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ORIGINAL RESEARCH OR TREATMENT PAPER

Colorimetric Evaluation of Pictorial Coatings in Conservation of Plasterworks from the Islamic Tradition

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ABSTRACT
The main aim of our current investigation is the colorimetric evaluation of protective treatments (consolidants) applied to traditional Islamic plasterworks, under natural ageing conditions. From analyses of the original pictorial plaster remains in the Courtyard of the Maidens of the *Real Alcázar* in Seville, Spain (a World Heritage Site) we prepared test specimens, using materials and techniques similar to the original ones. We analysed 56 test specimens painted with four pigments (yellow, green, blue, and red), using two different binders (animal glue and gum arabic), onto which five representative consolidants were applied: barium hydroxide, acrylic copolymer, polyvinyl butyral, ethyl silicate, and bacterial carbonatogenesis. The test specimens were subjected to natural ageing for one year (indoors and outdoors), enabling a colorimetric assessment to be made of the changes of the polychrome surfaces. The colorimetric heterogeneity of the 56 specimens after ageing registered an average value of 2.7 CIELAB units, assessed using the mean colour difference with respect to the mean. In the aged specimens, the addition of consolidants resulted in average colour differences (mainly lightness differences) of 10.7 and 6.7 CIELAB units, considering as a reference the specimens without consolidants aged indoors and outdoors, respectively. These colour differences were very similar for both binders but not for the four pigments, higher values being found for the blue and red pigments. Considering as reference the samples without consolidants aged outdoors, we found no statistically significant colour differences, either among the five consolidants ($p = .094$) nor the two binders ($p = .674$) used. In addition to the magnitude of colour differences, the choice of the most appropriate consolidants must also consider aspects related to performance and effectiveness. Overall, for the type of paints tested, the polyvinyl butyral consolidant appeared to perform the best, followed by the ethyl silicate.

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Introduction

The conservation or restoration of architectural ornamentation requires the treatments applied to be monitored. Treatments involving the consolidation of polychromed plasterwork coatings can entail complex problems. In addition to the difficulties of conserving polychrome decoration, due to binder deterioration, re-polychroming combined with restoration repeated over the years raises additional challenges. The historical and artistic importance of the chromatic finishes on traditional Islamic plasterworks, combined with the complex conservation challenges they present, led us to conduct a colorimetric evaluation of the effectiveness of protective treatments applied to these plasterworks, as the main goal of the current article.

While previous studies have examined the effectiveness of consolidation treatments, most research focuses on controlling the effectiveness of mortars (Mateos et al. 2006; Rubio 2006, 2010; Villegas 2007; Jroundi et al. 2013; Sassoni et al. 2013), and very few works investigate polychrome colour changes resulting from such treatments (García-Talegon et al. 1998; Gilabert 2012; Ferreira and Delgado 2014).

The present study forms part of more extensive research undertaken by our team on various architectural elements of the *Real Alcázar* in Seville (Spain), including the plasterwork and tilework in the Courtyard of the Maidens (Melgosa et al. 2015; Calero-Castillo et al. 2016), the façade and eaves of the Palace of King Don Pedro (López et al. 2015), and other plasterworks (Bueno and Medina Flórez 2004; García Bueno, Medina Flórez, and González Segura 2010). Brightly coloured plasterwork in the Palace of King Don Pedro (Figure 1, left) by master craftsmen from Toledo, Granada, and Seville dates to 1355–1366 (Marín Fidalgo 1990; Almagro 2007). The conservation of these decorative elements is currently proving difficult due to successive interventions in later periods (including re-polychroming, whitewashing, and aggressive cleaning), which have led to disintegration. Consequently, the original polychromes that survive are at risk of being lost altogether (Figure 1, right). Thus, we have developed a methodology to restore and

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Figure 1. Left: Plasterwork on the south-eastern wall of the Courtyard of the Maidens, where the original blue polychrome persists. Right: Detail of results of a cleaning test on one area of this plasterwork, which decorates the main entrance to the *Salón del Techo de Carlos V*, where some remains of the original red polychrome can be seen.

preserve, as far as possible, the original polychrome finishes that remain.

More specifically, the present research seeks to assess the colour changes that took place over a 12-month period of natural ageing (indoor and outdoor), using a series of trial samples treated with five representative consolidants. The samples, made of layers of polychrome on gypsum plaster, simulate traditional Islamic materials and techniques previously identified by the team (Bueno and Medina Flórez 2004; García Bueno, Medina Flórez, and González Segura 2010; Calero-Castillo et al. 2016). Preparation for the study took into account the state of conservation of the plasterwork polychrome at the Courtyard of the Maidens and also the variety of products currently available for fortifying and protecting colour pigments. The present work employs colorimetric analyses for assessing the deterioration of pictorial coatings after restoration treatments, in order to select the most suitable ones, both for the plasterwork under study, and for works of a similar typology and age.

Materials and methods

The experimental method was based on the following three processes: preparation of test samples; natural ageing test; and colorimetric analyses, as explained below.

Preparation of test samples

An effort was made to ensure that the materials and techniques used in the preparation of the test specimens matched those typifying polychromes of the Moorish tradition. Selections were based on scientific analyses of 155 original microsamples taken from the plasterwork of the Courtyard of the Maidens (Calero-Castillo et al. 2016), together with information from other studies on coatings from a period similar to that of the *Real Alcázar,* Seville (López et al. 2015), the *Alhambra*, Granada (Cardell-Fernández and Navarrete-Aguilera 2006), the *Madraza* oratory, Granada (García Bueno, Medina Flórez, and González Segura 2010), and the *Cuarto Real de Santo Domingo*, Granada (Bueno and Medina Flórez 2004). Of the 155 microsamples analysed, 133 were studied to determine the sequence of paint layers, 10 were analysed to establish the composition of the mortar, and 12 were examined for binders and fixatives. The methods used in the study of these microsamples included (Calero-Castillo 2016) stereoscopic microscopy of untreated samples; light microscopy with polarizing light, via microchemical trials and selective colouring of layers of tempera and oil paint; scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDX); Fourier-transform infrared spectroscopy (FTIR) for the components of coatings and varnishes; gas chromatography and mass spectroscopy (GC-MS) to determine lipophilic (drying oils, resins, and waxes) and hydrophilic substances (proteins and polysaccharide gums, such as gum arabic and related products); and X-ray diffraction spectrometry (XRD) for the characterization of the mortars.

The analysis of the original plasterwork microsamples revealed that their main component was gypsum ($CaSO_4$ $2H_2O$), comprising 100% of three samples. Calcite ($CaCO_3$) constituted around 80% of two samples. Also, various mineral pigments of different origins appeared in small quantities (a maximum of 10%). The proportion of additional elements which were not identified in all samples analysed, mainly quartz (SiO_2), ranged between 1% and 4%. Previous analyses of plasterwork in other parts of the *Real Alcázar* (façade of the Palace of King Don Pedro) gave similar results (López et al. 2015). The analysis of the original microsamples of pigments identified iron oxide (Fe_2O_3), natural malachite [$Cu_2(CO_3)(OH)_2$], natural azurite [$Cu_3(CO_3)_2(OH)_2$], and cinnabar (HgS), among other substances. Finally, analyses of the original microsamples of binders (Calero-Castillo et al. 2016) revealed the presence of animal glue (e.g. glycine, proline, and hydroxyproline), a material that had previously been identified in other case studies, such as that of the plasterwork at the *Alhambra*, Granada (García, Medina, and Gómez 2006). Elsewhere, gum arabic (a polysaccharide formed by a main chain of ß-1,3-galactopyranose units, together with residues of ramnopyranoses, arabinopyranoses, arabinofuranoses, glucuronic acid, and 4-O-methylglucuronic acid) was used as a binder in plasterworks, as in works such as the *Cuarto Real de Santo Domingo*, Granada (Bueno and Medina Flórez 2004) and the *Camaranchón* of the *Real Alcázar*, Seville (Calero-Castillo 2016). This latter example is of particular significance for the present study. As well as being located within the palace itself, these are the only plasterworks with the original materials, having undergone no later re-polychroming (having remained hidden during the remodelling conducted in the period of the Catholic Monarchs, in the late fifteenth century). Other studies (García Bueno, Medina Flórez, and González Segura 2010) have confirmed that the plasterworks from al-Andalus were polychromed in tempera, the most frequently used binders being animal glue and gum arabic.

The materials chosen for the preparation of our trial samples were as follows: (1) support: ceramic bricks (23 × 11.5 cm); (2) plaster (2 cm thick) made with gypsum hemihydrate (95%) and slaked lime (5%); (3) polychrome layers produced with yellow (iron oxide), green (malachite), blue (azurite), and red (cinnabar) pigments, using animal glue and gum arabic as binders. Specifically, two layers were applied by brush. The technical specifications for the four above-mentioned pigments are shown in Table 1. Four XRD spectra (mineralogical analyses), four SEM-EDX spectra (chemical analyses) and four SEM images, corresponding to the four pigments employed, are provided as Supplementary Materials. The two binders, animal glue and gum arabic (®CTS Srl), had a concentration of 10%. Previous studies (Gueli, Pasquale, and Troja 2017) have confirmed that the binders used can influence the appearance of the colour layers. Hence, both binders were used in our trial samples in order to assess this chromatic influence.

After a literature review (Durán 1995; Borgioli and Cremonesi 2005; Jroundi et al. 2014), we selected five types of consolidants (both organic and inorganic), including those traditionally used in plasterwork restoration and those introduced more recently: barium hydroxide, polyvinyl butyral, acrylic copolymer (ethylmethacrylate/methyl-acrylate, EMA/MA), ethyl silicate, and bacterial carbonatogenesis. Barium hydroxide and ethyl silicate are widely used in restoration for being highly compatible with mortars (Pascoal et al. 2015), in this case gypsum. However, its action, based on the formation of new inorganic material, is difficult to control and often triggers changes in the refractive index of the pigments comprising the layers of paint. The acrylic copolymer and polyvinyl butyral present the disadvantage of forming an organic layer that can leave residues on the surface, forming a waterproof layer and thus provoking changes in appearance, mainly in the form of shiny patches, especially at high concentrations (Ferreira and Delgado 2014; Calero-Castillo 2016). The consolidant action of bacterial carbonatogenesis results from the formation of new calcite, following the application of a nutrient solution to encourage bacterial activity. In this case, although prior studies have demonstrated that the colour was not masked nor took a shine or other undesirable effects (Jroundi et al. 2014), the results of this consolidant on polychromes have not yet been thoroughly assessed and therefore were evaluated here. The technical specifications for the five selected consolidants are shown in Table 2.

The binder:pigment ratio was chosen in order to produce homogeneous and opaque paint layers that would be fluid enough for proper application on the gypsum plasters. Specifically, for both binders, the binder:pigment ratios were 10 ml:3 g, in the case of pigments P1 and P4, and 10 ml:5 g, in the case of pigments P2 and P3. The selection of the concentrations, solvents, and application methods for the consolidants used was performed as follows. Regarding the polymers, for the polyvinyl butyral and the acrylic copoymer, low concentrations were used in order to avoid the making of a harmful impermeable layer that would prevent the transpiration of the underlying bindings. The 5% concentration for the acrylic copolymer xylene solution was based on previous treatments used at the consolidation of the plasterworks of the façade of the Palace of King Don Pedro, in the *Real Alcázar* in Seville (Almagro et al. 2010). The polyvinyl butyral was prepared in an ethanol solution (5% concentration) in the way it was used by our team for the consolidation of plasterworks of the *Cuarto Real de Santo Domingo* and the Mihrab of the Mosque of Fiñana, Almería (Spain) (Bueno et al. 2006).

A total of 56 trial samples were prepared: 8 samples (4 pigments × 2 binders) without any consolidant for indoor exposure; 8 samples (4 pigments × 2 binders) without any consolidant for outdoor exposure; 40 samples (4 pigments × 2 binders × 5 consolidants) for outdoor exposure. Hereafter the following abbreviations will be used for the samples, according to the

Table 1. Characteristics of the four pigments used for the preparation of the test specimens.

Name	Pigment	Trade name	Mineralogical analyses (XRD)	Chemical analyses (SEM-EDX)	Granulometry
P1	Yellow	Iron Oxide Yellow Ref. 40301 (KREMER® Pigmente)	Goethite [α-FeO(OH)] Quartz (SiO_2) Kaolinite [$Al_2Si_2O_5(OH)_4$]	Fe Si Al	0–80 µ
P2	Green	Malachite natural, standard REF. 10300 KREMER® Pigmente	Malachite [$Cu_2CO_3(OH)_2$] Rutile (TiO_2)	Cu	0–120 µ
P3	Blue	Azurite natural, standard REF. 10200 KREMER® Pigmente	Azurite $Cu_3(CO_3)_2(OH)_2$	Cu	0–120 µ
P4	Red	Natural Cinnabar REF. 10620 KREME® Pigmente	Cinnabar (HgS)	S Hg	0–70 µ

conditions of exposure, pigments, binders, and consolidants: (a) for the exposure conditions, 'indoor exposure' (E1) and 'outdoor exposure' (E2); (b) for the pigments, yellow (P1), green (P2), blue (P3), and red (P4); (c) for the binders, animal glue (B1) and gum arabic (B2); and (d) for the consolidants: barium hydroxide (C1), polyvinyl butyral (C2), acrylic copolymer (C3), ethyl silicate (C4), and bacterial carbonatogenesis (C5). For example, the sample with the code E1.P2.B1 stands for the one exposed indoor (E1), with a green pigment (P2), animal- glue binder (B1), and no consolidant; while the sample coded as E2.P1.B1.C5 indicates the one exposed outdoor (E2), with a yellow pigment (P1), animal-glue binder (B1), and bacterial carbonatogenesis consolidant (C5). We will also note sets of samples such as E1.P1-P4.B1-B2 to indicate all samples exposed indoor, with any of the four pigments, either of the two binders, and no consolidants (i.e. a total of eight samples).

For easier handling of our samples, two types of pigments were applied to each brick (Figure 2). The scheme of Figure 2 shows a total of 14 samples because we considered only the binder B1 and two pigments (P1 and P2). If the other binder, B2, were considered, we would have 14 additional samples for these two pigments (P1 and P2). Finally, following this scheme, but using the two other pigments, P3 and P4, and the two binders, B1 and B2, we have 28 additional samples, for a total of 56 samples used in our analyses. For example, the samples E1.P2.B1 and E2.P1.B1.C5, mentioned above, are located in Figure 2 at the bottom left and the upper left corners, respectively.

Natural ageing test

Of the 56 trial samples, 48 (8 without consolidants and 40 with consolidants) were subjected to natural outdoor ageing, and 8 (without consolidants) underwent indoor ageing, all for 12 months. For the outdoor ageing process, environmental conditions (temperature, relative humidity, ultraviolet radiation, total solar radiation, rain, and wind) were monitored to assess the effectiveness of the treatments and their effect on the polychromes. For the indoor ageing process, only temperature and relative humidity were tracked.

Although accelerated ageing tests are sometimes used in these types of studies, in the present case, the size and quantity of test specimens made the use of an ageing chamber unfeasible. In addition, we took into account that long-term exposure to common environmental conditions, typical of natural ageing processes, would provide more realistic

Table 2. Characteristics of the five consolidants applied on the polychrome layers of the test specimens.

Name	Consolidant	Trade name	Chemical composition	Solvent/Concentration	Application
C1	Barium hydroxide	Barium hydroxide CTS	$Ba\ (OH)_2{\cdot}8H_2O$	Distilled water/Solution saturated	Apply to the surface by compress for c.10 min.
C2	Polyvinyl butyral	Mowital® B60H (CTS)	$(C_8H_{14}O_2)n$	Ethanol solution/5%	Apply by brush through Japanese tissue once per day on two consecutive days.
C3	Acrylic Copolymer (Ethylmethacrylate/methyl-acrylate, EMA/MA)	Paraloid® B-72 (CTS)	$C_6H_{10}O_2$	Xylene solution/5%	Apply by brush through Japanese tissue once per day on two consecutive days.
C4	Ethyl silicate	Bioestel 1200 (CTS)	$Si(OC_2H5)_4$	Undiluted	Apply by brush through Japanese tissue once per day for two consecutive days.
C5	Bacterial carbonatogenesis	–	*Myxococcus xanthus* (precultured in liquid medium CT). (Jroundi et al. 2013)	Sterile nutritive solution M3P: 1% Bacto Casitone, 1% Ca $(CH_3COO)24H_2O$, 0.2% K2CO3·½H_2O in a 10 mM phosphate buffer, pH 8) Liquid medium CT: 1% [wt/vol] Bacto Casitone and 0.1% [wt/vol] $MgSO_4\ {\cdot}7H_2O$ in a 10 mM phosphate buffer, pH 6.5	Spray and cover with a plastic film 1 day. Inoculation of the *Myxococcus xanthus* bacterium. 6 consecutive days. Application of the nutrient solution 3 times per day.

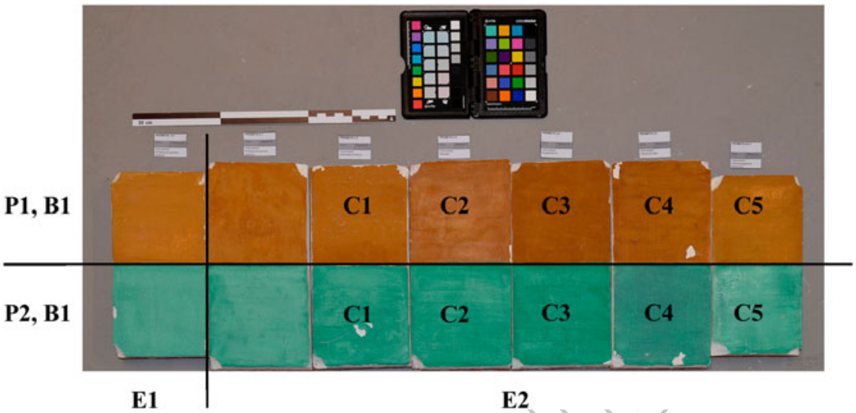

Figure 2. Example of a series of samples using pigments yellow (P1) and green (P2), binder animal glue (B1), and consolidants C1–C5, for indoor (E1) and outdoor (E2) exposures, prior to the ageing test.

information than exposure to the more severe conditions over a short time span in ageing chambers (Borgioli and Cremonesi 2005). We used a sufficiently long ageing period of 12 months, in order to achieve steady colorimetric results.

The samples aged outdoors were placed in a rust-proof structure exposed to the air, attached to the wall in a vertical position. The entire structure was protected by an overhang so that the position of the samples would simulate the placement in real life (Figure 3, left). To gather data on the environmental variables during the outdoor ageing process, we used the following equipment: Class I pyranometer (ISO 9060:1990), anemometer, wind vane, pluviometer, ultraviolet sensor, temperature sensor (ranging from −30°C to 50°C, accuracy ± 0.3°C at 25°C), and surface relative-humidity sensor (ranging from 0 to 100%, accuracy ± 2%). The samples aged indoors were horizontally placed in a laboratory cupboard and were monitored only for temperature and relative humidity (Figure 3, right). All of the environmental data were managed using SENSONET® software via in-built equipment in the weather station for measuring the signals.

Throughout the 12-month ageing process, the average temperature of the outdoor samples was 16.0°C (47.4°C maximum, −5.0°C minimum) vs. 20.0°C (30.0°C maximum, 7.0°C minimum) for indoor samples. The relative humidity outdoors was 38% (100% maximum, 10.9% minimum) and 42% indoors (48.3% maximum, 36.6% minimum). A non-negligible flow of harmful radiation was considered to affect the polychrome finishes during the ageing period of the outdoor samples. In particular, solar irradiance fluctuations (average value 107 w/m^2) are indicative of the harsh conditions attacking the outdoor test specimens. The UV radiation, stronger in the warmer months, was assumed to degrade the samples, especially the organic materials. The samples were often subjected to a westerly wind, the sensors marking an average speed of 3.7 km/h (a grade 1 or light breeze on the Beaufort land scale), with a maximum value of 49.0 km/h.

Colorimetric analyses

After one year of ageing, the colour was instrumentally measured on the 56 trial samples using a Konica Minolta spectrophotometer CM-2600d with diffuse illumination, 8° viewing angle, specular component excluded (SCE), target mask (illumination/measurement area) with a diameter of 8 mm, ultraviolet

Figure 3. Left: General view of the samples exposed to outdoor ageing. Top left, the equipment for monitoring environmental conditions: pyranometer, anemometer, wind vane, ultraviolet sensor, temperature sensors, pluviometer, and relative-humidity sensors. Right: Detail of a few samples exposed indoors, showing the device controlling temperature and relative humidity.

illumination of 0%, CIE D65 standard illuminant (ISO 11664-2:2007), and CIE 1964 standard colorimetric observer (ISO 11664-1:2007). The acquisition step was performed by the Spectramagic^{TM} NX Pro Color Data Software of the spectrophotometer provided by Konica Minolta. The instrument was calibrated using a white calibration plate (CM-A145) and a zero calibration box (CM-A32) provided by the manufacturer of the spectrophotometer.

The methodology was based on measuring at different points on each sample (between 4 and 8 points, depending on the colour heterogeneity of the samples) to determine the corresponding colour values in CIELAB colour space (ISO 11664-4:2008, CIE S 014-4/E:2007). This space enabled us to calculate the Cartesian coordinates (L^*_{10}, a^*_{10}, b^*_{10}), as well as the cylindrical coordinates (more associated with the main perceptual colour attributes), called lightness (L^*_{10}), chroma ($C^*_{ab,10}$), and hue-angle ($h_{ab,10}$). Lightness (L^*_{10}) represents the brightness of a surface compared to a white reference surface that is identically lit, and, for non-fluorescent samples, ranges from a value of 0 (for an entirely black surface) to 100 (for an 'ideal' reference white). Chroma ($C^*_{ab,10}$) refers to the colour intensity of a surface. Finally, hue-angle ($h_{ab,10}$) indicates the tone, expressed in sexagesimal degrees, such that hue-angles of 0°, 90°, 180°, and 270° correspond to red, yellow, green, and blue, respectively. Total colour differences, calculated using the CIELAB colour-difference formula ($\Delta E^*_{ab,10}$), can be divided into lightness (ΔL^*_{10}), chroma ($\Delta C^*_{ab,10}$), and hue ($\Delta H^*_{ab,10}$) differences (CIE 15:2004), which can be expressed as percentages of the total colour difference. Under certain 'reference conditions', the CIEDE2000 colour-difference formula (ISO/CIE 11664-6:2014) is currently recommended by the International Organization for Standardization (ISO) and the International Commission on Illumination (CIE) for an improved correlation between calculated colour differences and visually perceived differences by observers with normal colour vision. However, in the present study, several 'reference conditions' were not fulfilled (e.g. our samples were not homogeneous in colour and calculated colour differences often registered values above 5.0 CIELAB units). Therefore, only the CIELAB formula was used in the current study, taking into account also that it is the most widely used formula in cultural heritage research (Della Patria et al. 2007; Prestileo et al. 2007; Acquaviva AQ8 et al. 2010; Collado-Montero and Espejo-Arias 2015; Melgosa et al. 2015). For observers with normal colour vision, the magnitude of visual colour thresholds depends on the location of the reference colour in CIELAB space (Melgosa et al. 1994), and under optimal viewing conditions this may be even lower than 1.0 CIELAB unit (Melgosa et al. 1992; Huang et al. 2012). To determine the variability in measurements taken at 4–8 different points of each sample, we calculated the 'mean colour difference from the mean' (MCDM) in CIELAB units, according to the following equation (Berns 2000):

$$\mathrm{MCDM} = \left(\frac{1}{N}\right) \times \sum_{i=1}^{N} \left[(L_i^* - \overline{L^*})^2 + (a_i^* - \overline{a^*})^2 + (b_i^* - \overline{b^*})^2 \right]^{1/2}, \qquad (1)$$

where the subindices '*i*' represent the colour coordinates of each of the *N* measurements and the overlines represent their average values (AVG). MCDM enabled us to represent the dispersion of the three CIELAB coordinates in a single number, and therefore, evaluate the variability of the colours measured at different points on the same non-homogeneous sample: The lower the MCDM value, the less the colour variability (i.e. the more homogeneous the sample), and vice versa. MCDM in CIELAB units was determined, together with AVG and the standard deviation (SD) of the samples measured.

After the end of the one-year ageing process, we computed CIELAB colour differences to determine the ageing and the efficacy of different treatments applied. Specifically, CIELAB colour differences were computed for the following two sets of samples, in all cases considering sample pairs with the same pigments and binders:

(a) Comparison 1: Samples E1.P1-P4.B1-B2 vs. samples E2.P1-P4.B1-B2.C1-C5. A total of 40 colour pairs, fixed by the 40 samples in the set E2.P1-P4.B1-B2.C1-C5 (4 pigments × 2 binders × 5 consolidants), were compared with 8 samples (4 pigments × 2 binders in the set E1.P1-P4.B1-B2) replicated 5 times, corresponding to the measurements of samples aged indoor (E1). Therefore, in Comparison 1, we took into account both the effect of indoor (E1) and outdoor ageing (E2), as well as the effect of the 5 consolidants (C1–C5) vs. no consolidants. (b) Comparison 2: Samples E2.P1-P4.B1-B2 vs. samples E2.P1-P4.B1-B2.C1-C5. Again, 40 colour pairs, fixed by the 40 samples in the set E2.P1-P4.B1-B2.C1-C5 (4 pigments × 2 binders × 5 consolidants), were compared with 8 samples (4 pigments × 2 binders in the set E2.P1-P4.B1-B2) replicated 5 times, corresponding to measurements of samples aged outdoor (E2). Therefore, keeping the outdoor condition (E2) constant, in Comparison 2 we disregarded the effect of ageing and considered only the effect applying the five consolidants (C1–C5).

In summary, the same number of 40 colour pairs (fixed by the 40 samples in the set E2.P1-P4.B1-B2.C1-C5, corresponding to 4 pigments × 2 binders × 5 consolidants), were used in the Comparisons 1 and 2. The difference between Comparisons 1 and 2 was that in Comparison 1 we used as reference the samples

Table 3. Average and standard deviation of MCDM values (CIELAB units) for each set of samples, according to the ageing conditions (E1, E2), pigments (P1–P4), binders (B1, B2), and consolidants (C1–C5) used.

	Ageing		Pigments				Binders		Consolidants				
	E1	E2	P1	P2	P3	P4	B1	B2	C1	C2	C3	C4	C5
Average	2.0	2.7	1.5	2.3	4.5	2.6	2.6	2.9	2.8	1.6	2.7	2.3	4.9
Standard Deviation	0.8	2.1	0.7	1.4	3.1	1.7	2.1	2.3	1.6	0.9	2.9	1.6	3.3

aged indoor (and without consolidants), while in Comparison 2 we use as reference the samples aged outdoor (and without consolidants). For Comparisons 1 and 2 we performed an analysis of variance (ANOVA) test with two fixed factors (binder and consolidant) to analyse the results of the dependent variable $\Delta E^*_{ab,10}$.

Results and discussion

First, we studied the colour variability (heterogeneity) of samples using the MCDM in CIELAB units, according to ageing conditions, pigments, binders, and consolidants (Table 3).

Table 3 reflects that all AVG are higher than 1.0 CIELAB units, indicating that most of our samples are non-visually homogeneous in colour. More specifically, for the 56 samples tested, the average MCDM was 2.7 CIELAB units, with a standard deviation of 2.2 CIELAB units. Considering the ageing conditions (indoor vs. outdoor), we found that the average colour variability was slightly higher in the outdoor samples (E2) than in indoor ones (E1), implying that the outdoor conditions caused greater colour heterogeneity, as expected. Also, the standard deviation was considerably higher under outdoor (E2) than under indoor (E1) conditions. Regarding the pigment of the samples, the average MCDM values indicated that the greatest variability was found in the samples with blue pigment (P3), while the lowest one was found in the yellow pigment (P1), with a factor 3 between them. This suggests a relationship between the colour degradation of the sample and the type of pigment used. For binders, the average MCDM values were very similar in the specimens with animal glue and gum arabic (2.6 and 2.9 CIELAB units, respectively); hence, this parameter apparently exerts little influence on colour variability. With respect to the consolidants used, we found the greatest variability (highest average MCDM value) in the samples subjected to bacterial carbonatogenesis (C5), followed by C1, C3, C4, and C2, with a factor of around 3 between the highest and lowest AVG. Considering all of the factors together, we found that the samples that most varied in colour were those aged outdoors (E2) with a blue pigment (P3), using gum arabic (B2) as the binder and bacterial carbonatogenesis as the consolidant (C5). The samples presenting the least colour variability were those aged indoors (E1), with a yellow pigment (P1), using an animal-glue binder (B1) and the consolidant polyvinyl butyral (C2). The higher variability registered in the samples with consolidants bacterial carbonatogenesis (C5) and barium hydroxide (C1) was due not only to the consolidants themselves, which were aqueous and therefore partially dissolved the paint even before the ageing process (Figure 4), but also to the methods by which the consolidants were applied. In some instances, this caused significant

Figure 4. Left: Deterioration in the sample using pigments P3 and P4 and binder B1, following application of consolidant C5, prior to outdoor ageing. Right: Deterioration in the sample using pigments P1 and P2 and binder B2, following application of consolidant C1, prior to outdoor ageing.

losses and serious deterioration in the paint, particularly in the samples with yellow (P1) and green (P2) pigments, using gum arabic (B2) as a binder and barium hydroxide (C1) as the consolidant (Figure 4, right). From this, we deduce that certain consolidants in an aqueous medium, particularly those in which the application method requires prolonged contact with the surface (i.e. barium hydroxide, C1, and bacterial carbonatogenesis, C5), would not be suitable for polychromes using water-soluble binders. Other types of products that do not require lengthy contact with the surface but rather can be applied gently with a brush through Japanese tissue are preferable, e.g. the polyvinyl butyral (C2), acrylic copolymer (C3), and ethyl silicate (C4) used in the present study.

Next, we studied the magnitude of CIELAB colour differences ($\Delta E^*_{ab,10}$) designated in the previous section as Comparison 1 (i.e. samples E1.P1-P4.B1-B2 vs. samples E2.P1-P4.B1-B2.C1-C5) and Comparison 2 (i.e. samples E2.P1-P4.B1-B2 vs. samples E2.P1-P4.B1-B2.C1-C5), distinguishing the effects (AVG and standard deviations) for pigments (Figure 5), binders (Figure 6), and consolidants (Figure 7). Figures 5–7 show quite high CIELAB colour-difference values (i.e. values above 5.0 CIELAB units), in particular, higher values than those shown in Table 3, corresponding to the heterogeneity of individual samples. This means that outdoor ageing and consolidants produced marked colour changes in the samples tested. Also, Figures 5–7 show higher average colour differences for Comparison 1 than for Comparison 2 (with the only exception being Figure 7, consolidant C5), indicating that colour changes caused only by the use of consolidants (Comparison 2) were lower than those triggered by the combined effect of outdoor ageing plus consolidants (Comparison 1), as expected. Specifically, the colour-difference values (average ± standard deviation) were 10.7 ± 5.5 and 6.7 ± 4.2 CIELAB units for Comparison 1 and 2, respectively. On the average, the percentages of CIELAB lightness, chroma, and hue differences in these two total colour differences were rather similar: 56%, 33%, and 11% in Comparison 1, and 63%, 29%, and 8% in Comparison 2, respectively. From these percentages, we conclude that the main qualitative effect of adding consolidants is a change in lightness (specifically, the samples with consolidants become lighter than the original ones), together with a minimum change of hue, which should be considered the most dangerous effect for restored plasterwork in terms of visual observation.

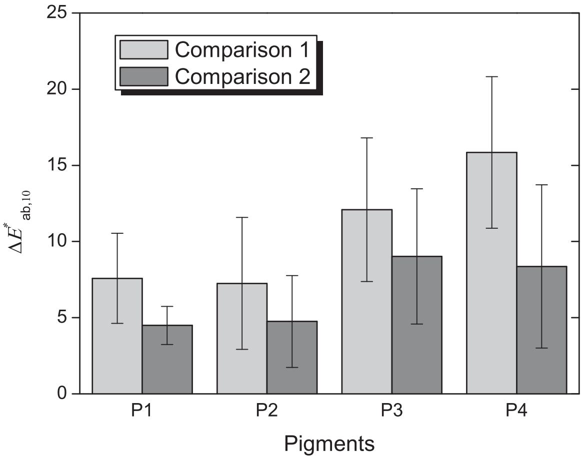

Figure 5. Average (bars) and standard deviation (error bars) values for CIELAB colour differences in Comparison 1 (E1/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5) and Comparison 2 (E2/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5), for each of the four pigments: yellow (P1), green (P2), blue (P3), and red (P4).

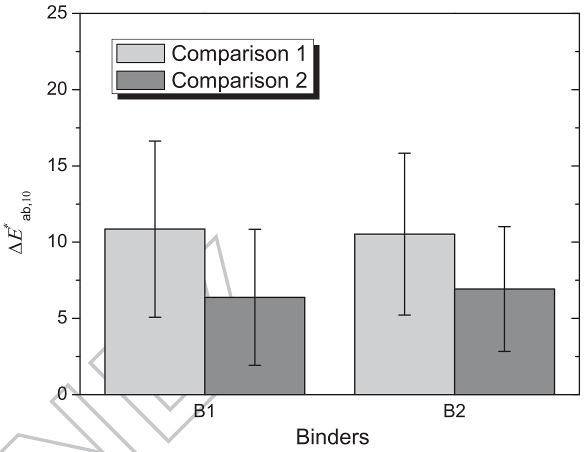

Figure 6. Average (bars) and standard deviation (error bars) values for CIELAB colour differences in Comparison 1 (E1/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5) and Comparison 2 (E2/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5), for the two binders: animal glue (B1) and gum arabic (B2).

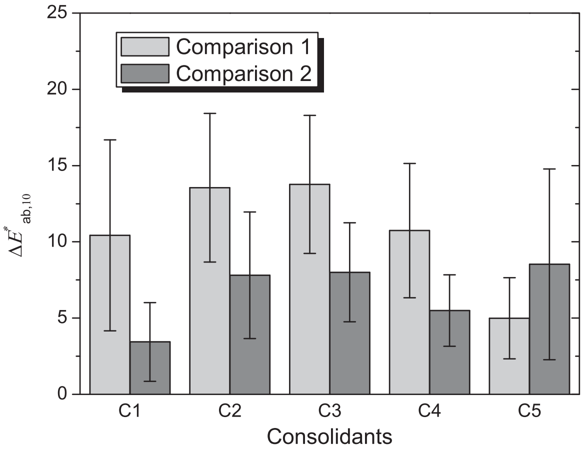

Figure 7. Average (bars) and standard deviation (error bars) values for CIELAB colour differences in Comparison 1 (E1/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5) and Comparison 2 (E2/P1-P4/B1-B2 vs. E2/P1-P4/B1-B2/C1-C5), for the five consolidants: barium hydroxide (C1), polyvinyl butyral (C2), acrylic copolymer (C3), ethyl silicate (C4), and bacterial carbonatogenesis (C5).

Figure 5 focuses exclusively on the influence of the four pigments used. The highest average $\Delta E^*_{ab,10}$ value in Comparison 1 presents in the samples with red pigment P4 (15.9 ± 5.0), followed by the blue pigment samples P3 (12.1 ± 4.7), while the lowest values registered were those of the green pigment samples P2 (7.3 ± 4.3), followed closely by those with yellow pigment P1 (7.6 ± 3.0). Therefore, marked differences appeared in Comparison 1, given that colour differences of around 1.0 CIELAB units were perceptible to observers with normal colour vision (Melgosa et al. 1992; Huang et al. 2012). In this regard, we can add that cinnabar red pigment, being by nature quite unstable, is significantly changed by humidity and sometimes light, transforming into black meta-cinnabar due to an alteration not of its chemical formula but rather of the crystalline pigment structure (Mora, Mora, and Philippot 2001). This phenomenon was recognized as early as ancient Rome, Vitruvius recommending this pigment for use only in locations not exposed to sunlight, and that it be protected with wax. We also see this transformation, among other examples, in the stone inscriptions at the *Hypogée des Dunes* in Poitiers (France), a building dated to the Early Middle Ages (Coupry and Palazzo-Bertholon 2011). In Comparison 2, the greatest average $\Delta E^*_{ab,10}$ value in Figure 5 was registered in the samples with blue pigment P3 (9.0 ± 4.4), followed closely by the value for the red pigment samples P4 (8.4 ± 5.4) while the lowest values presented in the yellow pigment samples P1 (4.5 ± 1.2), similar to those of the green samples P2 (4.8 ± 3.0). In short, the results in Figure. 5 indicate that, in both comparisons, the samples with red (P4) and blue (P3) pigments underwent the greatest shifts, while the samples with green (P2) and yellow (P1) pigments were those that underwent the least changes, although these latter ones are also easily perceptible by the human eye.

Figure 6, concerning the effects of the two binders used, indicates that the average $\Delta E^*_{ab,10}$ values in the Comparison 1 are very similar in both binders, animal glue B1 (10.9 ± 5.8) and gum arabic B2 (10.5 ± 5.3). Therefore, as also noted in our observations on MCDM (Table 3), no difference was identified between $\Delta E^*_{ab,10}$ values for the two binders used. In any case, there was more delamination and more losses in the specimens with animal glue (B1) than in those with gum arabic (B2), as the binder B1 creates a more brittle layer that tends to crackle and delaminate. In Comparison 2, the average $\Delta E^*_{ab,10}$ values in Figure 6 are again very similar for the samples with animal glue B1 (6.4 ± 4.5) and gum arabic B2 (6.9 ± 4.1) binders. Thus, we conclude that the binder exerts no significant effect in terms of colour variation.

Figure 7 focuses solely on the influence of the five consolidants used. Regarding consolidants, in Comparison 1 the greatest colour difference was found in the samples using the acrylic copolymer consolidant C3 (13.8 ± 4.5), followed closely by the samples with polyvinyl butyral consolidant C2 (13.6 ± 4.9). The lowest value was found in the samples with bacterial carbonatogenesis consolidant C5 (5.0 ± 2.7), followed by the samples with barium hydroxide C1 (10.4 ± 6.3). However, in Comparison 2 the greatest average CIELAB colour difference resulted in the samples using bacterial carbonatogenesis consolidant C5 (8.5 ± 6.3), followed by those with acrylic copolymer consolidant C3 (8.0 ± 3.2), while the lowest value was registered in the samples with barium hydroxide consolidant C1 (3.4 ± 2.6). We note that consolidant C5 presented the greatest colour difference in Comparison 2, and the lowest in Comparison 1.

However, the consolidant with the lowest colour-difference values (in our case, C5 in Comparison 1 and C1 in Comparison 2) cannot necessarily be considered preferable, as this depends also on other factors. Hence, it is demonstrated that, for paints using a water-soluble binder, such as animal glue and gum arabic, the consolidants C1 and C5 may cause worse deterioration, not only when being applied (as they dissolve the paint) but also after the ageing test (as they fail to prevent delamination and flaking in the polychrome). In the layers of paint with pigments P1 and P2 with both binders, consolidant C1 caused deterioration (delamination and losses) even prior to ageing (Figure 4, right), these effects being exacerbated by outdoor exposure. Also in the samples with pigments P1, P2, and P3, using both binders and the ethyl silicate consolidant C4, crackles and delamination occurred, mainly after ageing. This may have been caused by the action of this consolidant, which forms an extremely rigid post-drying film over the surface of the paint layer, and this layer becomes brittle (Jroundi et al. 2014; Calero-Castillo et al., 2016).

For CIELAB colour differences in Comparison 1, ANOVA tests with two factors (consolidants and binders) revealed statistically significant differences only for the group of consolidants ($p = .008 < .05$), whereas no statistically significant differences were detected among the two binders ($p = .834$). More specifically, the bacterial carbonatogenesis consolidant (C5) presented significant differences compared to polyvinyl butyral (C2) and acrylic copolymer (C3). Results from Comparison 2 showed that some variances significantly differed, and the ANOVA results should be interpreted with caution. From CIELAB colour differences in Comparison 2 we found no statistically significant differences either in the consolidant group ($p = .094 > .05$) or in the binder group ($p = .674$). The non-significance of the consolidants may be due to the different variability in the factor levels.

Conclusion

After previous analyses of 155 original microsamples extracted from the plasterwork of the Courtyard of the Maidens of the *Real Alcázar* of Seville, Spain (Calero-Castillo et al., 2016), we prepared a set of 56 test samples using four pigments (yellow, green, blue, and red), two binders (animal glue and gum arabic), and five consolidants (barium hydroxide, polyvinyl butyral, acrylic copolymer, ethyl silicate, and bacterial carbonatogenesis), which were subjected to natural ageing for one year both indoors and outdoors. Colour variability of individual test samples is reported, as well as colour differences caused by ageing, for different pigments, binders, and consolidants. The fact that some pigments (blue and red) undergo greater colour change, while others (green and yellow) change less may be related to their chemical and/or mineralogical characteristics. The binders used did not appear to influence the colorimetric results. The chromatic variations in a paint caused by a consolidant constitute an important factor, but not the only one, to take into account in evaluations. Equally important are the performance and efficacy of the consolidant. In the present trial, although consolidants C5 (in Comparison 1) and C1 (in Comparison 2) registered the lowest colour-difference values, they would not be ideal for water-soluble paints such as those tested (with animal glue and gum arabic binders), because such consolidants require significant amounts of water for application, which can partially dissolve the paint. Considering all aspects in terms of colour differences and effectiveness, for the type of paints tested, we conclude that polyvinyl butyral (C2) is the most suitable choice, followed by the ethyl silicate (C4), as these products do not alter the colour excessively, do not damage the paint during application, and act as adequate consolidants. The findings of the present work provide insights into the performance of pictorial coatings, from a colorimetric perspective, in response to the action of the consolidants studied, under natural ageing conditions.

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