Fabrication of low cost and low impact RH and temperature sensors for the Internet of Environmental-Friendly Things

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Abstract- Given the increasing number of connected devices as a consequence of the Internet of Things (IoT) revolution, the issue of the removal and recycling of electronics is becoming more and more urgent. In this context, biodegradable electronics is expected to be one of the biggest technological revolutions to tackle this problem. Following this direction, in this work we present the fabrication and characterization of temperature and humidity sensors based on biodegradable materials with the goal of making their removal easier as well as reducing their environmental impact. In particular, these multi-sensing devices were fabricated following a screen-printing process using a carbon-based paste and a conjugated polymer, both on paper and on a water soluble substrate. The results are more than promising and show how with our biodegradable sensors it is possible to obtain a sensitivity of 1 dec/20%RH to moisture content and around 0.04%/°C sensitivity to temperature. It is demonstrated that the simplicity and flexibility of the fabrication approach followed in this work paves the way to a set of new "green" IoT nodes that could be extended to wide range of sensing applications.

Keywords: carbon; humidity; paper; PVA; PEDOT:PSS; screen-printing; temperature

1. Introduction

Sensing technologies are one of the key fields in both consumer electronics and industrial environment. Today we are measuring everything; sensors are in cars, buildings, cell phones, watches, and many other things of our daily life. Thus, this trend towards an ubiquitous sensing has made that the research and development in this field becomes particularly interesting.

Humidity sensors are among the most studied in this area since they play a key role in several areas, such as agriculture, food industry or healthcare (e.g., pharmaceutical & bio-tech fabrication) [1], [2]. Humidity sensors are usually capacitive or resistive and, although there are further variations, they differ from others mainly in their preciseness, time response, size and costs [3]. However, given the circumstances imposed by the new technological applications, further aspects are becoming of special interest for their development, such as flexibility, biocompatibility or easy disposability [4]–[6]. This has led the novel fabrication methods to be focused not only on a highly scalable and economic production of devices, but also on their biodegradability in order to minimize, or even eliminate, their impact on the environment [7].

Printed sensors step exactly in this demand, as they allow the fabrication of cost-effective sensors based on fully biodegradable conductive pastes, sensitive materials and substrates [8], [9]. Therefore, several printable and organic materials have been proposed in this direction, such as conjugated polymers [9], graphene-based materials [10]–[12] and carbon-based pastes [13]. In the same way, different flexible substrates have also been used to deposit these materials, including paper [14]–[16] or liquid crystal polymers [17], [18], among others [19], [20].

Many of these materials and substrates has been already studied to develop temperature and humidity sensors, however, most of the implementations proposed in the literature resort to non-biodegradable materials to develop some elements of the final devices. For instance, Khan *et al*. presented a printed humidity sensor based on an egg albumin sensitive layer, but using silver electrodes on a PET substrate [21]. This approach was also followed in [22] and [12] using graphene oxide as sensitive layer and, in general, it can be found in many other works with different kind of materials [21], [23], [24]. Alternatively, Alrammouz *et al.* proposed a capacitive humidity sensor based on self-assembled graphene oxide sheets on a paper substrate, but with the drawback of using electrodes made of aluminum [25].

Within this context, in this work we present the fabrication and characterization of flexible humidity and temperature sensors by means of printing techniques and biodegradable materials (for both electrode and substrate) that are fully compatible with a cost-effective fabrication of eco-friendly devices. For that, a simple screen-printing process was used, demonstrating the feasibility of this approach through the combination of different conductive pastes and substrates. On one hand, we used standard paper as substrate since, in addition to being compatible with printing techniques and providing flexibility, its hydrophilic nature makes it a perfect candidate to act as a sensing material in RH sensors [26]–[28]. On the other hand, poly-vinyl alcohol (PVA) films were also used for this purpose, given that the dielectric and conductive properties of this hygroscopic polymer change when the hydroxyl groups (O-H) of its structure interact with the water molecules [29]. These two substrates were used to print on them different electrodes configurations also using fully biodegradable conductive materials, specifically poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) and a carbon-based paste. Each possible combination of these two conductive pastes and substrate materials was fabricated and characterized.

The manuscript is structured as follows: following this introduction, Section 2 summarizes the materials used for the fabrication of the sensors, together with the methodologies followed for their characterization. Section 3 presents the results of the temperature and humidity sensors fabricated, as well as a hybrid configuration to monitor both temperature and humidity on the same device. Finally, the main conclusions are drawn in Section 4.

2. Experimental Section

2.1. Materials

Two conductive inks were used for the fabrication of the printed sensors, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) and a commercial carbon-based paste, both of them providing flexibility and biodegradability. PEDOT:PSS at a weight content of 1.3 wt.% (viscosity: >14000 mPa.s) was acquired from Sigma Aldrich (St. Louis, MO, USA) [30], while the Loctite ECI 8001 E&C carbon-based paste (viscosity: 6500 mPa.s) was provided by Henkel AG (Düsseldorf, Germany) [31]. These pastes were printed on three different substrates: standard paper (DIN ISO 9706, 80g/m^2) [32], polyvinyl alcohol (PVA) films from Wagner Polymertechnik GmbH**,** and polyethylene terephthalate (PET) foils from DuPont (product name: Melinex® 506, Wilmington, DE, USA) [33]. This latter only for comparison and control since it is not biodegradable.

2.2. Devices Fabrication

A capacitive Interdigitated Electrode (IDE) structure was considered for the RH monitoring devices. Given that the dependence of the dielectric constant of the substrates with respect to the RH, the capacitance of these structures is also subject to the level of RH [34]. The advantage of the IDE layout lays in the simplicity of its geometry, and therefore, its easy application via printing on the flexible substrates [35]. After optimizing the four combination of pastes and substrates by printing lines of several widths, the combination carbon on PVA-L limited the resolution of our patterns with 800 µm as minimal reproducible dimension. The IDE structure used in work (Figure 1) has 10 fingers per electrode with a distance between consecutive fingers and width of 1 mm. These dimensions are large enough to minimize the possible effects on the capacitance as a consequence of the paste spreading over the substrate after the screen-printing process. Moreover, given that the resistivity of both of the conductive pastes used is temperature-dependent [31], [36], we opted for a resistive sensor for the temperature monitoring which consisted of a simple line with a length of 5 cm. In addition, in order study the change in resistance with respect to the temperature (sensitivity and hysteresis) at different average resistances, we tested two different lines widths (3 mm and 5 mm), as it is shown in Figure 1.

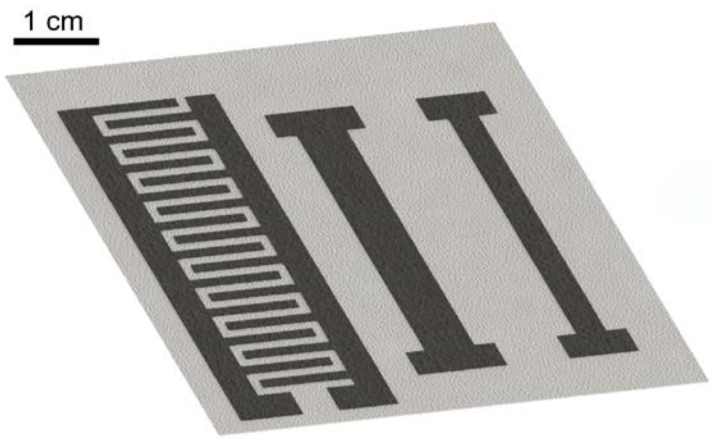

Figure 1. Left: Capacitive IDE layout. Right: Resistive line layouts with the same length (5 cm) and different widths (5 mm and 3 mm).

These layouts were screen-printed with the different pastes on the different substrates with a manual screen-printer (FLAT-DX200, from Siebdruck-Versand, Magdeburg, Germany). Before printing, the PET substrate was washed using deionized water and ethanol with the assistance of ultrasonic treatment in order to remove the impurities on the surface [37]. Moreover, pressurized air was applied to the paper substrate with the same purpose, whereas no cleaning process was carried out for the PVA films. No pretreatment was needed to screen printing the pastes on any of the substrates. The mesh used in this work was a 120 Threads per cm (T/cm) polyester mesh with a thickness of 65 µm, a thread diameter of 40 µm and an opening width of 47 µm which, according to the manufacturer, results in a theoretical wet film thickness of t_{theo} = 19.5 µm. After printing, the samples were dried using a UF55 oven (from Memmert, Schwabach, Germany) at 80 ºC in order to avoid the degradation of the substrates. We also considered two different curing times, 30 min and 60 min, to evaluate the influence of the drying process in the conductivity of the screen-printed patterns.

2.3. Characterization

The sheet resistance of the conductive layers was measured using a four-point probe head from Jandel (Leighton Buzzard, UK) connected to a source measuring unit (Keysight B2901A, Beaverton, OR, USA). The thickness of the samples was acquired using a DekTak XT contact profilometer (from Bruker Corporation, Billerica, MA, USA).

The characterization of the sensors under different environmental conditions (temperature and humidity) was performed using the setup shown in Figure 2. The sensors were connected to an impedance analyzer E4991A using a 42941A impedance probe kit (both from Keysight Technologies, Inc., CA, USA). Afterwards, each connected sensor was placed into a climate chamber VCL4006 (from Vötsch Industrietechnik GmbH, Balingen, Germany) and its impedance was measured from 1 kHz to 10 MHz under different values of temperature and humidity. The whole measurement setup was automatized using LabVIEW 2017 (from National Instruments Corporation, TX, USA).

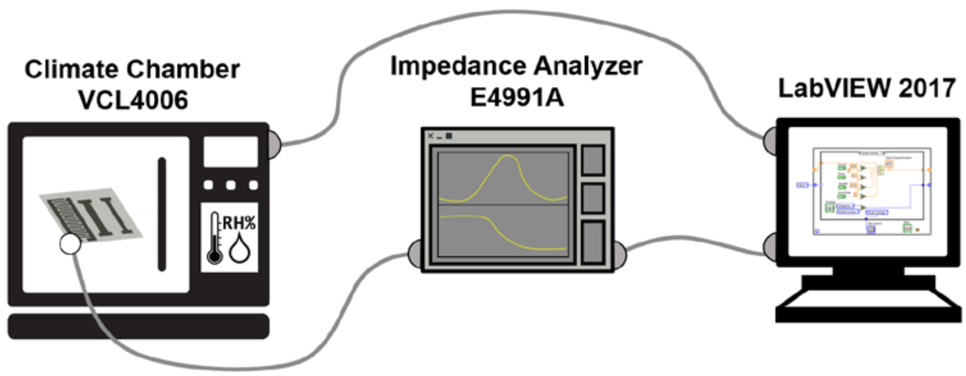

Figure 2. Characterization setup for humidity and temperature sensors.

The humidity sensors were tested as a function of both variating humidity and temperature. Firstly, the humidity sensors were characterized ranging the relative humidity level from 20%RH to 70%RH and vice versa at a temperature of 40 ºC. RH was changed in steps of 10%RH every 30 min to ensure the uniformity of RH value in the whole chamber's volume. Secondly, both sensors (resistive and capacitive) were characterized as a function of the temperature. For that, it was ranged from 15 ºC to 75 ºC and vice versa at a fixed 60%RH. In this case, the temperature was varied in steps of 5 ºC every 20 min.

3. Results and Discussion

Throughout this section, we show the results obtained for the characterization of both humidity and temperature sensors fabricated with the biodegradable carbon-based and PEDOT:PSS pastes on both paper and PVA films. The results are also compared with respect to those obtained using PET as substrate of reference.

3.1. Physical Characterization

All the fully-biodegradable RH sensors fabricated in this work are shown in Figure 3. After visual inspection, it can be clearly observed that all inks were properly transferred to the substrates, except for the carbon-based paste on the PVA substrate (Figure 3d). In that case, the printing of the carbon paste (with a thickness of 10 ± 1.2 μm) on the thin PVA substrate (30 μm) produces a non-flexible device. Thus, when this device is bent, cracks appear on the whole carbon layer surface. However, this does not happen on the paper substrate, whose surface roughness and greater thickness (~100 μm) enhance the adhesion of the carbon-paste (Figure 3c). On the contrary, the PEDOT:PSS layer, which is thinner (2 ± 0.3 μm), presents a good adhesion and flexibility on both paper and PVA substrates.

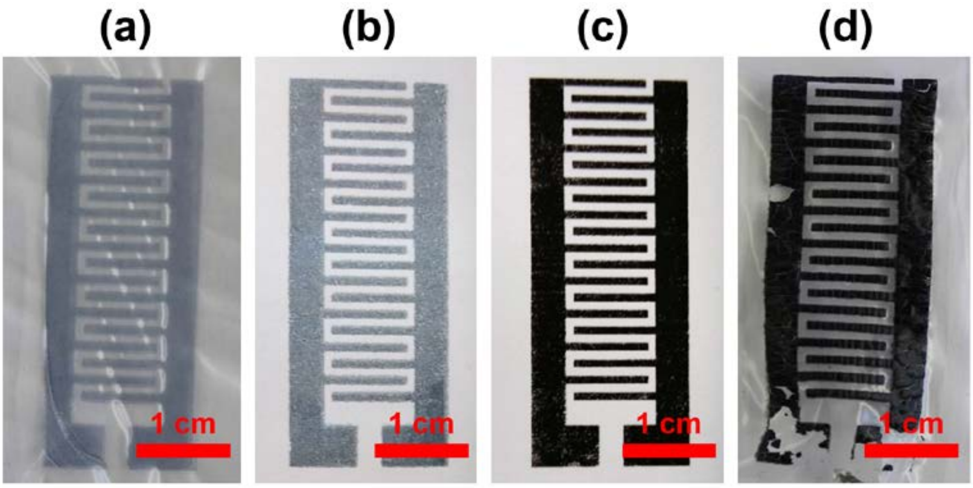

Figure 3. Real view of the screen-printed sensors: (a) PEDOT:PSS on PVA, (b) PEDOT:PSS on paper, (c) carbon-based paste on paper and (d) carbon-based paste on PVA. Scale bars: 1 cm.

The sheet resistances of each one of these patterns are presented in Table 1 for the different curing conditions. These values were obtained at ambient conditions, whereas errors were calculated as the standard deviation of five different samples. As seen, any substantial variation was observed when increasing the curing time. These values are also in accordance with respect to the obtained in other works, both for PEDOT:PSS [38]–[40] and for carbon-based pastes [41].

Table 1. Sheet resistance values for the different pastes and substrates under different curing conditions.

Paste	Substrate	Sheet Resistance (kΩ/sq.) (curing: 30 min at 80 ºC)	Sheet Resistance (kΩ/sq.) (curing: 60 min at 80 ºC)
Carbon	Paper	3.84 ± 0.86	3.73 ± 0.84
	PVA	3.89 ± 0.93	3.85 ± 0.93
PEDOT:PSS	Paper	1.20 ± 0.13	1.09 ± 0.08
	PVA	1.21 ± 0.16	1.09 ± 0.09

3.2. Characterization in relative humidity

Firstly, we characterized the impedance of the capacitive humidity sensors as a function of the RH. The results of these experiments are shown in Figure 4 for the PEDOT:PSS IDEs and in Figure 5 for the carbon-based ones. Reference calibration curves on PET can be found in supplementary Figure S1 and Figure S2, respectively.

On one side, the impedance of both carbon paste and PEDOT:PSS on PET shows no dependence on RH at any of the studied frequencies. In that case, the modulus and phase of the impedance remain almost constant for the different values of RH and decrease as the frequency increases. In the case of the phase, the decrease is less abrupt and, as capacitive structure, it is around -90º [42]. On the other side, it can be seen in Figure 4 that the same capacitive structure behaves differently on the other substrates. At low RH values, both on paper and PVA, the PEDOT:PSS structures behave as a capacitor (phase close to -90º). However, as RH increases, the phase decreases indicating an increase in the capacitor dissipation factor. This phenomenon can be related to an increase in the polymer conductivity as a consequence of the moisture accumulation on the sensing layer (i.e., the substrate), hence causing the apparition of electrical paths between electrodes. As a result of this, at high RH levels the devices start to behave more like a resistor (phase closer to 0º) than like a capacitor [42].

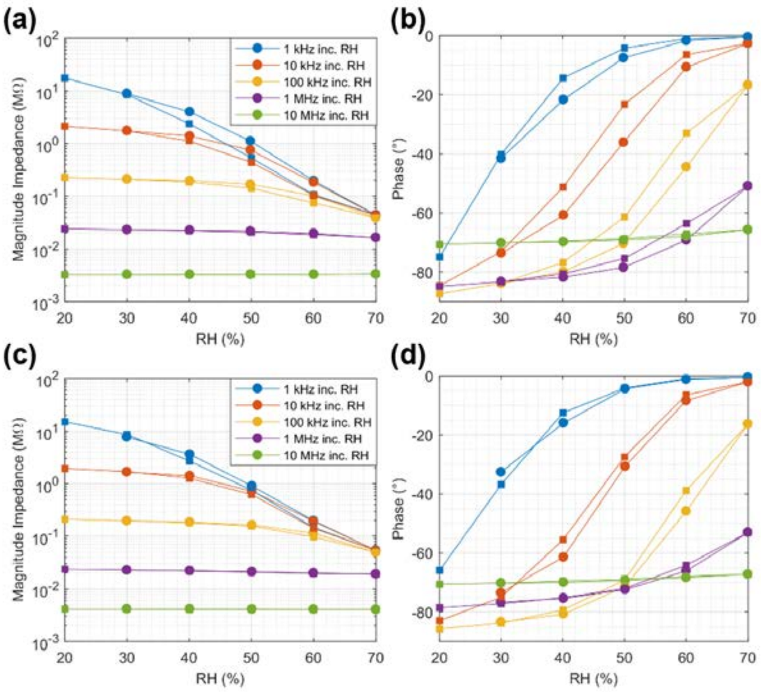

Figure 4. Impedance of the PEDOT:PSS IDEs on paper (magnitude: (a), phase: (b)) and on PVA (magnitude: (c), phase: (d)).

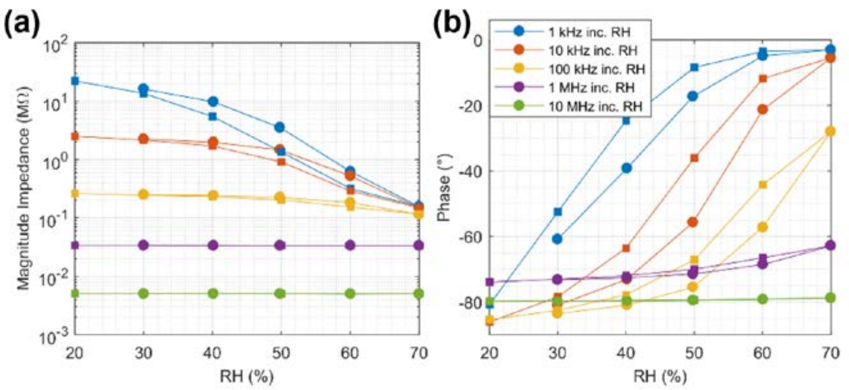

Figure 5. Impedance of the carbon IDEs on paper (magnitude: (a), phase: (b)).

Moreover, this behavior is similar to that obtained for the carbon-based IDEs on paper, as shown in Figure 5, which supports the theory that dependence with respect to the RH arises mainly from the properties of the sensing layer [43]. Finally, Figure S3 shows the results obtained for the the carbon- based paste on PVA, which demonstrates the non-viability of this combination for screen-printing (as it was shown in Section 3.1).

In general, in all cases a sensitivity as high as 1 dec/20%RH can be reproducibly achieved. In all the analyzed combination of materials, the impedance changes abruptly from 20 to 60%RH and then its responses is saturated. This behaviour is particularly significant up to 100 kHz. At low RH values, the devices are mainly capacitive (phase <80º) and when RH increases, their phases decrease, achieving phases above -10º at 60%RH (mainly resistive). However, there are differences related to the hysteresis of the sensors, the lowest value (below 3%) is found for PEDOT:PSS IDES on PVA, followed by PEDOT:PSS on paper (5%). In general, as it can be seen in Figure 4 and Figure 5, the paper substrate results in a higher hysteresis than the case of the PVA substrate.

Regarding the dynamic response of the sensors, we define the response time as t = τ, which corresponds to the 63% of the maximum value of magnitude reached at equilibrium (for every increasing step of RH). Table 2 summarizes the response time for each one of the analysed devices.

Table 2. Time response for RH

Substrate	Time response of PEDOT:PSS IDES (min)	Time response of Carbon IDES (min)
PVA	5.8	6.5
Paper	4.6	4.8

3.3. Characterization in Temperature

As it was already introduced, the resistivity of both PEDOT:PSS and carbon-based pastes used in this work is temperature-dependent. In the case of the PEDOT:PSS, it exhibits a Negative Temperature Coefficient (NTC) [36], [44], whereas the Loctite ECI 8001 E&C paste is a Positive Temperature Coefficient (PTC) ink [31]. On this basis, we characterized the impedance of the resistive patterns as a function of the temperature on a paper substrate. Looking at the behavior of single lines on paper using the same conductive materials (Fig. 6), we found that PEDOT:PSS showed a relative linear response at 1 kHz in the temperature range analyzed, as it was also demonstrated in other works [45], [46]. These sensors exhibited a sensitivity of around 0.04%/°C and 0.03%/°C (considering the relative resistance change as output variable) for the 3 mm and 5 mm lines width, respectively. Contrary to this, carbon lines exhibited a quite linear response at low temperatures, but surpassing the ~50 ºC their resistivity presented a sharp step response. It can be also seen that the response of the carbon-based paste presents a higher hysteresis than the PEDOT:PSS paste. These two effect are in accordance with the data provided by the manufacturer [31].

For these structures, the sensitivity can be estimated as 3%/°C, which is significantly high enough to be easy detectable with any low-cost measurement equipment. Furthermore, the results obtained for the lines with 5 mm of width (supplementary Figure S4) showed that, while the temperature response and hysteresis of the PEDOT:PSS patterns does not suffer significant changes, the hysteresis presented by the carbon-based paste is reduced as a consequence of the overall reduction of the pattern resistance.

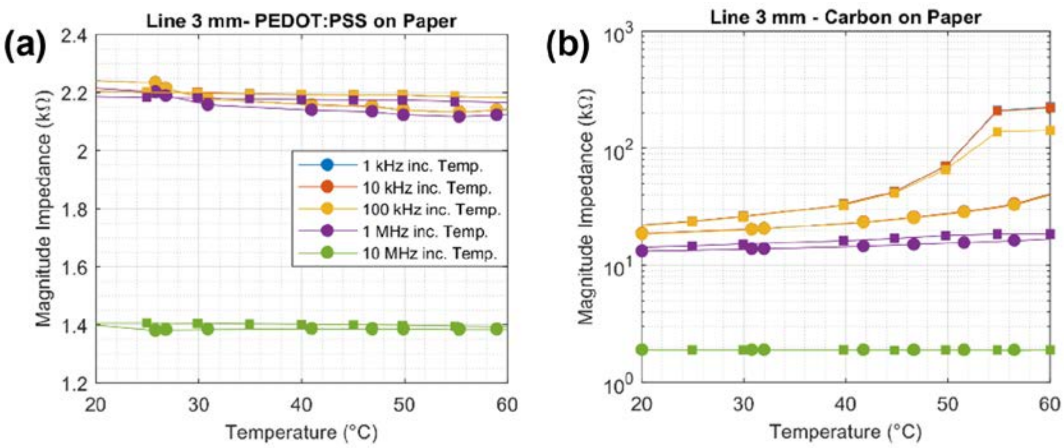

Figure 6. Impedance vs. temperature at different working frequencies of lines on paper made of PEDOT:PSS and Carbon with width of 3 mm.

Regarding the dynamic response of the sensors and following the same definition as the one employed for RH, Table 3 depicts the response time for each sensor characterized.

Table 3. Time response for Temperature

Substrate	Time response of PEDOT:PSS IDES (min)	Time response of Carbon IDES (min)
PVA	3.9	3.3
Paper	4.5	3.8

3.4. Comparison with similar sensors in the literature

In this section, we compare the sensors proposed in this work with other fully biodegradable sensors presented in the literature. The comparison was made in terms of type of sensor, materials, fabrication process, area and sensitivity to the variable to monitor (temperature or humidity), as presented in Table 4. In recent years, novel approaches on carbon nanotubes (CNTs) in their various forms, single-wallet and multi-wallet (SWCNTs, MWCNTs), are positioning this material among the most promising materials for the development of biodegradable sensors [47]. For instance, Liakos *et al.* [48] and Zhu *et al.* [49] used this conductive organic material to fabricate humidity sensors on different biodegradable substrates, achieving a quite similar sensitivity to humidity changes. However, although these sensors also provide interesting features such as flexibility, their fabrication processes do not allow to pattern surface of the substrates in order to optimize the sensing area. Contrary to this, printing techniques are able to transfer different kind of layouts on the substrates, such as the capacitive structures presented in this work, which allows to enhance the sensitivity of humidity sensors more than two orders of magnitude. In addition, some of the presented processes are not suitable for a large-scale and cost-effective production of devices, as it is de case of the one presented in [49].

Table 4. Comparison with other biodegradable sensors.

Reference	**Sensor Type**	**Electrode and Substrate Materials**	**Fabrication Process**	**Area (cm^2)**	**Sensitivity (Δ|Z|/ºC or Δ|Z|/%RH)**
Salvatore *et al.* [50]	Temperature	Mg/EcoFlex	UV lithography / etching	7	70 Ω/ºC
Yi *et al.* [51]	Temperature	Zn/Galactomannan	Drop Casting	~0.5	5 Ω/ºC
Liakos *et al.* [48]	Humidity	SWCNT, Sodium alginate, $CaCl_2$	Immersion/Coating	-	1210 Ω/%RH @ 1kHz
Zhu *et al.* [49]	Humidity	Cellulose Nanofibers, CNTs	Vacuum filtration	0.5	~1307 Ω/%RH @ DC voltage
Syrový *et al.* [52]	Humidity	Carbon Paste Cellulose nanofibril	Screen-Printing	1	Up to 122 kΩ/%RH @ 1kHz
Liu *et al.* [53]	Temperature	Laser-Induced Graphene (LIG)/Starch Film	Laser-Patterning and Transferring	-	~1.2 kΩ/ºC
Barras *et al.* [54]	Humidity	Carbon Filaments and carboxymethyl cellulose on Paper	Screen-printing	0.7	~2.6 kΩ/%RH @ DC voltage
This work	Humidity	PEDOT:PSS on Paper	Screen-Printing	4	217.2 kΩ/%RH @ 1kHz
		PEDOT:PSS on PVA			207 kΩ/%RH @ 1kHz
		Carbon Paste on Paper			204 kΩ/%RH @ 1kHz

Temperature	PEDOT:PSS on Paper	Screen-Printing	1.5	25 Ω/ºC @ 10kHz
	Carbon Paste on Paper			75 Ω/ºC

In the case of biodegradable temperature sensors, authors like Salvatore *et al.* [50] and Yi *et al.* [51] also opted for the pattering of resistive layouts on biodegradable substrates; one of them following a UV lithography and etching process, after which the pattern is transferred to the substrate (which is a similar process to the one followed in the LIG-based sensor presented by Liu *et al.* [53]); and the other following a mask-aided drop casting method. It can be seen that the sensitivities of the temperature sensors presented in this work are higher than that obtained with the Mg-based and Zn-based sensors, even with smaller areas (e.g., when compare the carbon-based sensor with respect to the Mg-based one). Regarding printed sensors, although there are numerous screen-printed as well as inkjet-printed sensors reported in the literature, most of them are based on non-biodegradable materials (either the electrodes or even both electrodes and substrates). Among the few fully biodegradable sensors that can be found are the screen-printing approaches presented by Syrový *et al.* [52] and Barras *et al.* [54], which demonstrate the trend towards the use of carbon materials on cellulose-based substrates for the fabrication of fully biodegradable and printed sensors.

3.5.Characterization as Hybrid Sensors

Based on the results obtained for both RH and temperature sensors, the devices fabricated could be successfully employed in a hybrid sensor, where the RH is extracted by measuring the impedance between the terminals of IDE structure, and the temperature through one of the junctions of the IDE structure, i.e., the line that links all fingers of one IDE terminal [55]. To exemplify this concept, in Figure 7 we show a comparison between the impedance of a carbon-based line of 3 mm of width and the impedance of a carbon-based IDE structure on the same substrate (paper). These results show how at 1 kHz and 10 kHz the combination of the two measurements, at the IDE terminals and at the line ends, would provide sufficient data to isolate the effects of temperature and humidity, resulting in a biodegradable, compact, simple and cost-effective multi-sensing method.

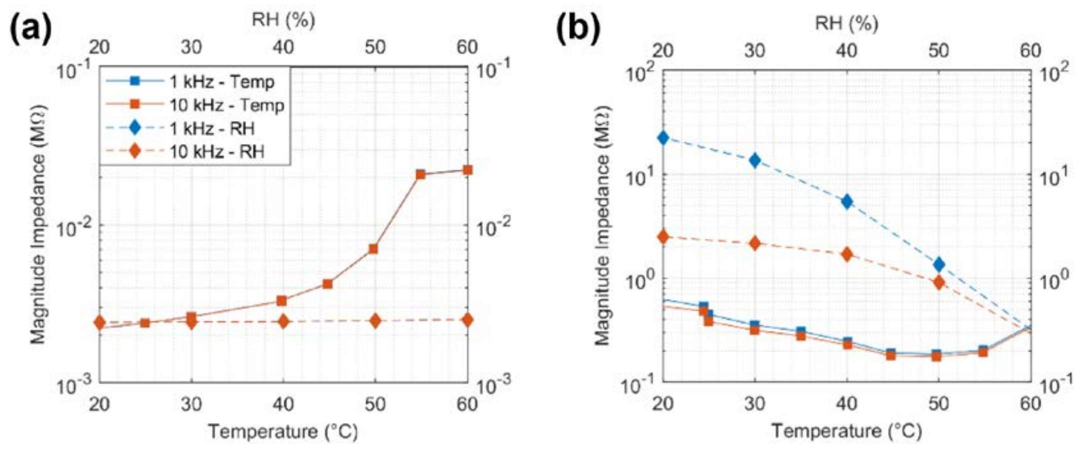

Figure 7. Impedance as a function of temperature and RH for a carbon-based line with 3 mm of width (a) and a carbon-based IDE structure (b), both on paper.

4. Conclusions

With this contribution, we intend to shift the focus of research in printed electronics and sensors towards a real sustainability of printed Internet of Things nodes. For this reason, we designed, fabricated and characterized a set of different sensors with biodegradable materials (both conductive materials and insulating substrates), in order to measure the temperature and humidity content in the environment. Our low-cost, sustainable and high throughput method allowed us to produce multi-sensing devices made of conductive polymers and a carbon-based paste on both paper and PVA substrates. The performances of these sensors are remarkable and detectable with low-cost equipment (0.04% change in relative resistance per degree Celsius, 1 dec change in impedance per 20% RH). This factor, in combination with the facile production method and the significantly reduced environmental impact, makes this new class of sensors promising candidates for a sustainable development of sensors within the Internet of Everything paradigm.

Acknowledgements

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Fabrication and characterization of fully printed biodegradable sensors for humidity and temperature sensing. These sensors will contribute enormously to pave the way to more cost-effective and environmental friendly devices.

Keywords: carbon; humidity; paper; PVA; PEDOT:PSS; screen-printing; temperature

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