

Effect of newly developed pigments and ultraviolet absorbers on the color change of pigmented silicone elastomer

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Abstract

Aim and Objective: The aim and objective of the study is to evaluate effect of ultraviolet (UV) stabilizer (UV absorber Chimassorb 81) on color change of pigmented silicone elastomer when commercially available (red and yellow), and newly developed pigments (sicotrans red and sicopal brown) were used.

Materials and Methods: Two commercially available pigments – red (P112 Brilliant Red) and yellow (P106 Yellow) and two newly developed pigments – sicotrans red and sicopal brown were studied. In total eight groups made up of nine samples each were fabricated using elastomer with the combinations of the above pigments and UV stabilizer (Chimassorb 81). Groups 1, 3, 5, and 7 contain elastomer in combination with sicotrans red, sicopal brown, yellow and red pigments, respectively. Similarly, groups 2, 4, 6, and 8 along with elastomer and pigments (sicotrans red, sicopal brown, yellow and red, respectively) contain the UV stabilizer (Chimassorb 81). Samples were subjected to aging in an accelerated weathering chamber (Weather-Ometer). Color values CIE (Commission Internationale d'Eclairage) L*, a*, and b* were measured at baseline and after 1000 h of weathering. Change in color (Delta E) was calculated.

Results: All groups showed a significant color change at 1000 h. Groups 1, 2, 3, and 4 showed a statistically significant less change in both colors (sico trans red and sicopal brown) compared to groups 5,6,7, and 8 (commercial pigments-Red and Yellow). Overall, the change in the color in groups with the UV stabilizer (Chimassorb) was less when compared to the groups where the stabilizer was not used.

Conclusion: The newly developed pigment led to increased color stability as compared to commercially available pigments. Addition of UV stabilizer, Chimassorb led to a further reduction in color change of the pigmented elastomer.

Keywords: Color stability, elastomers, ultraviolet stabilizers

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INTRODUCTION

Extraoral defects that are present from birth or as a result of trauma, burns, and surgical resections can be successfully rehabilitated using silicone elastomers.^[1] A major shortcoming of these pigmented elastomers is their discoloration over a period leading to the prostheses becoming unesthetic, thereby requiring a remake of the prostheses every 6 months– 2 years. Unfortunately, this leads to an increased cost of rehabilitation.^[2-8]

A common problem affecting prosthesis longevity is color instability.^[5-7] The deterioration of color of prostheses is a result of multiple factors including natural climatic conditions, environmental conditions such as humidity, air pollutants, ultraviolet (UV) radiation, rain, and human body secretions. In addition, the inherent color instability of silicone elastomers in their nonpigmented state adds to the overall color instability and discoloration of prostheses.^[9-15]

Research strategies have focused toward improving color stability and mechanical properties of pigmented elastomers. Various authors in the past have attempted the addition of UV stabilizers, thermochromic pigments, and opacifiers to improve the color stability of the pigmented elastomers, but the outcome of these studies have been inconclusive.^[16-20]

The term UV stabilizer is a broad expression that includes ultraviolet absorbers (UVAs) and hindered amine light stabilizers (HALS), both having different modes of actions. UV stabilizers have been used routinely in the past to increase the shelf life and prevent color degradation of the products such as polymers, wood, cellulose fabrics, and paints.^[21-23]

To improve the color stability of silicone prostheses, it is of prime importance to analyze the effect of the elastomer and the pigments used in the fabrication. Gary *et al.*, Hatamleh and Watts have stated that color degradation of the prosthesis can be attributed to the continuous polymerization of the elastomer alone.^[24,25] Alterations in the chromatic pattern of the elastomer occur due to continuous release of subproducts during the polymerization of silicones. However, in a study by Kheur *et al.*, it was observed that the color change in an elastomer can be reduced to a great extent by the use of UV stabilizers.^[26]

Tran *et al.* analyzed the effect of UV stabilizers Tinuvin 213 and Tinuvin I23 on organic and inorganic pigments. They observed color change in all samples at two different

weathering locations (Miami and Phoenix), but the amount of color change was significantly lower in specific pigmented groups (i.e., burnt sienna and hansa yellow).^[18]

The authors of this current study have previously analyzed the effect of UV stabilizers on the color change of pigmented silicone elastomer and observed that the addition of UV stabilizers leads to increased color stability for red and yellow pigments.^[27] However, the authors noted that pigments play a vital role in degradation and color instability in elastomers and, therefore, there is a need for the development of newer pigments which would improve the color stability of the elastomers.

This study analyzed the effect of a UV stabilizer (UVA Chimassorb 81) on the color change of a platinum-based pigmented silicone elastomer on commercially available (red and yellow) and newly developed pigments (sicotrans red and sicopal brown).

MATERIALS AND METHODS

Two newly developed pigments – sicotrans red and sicopal brown (BASF, India) and commercially available pigments (red and yellow) were tested in this study. An Food and Drug Administration (FDA) approved UV stabilizer Chimassorb 81 (BASF, India) was used as an additive.

A platinum-based maxillofacial silicone elastomer Z004 (Technovent Pvt. Ltd., UK) mixed in a 1:1 base: Catalyst ratio was used for the study.

A total of 8 groups of 9 samples each were fabricated using the above pigments and stabilizer. Commercially available and newly developed pigments were divided into groups as seen in Table 1.

A stainless steel mould was prepared for the fabrication of samples as described in a previous study by Kheur *et al.*^[27] Pigmented silicone samples rectangular in shape, of dimensions 1.5 cm × 2 cm × 0.5 cm were fabricated.

In groups 1, 3, 5 and 7 only pigments were added, and no UV stabilizer was used.

Table 1: Distribution of groups

Group number	Pigments used
Group 1	Elastomer + sicotrans red
Group 2	Elastomer + sicotrans red + chimassorb 81
Group 3	Elastomer + sicopal brown
Group 4	Elastomer + sicopal brown + chimassorb 81
Group 5	Elastomer + yellow
Group 6	Elastomer + yellow + chimassorb 81
Group 7	Elastomer + red
Group 8	Elastomer + red + chimassorb 81

In groups 2, 4, 6, and 8 the UV stabilizer was added 1% by weight, and 0.2% by weight of the respective pigment were added to silicone elastomer in all eight groups. These were as per recommendations by Tran *et al.*, Beatty *et al.*, and Gary *et al.*^[18,24,32] A digital scale was used to weigh the stabilizer and pigments for groups 2, 4, 6, and 8 and the pigment alone for groups 1, 3, 5, and 7. The pigments and stabilizers were thoroughly spatulated together with the silicone (base + catalyst) for 5 min to obtain a homogenous mix. The silicone was vacuum mixed for 20 min under 30 inch/Hg pressure. After the addition of silicone, the molds were carefully loaded and clamped under pressure for 24 h to allow the silicone to polymerize. Samples were examined carefully for surface porosities or impurities postcuring. Acetone and cotton were used to clean the obtained samples. They were tested at 0 h to obtain the baseline Commission Internationale d'Eclairage (CIE) L*, a* and b* values.

A Weather-Ometer (Xenon Arc Ci 4000, Atlas Material Testing Technology, USA) was used for aging of samples. Alternating light and dark cycles for a total of 180 min completed one cycle. The light cycle (120 min) was performed using an irradiance of 340 nm of 0.55W/m², humidity of 50%, and a chamber temperature of 47° C with water spray for 60 min. This was followed by 60 min without water spray. Whereas, the dark cycle lasted for 60 min with a temperature of 38 degrees Celsius, humidity of 95%, and irradiance at 340 nm of 0.55W/m². Tropical climatic conditions were kept in mind while selecting these parameters.

Testing of the samples was carried out at intervals of 0 and 1000 h. Cotton and acetone were used for cleaning of samples before testing. Each sample was measured three times, and an average of the readings was considered for its analysis.

A spectrophotometer (TES-135 Color Meter, Instruments and Machinery Sales Corporation, Mumbai, India) was used to measure the color variables L*, a*, b* according to the CIE Laboratory system before and after aging.

The L* parameter denoted the degree of lightness and darkness (100 ideal white, 0 ideal black) while a* and b* coordinates correspond to red or green chroma (+a = red, -a = green) and yellow or blue chroma (+b = yellow, -b = blue), respectively. The Delta E (change in color) was calculated for each sample using a software with the formula $\Delta E = \sqrt{[\Delta L^*]^2 + [\Delta a^*]^2 + [\Delta b^*]^2}$ / 2, where Delta L*, Delta a*, and Delta b* are the difference in L*, a*, and b* values before and after aging.

For all the statistical tests, $P < 0.05$ was considered to be statistically significant, keeping α error at 5% and β error at 20%, thus giving a power to the study as 80%.

RESULTS

Average Delta E values obtained for all the Groups are seen in [Table 2] and represented in Figure 1.

A one-way ANOVA test was applied to compare the mean values of the 8 groups. A further analysis, the *post hoc* Bonferroni's test was carried out for intergroup comparison [Table 3]. The significance level was considered at 0.05.

At 1000 h, for the yellow pigment, the color change between Group 6 (yellow + chimassorb 81) and Group 5 (elastomer + yellow) was significant ($P < 0.05$) [Table 3].

Similarly, for the red pigment, the color change between Group 8 (red + chimassorb 81) and the Group 7 (elastomer + red) was highly significant ($P < 0.01$) [Table 3].

On intergroup comparison of the newly developed and commercially available pigments, the following was noted [Table 3].

At 1000 h, when sicotrans red (Group 1) and commercially available red (Group 7) were compared, the color change between sicotrans red and commercially available red pigment was highly significant ($P < 0.01$).

Table 2: Average color change (ΔE-values) after 1000 h of ageing

Groups (n=9)	ΔE 1000 h (n=9)	
	Mean	SD
Group 1 (sicotrans red)	2.29	0.94
Group 2 (sicotrans red + chimassorb)	1.51	0.60
Group 3 (sicopal brown)	1.78	0.49
Group 4 (sicopal brown + chimassorb)	1.55	0.42
Group 5 (yellow)	5.92	1.02
Group 6 (yellow + chimassorb)	4.28	1.09
Group 7 (red)	9.58	1.14
Group 8 (red + chimassorb)	5.49	0.83

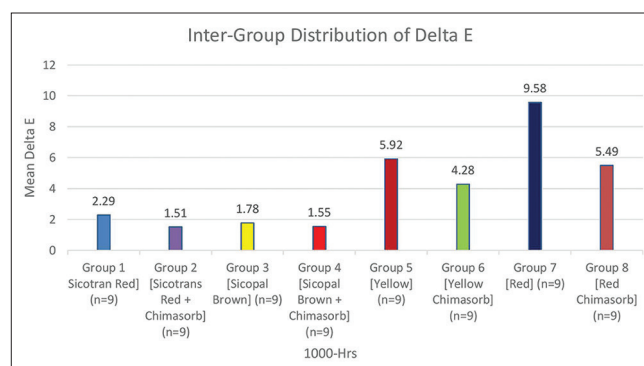


Figure 1: Color change at 1000 h

Table 3: P values by one-way analysis variance with *post hoc* Bonferroni's correction for multiple group comparisons

ΔE	Inter-group comparisons (P)						
	Group 1 versus Group 2	Group 1 versus Group 3	Group 1 versus Group 4	Group 1 versus Group 5	Group 1 versus Group 6	Group 1 versus Group 7	Group 1 versus Group 8
1000 h	0.999	0.999	0.999	0.001***	0.001***	0.001***	0.001***
ΔE	Group 2 versus Group 3	Group 2 versus Group 4	Group 2 versus Group 5	Group 2 versus Group 6	Group 2 versus Group 7	Group 2 versus Group 8	Group 3 versus Group 4
	1000 h	0.999	0.999	0.001***	0.001***	0.001***	0.999
ΔE	Group 3 versus Group 5	Group 3 versus Group 6	Group 3 versus Group 7	Group 3 versus Group 8	Group 4 versus Group 5	Group 4 versus Group 6	Group 4 versus Group 7
	1000 h	0.001***	0.001***	0.001***	0.001***	0.001***	0.001***
ΔE	Group 4 versus Group 8	Group 5 versus Group 6	Group 5 versus Group 7	Group 5 versus Group 8	Group 6 versus Group 7	Group 6 versus Group 8	Group 7 versus Group 8
	1000 h	0.001***	0.004**	0.001***	0.999	0.001***	0.106

P < 0.05 is considered to be statistically significant. *P < 0.05, **P < 0.01, ***P < 0.001

Similarly, for the yellow pigment, a highly significant difference was noted in Delta E values for sicopal brown (Group 3) and commercially available yellow pigment (Group 5) ($P < 0.01$).

From the above results, it was observed that newly developed pigment showed less color change as compared to commercially available pigments.

At 1000 h, for the sicotrans red pigment, color change (mean delta E) between Group 1 (Sicotrans red) and Group 2 (Sicotrans red + Chimassorb) did not differ significantly ($P > 0.05$).

Similarly, for the sicopal brown pigment, color change between Group 3 (sicopal brown) and Group 4 (sicopal + chimassorb) was nonsignificant ($P > 0.05$).

On intergroup comparison, it was observed that the mean Delta E at 1000-H did not differ significantly across four study groups ($P > 0.05$ for all).

DISCUSSION

Silicone elastomers have been shown to degrade due to poor color stability and deterioration of their mechanical properties during the course of their use. This deterioration of silicone is multifactorial in nature, primarily due to the exposure to UV rays.^[28] Exposure to UV rays leads to a subsequent change in mechanical and optical properties of the silicones. The change is dependent on the duration, extent, and intensity of the UV exposure.^[8-12]

To achieve color stabilization of elastomers, the use of UV stabilizers, thermochromic pigments, and opacifiers have been attempted in the past.^[9-12,16] UV stabilizers have been extensively used in polymer, paint, cosmetic, and plastic industries as well as for wood and cellulose fabrics

to enhance color stability and increase the longevity of the products.^[21-23]

A UV stabilizer generally includes UVAs and HALS, both of which have different mechanisms of actions. The prerequisites for optimal action of the stabilizers are high solubility, minimal diffusion, and high distribution homogeneity.^[21] However, attempts at stabilization of elastomers have yielded mixed results. Kiat-Amnuay *et al.* analyzed various concentrations of opacifiers and found that they prevented color degradation overtime.^[16] Kantola *et al.* observed that thermochromic pigments were sensitive to UV radiation and lead to color instability. Therefore, it was concluded that thermochromic pigments are not suitable for prosthetic application.^[17]

In this study, the effect of Chimassorb 81, a benzophenone UVA, was analyzed. In the past, varying concentrations of UV stabilizers have been documented by researchers, but the results of these studies have been inconclusive. Chu and Fisher in their study used 1.5% weight UVA and found that it was effective in reducing color degradation of the elastomer.^[19] Tran *et al.* used 0.75% by weight UVA and HALS and found that addition of UV stabilizer can lead to increased color stability for certain pigmented groups.^[18]

In this study, 1% by weight of UVA (Chimassorb 81) was used for Groups 2, 4, 6, and 8, respectively. These additives are biocompatible and are approved by the FDA. An accelerated weathering chamber was used in the study to simulate weathering conditions and parameters such as UV lighting and radiation, water spray, humidity, and temperature were included to test the effectiveness of UV stabilizer. A combined effect of these would simulate real-time conditions and can probably cause a more pronounced change compared to that produced by one parameter alone.^[24]

UV irradiation leads to enhanced cross-linking resulting in breaking down of chain bonds and decomposition of the elastomer. This in turn reduces polymerization and causes decomposition of elastomer. All these factors lead to color instability of the elastomer.^[13,30]

Kheur *et al.* analyzed the effect of the UV stabilizer on plain elastomer and concluded that color change of the elastomer itself can be attributed to inherent chemical alterations in the silicone.^[26]

Goiato *et al.* and Mancuso *et al.* noted that Chimassorb 81 compound absorbs UV rays from sunlight and dissipates this energy throughout the polymer matrix, thereby preventing degradation.^[29,30] Chimassorb 81 soaks up harmful UV rays from sunlight converting this energy into heat energy which is then dissipated. It also prevents the formation of harmful free radicals. Chimassorb 81 prevents photosensitization by absorbing heat energy. It has a stabilizing effect as its color is transparent to visible light, and this prevents alteration in the appearance of the elastomer.^[31]

Red and yellow pigments are commonly used in color formulations to obtain skin colors. However, in this study, it was observed that the red pigment degraded to a larger extent than yellow. The results of this study are in agreement with previous studies by Kiat-Amnuay *et al.* and Beatty *et al.*^[31,32] This could be due to the organic nature of the red pigment being more affected by irradiation.^[31,32] Organic colorants rely on the placement of double and triple bonds to impart color to the molecule. Since these bonds tend to be relatively reactive, these colorants are considered to be unstable.^[3] Inorganic pigments are generally more color stable but are often not considered as these pigments are not as bright as organic pigments making it difficult to achieve a good color match.^[16]

The authors analyzed two newly developed (sicotrans red and sicopal Brown) pigments in this study. On comparison of sicotrans red and the commercially available red pigment, it was observed that sicotrans red had a higher color stability as compared to the commercially available red pigment. Similarly, when sicopal brown and yellow pigment were compared, it was noted that sicopal brown underwent minimal color degradation as compared to the commercially available yellow pigment. The findings of this study suggest that sicotrans red and sicopal brown could be used as an alternative to commercially available pigments. The increased color stability observed with newly developed pigments (sicotrans red and sicopal brown) could be due to their reduced particle size. Akash *et al.* in a study

analyzed the effect of incorporation of nano-oxides on color stability of maxillofacial silicone elastomer subjected to outdoor weathering. In this study, it was observed that ZnO-incorporated Cosmesil M511 silicone specimens showed minimal or no color change and proved to be most color stable after being subjected to outdoor weathering. This was attributed to the small particle size exhibited by ZnO oxide (<50 nm). With increased particle size, there is poor dispersal of the pigments in the elastomer matrix.^[33] A higher tendency to agglomerate decreases UV shielding. This leads to an increased color change.^[33]

When UV light is incident on the nanoparticles, their electrons are strained to vibrate. Since the nanoparticle size is smaller than the UV wavelength, part of the UV light is scattered and partly absorbed by nanoparticles simultaneously. Thus, UV shielding ability of the nanoparticles is a result of absorption and scattering. Since the particle size of sicotrans red and sicopal brown (<25 nm) is extremely small, these pigments are homogeneously dispersed in the silicone matrix, they are less likely to show migration, and thereby exhibiting greater color stability.^[20]

When sicotrans red, sicopal + Chimassorb 81, Sicopal brown, and sicopal brown + Chimassorb 81 were analyzed for color stability, it was observed that the addition of UV stabilizer (Chimassorb) led to improved color stability. The delta E values for all these groups studied were <2, thereby ensuring that the color change would not be detectable to the human eye. However, no statistical significant difference was observed between these groups.

This is an *in vitro* study evaluating the color stability of pigmented elastomers using simulated accelerated artificial weathering. More studies using clinical setups done in actual climatic conditions using prosthesis pigmented with the new color stable pigments can give a better insight on the performance of these pigments.

Further research should evaluate the effect of different types of UV stabilizers, their combinations and concentrations on color stability of pigmented elastomers. Evaluation over longer periods of time, different methods of aging to simulate natural conditions is also an exciting area of potential research.

CONCLUSION

Within the limitations of this study, it can be concluded that the newly developed pigments demonstrated superior color stability after 1000 h of weathering as compared to commercially available pigments.

The addition of UV stabilizer Chimassorb 81 resulted in a consistently lesser color change in all the groups.

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Conflicts of interest

There are no conflicts of interest.

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