.g. ash) and other compounds, known as *extractives*. The total amount of cellulose, hemicellulose, and



		Olive Leaves		Olive Pruning		Olive Wood	
		Avg	SD	Avg	SD	Avg	SD
Insoluble lignin, %		39.60	0.19	27.43	0.93	23.50	0.81
Soluble lignin, %		0.21	0.00	0.21	0.00	0.15	0.01
	Kit	8.54	0.31	20.24	0.90	31.48	0.61
Cellulose*, %	GC	5.72	0.54	18.57	1.17	31.90	0.47
	Somogyi	5.37	0.90	10.66	1.70	11.33	0.72
Hemicellulose*, %	GC	3.78	0.32	11.30	1.55	15.45	3.43
Character 0/	Kit	9.49	0.34	22.49	1.00	34.97	0.34
Glucose, %	GC	6.36	0.54	20.63	1.17	35.45	0.47
Xylose (GC), %		0.94	0.14	1.83	0.86	4.26	2.02
Rhamnose (GC), %		0.44	0.08	1.27	0.17	1.48	0.16
Fucose (GC), %		0.02	0.01	0.01	0.01	0.01	0.01
Arabinose (GC), %		1.27	0.25	2.16	0.98	2.71	0.93
Mannose (GC), %		0.94	0.10	4.57	0.73	6.10	2.59
Galactose (GC), %		0.73	0.06	0.99	0.35	0.99	0.40
Glucosamine (GC), %		0.00	0.00	1.14	0.00	1.14	0.00
Galactosamine (GC), %		0.00	0.00	0.88	0.00	0.89	0.01
Total, %**		49.31	0.66	57.51	5.91	71.00	3.55
Ash content, %		14.17	0.29	5.50	0.06	1.43	0.00
Extractives, %***		36.52	0.72	36.99	5.91	27.57	3.55

Table 25. Composition a	analysis of the different raw	materials
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GC= samples analyzed by gas chromatography

Kit: samples analyzed by the Glucose-TR kit

Somogyi: samples analyzed indirectly by Somogyi-Nelson method

* Cellulose and hemicellulose estimations using the anhydro correction.

** This value was obtained as the sum of lignin, cellulose, and hemicellulose percentages (sugars analyzed by GC).

*** Extractives content has been calculated as the remaining components after calculating the ash and major fractions (cellulose, hemicelluloses, and lignin).

lignin was lower in the case of olive leaves (Table 25) because of their higher ash content (14.17%), which is directly related to the high lignin content (39.81%) of the samples (Demirbas, 2001; Stelte et al., 2011).

The extractives in olive tree prunings are mainly due to the presence of leaves (Ballesteros et al., 2011). Not surprisingly, a higher percentage of extractives was found in the olive leaf samples (36.52%), followed by pruning residues (partially composed of leaves), and wood (Table 25). Similar values for the extractives in olive tree samples were obtained by Romero et al. (2008), who found that the content of extractives was in the range of 14–31% for olive tree prunings, depending on the quantity of leaves and the extraction method.

By definition, extractives are chemical components that can be extracted from lignocellulosic biomass with various solvents, and which are divided into resins (lipophilic compounds) and phenolics (Ekman et al., 2000). Their content and composition vary greatly, depending on the plant species and geographic location (Ballesteros et al., 2011). Previous studies (Sluiter et al., 2005; Chen et al., 2007) have reported that water and ethanol extractives can not only contain organic acids, inorganic materials, nitrogenous material, chlorophyll, and waxes but also non-structural sugars.

22.2. Thermogravimetric analysis (TGA)

The nature of the TGA curve in combination with the corresponding derivative of the TGA (DTG) curves/peaks indicates the number of stages of thermal degradation, which is related to the chemical composition. Figure 28 shows the weight loss curves obtained in the form of TGA curves and the DTG curves from the different residual biomass from olive trees. According to various studies (Liu et al., 2002; Grioui et al., 2006; Marcilla et al., 2009), these curves reflect different decomposition steps that are directly related to the decomposition of the main components.

The first stage corresponds to a wood drying phase during which moisture and certain volatile compounds are eliminated at temperatures ranging from room temperature to 180°C. In the case of the lignocellulosic materials, the degradation



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temperature and the thermal degradation rate of the hemicellulose and lignin were lower than those found for the cellulose (Milne, 1981). Therefore, the initial degradation temperature was higher for the olive prunings and wood since both contain more cellulose in comparison to the leaves (Table 25). In addition, it should be pointed out that a fuel with a higher percentage of ash and impurities in its composition has lower initial degradation temperatures (Huang and Li, 1998). This effect was evident in the case of the leaves, which generate a correspondingly large quantity of ash.



Figure 28. TGA/DTG curves for olive leaves (OL), olive prunings (OP) and olive wood (OW)

Consequently, polymer decomposition was indicated by a large weight loss beginning at 180°C at which the hemicellulosic component began to break down. The rate of weight change rapidly increased at temperatures above 200°C since the hemicellulose was essentially converted into gases and acetic acid (Grioui et al., 2006) and the cellulose decomposed (Liu et al., 2002; Shen et al., 2009).

The maximum mass loss took place in the region of fast combustion. For olive prunings and wood, the loss velocity became significantly slower at temperatures between 300 °C and 400 °C for the olive prunings and wood. The behavior of the olive leaf samples differed from the behavior of the other two samples because of their lower glucose and hemicellulose contents (Table 25). This stage was represented by the

shoulder peak of volatilization at a lower temperature (330 °C), which reflected the decomposition of hemicelluloses and cellulose. After this point, the burning ratio first decreased and then after a short time, it began to increase again. The steepest peak at a high temperature (430-460 °C) (Table 26) corresponds to the oxidizing process of the char (Safi et al., 2004; Gani and Naruse, 2007). The olive leaves were the type of waste that took the longest to react to the temperature. In fact, such a reaction did not occur until the temperature was 30 °C higher than that of the olive wood.

Sample	$T_{p1} \left({}^{o}C \right)^{a}$	$T_{p2} \\ (^{o}C)^{a}$	$T_f({}^oC)^b$	Moisture (%)	Volatile (%)	Ash (%)
Olive leaves	330	460	650	7	81	12
Olive prunings	330	450	650	7	91	2
Olive wood	330	430	650	7	91	2

Table 26. Features of the thermogravimetric curves for olive tree biomasses in air

a Temperatures of DTG peaks (for each sample there are two peaks) b Final decomposition temperature

According to Gani and Naruse (2007), biomass burns in flowing steps. First, the cellulose components in the biomass are volatilized, such that the porosity in the char particles of biomass increases and the oxygen easily diffuses into the char particles. Next, the lignin components in the biomass can also react with diffused oxygen even if the reactivity of lignin itself is low.

Both peaks were lightly overlapped because of the oxygen (Orfao et al., 1999). When thermal decomposition occurs in an air atmosphere, the presence of oxygen causes additional complexities. It enhances the degradation of the material at low temperatures (Shafizadeh and Bradbury, 1979); causes the appearance of gas-phase reactions between the volatile compounds released and oxygen; and when the temperature is high enough, oxygen is likely to promote the combustion of the char residue generated in the early stages of the solid degradation (Shafizadeh and Bradbury, 1979). Consequently, combustion and pyrolysis may coexist (Liu et al., 2002).

The rate of mass loss reached a maximum at 440-450°C. At that temperature, the cellulose had largely decomposed while the lignin components continued to break down



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(Byrne and Nagle, 1996). The lignin decomposition continued at a low rate above 450°C, which corresponded to the slow decomposition of the solid residue resulting from the previous step. The decomposition rate was very low at temperatures of 450-650°C, at which it became negligible (Table 26). At that temperature (T_f), a residue only composed of ashes was obtained, which was free of any carbon content. At higher temperatures, reductions of the mass can occur, which can be attributed to volatile metal loss and carbonate decomposition (Ross et al., 2008).

The mass loss was lower for olive leaves because of their low glucose and hemicellulose content, whereas their higher ash content was directly related to their high lignin content (Demirbas, 2001; Stelte et al., 2011). In this regard, the results were similar to those obtained for fir leaves (Liu et al., 2002). This behavior is usual since the ash yield from wood decomposition is generally lower than the yield from leaf decomposition because of the difference between the chemical leaf and wood constituents (Liu et al., 2002). This result means that the lignin in biomass controls the reaction rate during combustion.

23. Conclusions

The glucose content was found to be lower in olive leaves. As a result, their initial decomposition temperature was higher since there was an inverse relation between decomposition temperature and cellulose content.

Regarding hemicelluloses, olive leaves were found to contain a greater percentage of arabinosa, whereas in olive prunings and wood, mannose was the predominant hemicellulosic sugar. In this part of our study, the Somogyi method was found to be adequate to estimate the hemicellulose in leaves and pruning samples though it was not effective in wood.

The mass-loss fraction of olive leaves was lower at all temperatures, which indicated that the woody materials had a more rapid mass loss than the leaves because they are richer in glucose and hemicelluloses. Since the lignin decomposition temperature was high, it was responsible for the higher ash content of olive leaves and their low reaction rate.

PART 4

QUALITY OF PELLETS MADE FROM RESIDUAL BIOMASS FROM OLIVE TREES¹⁰

¹⁰ The results shown in this part were presented in: García-Maraver A, Rodriguez ML, Diaz LF, Zamorano M. Quality of pellets made from residual biomass from olive trees. Submitted to Biomass and Bioenergy



24. Introduction

In recent years, the production of thermal energy by means of biomass on a small scale has shown a clear trend towards densified biofuels (pellets) (Verma et al., 2009). This fact is due to their homogeneous size, which facilitates an automatic or semi-automatic treatment and thus, resolves the disadvantages of the traditional domestic use of biomass (Holm et al., 2006; Miranda et al., 2012). The use of densified biofuels also reduces the costs associated with handling, storage and transportation, due to the increase in bulk density associated with the densification process.

In general, pellets' quality depends on the properties of the feedstock and quality management of the manufacturing process (Gilbert et al., 2009). Considering that wood pellets from forestry residues already has successfully established technologies and markets for production and consumption in some European countries, it is necessary to focus on studying the pelletization of agricultural biomass, which is periodically planted and harvested and then may have a great energy potential especially in rural areas.

Despite the large generation of agricultural residues, the current level of their utilization as fuel is low. Generally, agricultural residues are produced locally and they often have a low bulk density (Werther et al., 2000; Carone et al., 2011). Other important factors which influence the level of energy usage of agricultural residues are the local availability, the fact that they are dispersed over a relatively large area, and the lack of sufficient information concerning fuel feeding, as well as the combustion and emission characteristics of these residues. This information is important for the design and efficient operation of combustion systems fully adapted to the biomass of each region.

In the Mediterranean areas of European Southwest, agricultural activities are very important, and a great amount of residues are produced from these activities. This is the case of olive tree pruning residues, which give rise to significant amounts of residual biomass, according to results explained in Part 1. The management of these residues involves a problem due to the costs associated to the treatments needed for their proper removal (Miranda et al., 2012). Therefore, pelleting this residual biomass would suppose the conversion of a residue difficult to manage into an energy resource especially indicated to be used in the areas where this grove exists.

The final quality of pellets varies depending on the raw material properties and the manufacturing process (Samuelsson et al., 2009). Inorganic and organic components of the different raw materials obtained after the olive pruning were analyzed and described in the previous part of the document. Nevertheless, there are variables dependent on the pelletization process that can be controlled to optimize production efficiency and improve the quality of finished product (Carone et al., 2011) according with the specific biomass studied.

Hence, the aim of this chapter is to analyze the mechanical, physical and chemical characteristics of the different olive pruning residues that were pelletized under different conditions of initial moisture, compression length and diameter of the pellet mill die and temperature of the pelletization process. With this purpose, the operational conditions have been optimized according to the final properties of the pellets produced. In addition, the quality parameters were discussed and compared with the EN 14961-2 (2011) in order to check if the pellets samples fulfill the guidelines established for non-industrial use (Table 14).

25. Materials and methods

25.1. Characterization of the raw materials

Olive pruning residues (leaves, prunings and wood) were obtained from an olive grove near Granada, Spain. Olive leaves (OL) were obtained from an olive oil industry in February 2012 with a moisture content of 24.4% (w.b.), prunings (leaves and small branches) (OP) were harvested as a whole in March 2012 with a moisture content of 26.7% (w.b.) and olive wood (OW) was collected in logs in March 2012 with a moisture content of 23.2% (w.b.).

Biomass was stored for 3 months uncovered, until its moisture content decreased approximately to 15% (w.b.). Then it was stored for a month in the laboratory in order to dry it progressively to the desire moisture contents (13, 11 and 9%).

Since there are properties dependent on the raw material itself, they were analyzed for the different materials studied before being pelletized. The methodology is described below.

25.1.1. Particle size distribution

5 kg of the milled material was screened through 2.5 and 4 mm circular holes and the distribution for classes <2.5mm, 2.5-4 mm and >4 mm was calculated according to EN 15149-2 (2010).

25.1.2. Moisture, ash and volatile content

The EN 14774-1 (2007), EN 14775 (2009) and EN 15148 (2009) test procedures were applied to three samples from each raw material to analyze their moisture, ash and volatile content respectively.

25.1.3. Elemental analysis

Three samples of each raw material were milled and mixed to obtain a homogenous sample of about 7 mg to determine the percentage of carbon, nitrogen, sulfur and hydrogen, according to EN 15104 (2011). A FISONS CARLO ERBA EA 1108 CHNSO detector type was used for this purpose.

25.1.4. Calorific value

The higher heating value (HHV) was determined three times for each raw material according to EN14918 (2009) by using a bomb calorimeter (Parr 6100) and the average value was calculated. The lower heating value (LHV) was calculated by correcting HHV by the heat energy required to vaporize water due to hydrogen released during combustion (Equation 1). The hydrogen content values ($%H_2$) were determined by elemental analysis.

LHV = $(HHV*4.184-206-\%H_2-23.05*\%H_2O)/4.184$ (Equation 1)

25.1.5. Chlorine content

After the determination of the calorific value, the soluble chlorine content was calculated by using a Mettler Toledo compact titrator G20 with silver nitrate as reaction agent.

25.1.6. Slagging and fouling tendencies

Slagging and fouling indexes resulting from investigations of bituminous and brown coal ashes can be calculated based on the composition of the ash and fusibility correlations (Pronobis, 2005) according to Equations 2 and 3. To determine the composition of the ash, 550°C ashes from the different olive tree pruning residues studied were analyzed by a sequential spectrometer with dispersive wavelength and X-Ray generator of 4 KW power, model PHILIPS Magix Pro (PW-2440).

Table 27 summarizes the classification of the indexes according to fusibility correlations.

SR (ratio slag viscosity index):

 $SR = SiO_2 \times 100/(SiO_2 + Fe_2 + CaO + MgO)$ (Equation 2)

Fu (fouling-index):

 $Fu = (B/A)(Na_2O + K_2O)$ (Equation 3)

where B/A is the ratio between the basic compounds (B) and acidic compounds (A) and is determined according to Equation 4:

 $B/A = (Fe_2O_3 + CaO + MgO + Na_2O + K_2O + P_2O_5)/(SiO_2 + Al_2O_3 + TiO_2)$ (Equation 4)

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S _R	$S_{R} > 72$	Low slagging inclination		
	$72 >= S_R > 65$	Medium slagging inclination		
	$S_R <= 65$	High slagging inclination		
Fu	$F_u <= 0.6$	Low fouling inclination		
	$0.6 < F_u <= 40$	High fouling inclination		
	$F_{u} > 40$	Extremely high fouling inclination,		
		tendency to sintering of deposits		

Table 27. Fusibility correlations (Pronobis, 2005)

Ash fusibility was also investigated using an imaging sintering point testing equipment (Leco AF-700). Ash samples were prepared by igniting the fuels in a muffle furnace at 550°C, and then shaped into pyramids. Ash fusion temperature measurements were carried out with a maximum temperature of 1500°C, in an oxidizing atmosphere. The measurements were carried out following international standard procedures (ISO 540, 1995; prEN 15370, 2005). During the testing process, the deformation temperature (DT), softening temperature (ST), hemisphere temperature (HT) and fluid temperature (FT) were recorded according to the specific shapes of the ash cylinders.

25.2. Densification of the raw materials

Since a particle size smaller than 5 mm was necessary for the pelletization process, the granulometric values of the raw materials had to be reduced. Therefore, a Retsch MM301 hammer mill was used during 5 min at 25 Hz, as well as a sieve of 4 mm size. In some cases it was also necessary to wet the samples by sprinkling water on them to get a homogeneous final moisture content for the tests (9, 11 and 13%).

The pellets were produced by means of a pelletizer KAHL 14-175 (Figure 29). This pelletizer has a nominal power of 3 kW. Its production reached approximately 100 kg/h for OL and OP, while the production capacity was 50 kg/h for OW.

Three different flat dies that differed in the size of their cylindrical holes were used for the production of the pellets:

D1: diameter 6 mm; compression length 20 mm

D2: diameter 6 mm; compression length 24 mm

D3: diameter 8 mm; compression length 32 mm



Figure 29. Pellet press used in this study

Several runs of pellets were obtained supplying the pellet press continuously with the milled and sieved biomass. Once the process stabilizes, the temperature of the material increases due to the friction between the stationary flat die, the driven roller and the residue (Figure 29). Therefore, the pelletizing runs were carried out according to an experimental design in the factors (i) raw material moisture content before pelletizing, (ii) raw material composition in three ingredients: OL, OP and OW, (iii) diameter and compression length of the pellet die, and (iv) temperatures reached during the pelletization process. The challenge in biomass pelletization is therefore to keep these parameters in the range where high quality pellets are produced, at a minimal energy input and at a high pellet mill capacity (Stelte et al., 2011). However; these parameters are a function of the properties of the raw material (Arshadi et al., 2008; Relova et al., 2009).

Once the pellets were cooled at room temperature, a representative sample was taken for the analyses described below.



25.3. Characterization of the pellets

Before laboratory analysis and due to the large quantity of pellets produced, it was necessary to use a sample splitter (Figure 30) for subdividing the sample and thus ensure a representative sample. Once the sample was reduced, the parameters described below were analyzed three times for each type of pellet.



Figure 30. Sample splitter used in this study

25.3.1. Durability

The pellet durability of about 100g samples of each type of pellet was determined according to EN 15210-1 (2009). The samples were subjected to a 60 second controlled air stream (70 mbar) which acted as the sieving process causing the pellets to collide with each other and also the hard surfaces within the perforated hopper. Upon completion of the cycle the pellets were removed and re-weighed. This was then compared against the sample weight prior to sieving, and the difference was then expressed as a percentage which corresponds to the durability of the pellets.

25.3.2. Hardness or compression strength

The hardness of a piece of pellet chosen at random from each type was measured using a hand-operated durometer (Amandus Khal mod. 21465): a single pellet was

located between two plates and a progressively increasing load was applied until fracturing (Figure 31).



Figure 31. Hardness tester used in this study

This test simulates the compressive stress due to the weight of the pellets on top on the pellets underneath them during storage in bins or silos and crushing of pellets in a screw conveyor (Li and Liu, 2000; Gilbert et al., 2009).

25.3.3. Pellet size, single pellet density and bulk density

Pellet size was measured with a digital caliper (0.01mm resolution, model CD-56C, Mitutoyo Corp., Aurora, IL) and the results were used to calculate the ratio length/diameter.

Single pellet density was calculated according to CEN/TS 15150 (2005) and bulk density of the pellets was determined according to EN 15103 (2009) for solid biofuels and used for determining the energetic density (Equation 5).

Energetic density $(MJ/m^3) = Bulk$ density $(kg/m^3)*Lower$ Heating Value (MJ/kg) (Equation 5)

25.3.4. Moisture content

The EN 14774-1 (2007) test procedure was applied to three samples from each run of pellets.


25.4. Optimum parameters for the pelletization of residual biomass from olive trees

Once the pellet quality parameters were analyzed for the samples produced under different operating conditions, the variables that provide a better quality were calculated for the different raw materials. For that purpose, the ideal values of each parameter were fixed (Table 28) according to the interval of the results obtained in previous works for other pellets samples (Lehtikangas, 2001; Arshadi et al., 2008; Carone et al., 2011; Zamorano et al., 2011; Carroll and Finnan, 2012) and to the limits established for non-industrial pellets in EN 14961-2 (2011).

Table 28. Ideal values considered for quality parameters

Parameter	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m ³)	Bulk density (kg/m ³)	Moisture content (%)
Ideal value	100	22	40	1200	700	0
Range	0-100	0-25	0-40	0-1500	0-850	0-100

A Principal Component Analysis was done in order to try to reduce the number of variables to study. This reduction facilitates the determination of the optimum sample only if some indicators fulfill some established conditions. In this work, the KMO (Kaiser-Meyer-Olkin) measure of sampling adequacy was calculated by using IBM SPSS Statistics 20 software. The scale for this indicator is shown in the Table 29 below:

Table 29. Data adequacy scale for the KMO index

0.9 < KMO <= 1	marvelous	$0.6 < \text{KMO} \le 0.7$	mediocre
0.8 < KMO <= 0.9	meritorious	0.5 < KMO <= 0.6	miserable
0.7 < KMO <= 0.8	middling	KMO <= 0.5	unacceptable

26. Results and discussion

26.1. Characteristics of the raw materials

The results from the analyses are summarized in Table 30, which shows the average values of the parameters determined for OL, OP and OW before pelleting.

26.1.1. Particle size distribution

The granulometric values of the raw material had to be reduced not only to allow the pelletization process but also to work with representative samples of every material.

Although the same milling procedure was followed for the three raw materials, OL were easier to be reduced to less than 2.5 mm (84.42%) because of being not as hard as the OW (72.11%), which had a higher percentage of bigger particles than the rest of the samples analyzed (Table 30). OP showed intermediate values because this material is composed by a mixture of leaves and woody branches.

26.1.2. Moisture, ash and volatile content

The moisture content of the samples was adapted to do the different pelleting experiments. 9, 11 and 13 % were the moisture contents of the raw materials before pelleting. Moreover, this parameter had to be measured to calculate the lower heating value.

Regarding the ash content, and as it was described in the previous section of this document, the differences between the different materials are especially important due to the difference in their composition (Table 30). In the case of OL 14.17% of ashes were generated after being burnt at 550°C, considerably higher than the natural ash content values given in the literature (Vassilev et al., 2010) except for other non-woody materials such as rice husks (Madhiyanon et al., 2009). OP (5.50%) and OW (1.43%) showed common ash values for agricultural or herbaceous residues and woody materials respectively (Werther et al., 2000; Lehtikangas, 2001; Gilbert et al., 2009; Vassilev et al., 2010). Considering the EN 14961-2 (2011), only OW samples would fulfill the guidelines for non-industrial pellets because of having an ash content lower than 3% (Table 14). For the combustion of agricultural residues with high ash contents, consideration must be given to incorporate efficient ash removal equipment from the flue gas to eliminate or reduce particulate pollution, like in the case of coal combustion (Werther et al., 2000).



Agricultural residues are characterized by higher contents of volatile matter (Vassilev et al., 2010), and this fact was confirmed with the samples aim of this study (Table 30). This indicates that the residues are easier to ignite and to burn, although the combustion is expected to be rapid and difficult to control. The results obtained were similar to those obtained in other studies (Werther et al., 2000; Vassilev et al., 2010).

PARAMETERS		OL	,	OP)	OW		
PAKAMETERS	-	Avg	SD	Avg	SD	Avg	SD	
Douticle size	<2.5 mm	84.47	5.27	79.78	5.09	72.11	3.23	
distribution, % wt	2.5-4 mm	11.22	0.82	19.38	0.86	25.96	1.34	
	>4 mm	0.31	0.01	0.84	0.02	1.93	0.07	
Moisture, %		11.02	0.31	10.89	0.16	7.42	0.19	
Ash content, %		14.17	0.29	5.50	0.06	1.43	0.00	
Volatile content, %		63.74	0.09	79.80	0.10	90.15	0.00	
Fixed carbon, %		11.07	0.43	3.81	0.20	1.00	0.19	
C, %		45.71	0.68	47.12	0.47	46.49	0.11	
N, %		1.56	0.06	1.11	0.10	0.32	0.01	
Н, %		6.66	0.14	6.82	0.06	6.77	0.04	
S, %		0.11	0.00	0.05	0.00	0.00	0.00	
HHV, MJ/kg		19.64	0.07	18.20	0.01	17.53	0.32	
LHV, MJ/kg		18.01	0.07	16.55	0.01	16.06	0.32	
Chlorine content as received, %		0.03	0.00	0.02	0.00	0.01	0.00	
Na ₂ O, %		0.2050	-	0.1350	-	0.0920	-	
MgO, %		3.6870	-	3.8490	-	2.1420	_	
Al ₂ O ₃ , %		5.3700	-	0.5580	-	0.2680	-	
SiO ₂ , %		15.1400	-	1.7200	-	0.8130	-	
$P_2O_5, \%$		2.6000	-	3.4600	-	3.0000	-	
K ₂ O, %		5.1240	-	8.5390	-	18.4400	-	
CaO, %		38.5700	-	59.2300	-	52.1500	-	
TiO ₂ , %		0.4490	-	0.0590	-	0.0210	-	
Fe ₂ O ₃ , %		3.5330	-	0.6890	-	0.7720	-	
(B/A)		2.5630	-	32.4784	-	69.5063	-	
Fu		0.4810	-	3.7443	-	3.7506	-	
S _R		24.8482	-	2.6264	-	1.4550	-	
Initial deformation		1007		1011				
temperature (IDT), ℃		1225	-	1216	-	1165	-	
Spherical temperature (ST), °C		1241	-	1495	-	1493	-	
Hemispherical temperature (HT), °C		1246	-	>1500	-	>1500	-	
Fluid temperature (FT), °C		1256	-	>1500	-	>1500	-	

Table 30. Characteristics of the raw materials before pelleting

26.1.3. Elemental analysis

According to other studies (Vassilev et al., 2010), the main element of this analysis was carbon, with a value slightly lower than 50%; besides, a hydrogen value of slightly lower than 7% was observed. However, differences were found regarding the nitrogen content (Table 30). OL and OP showed similar values to those obtained from coniferous wood (Tillman, 1991), but since they were higher than 1%, these raw materials could not be used in non-industrial boilers (Table 14). OW presented 0.32% of nitrogen content, comparable to other deciduous trees (Tillman, 1991) and therefore could be categorized as A2 (EN 14961-2, 2011).

26.1.4. Calorific value

The high heating value (HHV) of densified biomass showed values between 19.64 MJ/kg for OL and 17.53 MJ/kg for OW (Table 30). Although the results were similar, there is a certain alteration due to the heterogeneity of the raw materials. This increase in the HHV can be caused due to the greater concentration of lignin in this type of biomass (Table 25), previously determined in Part 3, since lignin has a low degree of oxidation and a considerably high heat of combustion (Kataki and Konwer, 2001; Demirbas, 2004; Demirbas, 2005).

The heating values of the olive tree samples analyzed fall between the HHV of common oak and eucalyptus residues, which showed similar values to those obtained for OW, while grapevine and branches of cork oak (19.28 MJ/kg) showed approximately the same HHV than OL (Miranda et al., 2009).

Limits are established for the LHV, which has to reach a minimum of 16.0 MJ/kg to be used in non-industrial furnaces (EN 14961-2, 2011). Despite all the raw materials fulfill this requirement; OW would be classified as category C, while OL and OP are considered within category A1 in regards to this parameter (Table 14).



26.1.5. Sulfur and chlorine contents

The total chlorine contents were lower for OW (0.01%) and higher (0.03%) in OL. Due to its higher chlorine content, OL would be classified as category B, while OW and OP fulfill the requirement established for categories A1 and A2 (EN 14961-2, 2011).

The same tendency was found regarding the sulfur content with values ranging from 0.00 for OW to 0.11% for OL (Table 30). These values were close to the values reported in the literature for wood and woody biomass, being even in the case of OL lower than the mean values obtained for herbaceous and agricultural biomass (Lehtikangas, 2001; Vassilev et al., 2010). Nevertheless, only OW samples could be used in domestic systems (Table 14). The presence of leaves in OP samples makes this raw material show slightly higher sulfur content that the maximum allowed for a nonindustrial furnace and in consequence OP and OL would not be allowed to be used in this kind of furnaces.

26.1.6. Slagging and fouling tendencies

An ash-related problem normally experienced during the combustion of some agricultural residues is the low melting properties of the ash. The problems attributed to low melting temperatures of the ashes from these residues are fouling, slagging and corrosion of the heat transfer surfaces (Demirbas, 2005), which are due to the presence of very high contents of mineral oxides in some biomasses (Werther, 2000; Pronobis, 2005).

According to Equation 3, OP and OW presented high fouling inclination (0.6 < Fu <= 40), while OL presented a low tendency to cause fouling (Fu <= 0.6). On the other hand, and considering Equation 2, OL, OP and OW showed a high tendency to cause slagging (SR <= 65) (Tables 27 and 30). The results after applying these indices were inconsistent for the samples aim of the study because of being the woody portions more susceptible of producing slagging or fouling that the herbaceous samples (Bryers, 1996). In addition, Kuo et al., (2010) demonstrated that CaO improves the melting point

of the overall bed and inhibit sintering. Therefore, the presence of calcium has a positive effect on the suppression of agglomeration and defluidization.

Due to these facts and given that the indices used were originally designed for bituminous and a brown coal ash, another method for determining the fusibility of ashes was applied. By means of the sintering point testing equipment, it was possible to determine deformation temperature (DT), softening temperature (ST), hemisphere temperature (HT) and fluid temperature (FT). According to EN 14961-2 (2011) and despite the fact that no limits are still established, these temperatures should be declared for non-industrial pellets.

The results showed an inverse behavior except for the initial deformation temperature, which was higher for OL. Nevertheless, OL was the only sample with a fluid temperature lower than 1500°C (Table 30) and therefore the slagging and fouling tendencies were lower in the case of OP and OW samples despite having considerably higher K_2O and CaO content.



Figure 32. Samples of pellets from OL, OP and OW respectively

26.2. Characterization of the pellets

After the pelletization process, 81 different samples of pellets (27 samples for each raw material) were obtained (Figure 32). Their nomenclature is presented in Table 31, while the average values of quality parameters for every sample are shown in an annex in the end of this document (Table A1). Samples in Table 31 that do not appear in

		NTT T	THE A THOM		DAWATEDIAL	
MOISTURE CONTENT (M%)	TYPE OF DIE	LELL	EIIZATION	2	NAW MAIENIAL	
		TEM	PERATURE	TO	OP	MO
	¢	T1	<40°C	0L-M9-D1-T1	OP-M9-D1-T1	OW-M9-D1-T1
	D1 U=0 mm	17	40-60°C	0L-M9-D1-T2	OP-M9-D1-T2	OW-M9-D1-T2
		<u>р</u>	>60°C	0L-M9-D1-T3	OP-M9-D1-T3	OW-M9-D1-T3
		E		OL-M9-D2-T1	OP-M9-D2-T1	0W-M9-D2-T1
9M	$D_2 \frac{U=0}{1-24}$	1 2		0L-M9-D2-T2	OP-M9-D2-T2	OW-M9-D2-T2
	ШШ +7—Л	T 3		OL-M9-D2-T3	OP-M9-D2-T3	OW-M9-D2-T3
	Q	T1		OL-M9-D3-T1	OP-M9-D3-T1	OW-M9-D3-T1
	$D_{3} \frac{D-6}{1-20} mm$	T2		OL-M9-D3-T2	OP-M9-D3-T2	OW-M9-D3-T2
		T 3		OL-M9-D3-T3	OP-M9-D3-T3	OW-M9-D3-T3
		T1		OL-M11-D1-T1	OP-M11-D1-T1	OW-M11-D1-T1
	D1	T2		OL-M11-D1-T2	OP-M11-D1-T2	OW-M11-D1-T2
		$\mathbf{I3}$		OL-M11-D1-T3	OP-M11-D1-T3	OW-M11-D1-T3
		T1		OL-M11-D2-T1	OP-M11-D2-T1	OW-M11-D2-T1
M11	D2	T2		OL-M11-D2-T2	OP-M11-D2-T2	OW-M11-D2-T2
		$\mathbf{T3}$		OL-M11-D2-T3	OP-M11-D2-T3	OW-M11-D2-T3
		T1		OL-M11-D3-T1	OP-M11-D3-T1	OW-M11-D3-T1
	D3	T2		OL-M11-D3-T2	OP-M11-D3-T2	OW-M11-D3-T2
		$\mathbf{T3}$		OL-M11-D3-T3	OP-M11-D3-T3	OW-M11-D3-T3
		T1		OL-M13-D1-T1	OP-M13-D1-T1	OW-M13-D1-T1
	DI	T2		OL-M13-D1-T2	OP-M13-D1-T2	OW-M13-D1-T2
		13		OL-M13-D1-T3	OP-M13-D1-T3	OW-M13-D1-T3
		T1		OL-M13-D2-T1	OP-M13-D2-T1	OW-M13-D2-T1
M13	D2	T2		OL-M13-D2-T2	OP-M13-D2-T2	OW-M13-D2-T2
		13		OL-M13-D2-T3	OP-M13-D2-T3	OW-M13-D2-T3
		T1		OL-M13-D3-T1	OP-M13-D3-T1	OW-M13-D3-T1
	D3	T_2		OL-M13-D3-T2	OP-M13-D3-T2	OW-M13-D3-T2
		13		OL-M13-D3-T3	OP-M13-D3-T3	OW-M13-D3-T3

Table 31. Symbols for and explanations of different assortments

the final annex could not be produced under the corresponding pelletization

temperatures.

26.2.1. Durability

The durability values for the OL samples ranged between 14.58 % and 88.63%, not reaching in any case the minimum value for their use in non-industrial boilers (Table 32) because of generating more than 3.5 % of fines during transport and handling.

65.4 % of OP pellets produced could be used in domestic boilers, although 23.1% of them would be classified as category B because of producing between 2.6 and 3.5 % of fines (Table 14). OW were the samples with higher durability, being possible to use 91.7 % of the pellets produced in non-industrial furnaces as class A1/A2 pellets (EN 14961-2, 2011).

Table 32. Percentage of pellets samples that fulfill the requirements established in EN14961-2 (2011) for pellet quality

	Pellet samples (%)											
Parameter			OL			OP				()W	
	A1	A2	В	NC	A1	A2	В	NC	A1	A2	В	NC
Durability	(0	0	100	42.3 23.1		23.1	34.6	91.7 0		0	8.3
Diameter	100 0		100		0		100		0			
Length	100 0		100		0		100		0			
Bulk Density		0		100	38.5		61.5	83.4			16.6	
Moisture Content	63.2		36.8	84.6		15.4	83.4		16.6			

*NC: non categorized

26.2.2. Hardness or compression strength

Hardness is not limited by norm but it has been found to be especially important during the storage and feeding processes (Li and Liu, 2000; Gilbert et al., 2009).

Pellets made from OL reached a maximum of 13.33 kgf, being the average value of 7 kgf, a very low value when it is compared to those obtained for OP and OW, which showed values between 21 and 24.86 kgf, similar to those obtained in other studies (Stelte et al., 2011).

26.2.3. Pellet size, single pellet density and bulk density

Since all the pellets were made with pellet dies of 6 and 8 mm of diameter, they fulfilled the requirements established for this parameter. The same occurred with the pellet length, which ranged between 12.29 mm for OL and 28.74 mm for OW. Despite the differences, all the pellets tested showed a length within the range between 3.15 and 40 mm established by EN 14961-2 (2011).

The ratio length/diameter was calculated because of being a guideline in EN 14961-1 (2010), which establishes 5 as a maximum value for diameters of 6 mm and 4 for those of 8 mm of diameter. All the samples fulfilled this guideline, with an average value of 2.6 for OL and 3.5 for OP and OW.

Regarding the pellet density, no limits are established. Only OL samples showed particle densities lower than 1000 kg/m³, which is the average value obtained in other studies (Li and Liu, 2000; Lehtikangas, 2001; Rhén et al., 2005; Kaliyan and Morey, 2009), as well as for the OP and OW pellets.

Bulk density is an important parameter because of being related to the space required for storage and transport, as well as to the derived cost for transport. EN 14961-2 (2011) establishes a minimum for non-industrial pellets of 600 kg/m3 (Table 14). This requirement was not fulfilled by any of the samples from OL, while higher bulk densities were obtained by 38.5 % of pellets from OP and 83.4 % of pellets made from OW (Table 32), showing similar values than pellets from fresh logging residues (Vinterbäck, 2004), other wood pellets (Obernberger and Thek, 2004) or pellets from some herbaceous crops (Gilbert et al., 2009).

Energetic density takes into account not only the bulk density but also the low calorific value. It makes sense since the densification of biomass is a process that makes possible the storage of energy in less space than biomass not densified or pelletized. Nevertheless, no guidelines are established for this parameter. EN 14961-2 (2011) limits

the calorific value with a minimum of 16 MJ/kg and bulk density in 600 kg/m³ as the minimum value. Though bulk density was a parameter not fulfilled for OL, the energetic density of these samples can be higher than that that obtained by OW pellets despite having higher bulk densities. Obviously this is due to the fact that OL has a LHV higher than OW. Consequently, in the same space, it would be possible to store more energy with OL than with OW.

26.2.4. Moisture content

The moisture content of the pellets is limited by EN 14961-2 (2011) with a maximum value of 10% (Table 14). Pellet samples with moisture content higher than 10% were made from raw materials at 13% of moisture content and pelletization temperatures lower tan 60°C, impeding the water removal. 62.5% of pellets made from OL fulfill this requirement, while more than 80% of pellets made from OP and OW showed moisture contents lower than this limit (Table 32).

26.3. Optimum parameters for the pelletization of residual biomass from olive trees

For the determination of the optimum parameters for every raw material analyzed it was necessary to determine the KMO index.

Table 33 shows the KMO value for each raw material. Since no high enough KMO values (0.8-1.0) were obtained (Table 29), the statistical analysis has to consider all the initial variables.

Raw material	I	KMO
OL	0.783	Medium
OP	0.553	Low
OW	0.618	Mediocre

Table 33. KMO values obtained for the raw materials aim of study

The least squares method was applied since the number of samples is small and the ideal values are known (Table 28), so an exact answer can be obtained.

Firstly, all the variables were normalized considering the interval within each variable could range (Table 28), and they were changed to a unit interval.



Secondly, the most adequate sample was selected as the solution of a minimization problem. The minimization of the quadratic functional is shown in Equation 6, where $f(\bar{x})$ represents the scalar product of two vectors, \bar{x}_o represents the vector of optimum components and \bar{x} represents the independent variable.

$$f(\overline{x}) = (\overline{x} - \overline{x}_o, \overline{x} - x_o)$$
 (Equation 6)

Therefore, the problem to solve is: *Minimize* $f(\overline{x})$, $\overline{x} \in M$, where M is the set of analyzed samples. This problem has a solution because the set is closed and finite.

The solutions obtained for each raw material are collected in the Table A2 in the annex at the end of the document. The optimum conditions for the production of pellets from every raw material considered are the solution of the indicated problem, being also necessary the fulfillment of the requirements established in EN 14961-2 (2011).

It can be observed that in the case of OW, the sample solution of the problem (OW-M9-D1-T2) fulfills the quality parameters (Table 14) for non-industrial pellets. For OP samples, the solution (OP-M11-D1-T1) does not achieve the minimum value established for the bulk density (Table 14), so the following sample (OP-M9-D2-T3) was considered as the optimum OP pellet sample. In the case of pellets made from OL, none of the pellets fulfills the requirements established for bulk density and durability (Table 32), so the solution of the problem (OL-M9-D1-T3) has been considered as the optimum sample since the rest of the parameters fulfills the requirements established (EN 14961-2, 2011).

27. Conclusions

Regarding the chemical properties and due to the fact that these properties are dependent on the nature of the raw materials, OP and OL exceeded the values for ash, nitrogen and sulfur that the guidelines established for non-industrial pellets.

Though the calorific value of OL was higher than that of OP and OW, pellets made from OL did not reach the minimum value of bulk density established for nonindustrial pellets. Nevertheless, if the energetic density was considered, samples made with OL could show higher values than those from OP and OW. The results obtained after calculating the slagging and fouling indexes were not consistent when they were compared to the melting temperatures of the samples tested, being the fusion temperature higher in the case and OP and OW.

After the pelletization tests, no samples from OL fulfilled the requirements of durability and bulk density established for non industrial pellets, being the sample OL-M9-D1-T3 the one with the best quality of the rest of OL pellets.

While in the case of OW the sample solution of the least squares method fulfilled the requirements established in EN 14961-2 (2011) for non-industrial pellets, in the case of OP pellets, the solution could not be selected as the optimum sample from OP because of presenting a bulk density lower than 600 kg/m^3 .

From the optimum samples selected, it can be concluded that low moisture content (9%) is the most adequate for producing pellets from the different residual biomass from olive trees, while the compression length and the pelletization temperature has to be adapted to the biomass to be pelletized. Short compression lengths are suitable for producing high-quality pellets from olive trees residual biomass. In the case of OP and OL, temperatures higher than 60°C are necessary to improve the quality of the pellets, while temperatures within the range 40-60°C are adequate to produce pellets from OW.



PART 5

ANALYSIS OF THE COMBUSTION OF PELLETS MADE FROM RESIDUAL BIOMASS FROM OLIVE TREES IN A DOMESTIC BOILER ¹¹

¹¹ The results shown in this part were presented in: García-Maraver A, Zamorano M, Fernandes U, Rabaçal M, Costa M. Particulate emissions, combustion behavior and quality parameters of agropellets burned in a domestic pellet-fired boiler. Submitted to Applied Energy



28. Introduction

As it was described in the previous chapter, the final quality of pellets varies depending on the raw material properties and the manufacturing process (Gilbert et al., 2009; Samuelsson et al., 2009). Several studies (Gonzalez et al., 2005; Gonzalez et al., 2006; Tissari et al., 2008; Mediavilla et al., 2009; Garcia-Maraver et al., 2012; Rabaçal et al., 2013; Verma et al., 2012) have shown that non-conventional raw materials, such as agricultural residues or energy crops, have a considerable potential to be used in pellets fired boilers. However, these low-quality biofuels in terms of chemical composition can damage combustion equipment and produce undesirable effects such as slagging, fouling or corrosion (Lehtikangas, 2001). Furthermore, physical and mechanical properties such as the content of fine particles in the fuel, which is extremely dependent on mechanical durability as well as logistics and storage, can disturb the control of highly automated heating systems or interrupt automated fuel feeding (Li and Liu, 2000).

Regardless of the pellet type, pollutant gaseous emissions are significantly affected by inadequate operating conditions, specifically CO and HC, due to poor mixing, low temperatures and short residence times (Dias et al., 2004; Rabaçal et al., 2013; Verma et al., 2012). Moreover, since combustion of biomass fuels generates substantial amounts of fine particles (particles smaller than 1 μ m in aerodynamic diameter), the interest has shifted towards particle emissions due to a need of deep understanding on particle formation when burning different types of pellets. If no effective flue gas cleaning systems are in use, most of these particles are released into ambient air, affecting air quality and the climate system.

Particulate air pollution is an important health concern worldwide (Samet et al., 2000). Furthermore, it has been observed that in the case of combustion-related particles, the fine particle fraction (PM1 or PM2.5) is especially harmful to human health (Schwartz et al., 1996). Residential biomass combustion has been accused of producing harmful particulate pollutants, since it generates high amounts of fine particles at low emission heights (Titta et al., 2002; Boman et al., 2003; Karvosenoja

and Johansson, 2003). Consequently, many European countries have developed standards for the quality, storage, transport, and combustion of densified biomass fuels (García-Maraver et al., 2011). Nevertheless, no standard has been developed regarding particle emissions from biomass residential boilers.

The aim of this chapter is to evaluate the combustion behavior of pellets from different raw materials with special focus on agro-pellets and PM emissions. Commercial pine pellets from Portugal and Spain were selected as reference raw material. Cork pellets were produced by a company using residues from the cork producing process. Olive grove residues, which present a high potential for energetic valorization in Andalusia (Garcia-Maraver et al., 2012), were collected near Granada (Spain) and densified in a pilot scale pelletizer as it was described in the previous chapter.

Subsequently physical and chemical properties of pellets were analyzed and then burned in a small-scale pellet boiler. Gaseous and PM emissions concentration and size distribution were characterized.

29. Materials and methods

29.1. Pellet fuels tested

Olive prunings and olive wood were obtained from an olive grove near Granada (Spain) as residual biomass. Olive pruning was composed by leaves and small branches that were harvested as a whole, whereas olive wood was collected in logs, both in March 2012. Both types of pellets were made under the optimum conditions calculated in the previous chapter, assuring that physical and mechanical requirements established for non-industrial pellets were fulfilled (Table 14). Since no samples from olive leaves fulfilled bulk density and durability parameters (Table 32), they were not tested.

On the other hand, two samples of different commercial pellets made from pine sawdust, one from Portugal (Pine Port) and one from Spain (Pine ES), and one



commercial sample of cork pellets were also analyzed in terms of pellet quality and combustion behavior.

The characteristics of the pellets were tested according to the norms described in the previous chapter and they are summarized in Table 34 below.

Parameter	Pine Portugal	Pine Spain	Cork	Olive Wood	Olive Pruning
Proximate analysis (% wt, as					
received)					
Volatiles	82.3	78.1	79.3	82.0	74.7
Fixed Carbon	8.8	10.8	10.8	9.0	11.8
Moisture	8.1	10.1	7.9	7.6	8.0
Ash	0.8	1.0	2.0	1.4	5.5
Ultimate analysis (% wt, dry ash					
free)					
Carbon	46.0	45.7	55.9	46.5	47.1
Hydrogen	6.2	6.6	5.6	6.8	6.8
Nitrogen	0.5	0.3	0.4	0.3	1.1
Sulphur	< 0.01	0.02	0.12	< 0.01	0.05
Oxygen	47.3	47.4	37.9	46.4	44.9
Low heating value (MJ/kg)	17.1	16.9	19.1	16.0	16.5
Particle density (kg/m^3)	1198	1021	1204	1327	1268
Durability (%)	96.77	94.88	97.74	98.09	98.92
Bulk Density (kg/m ³)	627	544	640	638	610
Crushing resistance or hardness (kg)	22.16	20.33	21.83	23.17	21.83
Ratio L/D	3.24	1.52	3.20	3.64	3.86

Table 34. Characteristics of the pellets used in this study.

29.2. Combustion studies

29.2.1. Experimental set-up

Experimental tests for this study were carried in a domestic wood pellet-fired boiler with a maximum thermal capacity of 22 kW (Figure 33). The boiler is top fed with forced draught. The pellets are manually loaded into a hopper with a capacity of 45 kg and are fed to the burner through a screw feeder that works by impulses. The feeding rate of the pellets is regulated by the boiler load and is measured with the aid of a loss-in-weight technique, for what the boiler is mounted on a weighbridge (Fernandes and Costa, 2012).



Figure 33. Schematic of the experimental set-up (Fernandes and Costa, 2012)

The combustion of the pellets takes place within a hemispherical basket (brazier) with a diameter of 120 mm. The basket is top-fed with pellets by a screw. Ignition is accomplished with the aid of an electrical resistance placed close to the basket and the primary air is supplied by a dedicated fan to the basket through several small orifices located across the bottom of the basket. The secondary air is supplied through a vertical tube, located slightly above the basket, which injects the air in the same direction as the exiting flow of the basket. The resulting hot gases from the combustion exchange heat with water circulating in a heat exchanger located at the top of the combustion chamber.

It should be pointed out that a short cleaning period of the basket is programmed to occur once every 11.5 min. During the cleaning process the fuel supply decreases and the air supply increases during few minutes in order to remove the ash accumulated at the bottom of the basket (bottom ash).

The heat generated by combustion is transferred to an inner water circuit and it is dissipated through a plate heat exchanger with the aid of an external water circuit.



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The water flow rate circulating in the inner circuit of the boiler was measured with a rotameter and the inlet and outlet temperatures were measured with thermocouples type K, T1 and T2 in Figure 33, respectively. Temperature at the flue gas exit of the boiler, T3 in Figure 33, was also measured with a thermocouple type K.

29.2.2. Gaseous sampling and analytical techniques

Flue-gas was sampled for gaseous pollutants. The gases were collected by means of a stainless-steel probe and then conditioned by a sampling train that included a condenser, silica gel and a cotton filter before being fed to the analytical instrumentation: (i) a magnetic pressure analyzer for O_2 measurements, (ii) a nondispersive infrared gas analyzer for CO and CO₂ measurements, (iii) a flame ionization detector for HC measurements and (iv) a chemiluminescent analyzer for NO_x measurements.

Zero and span gas calibrations with standard mixtures were performed before and after each measurement session.

29.2.3. Particulate matter sampling and analytical techniques

For PM emissions characterization the flue gas was sampled isokinetically from the centerline of the exhaust pipe. Two low pressure impactors were used to collect PM by several particle cut sizes during the same sampling.

A three-stage low pressure impactor (LPI-3) TCR Tecora (Figure 34a) allowed collection within the following cut off sizes: PM with diameters above 10 μ m (PM10), PM with diameters between 2.5 μ m and 10 μ m, and PM with diameters below 2.5 μ m (PM2.5).

In order to avoid condensation along the line connecting the probe outlet to the impactor inlet and also inside the impactor, a heating jacket Winkler WOTX1187 was used during sampling, heating the sample up to 150°C. A constant suction flow rate of ~ 7 Nl/min was used. PM was collected on quartz microfiber filters (Figure 34b), which were dried in an oven and weighted before each test. After each test, the filters were again dried, to eliminate moisture, and weighted to determine the quantity of PM captured.



Figure 34. a) Section of the LPI-3 impactor used in this study; b) Quartz microfiber filters after testing

A 13-stage Dekati low-pressure cascade impactor DLPI (LPI-13) Dekati Ltd (Figure 35a) allowed to size-classify the PM according to the cutoff aerodynamic diameters listed in Table 35. The cut off diameter is defined as the size of particles collected with 50% efficiency. PM size fractions were determined by weighing each stage's substrates due to bounce and blow off some of the bigger particles might go to the lower stages and due to diffusion some of the smaller particles might be collected in upper stages than they should.



Figure 35. a) LPI-13 impactor used in this study; b) Aluminium film after testing



Greased thin aluminum films (Figure 35b) were used as impactor substrate in order to minimize the bouncing effect and collection time was short enough to avoid filter saturation, which could lead to the occurrence of bouncing effect, but long enough to collect a representative sample. Diffusion has a negligible effect on the results since the mass of smaller particles is often insignificant when compared to the actual impacted mass. A small fraction of PM may also be collected in other surfaces such as walls. The first (top) 6 plates operate close to atmospheric pressure, thereafter the pressure decreases over the final 8 plates through the impactor to a final pressure of 10 kPa. The flow through the impactor was kept constant at a value of ~10 Nl/min and outlet pressure of the impactor was also measured to ensure proper operation of the impactor throughout the tests.

Table 35. Calibrated cutoff diameters, d50, of the 13-stages Dekati low-pressure cascade impactor

Stage	1	2	3	4	5	6	7	8	9	10	11	12	13
d ₅₀ (μm)	0.0282	0.0559	0.0927	0.153	0.259	0.379	0.608	0.940	1.59	2.37	3.96	6.63	9.84

Fuels	Thermal input (kW)	O _{2 flue-gas} (% dry volume)	T _{flue-gas} (K)
	10	18.5	130.6
Pine Portugal	14	17.5	147.4
	17	18.0	159.0
	10	19.2	108.7
Pine Spain	14	18.4	143.6
	17	17.9	160.2
	10	18.8	128.8
Cork	14	17.8	159.4
	17	17.3	155.3
	10	18.8	126.3
Olive Wood	14	18.2	149.0
	17	17.4	166.4
Olive	10	18.6	128.0
Dilve	14	18.3	149.1
Pruning	17	17.4	164.0

Table 36. Operating conditions tested in this study

29.3. Conditions studied

Table 36 lists the operating conditions studied. Tests were performed for three operating conditions corresponding to low, medium and high thermal input, for each type of pellets. For all operating conditions, gaseous and particle emissions measurements were performed simultaneously. The LPI-3 was used in all operating conditions, whereas the LPI-13 was only used in the intermediate load.

30. Results and discussion

30.1. Gaseous emissions

Figure 36, Figure 37 and Figure 38 show, respectively, the CO, HC and NO_x emissions of the different types of pellets for all operating conditions studied.



Figure 36. CO emissions of the different pellets for all operating conditions

Pine based pellets consistently present lower emissions of all pollutants emissions than agro-pellets, as expected, in accordance with previous work (Boman et al., 2004; Dias et al., 2004; Rabaçal et al., 2013; Verma et al., 2012). Among agro-pellets, cork pellets present the lowest emissions followed by olive wood pellets and finally olive pruning pellets. Furthermore, pellet type has a more pronounced influence

on gaseous emissions than operating conditions, which was also observed in previous work (Boman et al., 2004; Dias et al., 2004; Rabaçal et al., 2013; Verma et al., 2012).



Figure 37. HC emissions of the different pellets for all operating conditions

In the case of CO emissions, pine based pellets and cork pellets show the same trend, presenting a minimum at medium load, whereas CO emissions from olive based pellets consistently decrease when the thermal input increases. HC emissions follow the same trend as CO, as expected since both species are related with incomplete combustion (Jenkins et al., 1998).

In fuels with a high volatile content, the volatile combustion phase is dominant, which is characterized by a rapid and significant heat release contrary to the long heterogeneous char phase (Jenkins et al., 1998) that occurs with fuels with lower volatile content, such as olive pruning (Table 34). However the volatiles in olive wood pellets have a lower calorific value than pine based pellets and cork pellets, as typified by the low heating value (Table 34). The higher low heating value of pine based pellets and cork pellets contribute to their superior performance as more energy is released during combustion.

Although cork pellets present the highest low heating value (Table 34), the results show a slightly lower performance when it is compared with pine based pellets

(Figure 37). This could be due to the higher particle density of cork pellets (Table 34), which can make necessary a longer burnout time (Obernberger and Thek, 2004). This fact also affects to the combustion performance of olive based pellets, which are the samples with higher particle density (Table 34). Note that this parameter that has an obvious effect on the combustion process is not limited by the European standards (Table 14) although Austrian, German and French norms establish a maximum of 1100-1400 kg/m³ (Tables 8, 10 and 12). In the case of olive wood pellets, their higher hardness value (Table 34) also resulted in a difficult operation of the boiler during the combustion tests due to the occasional blocking of the screw conveyor. Note that hardness, which is not limited by any pellet quality norm, negatively affects to the feeding process. In addition, particle density has an obvious effect on the combustion process but is not limited by the European standards (Table 14), although Austrian, German and French norms establish a maximum of 1100-1400 kg/m³ (Table 8, Table 10 and Table 12).



Figure 38. NO_x emissions of the different pellets for all operating conditions

 NO_x emissions show a clear dependence on fuel's nitrogen content and there is no significant influence of operating conditions, as seen in a previous work (Rabaçal et al., 2013). Fuel-N is the main mechanism of NO_x formation in this type of boilers, as temperatures inside the combustion chamber are too low for the occurrence of the

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thermal mechanism (Jenkins et al., 1998; Verma et al., 2013). Therefore, as it can be observed, NO_x emissions from olive pruning pellets were considerably higher than in the rest of samples tested due to the high nitrogen content of this raw material (Table 34).

30.2. PM LPI-3 emissions

Figure 39 shows the PM emissions distribution obtained with the LPI-3 of the different types of pellets for all operating conditions showing the total PM emissions divided by size fractions.

In the open literature it is possible to observe a wide range of total PM emissions (Ghafghazi et al., 2011; Verma et al., 2011; Fernandes and Costa, 2012; Limousy et al., 2012; Verma et al., 2012), which is also related to the burner configuration, ranging from 5 to 800 mgNm⁻³ (values corrected to 10% O_2 in the flue gas), but in average PM total emissions are typically between 50 and 100 mgNm⁻³ in the case of pine pellets.

The values observed in this work, between 20 and 200 mgNm⁻³, are consistent with related previous works (Verma et al., 2011; Fernandes and Costa, 2012; Limousy et al., 2012; Verma et al., 2012) and analogously to gaseous emissions presented in the previous sections, it is observed that the type of pellets has a more significant impact on PM emissions than operating conditions. It is possible to observe that pine based pellets produce the lowest total PM emissions and, among agro-pellets, cork pellets produce the lowest total PM emissions followed by olive wood and finally olive pruning. Nevertheless, total PM emissions by agro-pellets are somewhat sensitive to operating conditions, significantly more in the case of olive pruning (Figure 39).

It is significant that pine ES pellets, which showed the lowest durability (Table 34), not reaching the minimum value for non-industrial pellets (EN 14961-2, 2011), did not generate higher amounts of PM, as was expected because of being an indicator of the fine particles in the fuel (Li and Liu, 2000).

Typically, pellets fired boilers are prone to emit particulate emissions originated by poor local combustion conditions, soot articles and aggregates, so it is not surprising that, such as in the case of unburned gaseous emissions, the pellets with the lower volatile content produce higher total PM emissions than pellets with the higher volatile content. This correlation seems reasonable for each operating condition, but inspecting the evolution of total PM emissions it is observed that although at medium and higher operating load combustion conditions seem to improve, as typified by CO and HC emissions (Figure 36 and Figure 37), total PM emissions increase for all types of pellets (Figure 39).

It is well established (Wiinikka, 2005; Ghafghazi et al., 2011) that there are two main primary sources for particulate matter formation in biomass combustion: (i) incomplete combustion due to inadequate combustion conditions, originating soot and unburned char; and (ii) presence of inorganic matter that constitutes the ash content of biomass. In a previous study performed in this boiler burning pine pellets (Sippula et al., 2007) it was observed that PM>10 is mainly composed by unburned particles ejected from the brazier, hence it seems reasonable to assume that this is the case in this study. In the case of olive pruning, its much higher ash content caused the obstruction of the fuel bed, impeding air to flow. Therefore it was necessary to periodically shake the fuel bed, causing some disturbances and contributing to a high release of particles that got entrained in the air flow which explain the much higher emission of PM>10 when compared with the other types of pellets (Figure 39).

For all types of pellets, PM<2.5 are dominant and 2.5<PM<10 are marginal. It is very interesting to note that the increase of total emissions is due to a significant increase of PM<2.5. Given the high sensitivity of PM<2.5 emissions to the operating conditions and its negative impact to the environment and health (Schwartz et al., 1996; Samet et al., 2000), a detailed test was performed on this size fraction, presented in the section below.



30.3. PM LPI-13 emissions

Fine particles (equivalent aerodynamic diameter $< 2.5 \ \mu$ m) emitted from residential pellets combustion appliances may be divided into three typical classes based on chemical composition and morphology: (i) aggregated soot particles, (ii) spherical inorganic particles and (iii) aggregated inorganic particles. Furthermore, within this size range, a bimodal size distribution has been described systematically, with sub-micron and ultra-fine modes (Boman et al., 2004; Wiinikka, 2005; Wiinikka, 2006; Sippula et al., 2007; Fernandes and Costa, 2012).



Figure 39. PM LPI-3 emissions of the different pellets for all operating conditions

Figure 40 shows the particle mass size distribution obtained with the LPI-13 of the different types of pellets for the medium load operating condition. With exception of the case of olive pruning, the particle mass size distribution has a unimodal characteristic, as observed by other authors at the exhaust of combustion appliances (Boman et al., 2004; Wiinikka et al., 2006; Sippula et al., 2007; Bafver et al., 2009; Limousy et al., 2012; Roy and Corscadden, 2012). Olive pruning, on the other hand, produced particle emissions with a trimodal mass size distribution, obtained in all repeated tests and possibly as a consequence of fuel bed disturbances.

The particle mass size distributions were all dominated by fine submicron particles but with different dominating modes between pellets type. In the case of pine based pellets, particle mass size distribution was dominated by particles with mass median (aerodynamic) diameters (MMD) of 0.056-0.153 μ m (substrate 3), whereas in the case of cork a wide range of particles modes is observed with dominating MMD of 0.259 μ m. In the case of olive wood, the particle mass size distribution is dominated by particles with MMD of 0.153-0.608 μ m. Finally, in the case of olive pruning the three dominating MMD modes are 0.093, 0.259 and 0.608. As seen in Figure 39, cork, olive wood and olive pruning pellets lead to high emissions of PM, when compared to Pine PT and Pine ES.

Inorganic particles are initially formed by homogeneous nucleation, when inorganic vapors reach condensation temperature and particle concentration is low, which results in super fine particles. As particle concentration tends to increase, heterogeneous condensation in existing particles occurs as well as particle agglomeration (Jenkins et al., 1998). Therefore, in the case of cork, olive wood and olive pruning, due to the higher concentrations of emitted particles as seen in Figure 39, particle growth occurs, which explains the higher MMD dominating modes for these types of pellets observed in Figure 40.



Figure 40. PM mass size distribution emissions of all pellets for the medium load operating condition

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In order to better understand the formation mechanism behind the high PM emissions observed in olive based pellets, morphology and chemical analysis were performed to a number of representative substrates. Figure 41to Figure 43 show typical SEM images of different subtracts. Note that particles were collected at the exhaust where temperature is considerably lower than in the combustion chamber and at this point residence time is long enough to consider that any further condensation or agglomeration occurs when the particles are sampled.



Figure 41. Typical SEM images of substrate 3: a) Pine Portugal, b) Pine Spain

Figure 41 and Figure 42 show typical sub-micronic particles collected in subtract 3 for pine based pellets and agro-pellets, respectively. In both figures a large number of soot aggregates can be observed, but the presence of condensed inorganic species over soot aggregates is more noticeable in Figure 42 owing not only to higher particle concentration but also to a higher inorganic vapor concentration arising from the fuel bed (Jenkins et al., 1998) due to the high ash content of agro-pellets (Table 34).



Figure 42. Typical SEM images of substrate 3: a) cork, b) olive wood, c) olive pruning

Figure 43 shows typical SEM images of Olive wood – substrate 6 and Olive pruning – substrates 7 and 10. Again, soot aggregates are present but crystalline structures can be clearly identified. Note that in the case of Olive pruning – substrate 10 it is possible to identify spherical inorganic particles. The observed particles are a result from particle growth through aggregation and heterogeneous condensation (Jenkins et al., 1998; Verma et al., 2012).



Figure 43. Typical SEM images: a) Olive Wood substrate 6, b) Olive Pruning substrate 7, c) Olive Pruning substrate 10

Figure 44 and Figure 45 show the PM chemical composition for several substrates for Olive pruning, and substrate 3 for the remaining types of pellets. Coarse fly ash particles originate from fragments of ashes of the burning pellets in the fuel bed that are entrained in the flue-gas flow (Jenkins et al., 1998).

Figure 44 shows that the composition of PM under 1 μ m from Olive pruning combustion varies marginally, in terms of present elements and in relative amounts of each element. Besides the presence of carbon from incomplete combustion (Figure 36 and Figure 37), high amounts of K, S and Cl and a small amount of Na from the inherent inorganic composition of the Olive pruning (Table 30) are observed in Figure 44. The very high calcium content of olive pruning and olive wood (Table 30) is not reflected in the composition of the smallest particles because this component remains in the PM larger than 1 μ m (Figure 44 and Figure 45) and in the bottom ashes.

Figure 44 also shows that for PM larger than 1 μ m, besides the presence of the above-mentioned species, Mg, Ca, P, Fe and Si are present.





Figure 44. PM chemical composition in several substrates collected from the combustion of olive pruning pellets



Figure 45. PM chemical composition in substrate 3 collected from the combustion of pine based, cork and olive wood pellets

31. Conclusions

At maximum loads, the gaseous emissions related to an incomplete combustion (CO and HC) were reduced, but the particulate emissions increased, being necessary to find an equilibrium point. This behavior is affected by the fact that this boiler is not designed for the combustion of agricultural pellets with high ash contents, which was especially observed when olive pruning was burnt.

While $PM < 10 \mu m$ emissions were not affected by the boiler operating loads, $PM > 10 \mu m$ increased with the operating load of the boiler since their emission is related to the presence of unburnt particles.

The optimum operating conditions of the pellet fired boiler are bound to the raw material used as biofuels, having more influence than the boiler load on the gaseous and particulate emissions. Therefore, the combustion behavior observed makes evident the need to design a boiler for the type of biomass that will be used as fuel.



CONCLUSIONS

The most important contributions that deduced from this work can be classified in five sections:

- (i) The evaluation of the potential of residual biomass from olive trees and the benefits derived from its use as a renewable energy source in Mediterranean areas, more concretely in the case of Andalusia.
- (ii) Comparative analysis of the parameters used to establish the quality and use of pellets at a European level.
- (iii) The physical and chemical characterization of residual biomass from olive trees, according to the requirements established for the use of solid biofuels at a non-industrial scale.
- (iv) The determination of the optimum variables to produce pellets from residual biomass from olive trees.
- (v) Determination of the combustion behavior of pellets made from residual biomass from olive trees under real conditions in a domestic boiler.

Regarding the evaluation of the potential and benefits of residual biomass from olive trees as a renewable energy source in Andalusia, it has been concluded that:

- Andalusia is very rich in renewable resources, especially in biomass, with a potential of about 3327 ktoe/year, providing 6.3% of the toal primary energy consumption and 78.7% of the renewable energy consumption.
- Biomass potential from agricultural residues is estimated on 1434 ktoe, of which approximately 50% is olive tree residues (803 ktoe/year). In particular, there are 1.4 million hectares of olive crops in Andalusia producing an average of three tons of pruning residues per hectare, resulting in about four million tons per year in Andalusia. Most of them are now illegally burnt or left on the ground.



• Residual biomass from olive trees can be used to generate electricity, thermal energy and biofuels for transportation, depending on the characteristics of the raw material. Andalusian government objectives have been fulfilled regarding the use of biomass to produce electrical energy, however the use of biomass for thermal energy is still considerably below the levels desired. In consequence the production of densified biomass should be increased in Andalusia in order to meet the demand for space and domestic water heating in the building sector.

With regards to the comparative analysis of the parameters used to establish the quality and use of pellets at a European level, it has been concluded that:

- The only countries that have quality standards for pellets are Austria, Sweden and Germany, whereas France and Italy have published some recommendations. The comparative analysis of these standards and recommendations for pellets highlights significant differences in their quality parameters as well as in their guidelines.
- At a European level, the European standard EN 14691-2 (2011) regulates the quality parameters for non-industrial pellets. This common standardization will be beneficial for Europe since it will guarantee a market for high quality pellets.
- No guidelines have been established for pellets made from agricultural residues, which may significantly differ from those used as a reference for the limits established. Therefore this means that there is an important barrier for the use of agricultural residues.

Concerning the physical and chemical characterization of residual biomass from olive trees in comparison to other agricultural and forestry biomasses, and according to the established requirements for non-industrial pellets, the following conclusions were deduced:

• The use of residual biomass from olive trees results in different physical and chemical properties of pellets imposing restrictions on application of the pellets,

especially because of its higher ash content, also observed in other types of residual biomass from agricultural activities, which would lead to more maintenance problems in domestic heating systems.

- The characterization of the different fractions that are obtained from olive tree pruning showed differences among them, being higher the ash content for the leaves (8.9%), followed by the prunings (6.3%). In both cases biomass was not appropriate for domestic heating systems according to the current framework. Woody fraction showed a lower ash content (0.98%), fulfilling the requirements of EN 14961-2 (2011) for their use in non-industrial systems. This norm establishes three categories according to the values of some parameters; ash content is one of the parameters considered, being accepted to be within the maximum values of 0.7% for the category A1 and 3.0% for class B.
- The higher ash content of residual biomass from olive trees was not a result of the mineral contamination of Mg, Ca and K from the soil during the handling of biomass at the point of origin. The inherent composition of the biomass, with high lignin content, was responsible for this fact. Lignin increases the calorific value of the raw materials and favors the pelletization process since it works as a binder. Nevertheless, the decomposition temperature of lignin is higher that 900°C, and thus it can be considered as the main component responsible for the higher ash content of olive leaves and their low reaction rate.
- The results obtained after calculating the slagging and fouling indexes in the bibliography consulted were compared to the melting temperatures of the samples tested, and the results were inconsistent. Therefore it is necessary to establish new procedures to calculate these indexes.

In relation to the determination of the optimum operating conditions for pelletization regarding moisture content of the raw material, size of the die (diameter and compression length) and pelletization temperature, the following conclusions were obtained after the analysis of residual biomass from olive trees:


- It was possible to optimize the pelletization operating conditions in order to produce pellets from olive wood that fulfilled all the guidelines for the chemical, physical and mechanical parameters established in EN 14961-2 (2011) for non-industrial pellets.
- No pellets made from olive pruning could fulfill the chemical properties established in EN 14961-2 (2011) since their ash, sulfur and nitrogen contents were higher than the limits. Nevertheless, it was possible to optimize the production properties to achieve the physical and mechanical requirements.
- No pellets made from olive leaves fulfilled the chemical, physical and mechanical requirements, since they exceeded the limits established in EN 14961-2 (2011) concerning ash, nitrogen and sulfur contents, as well as durability and bulk density. Nevertheless, and given the fact that the calorific value of olive leaves was higher than that of olive pruning and olive wood, if the bulk density was substituted by the energetic density, pellets made from olive leaves would show higher values than those from olive pruning and olive wood.
- In relation to the optimum operating conditions for pelletization, low moisture content (9%) and 6 mm diameter were the most adequate parameters for producing pellets from the different residual biomass from olive trees, while the compression length and the pelletization temperature has to be adapted to the raw material to be pelletized. Pellets' quality was higher when they were produced with shorter compression lengths for all the types of residual biomass from olive trees. In the case of olive pruning and olive leaves, temperatures higher than 60°C were necessary to improve the quality of the pellets, while temperatures within the range of 40-60°C were adequate to produce pellets from olive wood.

Finally, through the analysis of the combustion behavior of pellets from olive residual biomass under real conditions in a domestic boiler it was possible to conclude that:

- The optimum operating conditions of the pellet fired boiler are bound to the raw material used as biofuels, having more influence than the boiler load on the gaseous and particulate emissions.
- Problems related to high values of hardness and particle density were observed. Harder pellets resulted in occasional blocks of the screw conveyor that feeds the pellets, while pellets with higher particle densities make it necessary to increase the burnout time. These facts make evident the need to include limits for these parameters in pellets quality standards.
- At maximum loads, the gaseous emissions related to an incomplete combustion (CO and HC) were reduced, but the particulate emissions PM>10µm increased since their emission is related with the presence of unburned particles. The combustion behavior observed makes evident the necessity of designing a boiler for agricultural biomass that presents higher ash contents to allow it to be use as fuel in domestic heating systems under optimum conditions that reduce the gaseous and particulate emissions as well as the problems derived from their characteristics.



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CONCLUSIONES

Las aportaciones más importantes que se han obtenido a partir de este trabajo pueden agruparse en cinco apartados coincidentes con los objetivos fijados en esta memoria:

- (i) La evaluación del potencial y los beneficios del uso de la biomasa residual del olivar como fuente de energía renovable en zonas mediterráneas, más concretamente en el caso de Andalucía.
- (ii) Comparativa de los parámetros utilizados para establecer la calidad y el uso de los pellets a nivel europeo.
- (iii) La caracterización física y química de la biomasa residual del olivar, de acuerdo a los requisitos establecidos para el uso de biocombustibles sólidos a escala no industrial.
- (iv) La determinación de las variables óptimas para la fabricación de pellets a partir de biomasa residual del olivar.
- (v) La determinación del comportamiento de combustión de los pellets de biomasa residual del olivar bajo condiciones reales en una caldera doméstica

En relación a la evaluación del potencial y los beneficios del uso de la biomasa residual del olivar como fuente de energía renovable en Andalucía, se ha concluido que:

- Andalucía es una región muy rica en recursos renovables, especialmente en biomasa, la cual presenta un potencial de 3327 ktep/año y proporciona el 6,3% de la energía primaria consumida y el 78,7% del consumo de energías renovables.
- El potencial de biomasa procedente de residuos agrícolas se estima en 1434 ktep, de los cuales aproximadamente el 50% proviene de residuos del olivar (803 ktep/año). En particular, existen 1,4 millones de hectáreas de olivar en



Universidad de Granada Andalucía, las cuales producen una media de tres toneladas de residuos por hectárea y generan por tanto alrededor de cuatro millones de toneladas al año en la región. La mayoría de esta biomasa residual es actualmente quemada de forma ilegal o dejada en los campos.

 La biomasa residual del olivar puede emplearse para generar electricidad, energía térmica o biocombustibles para el transporte, dependiendo su uso final de las características de la materia prima. Los objetivos planteados en Andalucía han sido logrados en lo relativo al uso de biomasa para aprovechamiento eléctrico, sin embargo, su uso en energía térmica es aún considerablemente inferior a los niveles deseados. Como consecuencia, la producción de biomasa densificada debe ser incrementada de forma que se alcance la demanda de calefacción y agua caliente del sector de la construcción.

En relación al análisis comparativo de los parámetros utilizados para establecer la calidad y el uso de los pellets a nivel europeo se ha concluido que:

- Los únicos países europeos que cuentan con estándares de calidad para los pellets son Austria, Suecia y Alemania, mientras que Francia e Italia han publicado recomendaciones. El análisis comparativo de éstos ha mostrado diferencias significativas tanto en los parámetros de calidad considerados como en los límites establecidos.
- A nivel europeo, la norma EN 14961-2 (2011) regula los parámetros de calidad que deben alcanzar los pellets de uso no industrial. Esta estandarización común beneficiará a Europa ya que garantizará un mercado del pellet de calidad.
- No existen directrices para pellets fabricados a partir de residuos agrícolas, o agropellets, los cuales pueden diferir significativamente de los fabricados con madera, usados como referencia para el establecimiento de estándares de calidad, lo que significa una importante barrera para su uso.

En relación a la caracterización física y química de la biomasa residual del olivar, en comparación con otras biomasas de origen agrícola y forestal, y de acuerdo a

los requisitos establecidos para el uso de biocombustibles sólidos a escala no industrial, se han obtenido las siguientes conclusiones:

- El uso de biomasa residual del olivar tiene como resultado unas propiedades físicas y químicas del pellet que imponen restricciones en su uso, especialmente por su mayor contenido en cenizas, característico de la biomasa residual agrícola, lo que conlleva mayores problemas de mantenimiento de los sistemas domésticos.
- La caracterización de las diferentes fracciones de la biomasa del olivar por separado, mostró diferencias entre ellas, siendo mayor el contenido en cenizas en la hoja (8,9%), y algo menor en la poda (6,3%), aunque en ambos casos inapropiada para su uso en sistemas domésticos de calefacción, con el marco regulador existente en la actualidad. La fracción maderera mostró menor contenido en cenizas (0,98%), en cumplimiento con la norma EN 14961-2 (2011) para su aplicación en sistemas no industriales, que establece tres categorías en función de los valores alcanzados, entre otros, para el contenido en cenizas, que puede oscilar entre 0,7% para pellets A1 y 3,0% para pellets de categoría B.
- El alto contenido en cenizas de la biomasa residual del olivar viene ocasionado, no por la presencia de compuestos minerales del suelo tales como Mg, Ca y K procedentes de la contaminación fruto de su manipulación en origen, sino por su propia composición, con un alto contenido en lignina. Este componente incrementa el poder calorífico de la materia prima y favorece su pelletización, actuando como ligante, sin embargo, su elevada temperatura de descomposición, superior a los 900°C, se ha considerado la responsable del alto contenido en cenizas de las hojas de olivo y poda, así como de su baja velocidad de combustión.
- Los resultados obtenidos tras calcular los índices de escorificación y aglomeración con los métodos recogidos en bibliografía consultada, fueron comparados con los correspondientes valores de temperaturas de fusión de las cenizas, resultando inconsistentes, por lo que es necesario establecer nuevos



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procedimientos de cálculo más adecuados para la determinación de estos índices.

En relación a la determinación de las condiciones óptimas para la fabricación de pellets a partir de las diferentes fracciones de la biomasa residual del olivar, en lo que se refiere a humedad de la materia prima, tamaño de la matriz (diámetro y longitud de compresión) y temperatura de pelletización, se han obtenido las siguientes conclusiones:

- Fue posible optimizar las condiciones de fabricación, para obtener pellets de leña que cumplieron con todos los criterios de calidad físicos y mecánicos establecidos en la norma EN 14961-2 (2011) para uso no industrial.
- Ninguna muestra de pellets fabricado con poda de olivar consiguió cumplir con los criterios de calidad químicos, superando los valores límites establecidos en la norma EN 14961-2 (2011) en lo relativo a contenido en cenizas, nitrógeno y azufre. No obstante, fue posible optimizar las condiciones de fabricación para obtener pellets que cumplieron con los criterios de calidad físicos y mecánicos establecidos en la citada norma y uso no industrial.
- Ninguna muestra de pellets fabricado con hoja de olivo consiguió cumplir con los criterios de calidad físicos, químicos y mecánicos, superando los valores límites establecidos en la norma EN 14961-2 (2011) en lo relativo a contenido en cenizas, nitrógeno, azufre, densidad aparente y durabilidad, para pellets aplicados en sistemas no industriales. No obstante, y dado que el poder calorífico de la hoja es superior que el de la poda y la leña, si la densidad aparente fuese sustituida por la densidad energética, los pellets fabricados con hoja alcanzarían valores superiores a aquellos fabricados con poda o leña.
- En cuanto a los efectos de las condiciones de fabricación de los pellets, un bajo contenido de humedad (9%) y un diámetro de 6 mm son los más adecuados para todas las fracciones de los residuos del olivar; sin embargo la longitud de compresión y la temperatura de pelletización más adecuada depende de la materia prima. La calidad de los pellets de biomasa residual del olivar fue mayor cuando estos fueron fabricados con longitudes de compresión menores. En el

caso de la poda y las hojas, temperaturas superiores a 60°C fueron necesarias para incrementar la calidad de los pellets, mientras en el caso de la leña la temperatura idónea fue el intervalo comprendido entre 40-60°C.

Finalmente, el análisis del comportamiento de combustión de los pellets de biomasa residual del olivar bajo condiciones reales en una caldera doméstica de 14 kW de potencia nominal ha permitido obtener las siguientes conclusiones:

- Las condiciones óptimas de operación de la caldera de pellet están ligadas a las características de los pellets utilizados, teniendo éstas más influencia en lo relativo a emisiones gaseosas y de partículas que la potencia a la que trabaja la caldera.
- Se observaron problemas derivados de valores elevados de densidad de partícula y dureza en los pellets. Pellets de dureza elevada ocasionaron bloqueos del tornillo sinfín que alimenta el quemador, así como densidades de partícula elevadas elevan el tiempo necesario para completar la combustión. Estos hechos hacen evidente la necesidad de incluir límites de estos parámetros en la normativa relativa a la calidad el pellet, no recogidos en la norma EN 14961-2 (2011) para sistemas no industriales.
- A potencia máxima (17kW), las emisiones gaseosas relacionadas con una combustión incompleta (CO y HC) se minimizan, pero la emisión de partículas mayores de 10 µm aumentó, como consecuencia de la presencia de partículas inquemadas. Este comportamiento se explica porque el diseño de la caldera no está preparado para agropellets. El comportamiento de combustión observado hace por tanto evidente la necesidad de diseñar calderas para biomasa agrícola con mayor contenido en cenizas lo que permitirá el uso de esta biomasa como combustible en sistemas de calefacción domésticos bajo condiciones óptimas que reduzcan las emisiones y los problemas derivados de sus características.



FUTURE LINES OF RESEARCH

From the work developed in this work, there have been observed some aspects that need a more detailed analysis. Therefore, they are proposed below as future lines of research:

- Improve the thermal behavior of residual biomass from olive trees, especially in terms of chlorine and ash content, as well as regarding ash-related problems such as the ash fusibility temperatures and the slagging and fouling indexes.
- Optimize the production process of pellets from agricultural biomass in accordance with the current combustion technologies for industrial and domestic use for their subsequent commercialization.
- Design quality standards for pellets from agricultural biomass for industrial and domestic use, considering the particularities of this type of biomass.
- Design domestic boilers and operating conditions adapted to the quality standards for pellets from agricultural biomass and paying special attention to the reduction of the emissions.



LINEAS FUTURAS DE INVESTIGACION

En el desarrollo de este trabajo, han surgido algunos aspectos derivados del mismo que necesitan de un análisis más detallado, por lo que se proponen a continuación como futuras líneas de investigación:

- Mejorar el comportamiento de la biomasa residual agrícola, especialmente en aspectos relativos a los altos contenidos en cloro, mayores contenidos en cenizas, bajas temperaturas de fusibilidad de las mismas e índices de escorificación y aglomeración, de forma que se obtenga un producto compatible con las tecnologías de combustión actuales, y con un precio competitivo.
- Optimización del proceso de fabricación de agropellets que sean compatibles con las tecnologías actuales de combustión, tanto a nivel doméstico como industrial, para su posterior comercialización.
- Diseñar estándares de calidad específicos para los agropellets, tanto para uso doméstico como industrial, que tengan en cuenta las particularidades de las mismas.
- Diseñar calderas domésticas, así como configuraciones de operación, adaptadas a los estándares de calidad que se pueden alcanzar con los agropellets, haciendo especial hincapié en la reducción de emisiones.

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ANNEX:

Table A1. Average values of pellets' quality parameters
Table A2. Results from the least squares method
Published Articles





Ángela García Maraver

	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m ³)	Bulk density (kg/m ³)	Moisture content (%)
OL-M9-D1-T1	38.46	4.33	17.59	764.11	368.79	9.04
OL-M9-D1-T2	74.46	11.67	23.47	896.58	427.25	8.76
OL-M9-D1-T3	87.13	13.33	25.56	971.50	467.59	8.88
OL-M9-D2-T1	62.08	4.33	21.58	665.80	359.64	10.22
OL-M9-D2-T2	57.65	6.33	23.78	946.35	333.95	6.99
OL-M9-D3-T1	83.22	7.33	18.73	789.48	451.26	7.02
OL-M9-D3-T2	70.43	12.33	20.29	718.12	391.39	6.85
OL-M11-D1-T1	56.41	9.00	21.51	977.40	455.99	8.39
OL-M11-D1-T2	14.58	3.33	21.28	845.01	353.49	10.09
OL-M11-D2-T1	77.66	7.17	18.41	863.75	447.09	8.15
OL-M11-D2-T2	88.63	12.33	19.32	969.88	462.06	10.03
OL-M11-D3-T1	57.88	6.67	13.76	736.27	426.85	9.71
OL-M11-D3-T2	70.43	10.67	15.04	912.55	421.28	8.71
OL-M13-D1-T1	31.54	1.00	15.68	852.68	312.72	11.61
OL-M13-D1-T2	21.26	4.67	12.29	697.01	280.68	10.42
OL-M13-D2-T1	39.89	3.67	17.19	632.88	297.72	10.21
OL-M13-D2-T2	84.79	11.50	19.69	902.83	344.98	12.01
OL-M13-D3-T1	36.93	1.00	13.21	586.76	279.90	8.71
OL-M13-D3-T2	32.84	2.33	13.57	699.48	297.28	8.53

Table A1. Average values of pellets' quality parameters

	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m3)	Bulk density (kg/m3)	Moisture content (%)
OP-M9-D1-T1	97.11	23.00	24.87	1248.72	584.26	9.23
OP-M9-D1-T2	97.49	23.00	25.18	1222.52	524.73	10.15
OP-M9-D1-T3	96.99	23.33	20.21	1218.52	545.94	9.10
OP-M9-D2-T1	96.19	23.67	24.77	1147.60	524.36	7.09
OP-M9-D2-T2	98.47	23.00	23.65	1275.12	607.55	8.11
OP-M9-D2-T3	98.92	23.17	24.69	1268.15	610.22	9.01
OP-M9-D3-T1	95.99	21.33	19.68	1252.21	657.33	7.65
OP-M9-D3-T2	98.60	21.00	20.73	1284.45	679.38	6.69
OP-M9-D3-T3	96.65	21.33	20.88	1270.95	662.62	6.67
OP-M11-D1-T1	97.86	24.00	25.50	1238.79	588.36	6.94
OP-M11-D1-T2	96.63	24.83	23.46	1203.73	514.73	9.98
OP-M11-D1-T3	94.64	23.33	18.11	1186.13	523.80	9.66
OP-M11-D2-T2	98.46	23.00	23.03	1242.01	641.12	7.81
OP-M11-D2-T3	99.76	22.83	21.85	1249.16	612.23	7.81
OP-M11-D3-T1	91.59	21.83	18.33	1223.90	693.10	7.94
OP-M11-D3-T2	98.19	21.50	18.56	1199.35	604.88	10.05
OP-M11-D3-T3	97.75	21.50	18.94	1113.88	592.30	9.71
OP-M13-D1-T1	98.15	23.17	24.37	1242.32	588.37	10.33
OP-M13-D1-T2	99.14	23.33	22.99	1247.65	590.58	9.60
OP-M13-D1-T3	97.27	23.17	22.40	1180.58	538.61	9.26
OP-M13-D2-T1	97.74	23.00	24.96	1262.88	573.28	9.46
OP-M13-D2-T2	90.67	22.83	19.48	1128.62	452.56	9.66
OP-M13-D2-T3	78.80	22.17	19.23	1082.97	453.30	8.43
OP-M13-D3-T1	95.89	21.67	19.21	1272.03	653.42	8.43
OP-M13-D3-T2	96.31	21.67	20.49	1186.81	575.42	10.22
OP-M13-D3-T3	95.62	21.50	19.34	1135.84	572.29	9.22

Table A1. Average values of pellets' quality parameters
	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m3)	Bulk density (kg/m3)	Moisture content (%)
OW-M9-D1-T2	98.09	21.83	28.74	1327.81	638.82	7.43
OW-M9-D1-T3	98.72	21.83	25.21	1357.84	638.95	7.42
OW-M9-D2-T2	98.42	23.50	22.87	1322.12	672.39	7.72
OW-M9-D2-T3	95.94	23.17	24.23	1378.69	663.52	6.96
OW-M9-D3-T2	97.95	23.33	17.50	1427.83	701.24	7.64
OW-M9-D3-T3	94.05	23.67	21.09	1376.24	641.00	6.54
OW-M11-D1-T1	98.83	22.33	25.12	1328.51	608.82	8.46
OW-M11-D1-T2	99.37	22.00	25.83	1319.27	607.02	8.79
OW-M11-D1-T3	99.13	22.00	27.22	1237.06	588.76	7.66
OW-M11-D2-T1	97.56	22.50	25.27	1365.48	615.23	8.01
OW-M11-D2-T2	98.72	22.50	16.60	1366.83	657.27	8.78
OW-M11-D2-T3	98.90	21.67	23.78	1327.03	624.73	7.34
OW-M11-D3-T1	98.60	22.33	17.58	1377.73	741.20	7.51
OW-M11-D3-T2	99.32	21.83	19.18	1366.13	671.11	8.14
OW-M11-D3-T3	99.47	22.17	18.61	1350.37	651.86	8.18
OW-M13-D1-T1	97.62	22.17	26.52	1281.15	584.68	10.01
OW-M13-D1-T2	98.69	22.00	25.59	1268.36	546.43	10.81
OW-M13-D1-T3	98.77	21.27	26.13	1244.68	546.50	9.76
OW-M13-D2-T1	98.13	22.17	25.86	1350.44	661.06	9.44
OW-M13-D2-T2	98.95	22.33	25.18	1328.65	626.15	9.99
OW-M13-D2-T3	98.24	22.17	25.23	1338.21	650.18	7.81
OW-M13-D3-T1	98.90	23.17	18.42	1351.44	646.15	10.32
OW-M13-D3-T2	99.41	22.67	18.78	1241.43	631.30	11.41
OW-M13-D3-T3	99.17	22.67	17.36	1367.06	643.58	9.01

Table A2. Results from the least squares method	L
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	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m ³)	Bulk density (kg/m ³)	Moisture content (%)
OL-M9-D1-T3	-12.8700	-8.6700	-14.4400	-228.5000	-232.4100	8.8800
OL-M11-D2-T2	-11.3700	-9.6700	-20.6800	-230.1200	-237.9400	10.0300
OL-M9-D1-T2	-25.5400	-10.3300	-16.5300	-303.4200	-272.7500	8.7600
OL-M13-D2-T2	-15.2100	-10.5000	-20.3100	-297.1700	-355.0200	12.0100
OL-M9-D3-T2	-29.5700	-9.6700	-19.7100	-481.8800	-308.6100	6.8500
OL-M11-D1-T1	-43.5900	-13.0000	-18.4900	-222.6000	-244.0100	8.3900
OL-M9-D3-T1	-16.7800	-14.6700	-21.2700	-410.5200	-248.7400	7.0200
OL-M11-D3-T2	-29.5700	-11.3300	-24.9600	-287.4500	-278.7200	8.7100
OL-M11-D2-T1	-22.3400	-14.8300	-21.5900	-336.2500	-252.9100	8.1500
OL-M9-D2-T2	-42.3500	-15.6700	-16.2200	-253.6500	-366.0500	6.9900
OL-M9-D2-T1	-37.9200	-17.6700	-18.4200	-534.2000	-340.3600	10.2200
OL-M11-D3-T1	-42.1200	-15.3300	-26.2400	-463.7300	-273.1500	9.7100
OL-M9-D1-T1	-61.5400	-17.6700	-22.4100	-435.8900	-331.2100	9.0400
OL-M13-D2-T1	-60.1100	-18.3300	-22.8100	-567.1200	-402.2800	10.2100
OL-M11-D1-T2	-85.4200	-18.6700	-18.7200	-354.9900	-346.5100	10.0900
OL-M13-D1-T1	-68.4600	-21.0000	-24.3200	-347.3200	-387.2800	11.6100
OL-M13-D3-T2	-67.1600	-19.6700	-26.4300	-500.5200	-402.7200	8.5300
OL-M13-D1-T2	-78.7400	-17.3300	-27.7100	-502.9900	-419.3200	10.4200
OL-M13-D3-T1	-63.0700	-21.0000	-26.7900	-613.2400	-420.1000	8.7100

	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m3)	Bulk density (kg/m3)	Moisture content (%)
OP-M11-D1-T1	-2.14	2.00	-14.50	38.79	-111.64	6.94
OP-M9-D2-T3	-1.08	1.17	-15.31	68.15	-89.78	9.01
OP-M9-D1-T1	-2.89	1.00	-15.13	48.72	-115.74	9.23
OP-M13-D2-T1	-2.26	1.00	-15.04	62.88	-126.72	9.46
OP-M13-D1-T1	-1.85	1.17	-15.63	42.32	-111.63	10.33
OP-M9-D2-T2	-1.53	1.00	-16.35	75.12	-92.45	8.11
OP-M9-D1-T2	-2.51	1.00	-14.82	22.52	-175.27	10.15
OP-M11-D2-T2	-1.54	1.00	-16.97	42.01	-58.88	7.81
OP-M9-D2-T1	-3.81	1.67	-15.23	-52.40	-175.64	7.09
OP-M13-D1-T2	-0.86	1.33	-17.01	47.65	-109.42	9.60
OP-M11-D2-T3	-0.24	0.83	-18.15	49.16	-87.77	7.81
OP-M9-D3-T3	-3.35	-0.67	-19.12	70.95	-37.38	6.67
OP-M13-D1-T3	-2.73	1.17	-17.60	-19.42	-161.39	9.26
OP-M9-D3-T2	-1.40	-1.00	-19.27	84.45	-20.62	6.69
OP-M11-D1-T2	-3.37	2.83	-16.54	3.73	-185.27	9.98
OP-M9-D3-T1	-4.01	-0.67	-20.32	52.21	-42.67	7.65
OP-M13-D3-T2	-3.69	-0.33	-19.51	-13.19	-124.58	10.22
OP-M13-D3-T1	-4.11	-0.33	-20.79	72.03	-46.58	8.43
OP-M9-D1-T3	-3.01	1.33	-19.79	18.52	-154.06	9.10
OP-M13-D3-T3	-4.38	-0.50	-20.66	-64.16	-127.71	9.22
OP-M11-D3-T3	-2.25	-0.50	-21.06	-86.12	-107.70	9.71
OP-M11-D3-T1	-8.41	-0.17	-21.67	23.90	-6.90	7.94
OP-M11-D3-T2	-1.81	-0.50	-21.44	-0.65	-95.12	10.05
OP-M11-D1-T3	-5.36	1.33	-21.89	-13.87	-176.20	9.66
OP-M13-D2-T2	-9.33	0.83	-20.52	-71.38	-247.44	9.66
OP-M13-D2-T3	-21.20	0.17	-20.77	-117.03	-246.70	8.43

Table A2. Results from the least squares method

	Durability (%)	Hardness (kgf)	Length (mm)	Single pellet density (kg/m ³)	Bulk density (kg/m ³)	Moisture content (%)
OW-M9-D1-T2	-1.91	-0.17	-11.26	127.81	-61.18	7.43
OW-M9-D2-T3	-4.06	1.17	-15.77	178.69	-36.48	6.96
OW-M13-D1-T3	-1.23	-0.73	-13.87	44.68	-153.50	9.76
OW-M13-D3-T2	-0.59	0.67	-21.22	41.43	-68.70	11.41
OW-M13-D1-T2	-1.31	0.00	-14.41	68.36	-153.57	10.81
OW-M11-D1-T3	-0.87	0.00	-12.78	37.06	-111.24	7.66
OW-M13-D1-T1	-2.38	0.17	-13.48	81.15	-115.32	10.01
OW-M11-D2-T2	-1.28	0.50	-23.40	166.83	-42.73	8.78
OW-M11-D3-T3	-0.53	0.17	-21.39	150.37	-48.14	8.18
OW-M13-D3-T3	-0.83	0.67	-22.64	167.06	-56.42	9.01
OW-M11-D2-T3	-1.10	-0.33	-16.22	127.03	-75.27	7.34
OW-M11-D3-T2	-0.68	-0.17	-20.82	166.13	-28.89	8.14
OW-M13-D3-T1	-1.10	1.17	-21.58	151.44	-53.85	10.32
OW-M11-D1-T2	-0.63	0.00	-14.17	119.27	-92.98	8.79
OW-M11-D1-T1	-1.17	0.33	-14.88	128.51	-91.18	8.46
OW-M9-D3-T3	-5.95	1.67	-18.91	176.24	-59.00	6.54
OW-M13-D2-T2	-1.05	0.33	-14.82	128.65	-73.85	9.99
OW-M11-D2-T1	-2.44	0.50	-14.73	165.48	-84.77	8.01
OW-M9-D1-T3	-1.28	-0.17	-14.79	157.84	-61.05	7.42
OW-M13-D2-T3	-1.76	0.17	-14.77	138.21	-49.82	7.81
OW-M11-D3-T1	-1.40	0.33	-22.42	177.73	41.20	7.51
OW-M9-D2-T2	-1.58	1.50	-17.13	122.12	-27.61	7.72
OW-M13-D2-T1	-1.87	0.17	-14.14	150.44	-38.94	9.44
OW-M9-D3-T2	-2.05	1.33	-22.50	227.83	1.24	7.64

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A comparative study of quality properties of pelletized agricultural and forestry lopping residues

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ABSTRACT

Biomass is an abundant renewable energy source; however, because of its characteristics it is not easily used in its original form. Pelletization technology represents mass and energy densification that provides easier biomass fuel handling and feeding. A privately owned pelletization industrial plant has been set up to produce pellets from agricultural and forestry lopping residues. The plant has produced so far pellets made of leaves of olive tree, a mixture of different parts of olive trees, almond trees, black poplar and holm oak trees. The characteristics of the resulting products have been analyzed in this research to foresee their behaviour in the combustion process. Parameters analyzed included particle and bulk density, moisture content, crushing resistance or hardness, number of particles of pellets per unit volume, size (including diameter, length and ratio of both values), percentage of carbon, nitrogen, sulphur and hydrogen, additives and ash content, and finally heating value. Relationships between various parameters and a comparison with guidelines established by some European norms have been also investigated in order to determine possible restrictions on the use of the pellets. Results have shown that the use of different types of raw materials from agricultural and forestry lopping residues results in different physical and chemical properties of the pellets defining their possible application. Relationships between several parameters of pellets and a comparison with some European norms guidelines were also investigated and different guidelines fulfilment were detected; however no pellets tested fulfilled all parameters from guidelines established in the norms considered.

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1. Introduction

Biomass resources include various natural and derived materials mainly categorized as agricultural residues, wood and wood wastes, animal dung or municipal solid wastes [1]. It is an abundant renewable energy source that entails significant socioeconomic and environmental benefits because it reduces the atmospheric emissions in which the net cycle of CO₂ does not contribute to the greenhouse effect. Moreover it also plays an important role in national economies, avoiding the import of fossil fuels.

Because of high moisture content, irregular shape and size, and low bulk density, biomass is very difficult to handle, transport, store, and utilize in its original form. Densification has prompted significant interest in developing countries in recent years as a technique for utilisation of residues as an energy source [1,2]. Pelletization of biomass is a mass and energy densification for materials that possess low bulk densities. Densification increases the bulk density of biomass from an initial bulk density of $40-200 \text{ kg/m}^3$ to a final bulk density of $600-800 \text{ kg/m}^3$ [3]. The process improves pellets' transportation, and storage and provides better handling and feeding of the biomass with less dust formation [2,4].

The pellets' market has increased during recent years for different raw materials [3–6]. The quality of pellets is determined by the end-user's requirements considering the heating system and the handling properties [6]. The quality depends on the properties of the feedstock, in terms of biomass type, moisture content and particle/ fibre size, and also on the quality management of the manufacturing process, in terms of operational conditions, pelletizer type and binding agents [4–6,8–10]. As a consequence an understanding of the physical and chemical properties of biomass residues is very important in order to determine their behaviour in a combustion process [2,4,6]. For example the physical properties, such as density, determine the particle heating rate and thermal efficiency; while the chemical properties, such as ultimate analysis (moisture content, volatile content, the free carbon remaining and the ash content) and proximate analysis (the composition of the biomass in wt% of





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Fig. 1. Scheme of pellets manufacturing process.

carbon, hydrogen, oxygen, sulphur and nitrogen), functional groups and calorific value, govern the combustion process.

Spain has been one of the countries with the highest dependency on imported energy resources in the European Union, so the use of biomass resources for power generation offers numerous benefits of interest to the political decision-makers. Furthermore, the use of residual biomass as a renewable energy source is situated within the framework of the main foundations of management set forth and in force through norms relating to waste and residue on the European level.

The purpose of this study has been to analyze physical and chemical characteristics of pellets produced in a pelletizing plant, to identify the quality of the products generated and to increase the knowledge of the effects of raw materials on properties of pellets. Five different types of pellets have been analyzed depending on the origin of the raw materials used: leaves from olive trees, olive trees (branches of different size and leaves), almond trees, black poplar and holm oak trees. Relationships between several parameters of pellets and a comparison with European norms guidelines established have been also investigated.

2. Materials and methods

2.1. The manufacturing process

Olive tree (OT)

The samples of pellets studied in this research were all produced at a privately owned industrial plant under the same manufacturing process conditions. Fig. 1 summarizes the manufacturing process. The raw material was received in a reception area where it was weighted and its degree of moisture measured. The working

Almond tree (AT)

capacity of the plant was 5000 kg/h. In a first phase the size of the material was reduced to 20-40 mm in a hammer splinter. Moisture content was reduced until a value lower than 15% in a rotary drier worked with an entrance drying temperature of 250 °C and exit temperature 85 °C. Magnets and screens were used to remove undesired particles before the dried material reached a hammermill that reduced the size of particles in the range of 6-8 mm. Pellets with a diameter of 6 mm and with no additives were produced with a Mabrik (PVR 220 2 N) press operating between 1500 and 1600 kg/ cm² and around 200 °C. Since the pelletizing process implied high temperatures, a cooling process was necessary to reduce the pellets' temperature to 20–25 °C before pellets storage.

2.2. Raw materials for pellets production

Lopping residues from almond trees (AT), black poplars (BP), branches of different sizes and leaves from olive trees (OT), holm oaks (HO) and only leaves from olive tree (OT_L) were used to make up pellets tested in this research. All the raw materials were delivered from agricultural activities in Granada (South of Spain). Fresh materials were collected and chipped in the point of origin with specific machinery and transported in trucks to the pelletizing plant to be processed to obtain pellets. Fig. 2 shows different types of pellets tested.

2.3. European standards related to quality of pellets

Quality pellets parameters and limiting values defined in Austrian (Ö NORM M 7135) [11], Swedish (SS 187120) [12] and German norms (DIN 51731 and DIN 15270) [13,14], Italian (CIT-R04/05) [15]



Black poplar (BP)



Fig. 2. Pellets tested in the study.

tree (OT_L)

		F									
Sample	Particle den m ³)	Particle density (kg/ m ³)		Bulk density (kg/m ³)		Moisture (%)		Crushing resistance (kg)		Compression resistance (kg)	
	Avg	SD	Avg	SD	Avg	SD	Avg	SD	Avg	SD	
AT	1187.2	57.48	575.3	3.23	4.78	1.56	23.33	0.58	21.33	0.76	
BP	1151.2	43.62	523.4	12.26	5.78	0.68	22.83	0.29	17.50	3.77	
НО	1124.5	56.77	558.5	8.23	6.50	2.01	23.33	0.29	12.50	1.50	
OT	1259.2	20.43	582.5	12.43	5.37	0.53	22.50	0.00	21.33	1.04	
OTL	1083.5	49.95	481.6	1.72	6.57	0.47	4.00	0.87	8.33	2.36	

 Table 1a

 Physical parameters of samples of pellets used in the study.

and French (ITEBE) [16] recommendations and guidelines of European Standard Committee (CEN/TC 145882:2004) [17] for densified biomass fuels have been considered in this research. They have been summarized by García-Maraver et al. [18].

2.4. Laboratory procedures

2.4.1. Particle density

The particle density of pellets was determined according to CEN/ TS 15150:2005 [19] by measuring dimensions, using a digital vernier caliper CLD-150, and weight of three randomly selected individual pellets per sample. The calculated particle density represents the average value of these measurements.

2.4.2. Bulk density

The volume and the weight of a pellet sample have to be measured in order to be able to determine the bulk density according to CEN/TS 15103:2005 [20]. The average bulk density was calculated from three measurement series per sample.

2.4.3. Moisture content

Three samples from raw material were milled to determine moisture content in an oven-dried at 105 ± 2 °C to a constant weight and according to CEN/TS 14774-2:2004 [21].

2.4.4. Crushing resistance or hardness

Three randomly chosen pellets of each material were chosen for calculation of crushing resistance or hardness (compressive resistance) defined as the maximum crushing load a pellet can withstand before cracking or breaking.

Compressive resistance and tensile strength of the densified products were determined by diametrical compression test using a KAHL tester working between 0 and 100 kg.

2.4.5. Number of particles of pellets

Three random samples of 100 g of each material were used to determine the number of pieces per 100 g of pellets.

2.4.6. Size

Forty randomly chosen pellets were used to determine the length and the diameter of the pellets (mm) using a digital vernier caliper CLD-150.

Table 1b

Physical parameters	of samples	of pellets	used in	the study.
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Pellets size distribution in percentage was studied screening the three random samples of 100 g of each type of pellets through 16, 10, 8, 4 and 2.5 mm circular holes.

2.4.7. Chemical composition

Three samples from raw material were milled and mixed to obtain a homogenous sample to determine percentage of carbon, nitrogen, sulphur and hydrogen according to CEN/TS 15104:2005 [22]. A FISONS CARLO ERBA EA 1108 CHNSO detector type was used.

2.4.8. Ash content

Three samples of pellets were chosen and the ash content was measured by measuring loss of ignition at 550 $^\circ$ C according to CEN/ TS 14775:2004 [23].

2.4.9. Heating value

Three samples from raw material were milled to determine heating value by using a bomb calorimeter IKA C 2000 and according to UNE 164001:2005 EX [24].

3. Results and discussions

3.1. Characteristics of pellets

The results of the analysis of densified biomass tested have been summarized in Tables 1a, b and 2, and analyzed and discussed in the subsections below.

3.1.1. Particle density

The variation in the particle density of pellets was between 1083 and 1259 kg/m³ for leaves of olive trees (OT_L) and olive trees (OT) respectively (Table 1a). Values obtained were similar to other wood pellets given in the literature [3,5,6,8], except for the samples produced from leaves of olive trees (OT_L) which had shown lower values. The energy density of the pellets defines the efficiency of energy storage, transport and energy production [1,2,4] as well as the combustion behaviour because dense particles show a longer burnout time [3]. Austrian and German standards, as well as French recommendations for pellets used in stoves and boilers have included guideline values for these parameters [18].

Some samples of pellets from almond tree (AT) fulfilled the limit established by the German standard and by the French

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Sample	Number per 100 g		Length (m	Length (mm)		Diameter (mm))	Particle size distribution (%)
	Avg	SD	Avg	SD	Avg	SD	Avg	SD	>16/16-10/10-7/7-4/4-2.5/<2.5 mm
AT	229	10.02	21.27	2.40	5.87	0.06	3.62	0.39	0.00/7.41/58.41/27.29/3.17/3.72
BP	273	18.23	20.99	7.63	5.83	0.23	3.64	1.47	0.00/4.25/67.38/26.88/1.27/0.21
HO	327	12.14	13.22	1.11	6.08	0.08	2.18	0.19	0.00/0.53/85.10/10.04/3.15/1.18
OT	176	9.64	20.36	2.01	5.94	0.02	3.43	0.35	0.22/1.81/93.69/3.71/0.04/0.01
OTL	322	36.06	10.65	2.09	6.04	0.11	1.76	0.31	0.00/1.76/76.22/13.23/5.39/3.39

Table 2

Chemical parameters of samples of pellets used in the study.

Sample	Chemic	al comp	osition (S	%)	Ash cor (%)	ntent	Upper heating value (kcal/kg)		
	С	Ν	S	Н	Avg	SD	Avg	SD	
AT	44.29	0.78	0.00	8	3.35	0.26	4308	18.72	
BP	46.15	0.29	0.00	7.19	2.37	0.69	4428	5.51	
HO	47.02	0.34	0.00	7.62	3.32	0.25	4496	13.58	
OT	44.84	0.95	0.00	7.64	4.79	0.11	4411	8.14	
OTL	42.80	1.09	0.00	7.05	12.34	0.55	4438	14.50	

recommendations for pellets used in stoves and boilers; no sample of AT pellets fulfilled guidelines established by Austrian norm (Table 3). All samples of black poplar (BP) and holm oak (HO) pellets showed values that fulfilled the limit established by both German standards (Table 3); furthermore, all samples would not have density value following guidelines of Austrian standard and French recommendations. Density values of pellets from olive tree (OT) only fulfilled guidelines of French recommendations for pellets used in stoves and boilers (Table 3). Finally samples of pellets from leaves of olive trees (OT_L) fulfilled limits of both German standards (Table 3) and only some samples fulfilled limit established by Austrian norm; French recommendations were not fulfilled.

Table 3

Guidelines fulfilment corresponding to norms considered in the research (\times : guideline not fulfilled; \downarrow : guideline fulfilled; \pm : guideline fulfilled only by some samples; -: guideline not included in the norm).

Parameter		Ö NORM M 7135	SS 1	87120)	DIN 51731	DIN 15270	CTI-	R04/0	5	ITEBE				CEN/TS 14588:2004
			G.1	G.2	G.3			A.1	A.2	A.3	Stove	Boiler	Big boiler	Incinerator	(Annex A.2)
Particle density	AT BP HO OT OT _L	× ± ± × ±	- - - -	- - - -	- - - -	$\stackrel{\pm}{\checkmark}$ $\stackrel{\sqrt}{\checkmark}$ $\stackrel{\times}{\checkmark}$	$\stackrel{\pm}{\checkmark}$ \checkmark \checkmark \checkmark	- - - -	- - - -	- - - -	$\stackrel{\pm}{\times}$ $\stackrel{\times}{\checkmark}$ $\stackrel{\checkmark}{\times}$	$\stackrel{\pm}{\times}$ $\stackrel{\times}{\checkmark}$ $\stackrel{\checkmark}{\times}$	- - - -	- - - -	- - - -
Bulk density	AT BP HO OT OT _L		× × × ×	$\sqrt[]{}$ $\sqrt[]{}$ $\sqrt[]{}$ \times	\checkmark \checkmark \checkmark \checkmark \times	- - - -	 	× × × ×	× × × ×	\checkmark \checkmark \checkmark \checkmark \checkmark $×$	× × × ×	× × × ×	× × × ± ×	× × × ± ×	
Moisture content	AT BP HO OT OT _L	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	$ \begin{array}{c} \checkmark \\ \checkmark $	$\begin{array}{c} \checkmark \\ \checkmark \end{array}$	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark \checkmark	 	- - - -	
Diameter	AT BP HO OT OT _L		\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	$\begin{array}{c} \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark$	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	× × × ×	\checkmark \checkmark \checkmark \checkmark	× × × ×	× × × × ×	× × × × ×	
Length ratio L/D	AT BP HO OT OT _L		\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	$\checkmark \checkmark \checkmark \checkmark \checkmark$	$\begin{array}{c} \\ \\ \\ \\ \pm \end{array}$	$\stackrel{\times}{_{\scriptstyle \times}}$ $\stackrel{\checkmark}{_{\scriptstyle \pm}}$	$\sqrt[]{}$ $\sqrt[]{}$ $\sqrt[]{}$ $\sqrt[]{}$	$\sqrt[]{}$ \times $\sqrt[]{}$ \times	
Ν	AT BP HO OT OT _L	× √ × ×	- - - -	 	- - - -	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ \times	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ \times	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ \times	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ \times	- - -	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ $\stackrel{\times}{\times}$	$\stackrel{\times}{\checkmark}$ $\stackrel{\times}{\times}$ $\stackrel{\times}{\times}$	- - - -	- - - -	- - - -
S	AT BP HO OT OT _L		 	 	 	$\begin{array}{c} \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark$	$\begin{array}{c} \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark$	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	 	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	- - - -	- - - -	
Additives	AT BP HO OT OT _L	\checkmark \checkmark \checkmark \checkmark	 	 	- - - -	 	$\begin{array}{c} \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark\\ \checkmark$	- - - -	 	 	- - -	- - - -	- - -	- - - -	
Ash content	AT BP HO OT OT _L	× × × ×	× × × ×	× × × ×	× × × ×	× × × ×	× × × ×	× × × ×	× × × ×	 	× × × ×	× × × ×	- - - -	- - - -	× × × ×
Heating value	AT BP HO OT OT _L		\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark		$\begin{array}{c} \checkmark \\ \checkmark $	\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark		\checkmark \checkmark \checkmark \checkmark	\checkmark \checkmark \checkmark \checkmark \checkmark	$\begin{array}{c} \checkmark \\ \checkmark $	$\begin{array}{c} \checkmark \\ \checkmark \end{array}$	

3.1.2. Bulk density

This parameter varied in a range between 482 and 583 kg/m³ (Table 1a) corresponding to pellets made of waste from leaves of olive trees (OT_L) and olive trees (OT) respectively. Values obtained in this research were always lower than bulk density of pellets from sawdust, bark and logging residues chipped and stored for several months; but they were similar to bulk density of pellets from fresh logging residues [1], other wood pellets [3] or pellets from some herbaceous crops [4]. The bulk density is of importance with regard to pellet storage and transport [1,2,4]; thus some norms have included guideline values for this parameter to increase the efficiency and decrease the costs of storage and transport.

Swedish and Italian standards and French recommendations include guidelines related to the bulk density of pellets. The comparison between bulk density of pellets analyzed and guiding value of these standards (Table 3) showed different fulfilment grades. Pellets produced from leaves of olive trees (OT_L) did not fulfil any of the established limits for this parameter because values were always lower than it; pellets from black poplar (BP) lopping waste only fulfilled guidelines established by Swedish norm in groups 2 and 3; olive tree (OT) samples satisfied several guidelines, including Swedish (G.2 and G.3) and Italian (A.3) standards. The average value of bulk density also fulfilled limits established in French recommendations for big heating and incinerators; samples from almond trees (AT), holm oak (HO) and olive tree (OT) fulfilled several limits established by Swedish and Italian standards [18], depending on type of pellets; French recommendations have been only fulfilled by some samples from olive tree (OT) and big heating and incinerators.

3.1.3. Moisture content

The water content has an influence on the net calorific value, the combustion efficiency, the temperature of combustion and the equilibrium moisture content with the ambient moisture content, affecting storage conditions [25]. Table 1a shows average value and standard deviation of moisture content of samples tested which were between 4.78 and 6.57% for almond tree (AT) and leaves of olive trees (OT_L) respectively; these values were similar to other wood pellets [3] although lower than moisture content of pellets from fresh sawdust, logging and bark, with values between 7.9 and 21.3% [6].

All samples tested had moisture content according to the guideline values of standards considered in this study (Table 3).

3.1.4. Crushing resistance or hardness

This parameter has not been included in standards considered in the study, however some literature includes compressive resistance test to simulate the compressive stress due to weight of the top pellets on the lower pellets during storage in bins or silos and crushing of pellets in a screw conveyor [4,5]. The compressive resistance test provides a quick measure of the quality of pellets as soon as the pellets are produced from the pellet mill and aids in adjusting the pelleting process to improve the pellet quality.

Pellets tested showed similar crushing resistance values except for the pellets from leaves of olives tree (OT_L) , which showed significantly lower value. Values of tensile strength showed important differences between different pellets tested (Table 1a). It was detected that higher moisture content, lower densities and lower length values could reduce tensile strength of pellets (Figs. 3 and 4). This could be explained by the fact that the tensile strength is related to the adhesion forces between particles at all contact points in the agglomerate [26].

3.1.5. Pellet number density

Table 1b shows average value and standard deviation of the number of pellets in a 100 g sample of pellets. Samples of pellets



Fig. 3. Relation between tensile strength, particle density and moisture content.

from leaves of olive tree (OT_L) and holm oak (HO) had higher while pellets from olive tree (OT) showed lower pellet number density. These results could be related to the tensile strength of samples and density of particles (Fig. 5). Pellets with lower values of tensile strength and density of particles were broken down easily during handling, transportation, and storage increasing the number of particles [2,6,25].

3.1.6. Size

Visual observations of pellets tested showed differences in size (Fig. 2). These observations were confirmed by results of the study of dimensions of pellets tested (Table 1b). The main part of the pellet samples obtained had a diameter of about 6 mm because all the samples were produced in the same industrial plant and under the same conditions; however some differences had been detected in relation to their length. Pellets from leaves of olive tree (OT_L) and holm oak (HO) showed lower length values than the rest of samples.

Some relationship between length, density of particle and moisture content had been detected. Pellets more densified and with lower moisture content broke down less than pellets with lower particle density values (Fig. 6), confirming conclusions explained in subsection about number of pieces, and they should be considered in relation to pellet storage and transport [25].

The dimensions of the pellets, both diameter and length, are important factors with respect to combustion [3,6]. As a consequence all the standards considered in this study include a guideline value about pellets' sizes or relation length/diameter [18].



Fig. 4. Relation between tensile strength and particle length.





In relation to the diameter of the pellets, all the samples fulfilled requirements except for the limit established by the Italian norm for group A.3 and French recommendations for pellets used in boilers, big boilers and incinerators (Table 3).

In relation to the length of pellets only French recommendation includes a limit value not related to the value of the diameter of pieces of pellets. The value of these parameters of pellets made from almond tree (AT), black poplar (BP) and olive tree (OT) waste fulfilled requirements established by French recommendation except for the application in boilers because length of pellets was higher than the guidelines (Table 3). Samples of HO only fulfilled requirements for stove and boiler; OT_L pellets did not fulfil them.

The analysis of values of the ratio length/diameter of pellets tested (Table 1b) showed that samples of OT_L pellets had the lowest ratio and samples from BP the highest ratio. However, all samples fulfilled guiding values established in the standards.

Finally the particle size distribution of pellets' length was also analyzed (Fig. 7). Results showed the higher percentage of particles were between 4 and 10 mm for all the pellets although percentages depended on different materials. Samples from OT_L had the higher percentage of particles lower than 2.5 mm and OT pellets the lowest one. These results are related to the conclusions obtained regarding the number of particles, density, length and crushing resistance or hardness.



Fig. 6. Relation between length, particle density and moisture content.



Fig. 7. Particle size distribution of pellets.

3.1.7. Chemical composition

Percentage of sulphur, nitrogen, carbon and hydrogen content in different types of pellets was determined (Table 2). Values obtained were similar to the values reported in the literature [3] except for OT and OT_I pellets which showed higher percentage of nitrogen.

Norms considered in this research include different chemical parameters. All of them include guidelines for nitrogen and sulphur content except for the Swedish norm. Requirements established for nitrogen were fulfilled only by BP pellets because in the rest of samples the values were higher than limits established (<0.3%). Limits established for sulphur content were fulfilled by all the samples (Table 3).

Manufacturing process of pellets did not use additives so requirements established in relation to this parameter were fulfilled in all cases.

3.1.8. Ash content

Table 2 shows some differences in relation to ash content of different pellets tested. BP pellets were the one that showed lower value of ashes at the end of the burning process; samples from OT_L and OT generated the highest percentage. It was noticeable that all values exceeded the usual ash content of softwood (0.4-0.8%), hardwood (1-1.3%) [3] and sawdust (lower than 0.5%) [6]; however values were similar to ash content from fresh bark (2.65%), fresh logging residues (2.36%), switchgrass (3.49%), bark (3%) and logging residues stored for months (3.81%) [6,27] for AT, BP and HO samples and to values of pellets from torrefied switchgrass (4.8%), wheat straw (5.28%) [3], and olive husk (between 4.6 and 5.8%) [28,29] for OT pellets. Finally ash content from OT_L pellets had higher value than the rest of materials tested including values given in the literature.

Increasing ash content lowers the heating value and implies the risk for sintering and dust emissions [3,29,30]. In order to provide operation comfort for end users in the residential heating sector, all the European standards and recommendations included in this study provide guiding limits related to ash content in the case of residential heating sector [18]. If the pellets are used in medium and large-scale applications (A.3 group of the Italian norm and French recommendations for big installations and incinerators), due to the higher robustness as well as to the more sophisticated combustion and process control technology, pellets' ash content could have higher values [3] and guidelines for this parameter have not been included.

The analysis of results obtained in the research showed that no samples fulfilled guidelines established in the norms (Table 3). The higher values of ash contents of samples could possibly be an indication that the producers had not exclusively used chemically untreated raw material or had problems with mineral contamination of the raw material used during storage and handling [4,6]. In consequence both raw materials collection and drying fuels/ processes should be carefully treated to avoid mineral contamination in order to minimize ash content.

3.1.9. Heating value

Table 2 shows average and standard deviation of upper heating value of different types of pellets tested and no differences between them were detected. The gross calorific values for the different samples of pellets were within the typical ranges from literature consulted [4,6,7,27].

This parameter is included in Austrian, Swedish, German and Italian standards, and even in the experimental European Standard specifications for pellets applied for domestic uses. Results obtained for all the samples tested were within the specified limit (Table 3).

3.2. Comparison of the characteristics of pellets

Table 4 summarizes percentage of parameters that each type of pellets has fulfilled for each norm, considering only requirements included in this study as well as average value of parameters. It is possible to conclude that samples of pellets made from lopping waste from BP fulfilled the higher percentage of parameters (70.3%) followed by samples of pellets from OT (66.1%); finally OT_L pellets had fulfilled the lowest percentage of parameters.

According to the norms considered in this study, Table 4 shows that CEN/TS 14588:2004 published by the European Standard Committee CEN/TC 335, containing the specifications for pellets applied for domestic use, is the norm with the highest percentage of parameters satisfied (85.7%) by different pellets. Otherwise French recommendations (ITEBE), for big boilers and incinerators, have shown the lower percentage of fulfilled guidelines (40%).

Table 5 shows that some requirements have been fulfilled by all the samples, moisture, S content, additives content and heating value. The requirements for diameter of pellets have been fulfilled by 71.4% and length by 88.6% of the samples. The limits established on ash content have not been fulfilled by any of the samples analyzed. In addition requirements about particle density (46%), bulk density (24%) and N content (20%) have been fulfilled by less than half of the samples.

Table 4

Percentage of samples fulfilling guidelines established by norms considered in the study

Norm		Perce	ntage of	sample	es (%)		Average (%)
		AT	BP	HO	OT	OT_L	
Ö NORM		66.7	77.8	66.7	66.7	66.7	68.9
SS 187120	G.1 G.2 G.3	66.7 83.3 83.3	66.7 83.3 83.3	66.7 83.3 83.3	66.7 83.3 83.3	66.7 66.7 66.7	66.7 80.0 80.0
DIN 51731 DIN 15270		62.5 66.7	87.5 88.9	75.0 77.8	62.5 66.7	75.0 77.8	72.5 75.6
CTI-R04/05	A.1 A.2 A.3	62.5 62.5 75.0	75.0 75.0 50.0	62.5 62.5 75.0	62.5 62.5 75.0	62.5 62.5 50.0	65.0 65.0 65.0
ITEBE	Stove Boiler Big boiler Incinerator	55.6 33.3 50.0 50.0	66.7 44.4 50.0 50.0	55.6 55.6 25.0 25.0	66.7 44.4 50.0 50.0	44.4 33.3 25.0 25.0	57.8 42.2 40.0 40.0
CEN/TS 1458 (Annex A.	85.7	85.7	85.7	85.7	85.7	85.7	
Average (%)		64.6	70.3	64.3	66.1	57.7	

Table 5

Percentage of samples fulfilling guidelines established for parameters included in the study.

Parameter	Percentage of samples (%)
Particle density	46.0
Bulk density	24.0
Moisture	100.0
Diameter	71.4
Length or ratio L/D	88.6
N content	20.0
S content	100.0
Additives content	100.0
Ash content	0
Heating value	100

4. Conclusions

Physical and chemical characteristics of samples of pellets, produced in a pelletizing plant with different raw materials, have been analyzed and compared to guidelines established by seven European norms.

The use of different type of raw materials from agricultural and forestry lopping residues results in different physical and chemical properties of pellets imposing restrictions on application of the pellets. It was concluded that samples of pellets made from lopping waste from holm oak have higher quality, followed by samples of pellets from black poplar, almond tree and olive tree; while pellets made from leaves of olive tree have shown the lowest quality.

Some relations have been observed between some of the parameters analyzed. These relations could be explained by the fact that tensile strength is related to the adhesion forces between particles at all contact points in the agglomerate and in consequence to density of particles. Density of particles could be also affected by other parameters, for example moisture content. Pellets with lower density and tensile strength were broken down easily during handling, transportation, and storage, reducing length of pellets and producing higher dust emissions.

No pellets tested fulfilled all parameters from guidelines established in the norms considered in the research. The higher ash content is a common problem in analyzed samples, although it is higher in pellets made from olive tree waste.

More research is needed on the effects of raw material characteristics, seasonal variations, collection and storage of raw material as well as manufacturing process in order to design a production line which would produce pellets with the required quality established by norms while considering the specific application of pellets.

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Data Bank A review of European standards for pellet quality

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ABSTRACT

The quality of pellets is directly related to their physical, chemical, and mechanical properties. It affects the emissions resulting from their combustion and also their use in stoves and boilers. Certain European countries have developed standards specifying control parameters and guidelines with a view to guaranteeing the effective and environmentally-friendly combustion of pellets. This paper presents a comparative study of regulations and standards created by government authorities in Austria, Sweden, Germany, Italy, and France. Also considered are the guidelines of the European Standard Committee CEN/ TC 335 for densified biomass fuels. The comparison of these standards provides evidence of significant differences in the quality parameters and guidelines for the manufacture and use of pellets in Europe. The results obtained could be used as a technical reference for all issues related to the production, sale and research on pellets.

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1. Introduction

The pelletisation of biomass involves the mass and energy densification of materials that possess low bulk densities such as sawdust, straw, and other herbaceous energy crops. This process reduces transportation costs, and provides better handling and feeding of the biomass with less dust formation. In general, pellet quality depends on the chemical, mechanical, and physical properties of biomass in terms of thermal utilization [1,2]. Some of these parameters are related to the raw materials used [3,4], whereas others are related to the quality management of the manufacturing process [5–7].

Apart from the energy content of the pellets, end-consumers are mainly concerned with two problem areas: (i) emissions generated by pellet combustion; (ii) the use of pellets in boilers and stoves. This is understandable since emissions and low-quality biofuels can damage combustion equipment and produce undesired effects such as slagging, corrosion, and interference with process control [6]. Furthermore, the content of fine particles in the fuel, which is extremely dependent on mechanical durability as well as logistics and storage, can disturb the regulation of highly automated heating systems or interrupt automated fuel feeding [7]. In addition, fine particles burn more quickly, and these higher temperatures can favour ash melting.

Consequently, many European countries have developed standards for the quality, storage, transport, and combustion of densified biomass fuels. These include parameters and guidelines pertaining to the following: particle and bulk density, moisture content, crushing resistance or hardness, particle number, particle size (length and diameter), chemical composition, ash content, and heating value. As trade between countries becomes more widespread, it is also necessary to create international standards to facilitate the purchase and sale of biomass fuels. Accordingly, the European Committee for Standardization, CEN, received a mandate from the European Commission to develop standards for solid biofuels within the Technical Committee (TC) 335 Solid Biofuels.

Nevertheless, current national standards regarding pellet quality are very heterogeneous. This is reflected in the differences detected in their quality control parameters and guidelines. For this reason, this paper compares the parameters and the limit values for densified biomass fuels, as defined in different European standards.

2. Pellet quality: standards and certification

2.1. Austrian standard: ÖNORM M 7135

The Austrian standard for energy pellets is the ÖNORM M 7135, which establishes requirements and test specifications for compressed wood or compressed bark in its natural state – pellets and briquettes [8]. Furthermore, it also includes requirements for non-wood biomass pellets (i.e. M 7139 for energy grains, C 4000 for Miscanthus pellets, and C 4002 for straw pellets).

The main parameters and guidelines of the ÖNORM M 7135 are summarized in Table 1. Compliance with this standard is certified



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Table 1

Parameters and guidelines included in Austrian standards for energy pellets ÖNORM M 7135 [8].

Parameter		Limit values
Physical	Diameter (mm)	4-10
	Length (mm)	<5D
	Particle density (kg/dm ³)	<1.12
Mechanical	Durability (%) ^a	<2.3
Chemical	Moisture content (%)	<10
	Ash content (%)	<0.5
	Heating value (kcal/kg)	>4302
	N (%)	<0.3
	S (%)	< 0.04
	Cl (%)	< 0.02
	Additives (%)	<2

^a Durability has been defined in terms of the percentage of fines after testing.

by the *ÖNORM tested label* for high-quality pellets. Other pelletrelated standards in Austria are those pertaining to pellet logistics (M 7136) and pellet storage tanks (M 7137).

2.2. Swedish standards: SS 187120

Sweden was one of the first countries to establish pellet quality standards in its SS 187120 [9]. This standard includes three categories of pellet quality, depending on the parameter limit values considered. The physical, mechanical, and chemical parameters, as well as the guidelines in this standard are listed in Table 2.

2.3. German standards: DIN 51731 and DIN EN 15270

Germany has two standards: DIN 51731 for pellets and briquettes [10], and DIN EN 15270 for high-quality pellets [11]. The first standard for compressed wood was introduced in Germany in 1996. The use of this standard in the small-scale pellet heating sector is limited because of its high maximum ash content (<1.5%) and the absence of an abrasion limit (see Table 3).

In consequence, the DINplus certification, based on the DIN EN 15270 standard for high-quality wood pellets, was subsequently developed. This standard has contributed significantly to the success of the residential pellet market in Germany. In fact, today it is the most important quality certification for wood pellets worldwide. Its parameters and limit values are listed in Table 3.

2.4. Italian standard: CTI-R04/05

The Italian standard CTI-R04/05 establishes quality parameters for energy pellets from biomass [12]. It classifies pellets in the following three categories, depending on their composition:

Table 2

Parameters and guidelines included in the Swedish standard for biofuels and peat SS 187120 [9].

Parameter		Pellet categ	gory	
		Group 1	Group 2	Group 3
Physical	Diameter (mm)	<25	<25	<25
	Length (mm)	<4D	<5D	<5D
	Bulk density (kg/dm ³)	>600	>500	>500
	Moisture content (%)	<10	<10	<10
Mechanical	Durability (%) ^a	<0.8	<1.5	<1.5
Chemical	Ash content (%)	<0.7	<1.5	<1.5
	Heating value (kcal/kg)	>4039	>4039	>3609
	S (%)	< 0.08	< 0.08	_
	Cl (%)	<0.03	<0.03	-

^a Durability has been defined in terms of the percentage of fines after testing.

Table 3

Parameters and guidelines included in German standard DIN 51731, for pellets and briquettes [10], and DIN EN 15270, for high-quality pellets [11].

	Parameter	DIN 51731	DINEN 15270
Physical	Diameter (mm)	4-10	_
	Length (mm)	<5D	<5D
	Particle density (kg/dm ³)	<1.2	<1.2
Mechanical	Durability (%) ^a	-	<2.3
Chemical	Moisture content (%)	<12	<10
	Ash content (%)	<1.5	<0.5
	Heating value (kcal/kg)	3705-4661	>4302
	N (%)	<0.3	<0.3
	S (%)	<0.08	< 0.04
	Cl (%)	<0.03	< 0.02
	Additives (%)	-	<2
	As (mg/kg)	<0.8	<0.8
	Cd (mg/kg)	<0.5	<0.5
	Cr (mg/kg)	<8	<8
	Cu (mg/kg)	<5	<5
	Hg (mg/kg)	<0.05	< 0.05
	Pb (mg/kg)	<10	<10
	Zn (mg/kg)	<100	<100

^a Durability has been defined in terms of the percentage of fines after testing.

- A.1. Deciduous and conifer tree trunk without bark; untreated wood from wood-yielding industry; untreated wood and wood without bark after being used; a mixture of these materials.
- A.2. Raw materials not included in category A.1; untreated herbaceous biomass; a mixture of these materials.
- A.3. Raw materials not included in category A.2

Table 4 summarizes the parameters, values, and guidelines for each pellet category.

2.5. French recommendation: ITEBE

Even though France has no official standard for wood pellets, the government has established certain quality controls (ITEBE) [13], which classify pellets according to their usage context (stove, boiler, large boiler, and incinerator). Table 5 summarizes the control and parameter values in these recommendations.

2.6. European standard committee CEN/TC 335

Finally, the European Standard Committee CEN/TC 335 is the technical committee that has developed the draft standard to describe all forms of solid biofuel in Europe, including wood chips,

Table 4

Parameters and guidelines included in Italian recommendation CTI - R 04/5 [12].

Parameter		Pellet category			
		A.1	A.2	A.3	
Physical	Diameter (mm)	$6\pm0.5{-8}\pm0.5$	$6\pm0.5{-8}\pm0.5$	$10 \pm 0.5 {-}25 \pm 1.0$	
	Length (mm)	-	<5D	—	
	Bulk density	620-720	600-720	≥550	
	(kg/dm ³)				
Mechanical	Dust emission	≤ 1	≤ 1	-	
	(%)				
Chemical	Moisture	≤ 10	<10	≤15	
	content (%)				
	Ash content (%)	≤0.7	<1.5	-	
	Heating value	>4039	>3870	-	
	(kcal/kg)				
	N (%)	≤0.3	≤0.3	-	
	S (%)	≤0.5	\leq 0.5	-	
	Cl (%)	≤0.03	-	-	
	Additives (%)	banned	-	-	

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 Table 5

 Parameters and guidelines included in the French recommendation ITEBE [13].

Parameter		Type of combustion installation				
		Stove	Boiler	Large boiler	Incinerator	
Physical	Diameter (mm)	6 ± 1	$8{-}10\pm1$	>16	>16	
	Length (mm)	10-30	10-15	>16	>16	
	Bulk density (kg/dm ³)	>650	>650	>580	>580	
	Particle density (kg/dm ³)	1.2 - 1.4	1.2 - 1.4	_	_	
Chemical	Moisture content (%)	<10	<10	_	_	
	Ash content (%)	<10	<10	_	_	
	Heating value (kcal/kg)	>4052	>4052	_	_	
	N (%)	<0.3	<0.3	_	_	
	S (%)	< 0.08	< 0.08	_	_	
	Cl (%)	<0.3	_	_	_	
	Na (ppm)	<300	-	-	-	

wood pellets and briquettes, logs, sawdust, and straw bales. The result is the experimental technical specification CEN/TS 14588:2004 for solid biofuels, which includes and specifies the relevant terminology, definitions, and descriptions [14].

The guidelines and pellet classification in the European standard are different from those in the national standards. More specifically, it classifies pellets in three general categories, depending on the origin of the raw materials that pellets are made of: (1) woody biomass; (2) herbaceous biomass; (3) biomass from fruit and a mixture of the previous types of biomass. Each category includes various subcategories that are specified to up to four levels.

For example, the category of woody biomass includes the following subcategories: (1.1) forest, plantation and other virgin wood; (1.2) by-products and residues from wood processing industry; (1.3) used wood; (1.4) blends and mixtures. Subcategory 1.1 has seven subcategories: (1.1.1) whole trees without roots; (1.1.2) stemwood; (1.1.3) logging residues; (1.1.4) stumps/roots; (1.1.5) bark (from forestry operations;(1.1.6) segregated wood from gardens, parks, roadside maintenance, vineyards and fruit orchards;(1.1.7) blends and mixtures. Finally each of these subcategories also contains various subdivisions. For instance, category 1.1 is subdivided as follows: (1.1.1) deciduous; (1.1.2) coniferous; (1.1.3) bushes; (1.1.4) blends and mixtures.

Each parameter also has various subdivisions, depending on their limit values (Table 6). Labels should provide information about the characteristics of each type of pellet. Furthermore, the annexes of this draft standard also include recommendations for the potential uses of pellets. For example, Table 7 shows the recommended parameter values for pellets used in domestic heating. In this case, guidelines are stricter because (i) domestic equipment does not employ advanced cleaning and control mechanisms; (ii) the equipment is not handled by professional staff; (iii) the combustion of pellets takes place in inhabited areas [14].

3. Discussion

The standards for pellet quality developed by various European countries differ not only in their basic requirements, but also in their limit values and their mode of application to the pellets. Pellet size, moisture content, heating value, and ash content are the pellet quality parameters that appear most frequently in standards and recommendations. However, pellet size (diameter, length and/or length/diameter ratio) is the only parameter that is included in all the norms.

Most of the standards specify moisture content limit values except in the case of large boilers and incinerators in the French recommendations [13]. Bulk density is also included in practically all the norms with the exception of the German standards [10,11].

Table 6

Classification of parameters included in CEN/TS 14588:2004 published by the European Standard Committee CEN/TC 335 including specifications for solid biofuels and analytical techniques [14].

Parameter	Classification
Size (diameter and length)	D06: $D \le 6 \pm 0.5$ and $L \le 5D$
(mm)	D08: $D \le 8 \pm 0.5$ and $L \le 4D$
	D10: $D \le 10 \pm 0.5$ and $L \le 4D$
	D12: $D \le 12 \pm 1.0$ and $L \le 4D$
	D25: $D \le 25 \pm 1.0$ and $L \le 4D$
Moisture content (%)	M10: ≤10%
	M15: ≤15%
	M20: ≤20%
Ash content (%)	A0.7: ≤0.7%
	A1.5: ≤1.5%
	A3.0: ≤3%
	A6.0: ≤6%
	A6.0+: >6%
N (%)	N0.3: ≤0.3%
	N0.5: ≤0.5%
	N1.0: ≤1%
	N3.0: ≤3%
	N3.0+: >3%
S (%)	N0.05: ≤0.05%
	N0.08: ≤0.08%
	N0.1: ≤0.1%
	N0.2+: >0.2%
Cl (%)	CL0.03: ≤0.03
	CL0.07: ≤0.07
	CL0.1: ≤0.1
	CL0.1+: >0.1
Durability ^a	DU97.5: ≥97.5
	DU95.0: ≥95
	DU90: ≥90
Fines content (%<3.15 mm)	F1.0: ≤1%
	F2.0: ≤2%
	F2.0 +: >2%
Bulk density (kg/m ³)	Recommended value should be included
	by manufacturer
Heating value (kcal/kg)	Recommended value should be included
	by manufacturer
Additives	Binding materials and ash inhibitory should
	be included in the label

^a Durability has been defined in terms of the percentage of whole pellets after testing.

Nitrogen and sulphur limit values are provided in the Austrian norm [8], the German standards [10,11], the Italian standard (for A1 and A2 pellets) [12], and the French recommendations (for the use of pellets in stove and heating) [13].

Heating limit values appear in all the norms except in the Italian standard for pellet type A3 [12]. Most standards also include ash

Table 7

Classification of parameters included in Annex A.2 of CEN/TS 14588:2004 published by the European Standards Committee CEN/TC 335 in relation to guidelines for pellets applied for domestic heating use [14].

Parameter	Classification
Size (diameter and length) (mm)	D06: $D \le 6 \pm 0.5$ and $L \le 5D$
	D08: $D \le 8 \pm 0.5$ and $L \le 4D$
Moisture content (%)	M10: ≤10%
Ash content (%)	A0.7: ≤0.7%
N (%)	No guideline is included
S (%)	S0.05: ≤0.05%
Cl (%)	No guideline is included
Durability ^a	DU97.5: ≥97.5%
Fines content (% < 3.15 mm)	F1.0: ≤1%
	F2.0: ≤2%
Bulk density (kg/m ³)	No guideline is included
Heating value (kcal/kg)	E4.7: \geq 4.7 kWh/kg = 4042 kcal/kg
Additives	<2%

^a Durability has been defined in terms of percentage of whole pellets after testing.

content limit values except in the case of A3 pellets in the Italian standard [12] and pellets for large boilers and incinerators in the French recommendations [13].

Finally, there are other parameters that appear in only a few of the standards. For example, the German standards include limit values for the As, Cd, Cr, Cu, Hg, Pb and Zn content of pellets. This is true even in standard DIN 51731 for lower quality pellets [10,11]. Particle density is specified in the German standards and the French recommendations (i.e. for the use of pellets in stove and boilers) [10,11,13]. Nevertheless, the German standard DIN 15270 is the only one that includes limit values for additives, though this parameter is restricted to higher quality pellets [11].

In reference to the limit values of the various parameters, no important differences were observed, except in the case of ash content, durability, and heating value. In these cases, the German standard DIN EN 15270 [11] and the Austrian norm [8] were found to be the most restrictive.

Finally, there are norms that establish quality parameters, according to a pre-established classification of pellets. For example, the Swedish standard specifies three categories of pellet quality depending on the limit values set. The set of values for group 1 defines pellets of the highest quality; the set for group 2 defines pellets of medium quality; and the set for group 3, pellets of lesser quality [9]. The classification of pellets in the Italian recommendation CTI - 04/5 is based on the origin of the raw material, and the limit values are more restrictive for pellets made with higher quality raw materials, such as deciduous and conifer tree trunks without bark, which belong to group A.1 [12]. For pellets made of untreated herbaceous biomass (group A.2), a lower heating value is specified, and the ash content can be higher. Group A.3 has even fewer restrictions, and, the guidelines only specify values for the diameter, bulk density, and moisture content of the pellets. French quality controls establish four groups of pellets depending on their final use. Accordingly, pellets of higher quality should be used in stoves or boilers, while less exacting requirements are applicable to pellets used in large boilers and incinerators [13].

Given the heterogeneous nature of national pellet quality standards, the experimental technical specification CEN/TS 14588:2004 for solid biofuels created a set of guidelines for pellet quality and use. It also elaborated a very specific categorization of pellets, based on the classifications in national norms. As previously mentioned, like the Italian standard, the CEN/TS 14588:2004 also classifies pellets depending on the origin of the raw materials. However, each quality parameter considered is subdivided into different categories depending on its value and therefore on its final quality (Table 6). Moreover, the annexes of this norm include recommendations concerning the final use of pellets. The most restrictive limit values are for pellets used in domestic heating devices (Table 7) [14].

4. Conclusions

In the vast majority of the European countries, there are very few regulations for the manufacture and quality of pellets. All too often, pellets only come under the jurisdiction of very general biomass laws. The only European countries with official standards for compacted biomass fuels are Austria, Sweden, and Germany. A few other countries, such as France and Italy have published recommendations or guidelines.

The comparative analysis of these standards and recommendations for pellets highlights significant differences in their quality parameters as well as in their guidelines. National standards that specify pellet quality requirements at an individual level have not been accepted by all stakeholders in Europe, and this has contributed to the development of national pellet types, whose characteristics depend on the country where they are manufactured. The situation will doubtlessly change when the application of the European standard for solid biofuels becomes official. A common standardization will be beneficial for Europe since it will remove an important barrier to the further development of European pellet and combustion technology market in general and will increase international pellet trade in particular.

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Analysis of olive grove residual biomass potential for electric and thermal energy generation in Andalusia (Spain)

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ABSTRACT

As fossil fuels are not only a limited resource, but also contribute to global warming, a transition towards a more sustainable energy supply is urgently needed. Therefore, today's environmental policies are largely devoted to fostering the development and implementation of renewable energy technologies. One important aspect of this transition is the increased use of biomass to generate renewable energy. Agricultural residues are produced in huge amounts worldwide, and most of this residue is composed of biomass that can be used for energy generation. Consequently, converting this residue into energy can increase the value of waste materials and reduce the environmental impact of waste disposal. This paper analyses the situation of biomass is the renewable source which most contributes to Andalusia energy infrastructure. The residual biomass produced in the olive sector is the result of the large quantity of olive groves and olive oil manufacturers that generate byproducts with a potentially high energy content. The generation of agricultural and industrial residues from the olive sector produced in Andalusia is an important source of different types of residual biomass that are suitable for thermal and electric energy since they reduce the negative environmental effects of emissions from fossil fuels, such as the production of carbon dioxide.

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1. Introduction

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Energy is essential to economic and social development and a high quality of life. In consequence, energy consumption in developed countries grows at a rate of approximately 1% per year, and that of developing countries, at a rate of 5% per year [1]. Therefore, much of the world's energy is currently produced and consumed in ways that could not be sustained if technology was to remain at

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its present level and if the overall energy demand was to increase substantially [2]. However, oil and natural gas reserves can only cover consumption at this rate for the next 40 years in the case of oil, and for the next 60 years in the case of natural gas.

From a purely environmental perspective, the emissions currently generated by the use of fossil fuels are the source of serious environmental problems, such as acid rain, the greenhouse effect, and holes in the ozone layer, which in many cases are irreversible [1,3,4]. Solutions to control atmospheric emissions of greenhouse gases as well as other gases and substances require potential longterm actions for sustainable development. Such actions should be geared to enhancing energy production, transmission, distribution, and consumption [2]. In this regard, renewable energy resources appear to be one of the most efficient and effective solutions because they are both renewable and sustainable [3]. Even more important, renewable energy produces scarcely any waste products such as carbon dioxide or other chemical pollutants. It thus has a minimal impact on the environment [5].

The Kyoto Protocol to the United Nations Framework Convention on Climate Change, which was adopted in December 1997, was a crucial turning point in the international effort to promote the use of renewable energy worldwide. After the original Framework Convention was signed at the Earth Summit in Rio de Janeiro in 1992, growing evidence of climate change has spurred many countries to increase their support of renewable energy. Even more ambitious efforts to promote renewables can be expected as a result of the Kyoto pact, which includes legally binding emissions limits for industrial countries, and for the first time, specifically identifies the promotion of renewable energy as a key strategy for reducing greenhouse gas emissions [6].

For 2010, the European Union set ambitious targets for the share of renewable energies in total energy and electricity consumption and for biofuels. The specific energy targets in the European Union for 2010 (EU-25) were to increase the share of renewable energies from 6% to 12% of the gross energy consumption, of green electricity from 14% to 21% of the gross electricity production and of liquid biofuels to 5.75% of the total fuel consumption [7]. Amongst renewable energy sources, the largest contribution (63%) comes from biomass. Today, energy from biomass already provides approximately 4% of the total EU energy supply. This energy is predominantly used for heat, and to a lesser extent, for combined heat and power (CHP) applications. By 2010, it was expected that biomass would provide as much as 8% of the total EU energy supply. The European Commission estimates that reaching the target of a 12% share of renewable energy in total energy consumption in Europe will require around 130 Mtoe of biomass [8].

In this regard, Spain has taken up the challenge in this effort to establish a new energy model, and is working to develop legal, regulatory, and budgetary tools to promote the transformation process that is crucial to the fulfilment of energy objectives. Amongst these were the Plan de Energías Renovables (PER) (2005-2010), that has now been followed by the Plan de Acción Nacional de Energías Renovables (PANER) (2010-2020). Both of these plans have replaced the Plan de Fomento de las Energías Renovables (2000-2010), which did not produce satisfactory results. The PER was enacted so that renewable resources would cover at least 12% of the total energy consumption (16.6 Mtoe) by the year 2010. In addition, new goals were incorporated, and adopted after the previous plan [9]. Furthermore, the implementation of this plan expands the scope of the energy policy. More specifically, it aspires to a new energy model that promotes structural changes in the system and the consolidation of an energy culture imbued with a collective awareness that energy is a scarce and valuable resource.

The structural reorientation of the Spanish energy system evidently requires a repositioning in regards to the best possible use of the country's energy resources and the overall definition of a



Fig. 1. Geographical location of Andalusia.

model for energy production and use. Such a model should be fully adapted to the climatic, cultural, and economic conditions of the different regions, where energy policies are generally implemented through regional development plans, such as the *Plan Energético de Andalucía* 2003–2006 (PLEAN) in Andalusia, which was followed by the *Plan Andaluz de Sostenibilidad Energética* (PASENER) 2007–2013.

In this regard, the PASENER is a new energy plan based on the commitments in the Kyoto Protocol. It incorporates changes in energy policies and envisages a new energy model that consolidates an energy culture that considers energy as a scarce and valuable resource. Its strategic objectives are the following: (i) to give priority to renewable energy sources in order to increase energy self-sufficiency in Andalusia; (ii) to protect the environment; (iii) to create an energy model adapted to the conditions of the region.

Evidently, Andalusia is in a privileged position because of its capacity for wind power production, as well as its abundant solar catchment. However, biomass is the renewable source which most contributes to Andalusian energy infrastructure [10] because of the availability of residual biomass. This is hardly surprising since farming is the main economic activity in Andalusia. In fact, there are 4.7 million agricultural hectares (57% of the territory). Of these hectares, 1.4 million are olive groves, which are the main source of residual biomass in the Autonomous Community [11,12]. This study analyzes the potential of olive grove residual biomass for the production of electrical and thermal renewable energy from biomass in Andalusia (Spain).

2. Energy situation in Andalusia

2.1. Geographic location and agricultural activity

Spain is located in southwestern Europe and comprises about 84% of the Iberian Peninsula (Fig. 1). Its total area is 504,782 km² of which 499,542 km² is land and 5240 km² is water. It is divided into 17 autonomous communities and 52 provinces.

The Autonomous Community of Andalusia is located in the south of Spain, between latitudes 37° and 42° N, with a surface area of $87,268 \text{ km}^2$, approximately 17.68% of the territory in Spain. The region is divided into eight provinces: Huelva, Seville, Cordoba, Jaen, Almeria, Granada, Malaga and Cadiz. It is the most densely populated autonomous community in the country (8,285,692 inhabitants in 2009), which amounts to 18% of the national population.

Of the various economic sectors, the primary sector (agricultural activities, hunting, silviculture and fishing) is more important in Andalusia than in the rest of Spain, and in 2003 represented 6% of the gross added value of Andalusian economy. This result is an excellent indicator of the importance of this sector in Andalusia, which generates a high percentage of employment (27% of all agricultural jobs in Spain) [12].

Table 1

Energy balance of Andalusia in 2008 [10].

Unit: ktoe	Coal and derivative products	Crude oil and derivative products	Natural gas	Renewable energies	Electric energy	Derivative energies (heat) ^a	Total
Production	23.4	0.0	15.4	1668.3	0.0	0.0	1957.1
Recoveries	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Exchange balance (imp-exp)	1823.0	10,135.8	6438.8	-58.2	286.1	0.0	18,625.5
Stock variation (initial-final)	-355.9	45.6	70.7	0.0	0.0	0.0	-239.6
Bunkers (maritime transport)	0.0	199.2	0.0	0.0	0.0	0.0	199.2
Gross internal consumption	1740.5	10,380.6	6524.9	1610.0	286.1	0.0	20,542.2
Available for final consumption	31.6	8871.3	2391.6	750.6	3134.9	33.9	15,213.9
Non-energetic final consumption	0.0	1312.7	276.7	0.0	0.0	0.0	1589.4
Energetic final consumption	31.6	7558.6	2115.1	750.6	3134.9	33.9	13,624.7

^a The column called "Derivative energies (heat)" shows the production of heat for sale.

Table 2

Energetic consumption distribution by sectors in Andalusia [10].

Unit: ktoe	Coal and derivative products	Crude oil and derivative products	Natural gas	Renewable energies	Electric energy	Derivative energies (heat)*	Total
Industry	31.6	682.4	1786.1	466.5	789.6	33.9	3790.1
Transport	0.0	5405.3	6.3	98.0	33.0	0.0	5542.6
Primary (agriculture and fishing)	0.0	896.7	88.2	7.5	126.1	0.0	1118.6
Service sector	0.0	33.9	122.5	56.3	1065.6	0.0	1278.3
Residential	0.0	540.3	112.0	122.3	1120.6	0.0	1895.2
Energetic final consumption	31.6	7558.6	2115.1	750.6	3134.9	33.9	13,624.7

*Quantity of produced heat used for sale.

2.2. Energy balance

Andalusia estimates its primary energy consumption at 20.5 Mtoe, based on the use of fossil fuels imported as crude oil (50.5%), natural gas (31.7%) and coal (8.5%). Fossil fuels are not produced in Andalusia because there is no crude oil. Since there are only five deposits of natural gas and four deposits of coal in the region [13], its self-sufficient energy supply is estimated at 9.1%.

Conversely, in this same region, 1668.3 ktoe of energy comes from renewable sources. Of this energy, 58.2 ktoe are exported. The gross internal consumption of renewable energies is thus 1610 ktoe, with 750.6 ktoe available for final energy consumption. This is 5.5% of the 13.6 Mtoe final energy consumption in the region (Table 1), which is mainly used by the industrial and domestic sectors (Table 2).

Of the various renewable energies, biomass is the source which most contributes to Andalusian energy infrastructure. It comes to 6.3% of the total primary energy consumption and 78.7% of the renewable energy consumption (Fig. 2). The progressive increase in renewable energy sources is mainly due to the use of biomass for energy. The rain calendar as well as the agricultural crops and harvests have a bearing on this contribution. In Andalusia, one of the most important crops is olives [14]. In 2008, the contribution of primary energy from biomass was 40.5% (365.1 ktoe), which was higher than in 2007 (Fig. 2).

2.3. Renewable energy infrastructure and its policy in Andalusia

Andalusia, while deficient in fossil energy resources, is very rich in renewable resources [13]. It is privileged in this respect because of its capacity for wind power production, and abundant solar catchment. It also has a large quantity of available residual biomass stemming from its great potential for producing energy-yielding crops. For these reasons, conversion to the industry of renewable energy, and more concretely, that of the generation of biomass from agricultural residues, is an opportunity that demands innovation as well as positive action geared towards the generation of knowledge.

Table 3 shows the installed power capacity for each renewable technology in Andalusia. It also specifies the attainment level



Fig. 2. Evolution of total renewable primary energy consumption in 2008 in Andalusia [10].

Table 3PLEAN and PASENER objectives [13,14].

		2006	PLEAN objectives 2006	Degree of compliance 2006 (%)	PASENER objectives 2007	PASENER objectives 2010	PASENER objectives 2013
Hydraulic (special regime)	MW	129.8	107.2	121.1	129.8	137.8	148.0
Hydraulic (ordinary regime)	MW	464.2	-	_	464.2	476	476
Wind	MW	607.9	2700	22.5	1284	4000	4800
Solar photovoltaic (insulated)	MWp	5.8	6.1	95.1	36.2	55.4	100
Solar photovoltaic (connected)	MWp	15.4	4.4	348.7			
Solar thermal	m ²	347,182	411.5	84.4	407,000	765,228	1,341,554
Solar thermoelectric	MW	11	100	11	60	250	575
Biomass thermal	ktoe	367.5	643	57.2	583.5	615.6	649.0
Biomass electric	MW	164.6	164	100.4	169.9	209.9	256.0
Biofuels consumption	ktoe	36.1	90	40.1	263.7	2000	2300

of the objectives established in the Plan Andaluz de Sostenibilidad Energética (PASENER) 2007–2013. The goal is for renewable energies to cover 12% of Andalusian energy consumption in 2010 and 25.4% of this consumption in 2013. In the case of biomass, it is necessary to distinguish between biomass for the generation of electricity and biomass for thermal use. Since the enactment of the Plan Energético de Andalucía (PLEAN) 2003-2006, the required levels for thermal biomass have not as yet been attained, though in the case of electricity biomass, the goal has been reached (Table 3). The reason for this was the transfer of the little fuel available in 2006 to electricity generation plants and the exportation of biomass during this same period to European countries [13]. When the Plan Energético de Andalucía (PLEAN) 2003-2006 was in force, biomass was exported to other European Union countries, mainly because of a compensation system in Spain, which could not compete with the prices of this biofuel in other countries. This situation was remedied with the passing of Royal Decree RD 661/2007, which was an important advance since it subsidized installations that used biomass to generate electricity [14].

At the end of 2009, the total power capacity available in installations in the Autonomous Community of Andalusia came to 14051 MW. Renewable sources provided 4419 MW (equivalent to 877.2 ktoe/year, which is the energy consumption of over two million homes). It is significant that 62% of this amount was produced with biomass [15].

3. Biomass potential in Andalusia

Biomass is the renewable energy that most contributes to the energy system in Andalusia (Fig. 2). This region has a cultivated land surface of 8,759,531.18 ha. Approximately 40% of this land is forest and 60% is farmland, of which certain crops cover a total surface of 1,937,107.76 ha [16].

3.1. Biomass potential

The biomass potential in Andalusia is about 3327 ktoe/year [13]. Of this annual amount of biomass, 1434 ktoe is obtained from agricultural residues, of which approximately 50% is olive tree residues (803 ktoe/year) whereas the other half is mainly obtained from sunflowers, cotton and fruit trees (Table 4). This was the capacity necessary to generate 5% of the primary energy consumption of Andalusia in 2007 [17,18]. On the other hand, since olive oil industries generate 77.25% of the industrial residual biomass (Table 4), the olive sector can be regarded as the greatest producer of residual biomass in Andalusia.

The energy exploitation of biomass allows the sustainable replacement of fossil fuels, increases self-sufficiency and energy diversification, and contributes to the development of rural areas. Furthermore, biomass is a renewable energy that can be used as an energy source for electricity, heating, and vehicle transport [17], and reduces $890 \text{ g CO}_2/\text{kWh}$ if it is used instead of oil. Biomass is a fuel of biological origin, such as woody biomass. If properly cultivated and managed, biomass does not contribute to climate change through emissions of CO₂ to the atmosphere because it absorbs the same amount of carbon in growing as it releases when consumed as fuel. In the future, CO₂ emissions from power and heat generation will have to be greatly reduced to meet environmental constraints and commitments [19].

The final use of biomass depends mainly on the characteristics of the raw material. Most of the biomass in Andalusia generates electricity (45%) and thermal energy (40%), which is mainly used in domestic heating systems (13%) and olive oil industries (14%) due to the abundance of olive groves and their proximity to this type of industry (Fig. 3).

3.2. Olive grove residual biomass in Andalusia

Olive groves are particularly relevant in Andalusia because of their importance in the region and more concretely, for the environmental management and energy use of the residues from olive oil industries and olive grove maintenance [20]. In fact, Andalusia has specific legislation concerning olive groves, which enhances the

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Table 4

Biomass potential in Andalusia [13,17].

	Kt0e/year	
Total biomass potential in Andalusia	3327	
Agricultural residues	1434	
Olive grove	803	
Fruit trees	86	
Corn	72	
Sunflowers	186	
Greenhouse residues	100	
Rice	43	
Cotton	143	
Industrial residues	589	
Olive oil industry	455	
Bark	35	
Rice husk	19	
Cotton	15	
Nuts	17	
Wood	41	
Olive stones	2	
Sugar refinery	4	
Cork	1	
Forestry residues	136	
Quercus	59	
Eucalyptus	53	
Pine	18	
Poplar	7	
Energy crops	559	
Cynara	559	
Biodegradable	609	
MSW	54	
Mud	43	
Animal waste	15	



Fig. 3. Biomass energy uses in Andalusia [10].

 Table 5

 Characteristics of olive trees residues [16].

	Olive pruning residues	Olive-pomace	Orujillo	Olive stones
Moisture content (%weight)	10.95	55.29	12.69	13.12
Lower heating value (kcal/kg dry basis)	4300	4250	4300	4500

efficient use of water and energy, and promotes the use of renewable energies, in particular of biomass [21]. These regulations were passed in order to conserve energy and generally improve energy efficiency in the olive sector. The goal is to foment the energy exploitation of agricultural and industrial residues, the production of energy from biomass, and the use of renewable energies.

There are 1.4 million hectares of olive crops in Andalusia [11], producing an average of 4 million tons of olives per year [17]. Of this quantity, 3.7 million tons per year are used to make olive oil (800,000 tons/year), and the rest to obtain olives (300,000 tons/year), to be consumed as table olives. Furthermore, olive crops generate a variety of energy residues, such as olive pruning residues (4.2 million tons/year of branches and leaves that can be pelletized for domestic heating), olive stones (suitable for domestic and industrial heating or electricity production), olive-pomace and *orujillo* [22,23], residues that have been used traditionally for domestic heating in rural areas.

One hectare of olive grove generates three tons of pruning residues (Fig. 4). This means that about four million tons are

Table 6

Electric energy generation plants with biomass as fuel in Andalusia [17]



Fig. 4. Biomass potential from olive grove [17].

generated per year in Andalusia, most of which are now illegally burnt or left on the ground to generate greenhouse effect gases [24]. Table 5 shows some of the physical and chemical properties of the residual biomass obtained from olive groves that determines their end use. The low heating value is similar for all the residues, but they have different moisture content, especially in the case of olive-pomace with 55.29% of moisture content in weight.

3.2.1. Electricity biomass

In Andalusia, there are 19 plants that use biomass to generate a total of 205.3 MW of electrical energy (Table 6). This amounts to 1.46% of the total power capacity of 14,051 MW available in existing installations in the Autonomous Community. This quantity is very near to the objectives established in the PASENER for the year 2010 (Table 3), signifies a reduction of approximately 8% of the CO_2 emissions [13]. The majority of these installations use olive trees residues as fuel though there are plants that consume greenhouse residues, wood, and sugar cane bagasse [17]. Olive-pomace, *orujillo*, and olive stones are the olive tree residues that are best for producing electrical energy because their lower bulk density, and in the case of olive-pomace, their higher moisture content (Table 5), make them less effective for domestic heating systems.

3.2.2. Thermal biomass

The demand for heating accounts for a significant portion of the total energy demand in the world today. The building sector consumes 35.3% of the final energy demand of which 75%

	Power (MW)	Fuel	Locality	Province
La Loma	16.0	Orujillo	Villanueva del Arzobispo	Jaen
Vetejar	12.9	Olive pomace	Palenciana	Cordoba
El Tejar Autogeneracion	5.7	Olive pomace	Palenciana	Cordoba
Agroenergética de Baena	20.0	Olive pomace	Baena	Cordoba
Hnos. Santamaria Muñoz E Hijos	1.7	Orujillo	Lucena	Cordoba
Extragol	9.1	Orujillo, pruning residues, energy crops	Villanueva de Algaidas	Malaga
Albaida Recursos Naturales I	1.7	Greenhouse residues	Nijar	Almeria
Albaida Recursos Naturales II	1.7	Greenhouse residues	La Mojonera	Almeria
Tradema	2.0	Wood	Linares	Jaen
Fuente de Piedra	8.0	Orujillo	Fuente de Piedra	Malaga
Biomasa Puente Genil	9.8	Orujillo	Puente Genil	Cordoba
Bioenergia Santamaria	14.3	Orujillo	Lucena	Cordoba
Bioenergetica Egabrense	8.0	Orujillo	Cabra	Cordoba
ENCE I	40.9	Wood	San Juan del Puerto	Huelva
ENCE II	27.0	Wood	San Juan del Puerto	Huelva
Agroenergetica de Algodonales	5.4	Olive pomace	Palenciana	Cordoba
Severaes	0.1	Olive tree pruning	Cañete de las Torres	Cordoba
Bioenergetica de Linares	15.0	Orujillo, agricultural and forestry residues, energy crops	Linares	Jaen
Aldebaran energia del Guadalquivir	6.0	Chips, cereal straw	Andujar	Jaen
Total	205 3			



Fig. 5. Olive groves and industrial pellets production plants operating in Andalusia [18].

is for space and domestic water heating [25]. Renewable energy sources used for heating and cooling purposes have received relatively little attention compared with those used to generate electricity and to produce transport fuels. This is surprising because the demand for heat consumes the largest share of the primary energy supply. Renewable energy sources can thus offer a practical alternative to fossil fuels under many circumstances [26]. The potential to increase the use of solar, geothermal, and biomass resources for renewable energy heating is therefore large [27].

In Andalusia, there is a regulatory framework of economic incentives to foment the environment and sustainable energy development. The objective of these measures is to improve environmental protection through energy saving in new and existing buildings. They stress the need to improve the efficient use of energy in the main consumption sectors, such as the industrial, service, and transportation sectors [28]. These economic incentives allow Andalusians to implement more environmentally friendly project in their businesses and homes, and thus reduce fossil fuel consumption.

Biomass consumption for thermal applications during the year 2008 was approximately 613.55 ktoe. This biomass was mainly obtained from olive-pomace and olive stones. This produced a carbon dioxide reduction of 1895.87 metric tons [29] in Andalusia. Moreover, the use of standardized pellets from olive pruning residues would permit the use of these residues in efficient combustion processes. Nevertheless, certain characteristics, such as the higher ash content of olive tree pellets [23] in comparison with other European pellets, have raised doubts concerning their use in thermal applications, especially in domestic heating systems.

Pelletized biomass made from agricultural and forestry products and residues are rapidly becoming an important renewable energy source for industrial and domestic purposes [30]. The pelletization of biomass is a mass and energy densification for materials that possess low bulk densities. The typical bulk density of biomass chips is less than 150 kg/m³ while that of wood pellets is typically over 600 kg/m³ [31]. In the pelletizing process, raw material is dried, ground, homogenized, and densified, which reduces transportation costs, improves storage, and results in better handling characteristics with less dust formation, besides providing potential storage for off-season utilization [31].

Andalusia is in the process of improving its infrastructure, and has become the first Spanish region in densified biofuel production, with 13 industrial pellet production plants. Six of these plants are in operation, and are producing 44 ktoe/year. All of them are strategically located near the olive groves to be able to use olive pruning residues (Fig. 5). There are currently three more plants under construction in Jaen that will produce 34.6 ktoe. This will be in addition to the energy generated by the other plants, which is steadily increasing.

4. Conclusions

The use of biomass as an energy resource entails significant socioeconomic and environmental benefits, such as the following: (i) biomass is an abundant resource, and its renewability is a guarantee of sustainable use; (ii) biomass reduces atmospheric emissions so that the net cycle of CO_2 does not contribute to the greenhouse effect; (iii) biomass is relevant to national economies since it eliminates the need to import fossil fuels.

Andalusia is very rich in renewable resources, especially in biomass, which in this region provides 6.3% of the total primary energy consumption and 78.7% of the renewable energy consumption. Much of this residual biomass is produced by the olive sector, which generates pruning residues and olive oil industrial residues, such as olive-pomace, *orujillo*, and olive stones. The management of these agricultural and industrial residues is actively encouraged by the Andalusian regional government, thanks to specific legislation pertaining to olive groves. These regulations foment the use of biomass with a view to improving energy conservation and efficiency in the olive sector.

Biomass can be used to generate electricity, thermal energy and biofuels for transportation. However, its final use depends on the characteristics of the raw material. Government objectives have been fulfilled regarding the use of biomass to produce electrical energy. In contrast, the use of biomass for thermal energy is still considerably below the levels desired. This situation is now being corrected, and the production of densified biomass has increased in Andalusia in order to meet the demand for space and domestic water heating in the building sector. Economic incentives are also offered to encourage people to implement environmentally friendly projects.

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