# Color stability and colorant effect on maxillofacial elastomers. Part III: Weathering effect on color

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**Statement of problem.** Maxillofacial prostheses are serviceable for approximately 6 months, after which they need to be refabricated because of the deterioration of color and physical properties.

**Purpose.** This third article in a 3-part series evaluated the color stability of commonly used colorant-elastomer combinations as a result of exposure to weathering.

**Methods and material.** Fifteen specimens were fabricated for each of the 3 elastomers (Silastic medical adhesive type A, Silastic 4-4210, and Silicone A-2186) and 6 colorants (dry earth pigments, rayon fiber flocking, artist's oil paints, kaolin, liquid cosmetics, and no colorant) for a total of 270 specimens (18 groups of 15 specimens). The 15 specimens of each elastomer-colorant combination were separated into 3 test condition groups (control, time passage, and natural weathering) of 5 specimens per test condition group. Control specimens were evaluated within 1 month of fabrication. The time passage group was sealed in glass containers and kept in the dark for 6 months before testing. The natural weathering groups were placed on the roof of the dental school for 6 months and exposed to sunlight and weathering. Color and optical density data for each elastomer-colorant combination were subjected to a 1-way analysis of variance to examine effects among test conditions (control, time passage, and weathering). When significant differences were observed, the Student-Newman-Keuls multiple range test was performed to identify differences in elastomer-colorant combinations among each test condition at a significance level of .05.

**Results.** Changes in color, as a result of weathering, were noted in many of the colorant-elastomer combinations. Also, color change occurred not only to the colored, but also to uncolored materials over time without exposure to weathering.

**Conclusion.** Clinically, the addition of colorants could have a stabilizing effect on the elastomer color when it is exposed to weathering. (J Prosthet Dent 1999;81:431-8.)

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### CLINICAL IMPLICATIONS

*The clinician must keep in mind the benefits and shortcomings of each material used in fabricating a facial prosthesis. Pure inorganic colorants are color stable but difficult to use clinically. Impure inorganic coloring agents, or inorganic colorants with a vehicle are easier to use clinically, but they are not as color stable. Rayon flocking is useful for mimicking surface blood vessels and telangiectasia, but as an organic colorant, it is not color stable. In addition, the more opaque the clinical prosthesis is, the greater its color stability.*

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The goal in maxillofacial prosthodontics is to restore the patient's appearance and protect the resection site, allowing improvement in self-esteem to help the patient lead as normal a life as possible. It is important that a prosthesis be fabricated with optimal esthetics and maintenance of its appearance over its service lifetime. Degradation in appearance due to changes in color and physical properties are reasons for refabrication of a facial prosthesis. Koran et al[1] evaluated the accelerated-aging testing with MDX 4-4210 that had been colored with

dry mineral earth pigments. There were small changes in luminous reflectance, contrast ratio, dominant wavelength, and excitation purity. The data did not support clinical observations of large changes in color over time. Beatty et al[2] evaluated color changes in Silicone A-2186 elastomer, uncolored and then colored with dry earth pigments, after exposure to ultraviolet-A and ultraviolet-B lighting. Silicone A-2186 elastomer underwent detectable color changes when stored in the dark for 45 days; cosmetic red and cadmium yellow pigments underwent significant color changes after 400 hours of exposure, and Mars violet and cosmetic yellow ochre remained color-stable after 1800 hours.[2] To determine the effects of a ultraviolet light absorber incorporated in the material, Lemon et al[3] evaluated Silastic 4-4210 and

Silastic medical adhesive type A colored with oil-based pigments after natural weathering and accelerated aging. A spectrophotometer was used to calculate color change from spectral reflectance measurements in the visual range of 400 to 700 nm. For a given amount of absorbed energy, artificial weathering caused a greater change than natural weathering.

This in vitro study is the third article in a 3-part series that examines the interactions between elastomers, colorants, and weathering as they influence properties that are related to the effective life span of these prostheses. The purpose of this study was to evaluate (1) optical density with an optical densitometer and (2) color (in the L*a*b* system) with a chromameter to examine commonly used colorant-elastomer combinations for facial prostheses, when exposed to natural weathering.

## MATERIAL AND METHODS

Four of the more commonly used intrinsic coloring agents based on a recent survey of both the American Academy of Maxillofacial Prosthetics and the American Anaplastology Associations were evaluated[4]: dry earth pigments, rayon fiber flocking, artist's oil paints and kaolin (Factor II, Inc, Lakeside, Ariz.), and 1 recently introduced method[5] using liquid facial cosmetic (Polished Performance liquid make-up, Alabaster Beige 18-N, Esté Lauder, New York, N.Y.). The maxillofacial elastomers evaluated were 3 commonly used elastomers, based on that same survey: Silastic medical adhesive type A (Dow Corning Corp, Midland, Mich.), Silastic 4-4210 (Dow Corning Corp), and Silicone A-2186 (Factor II, Inc).

A chromameter (chroma meter CR221, Minolta Camera Co Ltd, Osaka, Japan) was used to evaluate color in the L*a*b* color system. In this system, value (or gray scale) is represented by L*. The value a* indicates color in the red/green axis, and b* indicates color in the yellow/blue axis. An optical densitometer (Macbeth TD 501, Process Instruments, Newburgh, N.Y.) was used to evaluate optical density.

Fifteen specimens, 3 mm thick and measuring approximately $100 \times 20$ mm, were fabricated for each elastomer-colorant combination in improved dental stone molds (Silky-Rock, WhipMix Corp, Louisville, Ky.). A total of 270 specimens were fabricated; there were 90 of each of the 3 elastomers (Silastic medical adhesive type A, Silastic 4-4210, and Silicone A-2186). For each elastomer, the 90 specimens were divided into 6 colorant categories (5 pigmented samples and 1 with no coloring) of 15 specimens each. Within each colorant category, there were 3 test condition variables (control, natural weathering, and time passage) that included 5 test specimens for each test condition.

The materials were handled in strict compliance with the manufacturer's instructions. To achieve maximum consistency among specimens within an elastomer-colorant category, all 15 specimens were fabricated during 1 processing. For the 2-part room temperature vulcanizing systems (Silastic 4-4210 and Silicone A-2186), 273 g of base were mixed with 27 g of catalyst to achieve the recommended ratio of 10:1. Three hundred grams of medical adhesive A (1-part room temperature vulcanizing material) was used directly from the tube. Colorants then were added in amounts to achieve concentrations similar to those found in clinical prostheses; this procedure is described in part 1 of this study.[6]

Colorants were mixed with the elastomers by hand using wooden tongue blades in 5-quart paper paint pails for 5 minutes. Each mixture (both the 1- and 2-part systems) was de-aired under a vacuum of at least 30 in of mercury for 20 minutes. The mixture then was placed in the stone molds, which had been coated with 2 applications of tinfoil substitute (Al-Cote, Dentsply Trubyte, York, Pa.) and allowed to dry. Care was taken not to incorporate air bubbles into the mold space or mixture. The mold was closed and clamped with a 1-in web-type ratcheting clamp (Pony clamp, Adjustable Clamp Co, Chicago, Ill.), and placed in a 100°F dry oven (Imperial II radiant heat oven, Labline Instrument, Melrose Park, Ill.) for 16 hours to allow polymerization. After polymerization, the mold was carefully separated, specimens were removed, and flash was trimmed away with a sharp scalpel. Specimens were then separated into treatment groups. The control data were measured within 30 days after fabrication.

### Test conditions

For the natural weathering groups, specimens in these test condition groups were suspended from wooden racks by stainless steel suture material, and the assembly was placed on the roof of the dental school at Indiana University for 6 months. This period was selected because it is thought to be the average life span of a maxillofacial prosthesis.[7,8] At the end of the treatment period, specimens were removed and cleaned in an ultrasonic cleaner with tap water and liquid detergent (Dawn dishwashing detergent, Procter and Gamble, Cincinnati, Ohio) for 10 minutes. The specimens were rinsed in running tap water, wiped dry, and then tested for color and optical density.

For the time passage group, specimens in these test conditions group were placed in sealed glass containers and placed in a dark environment at ambient room temperature (72°F ± 50°F) and humidity (50% ± 10%) for 6 months. At the end of this time, specimens were removed and tested for color and optical density. This group served as a nonweathered control group that was compared with the natural weathering group.

For color and optical density measurements, all data collection was performed at ambient room temperature and humidity after the specimens had been held in
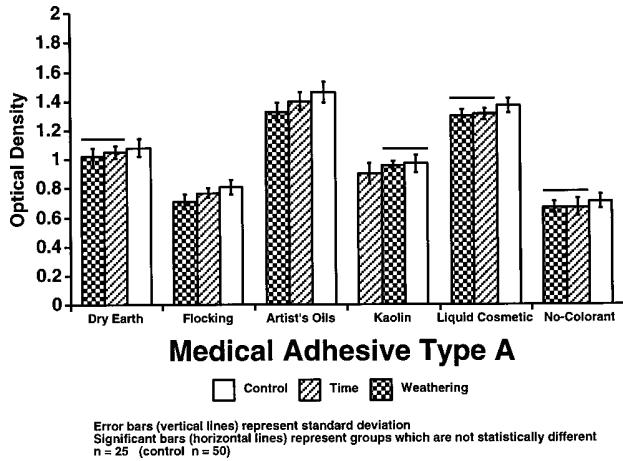
**Fig. 1.** Effects of weathering and time on optical density of medical adhesive A.

**Fig. 2.** Effects of weathering and time on optical density of Silastic 4-4210 elastomer.

these conditions for at least 24 hours. The chromameter (chroma meter CR-221, Minolta Camera Co, Ltd) was used to determine L*a*b* values at 3 locations on each specimen before and after subjecting the specimens to the test conditions. In this system, L* indicates gray scale, a* the red/green scale, and b* the yellow/blue scale. All readings were made at the same 3 relative locations on each specimen. The chromameter was calibrated with the standard white card before each group of specimens was measured, and the standard white card was used as a background when measuring all specimens. The mean measurement for each group was calculated. Because these specimens were able to serve as their own controls (were not damaged by measurement and could be measured before and after test conditions), the control data included 30 measurements, 15 from each test condition group.
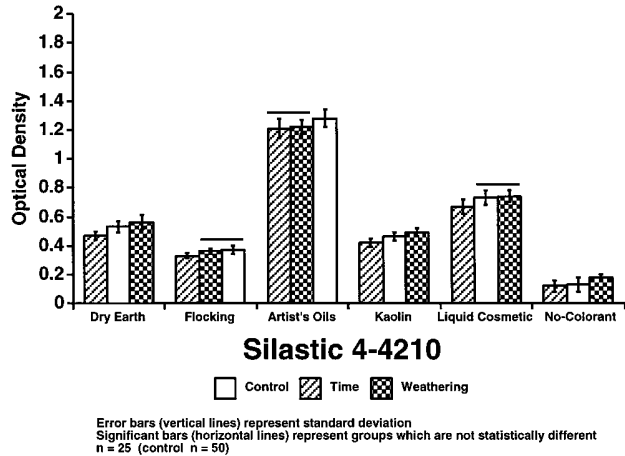
Color change or $\Delta E$ was calculated from the mean L*, a* and b* values for each group before and after subjecting the specimens to the test conditions by using the formula:

$$\Delta E = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{\frac{1}{2}}$$

The optical densitometer was used to measure light transmission through each specimen before and after subjecting the specimens to test conditions. These measurements were made at the same 5 relative locations on each of the specimens no closer than 6 mm from the edge, and at least 12 mm from the previous measurement. The mean of the 5 measurements for the 5 specimens (total of 25) in each group was recorded as the optical density for that group. Because these specimens were able to serve as their own controls (were not damaged by measurement and could be measured before and after test conditions), the control data included 50 measurements, 25 from each test condition group.

## Statistical analysis

Because colorants were added as representative values, and each elastomer may have required a different concentration of a given colorant to achieve a similar clinical effect, only pairings within each elastomer were of clinical relevance. Color and optical property data for each elastomer-colorant combination were subjected to a 1-way analysis of variance (ANOVA) to examine effects among test conditions (control, time passage, and weathering). When significant differences were observed, the Student-Newman-Keuls multiple range test was performed to identify differences in elastomer-colorant combinations among each test condition at a significance level of $\alpha \geq .05$. Changes from the control condition described in the text are statistically different.

## RESULTS
## Optical density

Optical density of medical adhesive type A without colorant added and then colored with dry earth pigment or liquid cosmetic was decreased by both time and weathering, but no statistical difference was noted between the time or weathering (Fig. 1). With rayon flocking or artist's oils were used to pigment type A medical adhesive, optical density decreased over time and to a greater extent with weathering. The addition of kaolin demonstrated a different result; there was no difference in optical density compared with the control specimens after weathering, but there was a statistically significant decrease with time.

The optical density of Silastic 4-4210 elastomer without colorant added when colored with dry earth pigments and kaolin decreased over time, but the optical density increased with weathering (Fig. 2). Optical density of Silastic 4-4210 elastomer colored with rayon flocking and liquid cosmetic remained unchanged with
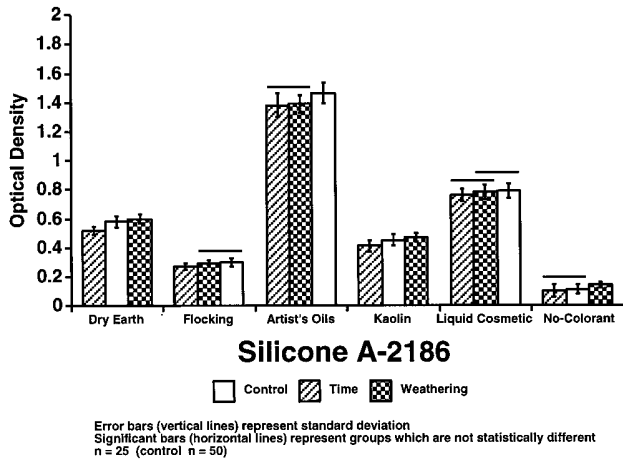
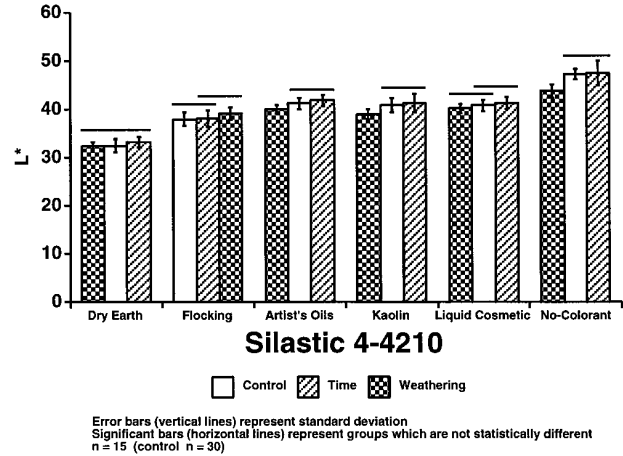**Fig. 3.** Effects of weathering and time on optical density of Silicone A-2186 elastomer.



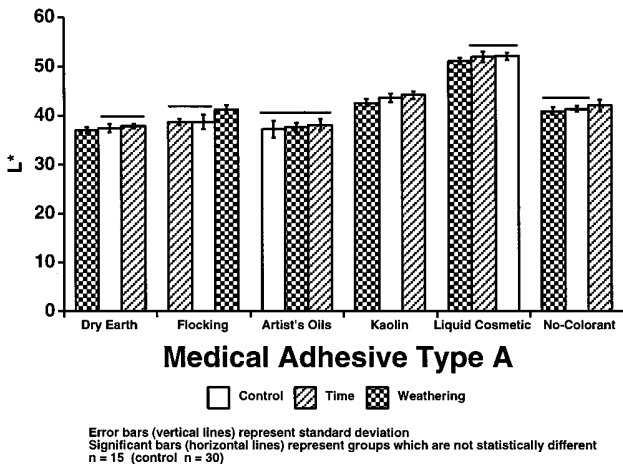**Fig. 5.** Effects of weathering and time on value of Silastic 4-4210 elastomer.



**Fig. 4.** Effects of weathering and time on value of medical adhesive A.
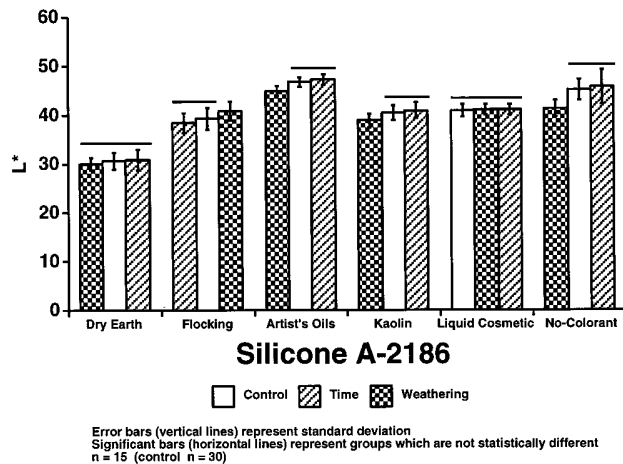


**Fig. 6.** Effects of weathering and time on value of Silicone A-2186 elastomer.

weathering, but optical density decreased with time. The optical density of that material with artist's oils decreased with both time and weathering.

Optical density results for Silicone A-2186 elastomer were similar to those for Silastic 4-4210 elastomer because dry earth pigment and kaolin increased the optical density with weathering and decreased the optical density with time (Fig. 3). Similarly, rayon flocking caused a decrease in optical density with time, but weathering had no effect. Artist's oils caused a decrease in optical density with time and weathering, but with no significant differences between time or weathering. The difference with Silastic 4-4210 was that without added color, Silicone A-2186 elastomer had no change in optical density over time, but weathering increased its optical density. When liquid cosmetic was used as a colorant, no statistical difference was found in any of the test conditions.

## Value (L*)

The L* value of medical adhesive type A without the addition of colorant increased as a result of time, but no difference was demonstrated for weathering (Fig. 4). No difference could be shown for artist's oils. For the dry earth pigment group, weathering decreased the L*, whereas no significant difference could be determined with time. Weathering increased the L* value for rayon flocking specimens, whereas time had no effect. Weathering decreased the L* value for the liquid cosmetic–colored specimens, whereas time had no effect. Kaolin-colored specimens increased in L* with time and decreased with weathering.

The L* of the Silastic 4-4210 and Silicone A-2186 specimens responded similarly to each other (Figs. 5 and 6). Specimens without colorant added and those
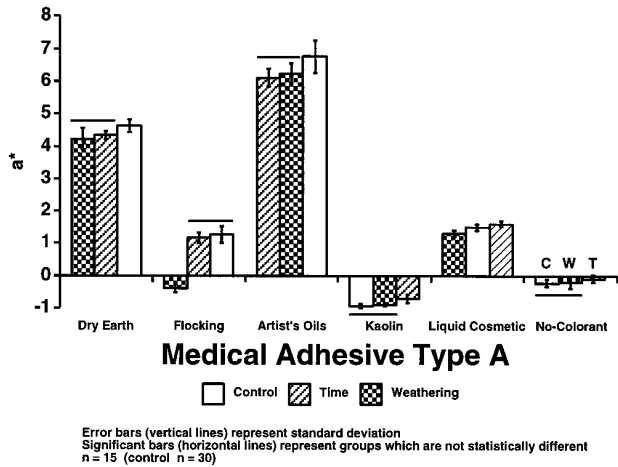
**Fig. 7.** Effects of weathering and time on red/green chroma of medical adhesive A.
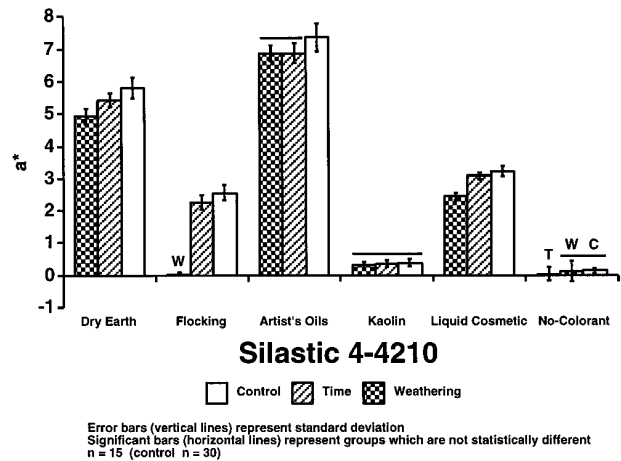


**Fig. 8.** Effects of weathering and time on red/green chroma of Silastic 4-4210 elastomer.

colored with artist's oils and kaolin decreased in L* with weathering and were not affected by time. Weathering increased the L* of rayon flocking–colored Silicone A-2186 elastomer. Differences could not be observed for the remaining polymer-colorant combinations.

## Red/green chroma (a*)

In the L*a*b* system, a positive a* value indicates a red chroma, whereas a higher positive a* indicates a more intense red chroma. A negative a* indicates a green chroma, whereas a higher absolute value of a negative a* (a more negative number) indicates a more intense green chroma. For medical adhesive A, both dry earth pigments and artist's oils were greener with time and weathering with no statistical difference between the 2 test conditions (Fig. 7). Weathering had no effect on kaolin specimens, but time caused them to become redder. Liquid cosmetic–colored specimens were greener with weathering, but redder with time. The greatest effect was seen with rayon flocking, which changed from the red to green end of the a* scale (numerical change of 1.67) with weathering yet no effect was seen with time. No effect of weathering could be shown for medical adhesive type A without colorant added, although these specimens did significantly shift in the red direction with time.

Weathering had no effect on the red/green scale for Silastic 4-4210 without colorant added, which became greener with time (Fig. 8). Both time and weathering made the artist's oil–colored Silastic 4-4210 greener with no statistical difference between the 2 test conditions. When Silastic 4-4210 elastomer was colored with dry earth pigments, rayon flocking, or liquid cosmetic, the test conditions were additive. Specimens were greener with time and even greener with weathering. The effect of weathering on flocking is noteworthy.
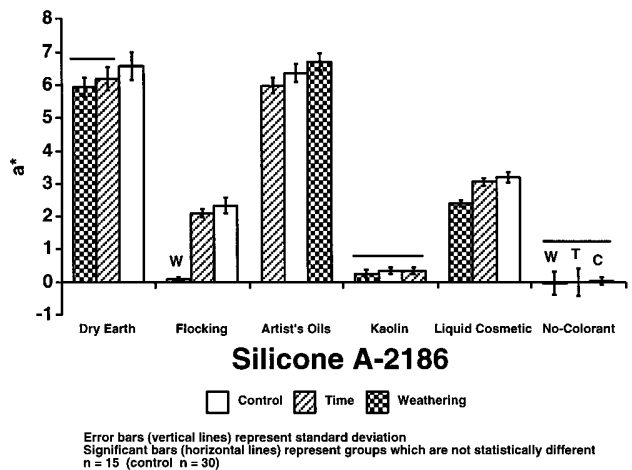


**Fig. 9.** Effects of weathering and time on red/green chroma of Silicone A-2186 elastomer.

The a* of Silastic 4-4210 decreased by 2.51 when the rayon flocking specimens were weathered. No difference could be demonstrated for kaolin specimens of this elastomer.

No difference in red/green chroma could be shown between test condition for Silastic 4-4210 or Silicone A-2186 elastomers colored with kaolin. No difference in test conditions could be shown with Silicone A-2186 specimens with no colorant (Fig. 9). Both weathering and time had the same greening effect on dry earth pigment–colored Silicone A-2186 as they had on Silastic 4-4210 elastomer. Again, additive effects could be seen when Silicone A-2186 elastomer was colored with rayon flocking and liquid cosmetic: time caused greening and weathering caused more greening. The greatest effect was seen with rayon flocking, which showed a
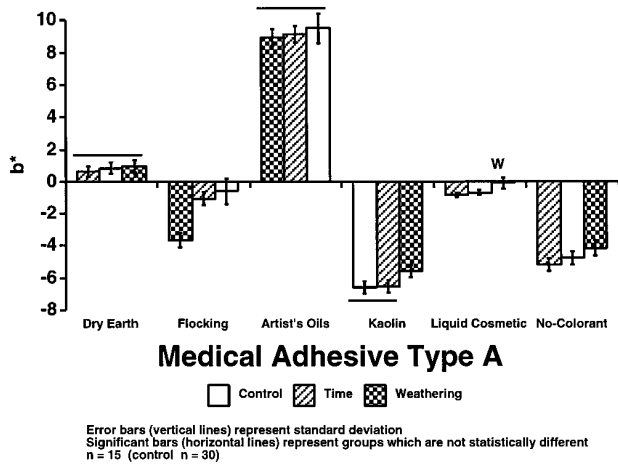
**Fig. 10.** Effects of weathering and time on yellow/blue chroma of medical adhesive A.
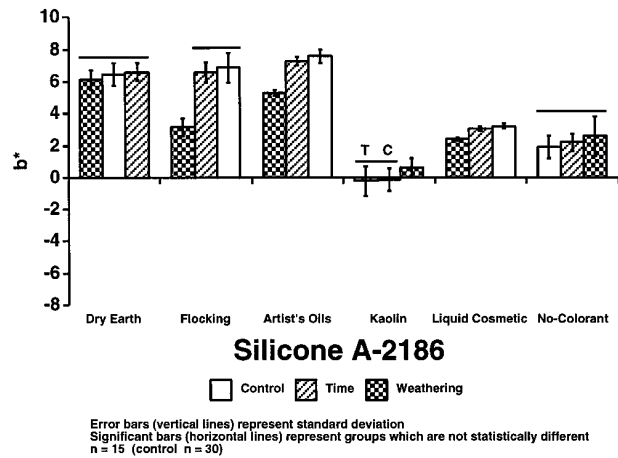


**Fig. 12.** Effects of weathering and time on yellow/blue chroma of Silicone A-2186 elastomer.
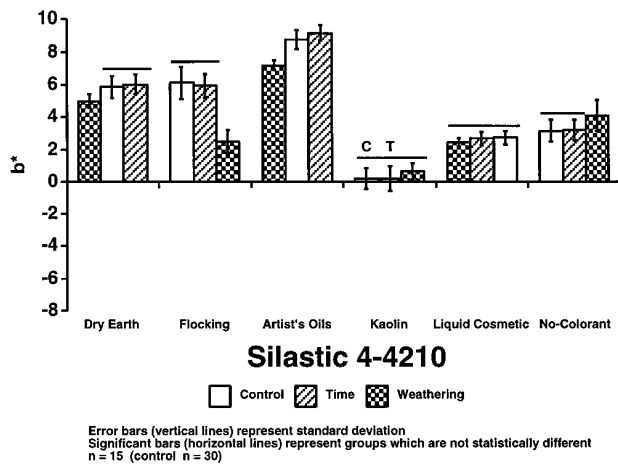


**Fig. 11.** Effects of weathering and time on yellow/blue chroma of Silastic 4-4210 elastomer.

2.25 numerical decrease in the a* scale when weathered. Silicone A-2186 colored with artist's oils became greener with time, but redder after weathering.

## Yellow/blue chroma (b*)

In the L*a*b* system, a positive b* indicates a yellow chroma, whereas a higher positive a* indicates a more intense yellow chroma. A negative b* indicates a blue chroma, whereas a higher absolute value of a negative b* (a more negative number) indicates a more intense blue chroma. When medical adhesive type A was used as the elastomer and when either dry earth pigments or artist's oils were the colorant, no difference could be demonstrated among the test conditions (Fig. 10). When kaolin was used to color medical adhesive type A, no difference in yellow/blue chroma was

observed with time, but weathering caused a yellowing. The medical adhesive type A without colorant added and the elastomer colored with liquid cosmetic became bluer with time and more yellow with weathering. Medical adhesive type A colored with rayon flocking became bluer with time, but the greatest effect was after weathering. The specimens turned bluer with a 3.04 numerical change in b*.

When Silastic 4-4210 elastomer was used as the elastomer, no difference among test conditions could be demonstrated when kaolin or liquid cosmetic was used as a colorant (Fig. 11). Weathering yellowed the Silastic 4-4210 with added colorant; time showed no effect. Artist's oil–colored Silastic 4-4210 specimens yellowed with time and became bluer with weathering. Dry earth pigment–colored specimens were not affected by time, but shifted to blue after weathering. Rayon flocking was not affected by time but shifted dramatically toward blue after weathering with a 3.65 numerical change in b*.

No statistical difference could be seen among the test conditions for Silicone A-2186 elastomer without added colorant; this elastomer was colored with dry earth pigments (Fig. 12). Kaolin-colored Silicone A-2186 elastomer was not affected in b* chroma by time but was yellowed by weathering. When Silicone A-2186 elastomer was colored with liquid cosmetic or artist's oils, the effect on b* was additive; time caused a bluing and weathering caused additional bluing. Silicone A-2186 specimens with rayon flocking were not affected in b* chroma with time, but a dramatic 3.68 numerical change in b* toward blue occurred when the flocking specimens were exposed to weathering.

## Change in color (ΔE)

In the L*a*b* system, a ΔE of >1 is considered visually detectable.[9] When medical adhesive type A was
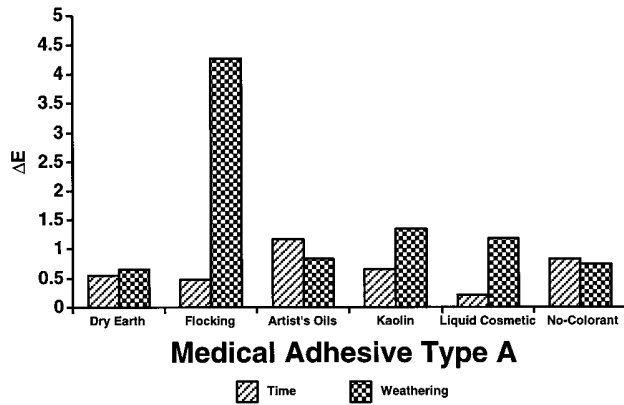
**Fig. 13.** Effects of weathering and time on change in color of medical adhesive A.



**Fig. 14.** Effects of weathering and time on change in color of Silastic 4-4210 elastomer.



**Fig. 15.** Effects of weathering and time on change in color of Silicone A-2186 elastomer.

used as the elastomer, the no-colorant specimens theoretically were not changed visually in color by time or weathering (Fig. 13). With $\Delta E$ values slightly over 1.0, medical adhesive type A was theoretically changed in color when colored by kaolin or liquid cosmetic and exposed to weathering, or when colored by artist's oils and stored for 6 months. The question arises, however, whether these changes would be enough to require the refabrication of a prosthesis, which would be of clinical significance. A dramatic $\Delta E$ of 4.27 occurred when medical adhesive type A with rayon flocking was exposed to weathering. This change was probably clinically significant.

When Silastic 4-4210 and Silicone A-2186 elastomers, both with no added colorant were exposed to weathering, $\Delta E$ values of 3.52 and 3.86, respectively, occurred, indicating a visual and perhaps clinically significant change (Figs. 14 and 15). With the exception of rayon flocking, the addition of colorants decreased the weathering effect on $\Delta E$. Rayon flocking, when added to these materials, showed the opposite effect with $\Delta E$ values of 4.59 and 4.60, again theoretically visually detectable and most likely clinically significant. With the exception of the artist's oil–colored medical adhesive A, which showed a $\Delta E$ of 1.17, all other groups had a $\Delta E$ of less than 1.0 with time.

## DISCUSSION

Degradation of the color and the physical properties of maxillofacial prostheses in clinical use require refabrication approximately every 6 months. Ideally, the elastomer-colorant combination should not only allow satisfactory esthetics to be achieved clinically, but also to maintain the esthetics indefinitely, or at least until tissue changes in structure, color, or esthetics necessitate refabrication of the prosthesis. The color also should be stable over time and aging.

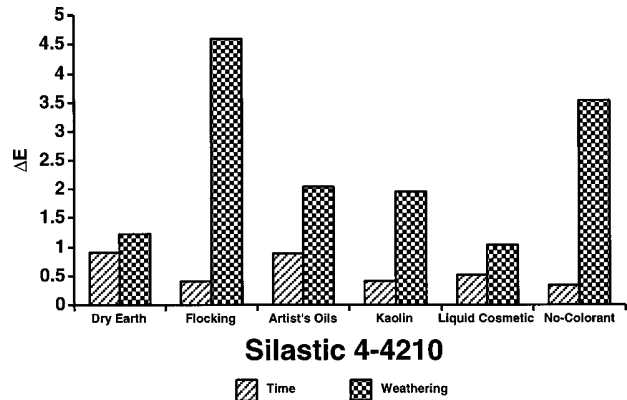Two types of colorants are available, inorganic and organic. The inorganic colorants are metallic oxides. These molecules are very stable as a result of their ionic bonds. These inorganic compounds are commonly used in products such as paints and tend to be very color stable, unless the components are washed away. Placement of double and triple bonds impart color to the organic colorant molecule. These bonds tend to be relatively reactive, and colorants are less stable. A common example of the use of organic colorants is in textiles.

The poor color stability of rayon fiber flocking with a numerical $\Delta a^*$ of as much as 2.51 and a $\Delta b^*$ of as much as 3.68, suggests the colorant is most likely organic. The clinician should keep this in mind when using rayon fiber flocking as a coloring agent in a facial prosthesis and should expect that the color will change over time. The clinician should reserve the use of rayon flocking for mimicking surface blood vessels and telangiectasia, because no suitable alternative exists.

Dry earth pigments and kaolin are forms of inorganic colorants. Judging from the relative stability of

color after weathering of artist's oil paints and liquid facial cosmetic compounds most likely contain inorganic coloring agents; the difference between them is how they are supplied. Some type of vehicle for the colorant is used in artist's oil paints and liquid facial cosmetics, which allows the colorant to be handled in liquid form. The products eventually harden, most likely through evaporation or absorption of the liquid vehicle.

Judging from the higher ΔE values of liquid cosmetic and artist's oil pigment compared with dry earth pigment, data suggest the vehicle in liquid cosmetic was not color-stable when exposed to weathering. Also, the vehicle in artist's oil pigment may affect the weathering-induced color instability of Silastic 4-4210 and Silicone A-2186 elastomers, but not affect the color stability of medical adhesive A. When using these colorants, the clinician must keep in mind that, although the liquid forms are easy to mix and are supplied in clinically useful colors and not as intense as the dry forms, liquid forms contain additional ingredients that can affect the long-term color of the prosthesis.

Kaolin was not as color-stable as anticipated, which could be attributed to organic impurities left in the material after the mining and refining process. Dry earth pigments, although more stable in color, presented their own shortcomings. These pigments are intense in color, which makes them difficult to use clinically.

One unexpected outcome of our study was an indication that the elastomers were not as stable as had been assumed by the profession. Changes in optical properties and color occurred in both colored and no-colorant specimens that had been sealed in containers and kept in unlighted conditions for 6 months. For example, the optical density of medical adhesive A colored with both dry earth pigment and liquid cosmetic decreased with time and weathering with no difference between the 2 test conditions. Both findings indicated the effects were inherent in the elastomers and not influenced by weathering. Possibilities are that the changes were caused by impurities incorporated during manufacturing, by reaction products, by initiators, or by some other mechanism. Determination of the reason for these changes could lead to more stable formulations, which could result in a longer length of service for a prosthesis.

Another unexpected outcome of our study was the apparent protective effect that the addition of coloring agents had on Silastic 4-4210 and Silicone A-2186 elastomers. The colorants actually decreased the apparent color deterioration compared with no-colorant specimens. As an example, the ΔE of weathered dry earth pigmented Silicone A-2186 elastomer was only 24% of the weathered no-colorant specimen. However, this was not observed with medical adhesive A, which had a much higher uncolored optical density, and the incorporation of the colorants increased the optical density

of the silicones. These findings supported the concept that the opacifying effect could protect the silicones from degradation. This effect should be the focus of further investigation, and studies should examine the effect of ultraviolet protectant and stabilizers on the color stability and the physical property stability of elastomers used in maxillofacial prosthesis.

## CONCLUSIONS

The following conclusions were drawn from this study:

1. Addition of colorants to the silicones altered the effect of weathering on optical properties and color. Colorants tended to "protect" the silicones from weathering, possibly by blocking light radiation to the elastomer.

2. Silicones were not as stable as has been assumed by the profession. Optical property and color changes occurred in both colored and no-colorant specimens, which had been sealed in containers and kept in unlighted conditions.

3. Inorganic coloring agents (dry earth pigments, kaolin, artist's oils, and liquid cosmetic) were the most color stable over time, whereas the organic colorant (rayon fiber flocking) was the least color stable.

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