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Color stability of non-pigmented and pigmented maxillofacial silicone elastomer exposed to three different environments

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Abstract

Statement of problem. The color degradation of maxillofacial prostheses in clinical service requires their frequent renewal.

Purpose. The purpose of this study was to investigate the color stability of a non-pigmented and pigmented maxillofacial silicone when stored in darkness and exposed to accelerated aging in a weathering chamber and natural outdoor weathering.

Material and methods. M511 silicone elastomer was colored with Spectromatch Pro colorants and stored in darkness, exposed to accelerated aging and natural outdoor weathering for 1500 hours. Test groups included non-pigmented samples (n=18), individually pigmented samples (n=90) and Caucasian Skin tone colored samples (n=18). L*, a* and b* values of test specimens were measured utilizing a spectrophotometer (Konica Minolta CM-2600d, Konica Minolta Sensing, Japan) at base line (0 hours) and then every 100 hours until 1500 hours of aging were completed. Color changes (ΔE) were calculated based on the recorded L*, a* and b* values of specimens. All data was analyzed using linear mixed models and Šídák's multiple comparison of means test (α=.05).

Results. For all samples and environments, there was a significant effect of time on ΔE of all test specimens ($p = 0.001$). All pigmented M511 specimens demonstrated good color stability with maximum ΔE below the Acceptability Threshold (AT) of 2 ΔE when stored in darkness and exposed to outdoor weathering. However, non-pigmented specimens clearly crossed this AT when exposed to outdoor weathering with maximum ΔE values of 3.65. Greatest color changes were observed for all specimens when exposed to accelerated aging and most exceeded the AT of 2 ΔE. Non-pigmented and Indian Yellow demonstrated highest color changes after 1500 hours with ΔE values of 4.86 and 5.20, respectively.

Conclusions. All environments resulted in visible color changes of non-pigmented and pigmented M511 elastomer. Least ΔE values were observed for samples stored in darkness and greatest for specimens exposed to accelerated aging. The organic pigment Logwood Maroon demonstrated best color stability with maximum ΔE values below the Perceptibility Threshold (PT) of 1 ΔE.

Clinical Implications. M511 maxillofacial silicone colored with Spectromatch Pro colorants demonstrated good overall color stability when stored in darkness and exposed to accelerated aging and outdoor weathering. Both materials are recommended to be used for the fabrication of maxillofacial prostheses.

Maxillofacial appliances provide an alternative treatment option for patients with surgically non-restorable facial defects.[1,2] It is most important that facial prostheses are not immediately recognized as such from a certain distance by an observer; and this is a direct indication for the success of maxillofacial prosthetic treatment.[3,4] Unfortunately, one major drawback of facial prostheses in clinical service is their color degradation over time which requires their frequent renewal and often within less than one year after provision.[5,6] This color instability has been described as a complex phenomenon and has been related to several environmental factors including ultraviolet (UV) light, humidity, air pollutants, personal habits of patients (cleaning routine, smoking), the color instability of elastomers and colorants as well as the loss of extrinsic coloring.[7-10]

The traditional materials used in the fabrication of facial prostheses are silicone elastomers as well as organic and inorganic pigments and currently, many products are commercially available to the professional. Professionals as service providers of maxillofacial prostheses do not have any influence on environmental factors and those related to patients' habits; however, they choose the base elastomer and colorant system when fabricating facial prostheses.

The color stability of maxillofacial silicones and pigments has been investigated over the last few decades. Studies involved assessment of non-pigmented elastomer (pure elastomer without incorporated pigments) in comparison to pigmented elastomer and concluded that the inherent color instability of non-pigmented elastomer contributed to the observed overall color changes of maxillofacial prostheses.[10-14] Several studies involved methods of accelerated aging in a weathering chamber[14-18] and natural outdoor weathering[9-11,19,20] of non-pigmented and pigmented silicone elastomer where artificial aging was performed in order to simulate environmental factors maxillofacial silicones are exposed to with the aim to get an indication on the color stability of these materials in clinical service. The applied research methodology varied in the reviewed literature and a direct comparison of one paper with another was therefore impossible. However, based on the results of the investigations, it was shown that accelerated aging generally caused higher color changes than natural outdoor weathering in both, non-pigmented and pigmented maxillofacial elastomer.

A few studies were conducted to see whether the exclusion of UV light, as an adverse environmental factor, would result in lower observed color changes of maxillofacial silicones and was generally confirmed based on provided results.[9,12,13,19] However, one study [21] reported extreme color changes of more than 20 ΔE for M511 and M522 maxillofacial silicone (Principality Medical Ltd., Newport, UK) following darkness storage for one year which was in large contrast with the reviewed literature.[9,12,13,19] However, a direct comparison was impossible due to varying research methodology. Furthermore, the adverse effect of sebaceous oil secretions and skin perspirations on the color stability of maxillofacial elastomer has also been shown in an in-vitro study.[19]

The color stability of inorganic and organic pigments and their application when coloring elastomer has also been investigated and it was concluded that inorganic pigments are generally more color stable than organic pigments as the latter are more subject to decay on aging and exposure to adverse environmental conditions.[22] Few studies have been conducted on the use of a variety of dry pigments, opacifiers, artist's oil paints, liquid cosmetics and silicone pigments and the results showed that their application had varying effects on different elastomers.[15-17,23] It was generally concluded that oil based pigments combined with opacifiers at varying concentrations, and certain silicone pigments protected maxillofacial elastomer. However, the use of Yellow silicone pigment significantly affected the color stability of A-2186 and A-2000 (Factor II Ltd., Lakeside, Ariz.) with maximum color changes of more than 10 ΔE.[17,23]

It has been shown that color changes of maxillofacial elastomer are apparent and these have been assessed applying instrumental color measurement including spectrophotometers and colorimeters. For calculation of color differences, the CIE L*a*b* 1976 equation [24] has frequently been applied. [8-21] However, what color changes of facial prostheses are visually perceptible and clinically acceptable is still unclear. Based on the literature, perceptible thresholds (PT) and acceptable thresholds (AT) of color changes of maxillofacial elastomer have been investigated and as a result, different values have been stated.[25,26] For fair skin, a PT of 0.8 ΔE and 1.1 ΔE has been reported, whereas the AT ranged from 1.8 ΔE for fair skin to 4.4 ΔE for dark skin tones. In this current study, a PT of 1 ΔE and AT of 2 ΔE has been considered as it was frequently used in investigations on color stability of maxillofacial elastomers and is based on accredited literature.[27,28]

Review of the literature showed that numerous investigations have been conducted on the color stability of maxillofacial silicones. However, the investigated materials had been chosen based on introduction of new elastomers and colorants available to professionals within the field of maxillofacial prosthetic rehabilitation. There was no elastomer/colorant combination that was specifically designed to be used together in order to minimize material interactions that could adversely affect the color stability of maxillofacial elastomer.

The aim of this study was to investigate the color stability of M511 maxillofacial elastomer (Technovent Ltd., Bridgend, South Wales, UK) colored with pigments dispersed in the same base elastomer, namely Spectromatch Pro (Spectromatch Ltd., Bath, UK) and involved storage in darkness, accelerated aging and outdoor weathering. The null hypothesis was that the color stability of non-pigmented and pigmented M511 silicone elastomer is not adversely affected when exposed to the above environments.

MATERIAL AND METHODS

Five different pigment pastes (Spectromatch Ltd., Bath, UK) were investigated in this study: Indian Yellow, Alizarin Crimson, Logwood Maroon, Malachite Green (all organic pigments) and MeSi Green (inorganic pigment) and were used to color M511 maxillofacial elastomer (Technovent Ltd., Bridgend, South Wales, UK). Seven test groups were designed and included non-pigmented M511 silicone specimens (pure elastomer without incorporated pigments), specimens colored with the pigments listed above and a Caucasian Skin tone, which was established by mixing individual Spectromatch Pro pigments. Six specimens were produced per test group and environment.

For fabrication of test samples, the base polymer and cross-linker of M511 were mixed according to the manufacturer instructions with a ratio of 10:1. For manufacture of colored specimens, 2% by weight of each individual colorant was added to the elastomer. For fabrication of skin colored specimens, a color recipe for a typical Caucasian skin tone was used from the anonymous skin color data bank (Spectromatch Ltd., Bath, UK) and the pigment loads weighed and added to the elastomer accordingly. For all test specimens, elastomer and pigments were weighed on a high precision scale (GR-120, AND Instruments, UK) and subsequently centrifugally mixed three times for 30 seconds at 1800 rpm using the Speed Mixer DAC 150 FVZ-K (Hauschild Engineeering, Hamm, Germany). A two piece aluminum mold, containing an inner polytetrafluorethylene (PTFE) layer, was used for processing of all test specimens. The elastomer was cured at 85 °C for 1.5 hours; all fabricated test specimens measured 40 mm length, 20 mm width and 8 mm thickness.

Samples were stored in darkness and exposed to accelerated aging and natural outdoor weathering for the duration of 1500 hours. For darkness storage, samples were kept inside a filing cabinet where room temperature was maintained at 22 ± 2 ˚C. Accelerated aging was performed using The Q-Sun/1000 XENON Test Chamber (Q-Panel Lab Products, Cleveland, OH) where all specimens were exposed to 1.10 Wm^{-2} xenon light source, with the temperature of the chamber maintained at 40 °C and was equivalent to a black panel temperature of 63˚C, and a relative humidity of 38 ± 2 %. Natural outdoor weathering was conducted on the roof of a five stories building on the Guy's Campus, King's College London, and in accordance to ASTM G24-94 utilizing a glass covered wooden cabinet. [29] Average monthly outdoor weathering conditions are presented in Table 1.

Prior to color measurement, test specimens were cleaned with distilled water and a detergent (Procter & Gamble, Weybridge, UK), to remove surface grime, wiped dry and then conditioned at a room temperature of 22 ± 2˚C for 30 min. A spectrophotometer (Konica Minolta CM-2600d, Konica Minolta Sensing, Japan) was utilized to measure the color of all test specimens at base line (0 hours) and then every 100 hours until a total of 1500 hours was completed. Prior to measuring at any time period, the spectrophotometer was calibrated according to the manufacturer instructions by using the supplied calibration standard. The instrument settings for this study involved a D65 standard illuminant, a viewing geometry of 8° and a 10° standard observer. A measuring head aperture of 8 mm was used, and a xenon flash light diffusely illuminated the samples to be assessed.

A custom designed sample holder ensured the light reflectance readings to be taken at the same location. All test specimens were measured three times over a white (W) and three times over a black standard background (B) which represents an accredited methodology in color stability testing and has been applied by various authors.[30-32] The color change values (ΔE) for all test specimens were calculated using the equation below.[24]

$$\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{0.5}$$

Statistical analysis was performed with Stata 14.2. (Stata Corp. 2016, Stata Statistical Software: Release 14, College Station, Tx.). Statistical significance was predetermined at $\alpha = 0.05$ for all hypothesis tests. As each specimen was measured three times and at 15 time periods the results are repeated measures and the data is correlated; to allow for this, the effect of time, pigment, background and environment were analyzed using linear mixed models (LMM), Stata program '`mixed`'. Where appropriate, Šídák's multiple comparison of means test was used in the comparison of groups. For conciseness, the results of the LMM statistical analysis are reported as the probabilities, rounded to 3 decimal places, related to the main factors and their interactions where appropriate.[33,34]

RESULTS

An initial analysis involving all independent variables showed all main factors (time, pigment, environment and background) to be statistically significant together with several of the interactions (Table 2). This makes a meaningful interpretation of the data difficult and consequently the data was split by background; a pairwise comparison of the environments for each background is summarized in Table 3. The comparison of pigments within each environment # background combination is provided in Table 4 and a univariate data summary of ΔE after 1500 hours of weathering for all test groups and environments in Table 5. All color change data is evaluated and shown in relation to the PT of 1 and AT of 2 ΔE.

Least color changes were observed for test specimens stored in darkness (Figure 1) and most for samples exposed to accelerated aging (Figure 2) and all other color change results were intermediate. Figures 1 and 2 illustrate the mean ΔE for all test specimens and associated 95 % confidence intervals for the above environments. Logwood Maroon was the most color stable pigment and did not cross the PT of 1 ΔE for the entire testing period. Highest color changes were obtained for Indian Yellow with maximum ΔE of 5.2 (W). However, the same pigment demonstrated good color stability when exposed to outdoor weathering with highest ΔE of 0.78 (B). There was no statistically significant difference between darkness and outdoor weathering for Indian Yellow (B, W). For MeSi Green colored specimens, all environment # background combinations demonstrated no statistically significant difference.

DISCUSSION

It is a known fact that maxillofacial prostheses exhibit two major clinical limitations which include their gradual discoloration in clinical service and degradation of physical and mechanical properties.[7,11,18,19] The color stability of non-pigmented and pigmented M511 silicone elastomer when stored in darkness and exposed to accelerated aging and natural outdoor weathering was investigated in this study. The results showed that specimens of all test groups underwent varying amounts of color changes regardless of the environment. Accordingly, the null hypothesis was rejected and these findings were in agreement with other studies.[9,10,13,18-21]

Throughout this study, the generally accepted and frequently applied PT of 1 and AT of 2 ΔE was used when evaluating the color changes of both, non-pigmented and pigmented elastomer. For all test specimens and environments, there was a significant effect of time on the ΔE ($p = 0.001$) of all test samples for both, black (B) and white (W) backgrounds. From the three investigated environments, specimens stored in darkness caused smallest color changes (max. 1.31 ΔE), whereas accelerated aging resulted in highest color changes (max. 5.20 ΔE) of specimens and was in agreement with other studies.[12,18,19]

Non-pigmented elastomer stored in darkness exhibited just visually perceptible color changes of 1.16 ΔE (W) but was higher than most of the pigmented test specimens. Within the group of pigmented specimens stored in darkness, only Alizarin Crimson and Indian Yellow crossed the PT with maximum color changes of 1.24 and 1.31 ΔE, respectively. It has been suggested that the observed color changes for elastomer stored in darkness is inherent in the elastomers as the known effect of UV radiation was excluded. It has further been indicated that these color changes may result from additional cross-linking caused by continued chemical polymerization of the silicone or by side reactions among impurities present within the silicone. Platinum compounds used as catalysts in addition curing silicones are especially known for their sensitivity to impurities.[9,12,19] A PTFE mold was used in this current study to prevent any influence caused by impurities and may have been a reason for the observed smaller color changes.

Within the test group of accelerated aging, non-pigmented specimens demonstrated maximum color changes of 2.99 ΔE (B) and 4.86 ΔE (W) after 1500 hours. It has been suggested that an increase in cross-linking within the elastomer network may be a response to UV light exposure and results in modifications within the polymer structure. These structural changes may in turn influence the transmission and scattering of light within the elastomer and thereby contribute to the observed color changes of maxillofacial elastomer.[12-14,19] For pigmented specimens, the organic pigment Logwood Maroon was the most color stable colorant and demonstrated minimal color changes below the PT of 1 ΔE throughout the entire testing period. The only investigated inorganic pigment, Me Si Green, exhibited good color stability with maximum color changes just crossing the PT, with a ΔE of 1.21 (W) and 1.16 (B). All remaining tested pigments as well as the combination of pigments in a Caucasian Skin tone demonstrated clearly visible color changes with crossing the AT of 2 ΔE. Indian Yellow was the least color stable colorant with maximum ΔE values of 5.2 (W) and 4.88 (B) at the end of the testing period of 1500 hours. These results support the known fact that inorganic pigments are generally more color stable than organic pigments.[22]

It has been stated that organic pigments have a limited life span and are more subject to decay on aging and exposure to adverse environmental conditions.[11,19,22] With this knowledge in mind, the professional should consider the application of more color stable pigments in order to minimize color changes of facial appliances. However, it was shown in the literature that a series of pigments demonstrated good color stability in the presence of UV radiation but showed significant color changes when mixed into the silicone base elastomer. It has been postulated that either a chemical interaction or a chemical incompatibility between pigments and elastomer were responsible for the observed color changes but this will require further research.[13]

For natural outdoor weathering, non-pigmented test specimens demonstrated color changes ranging from 2.42 ΔE (B) to 3.65 ΔE (W) after1500 hours of weathering and were significantly smaller when compared with the measurements recorded for artificial aging with 2.99 ΔE (B) and 4.86 ΔE (W). Based on the results of this part of the study it can be stated that for all non-pigmented and pigmented test specimens, apart from MeSi Green, there was a statistically significant difference of color changes when comparing outdoor weathering with accelerated aging in a weathering chamber ($p=0.001$). Highest color changes were observed for Alizarin Crimson outdoor weathered specimens with 1.48 ΔE (W) and Caucasian Skin tone with 1.23 (B) and lowest values were calculated for MeSi Green with 0.38 ΔE (B) and 0.39 ΔE (W). Interestingly, specimens colored with Indian Yellow proofed being very color stable with color change values below the PT but crossing this threshold only once at 1100 hours with 1.27 ΔE (W).

Investigations on outdoor weathering including this current study involved the following locations: Florida, Arizona[11], Indiana [9] (all USA), Athens (Greece)[10], Dammam (Saudi Arabia)[20], Manchester[21] and London (both UK). Outdoor weathering was performed for three months[11], six months[9,19,20] and one year.[10] Color changes ranged from around 1 ΔE[11], 2.5 to 3.5 ΔE[10] to 3.89 ΔE[19] for non-pigmented elastomers. For pigmented specimens, color changes from around 2 ΔE using dry pigments[9], 6.68 ΔE[20] compared to 8.30 ΔE[19] using both a pre-blended rose-pink skin shade (P409; Principality Medical) and 9.33 ΔE[11] when applying dry pigments were obtained. It was impossible to compare one study with another as different materials and research methodology had been applied. However, it was shown that local weather condition has an influence on the observed color changes of maxillofacial elastomer. Interestingly, the observed color changes following outdoor weathering performed in the British climate[19] were much higher than those recorded in a hot and humid climate.[20] Same elastomer and colorant as well similar methodology were applied in both studies and this suggests that humidity and rainfall seem to have a greater effect on colored elastomer than does heat and sun. However, this requires further investigations in order to draw substantial conclusions. The color changes observed in this current study were significantly lower than those measured in the same climate [19] but similar to the results stated for a much warmer climate [20] which may be related to the different elastomers and colorants used and their ability to better withstand color changes during outdoor weathering.

It may be argued that outdoor weathering of silicones represents more the natural environment of facial prostheses in service and that the color changes observed following outdoor aging reflect the expected color changes of facial appliances in a service environment. However, the greater changes in color following accelerated aging are more similar and closer to those observed by professionals on clinic. Overall, based on the results of this current study it can be stated that M511 elastomer used with Spectromatch Pro pigments demonstrated better color stability than other elastomer and colorant combinations when investigated in a similar climate.[9,19]

The application of Spectromatch pigments in this current study resulted in varying degrees of color changes and ranged from as little as 0.19 ΔE for Caucasian Skin stored in darkness to as high as 5.20 for Indian Yellow exposed to accelerated aging. Spectromatch Pro pigments can be grouped with silicone pigments as they are dispersed in base elastomer. Few investigations were conducted on color stability of dry pigments, oil based pigments and silicone pigments.[15-17,23] It was shown that application of certain percentage levels of opacifiers in combination with certain pigments resulted in lower color changes of elastomer when exposed to accelerated aging. The use of red dry pigment in combination with 15 % Georgia kaolin resulted in maximum ΔE of 49.57, whereas the same pigment combined with 15 % titanium white dry pigment as opacifier only demonstrated maximum ΔE of 16.59. The application of silicone pigments achieved significantly less color changes and maximum ΔE were reported for the yellow silicone pigment with 10.3 ΔE combined with 5 % silicone white pigment as opacifier[17] and 8.4 ΔE combined with 2.5 % Nano-Cerium Dioxide (CeO_2). These observed color changes are higher than those observed for Spectromatch Pro silicone pigments in this current study. However, a direct comparison of the above investigations with the current study is impossible due to varying materials and research methodology.

The adverse effect of sunlight on the color stability of pigments is a generally known fact.[4,7,8] However, another speculation on the observed color changes of maxillofacial silicone involves the movement of pigments within the elastomer. It has long been suggested that pigments may change their location within the elastomer over time as they are not chemically bonded to the elastomer. Hence, the color changes observed in a service environment may be as well related to relocation of pigments, their movement towards the surface of the silicone elastomer and eventual loss. However, currently this is a speculation and there is no scientific research available on this subject.

Based on the results of this current study, M511 maxillofacial silicone elastomer colored with Spectromatch Pro pigments demonstrated good overall color stability when stored in darkness and exposed to UV light utilizing accelerated aging in a weathering chamber and natural outdoor weathering. However, these statements are based on investigation of one single environmental factor and may be considered a limitation. In order to better predict color changes of facial prostheses in a service environment, other factors such as humidity, air pollution, body secretions, extrinsic coloring, personal habits of patients and naturally occurring color changes of skin need to be evaluated. Ideally, research on the color stability of maxillofacial elastomers should be carried out as in-vivo studies. Investigation of maxillofacial non-pigmented and pigmented elastomer in direct contact with human skin over a longer period of time will involve all environmental factors at the same time and only then the real effect of those can be assessed and a conclusion drawn on the prediction of color stability of maxillofacial silicones in service. However, this research approach would be most desirable but also most difficult to conduct.

CONCLUSIONS

Within the limitations of this study, storage in darkness resulted in smallest visible and exposure to accelerated aging in a weathering chamber in highest visible color changes of non-pigmented and pigmented M511 maxillofacial silicone. The material combination of M511 silicone base elastomer and Spectromatch Pro colorants demonstrated good overall color stability in this in-vitro study.

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TABLES

Table 1 Monthly average climate data and radiation during outdoor weathering

Month	Temperature (˚C)			Humidity	Global Radiation	Sunshine
	Max	Min	Average	(%)	(kJm^{-2})	(hours)
October 2011	18.1	10.1	14.1	76.0	7672	4.5
November 2011	13.6	7.3	10.4	87.2	3126	1.8
December 2011	9.9	3.8	6.8	81.2	2262	2.0
January 2012	9.8	3.4	6.6	80.4	2807	2.2
February 2012	8.0	1.3	4.6	76.7	5473	2.9
March 2012	14.7	4.7	9.7	73.6	10971	5.8
April 2012	13.3	4.9	9.1	74.5	12973	4.7
May 2012	18.2	9.7	13.9	72.2	16185	5.4
June 2012	19.4	11.6	15.5	74.5	15581	4.0

Table 2 Summary of the mixed model analysis for the dependence of ΔE on all the independent variables

Factor	p
time	0.001
pigment	0.001
environment	0.048
pigment # environment	0.001
background	0.021
pigment # background	0.065
environment # background	0.490
pigment # environment # background	0.962

Table 3 Probabilities, p(), for the comparison of environments for each pigmen t# background combination

Pigment	Background					
	Black			White		
	p(D-O)	p(D-A)	p(O-A)	p(D-O)	p(D_A)	p(O-A)
Caucasian Skin	0.001	0.001	0.001	0.001	0.001	0.001
Logwood Maroon	0.005	0.001	0.001	0.006	0.001	0.001
Alizarin Crimson	0.559	0.001	0.001	0.412	0.001	0.001
Indian Yellow	0.986	0.001	0.001	0.615	0.001	0.001
MeSi Green	0.469	0.256	0.063	0.300	0.460	0.076
Malachite Green	0.241	0.001	0.001	0.006	0.001	0.001
Non-pigmented	0.001	0.001	0.001	0.001	0.001	0.001

Legend: p(D-O) Darkness-Outdoor, p(D-A) Darkness-Accelerated, p(O-A) Outdoor-Accelerated

Table 4 Comparison of pigments for each environment # background combination over the entire exposure period using Šídák's multiple comparison test. Pigments sharing the same letter are not statistically significantly different.

		Background	
Environment	Pigment	Black	White
Darkness	Caucasian Skin	A	A
	Logwood Maroon	B	A
	Alizarin Crimson	A	DE
	Indian Yellow	A	CD
	MeSi Green	C	B
	Malachite Green	AC	BC
	Non-pigmented	A	E
Outdoor Weathering	Caucasian Skin	A	CD
	Logwood Maroon	C	BC
	Alizarin Crimson	B	A
	Indian Yellow	AB	AD
	MeSi Green	C	B
	Malachite Green	AB	A
	Non-pigmented		
Accelerated Aging	Caucasian Skin	AD	C
	Logwood Maroon	B	A
	Alizarin Crimson	CD	BC
	Indian Yellow	A	
	MeSi Green	B	A
	Malachite Green	C	B
	Non-pigmented	A	

Table 5 Mean ΔE and standard deviation at each environment # background # pigment combination at 1500 hours, and the probability, p, that ΔE is the same at each time period.

Environment	Pigment	B 1500 h $\overline{\Delta E}$, sd	p	W 1500 h $\overline{\Delta E}$, sd	p
Darkness	Caucasian Skin	0.19, 0.04	0.001	0.19, 0.05	0.001
	Logwood Maroon	0.14, 0.04	0.224	0.15, 0.04	0.390
	Alizarin Crimson	1.01, 0.66	0.001	1.24, 0.19	0.001
	Indian Yellow	1.26, 0.34	0.001	1.31, 0.35	0.001
	MeSi Green	0.66, 0.08	0.002	0.70, 0.09	0.001
	Malachite Green	0.51, 0.14	0.914	0.56, 0.17	0.404
	Non-pigmented	0.76, 0.06	0.529	1.16, 0.34	0.014
Outdoor Weathering	Caucasian Skin	1.23, 0.17	0.001	1.26, 0.15	0.001
	Logwood Maroon	0.66, 0.04	0.001	0.66, 0.05	0.001
	Alizarin Crimson	1.22, 0.08	0.001	1.48, 0.09	0.001
	Indian Yellow	0.78, 0.15	0.021	0.51, 0.09	0.037
	MeSi Green	0.38, 0.06	0.349	0.39, 0.06	0.296
	Malachite Green	0.85, 0.10	0.001	1.04, 0.09	0.001
	Non-pigmented	2.42, 0.12	0.001	3.65, 0.19	0.001
Accelerated Aging	Caucasian Skin	3.25, 0.67	0.001	3.26, 0.67	0.001
	Logwood Maroon	0.72, 0.16	0.053	0.74, 0.17	0.079
	Alizarin Crimson	2.38, 0.26	0.001	2.50, 0.25	0.001
	Indian Yellow	4.88, 0.49	0.001	5.20, 0.35	0.001
	MeSi Green	1.16, 0.26	0.001	1.21, 0.27	0.001
	Malachite Green	2.35, 0.30	0.001	2.57, 0.25	0.001
	Non-pigmented	2.99, 0.39	0.001	4.86, 0.13	0.001

FIGURES

Figure 1 Mean ΔE values and associated 95 % confidence interval at each time period

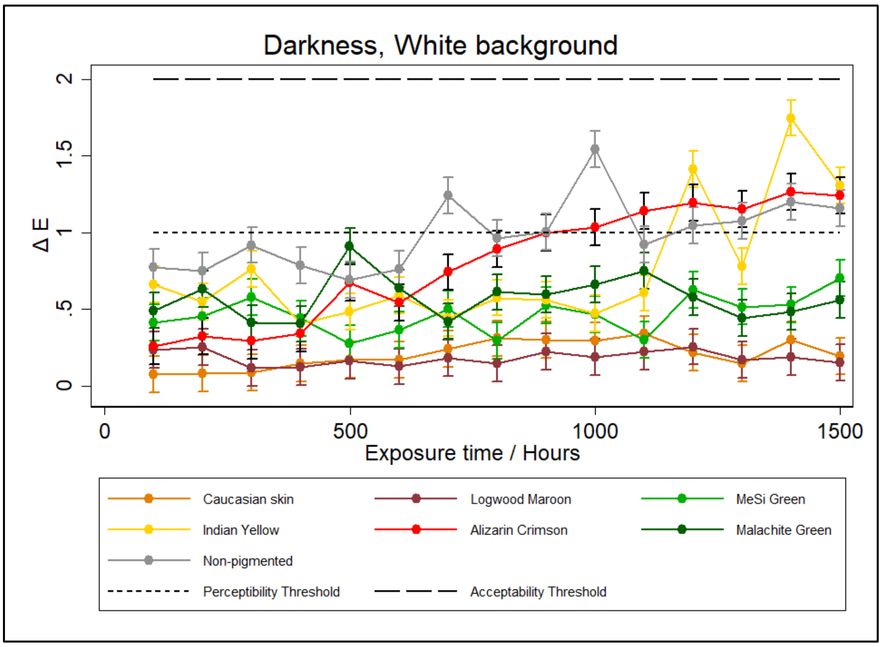

Figure 2 Mean ΔE values and associated 95 % confidence interval at each time period