The Color Accuracy of the Kubelka-Munk Theory for Various Colorants in Maxillofacial Prosthetic Material*

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The reflectance model developed by Kubelka and Munk was evaluated for agreement in color prediction of thick pigmented samples and for linearity of optical absorption and scattering coefficients with concentration of colorant in maxillofacial elastomer. The colorants tested were generic opacifiers, dry mineral earth pigments, and fibrous colorants. Significant linear relationships were commonly found between the optical coefficients and the concentration of the colorants. These relationships indicated occasional optical interaction between the colorants and the elastomer. Color differences between theoretical and observed colors of the thick samples averaged 2.96, 3.47, and 1.60 for the opacifiers, mineral earth pigments, and fibrous colorants, respectively, when measured using the CIELAB uniform-color space. The agreement between theoretical and observed colors was significantly closer for the fibrous colorants than for the dry mineral earth pigments of the same labeled color.

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Introduction.

The solution of an optical reflectance theory by Kubelka and Munk (1931) and its subsequent simplification by Kubelka (1948) gave rise to the application of this theory to describe the color and translucency of colored materials within the paint (Billmeyer and Abrams, 1973) and plastic (Billmeyer and Abrams, 1972) industries. Judd (1937) initially applied the Kubelka-Munk theory to dental materials, and O'Brien and his co-workers have advanced this theory in its application to dental composite (Miagawa *et al.*, 1981) and porcelain materials (Woolsey *et al.*, 1984; O'Brien *et al.*, 1985).

Kubelka and Munk (1931) solved the simultaneous flow of two diffuse light fluxes (forward and backward) within a turbid medium in optical contact with a reflective backing. An assumption involved in the development of the Kubelka-Munk theory is that no reflections occur at the interfacial boundaries between the turbid material and the air at the front surface, or between the turbid material and the backing. Corrections for the reflections which do occur have been presented by Duntley (1942) and Saunderson (1942), but the value for internal reflection proposed by Mudgett and Richards (1973) enables these corrections to provide better agreement between observed reflectance and the Kubelka-Munk theory (Johnston *et al.*, 1986).

The Kubelka-Munk theory, corrected as above, and the linear dependence of the optical absorption and scattering coefficients upon colorant concentration have provided the basis for colorant formulation for materials in general (Johnston, 1973; Kuehni, 1975), but Kuehni (1975) warns of possible optical or mechanical interactions of the coloring pigments and the substrate. However, Billmeyer and Richards (1973) state that, for translucent plastics, "two-flux turbid-medium theory is inadequate.... Four-flux theory is adequate ... but better accuracy can be obtained by the use of six-flux or many-flux calculations". Theories involving more than two diffuse light fluxes incorporate collimated light fluxes at specific angles to interfacial boundaries of the turbid medium. The purposes of this study were to ascertain the ability of the Kubelka-Munk theory to describe the color induced by different types of colorants in maxillofacial elastomer and to study the influence of concentrations of these colorants.

Materials and methods.

Three categories of colorants were chosen for this study: six generic opacifiers, nine dry mineral earth pigments (Artskin Products Co., 200 Circle E Bldg., Military Circle, Norfolk, VA), and six colored nylon fibrous colorants known as "flockings" (Claremont Flock Corp., Claremont, NH). The generic opacifiers were titanium dioxide (Batch 33578, J.T. Baker Co., Phillipsburg, NJ), tin (IV) oxide (Batch 2332, Allied Chemical & Dye Corp., New York, NY), zinc oxide (manufacturer's No. 8832, batch BVY, Mallinckrodt, Inc., St. Louis, MO), kaolin (manufacturer's No. G-1001, Factor II Products, Lakeside, AZ), zinc stearate (Batch 41585, Merck & Co., Inc., Rahway, NJ), and talc (Batch 23652, Johnson's Baby Products Co., Skillman, NJ). The mineral earth pigments were labeled black, white, red, blue, dark buff, light brown, medium brown, red brown, and yellow. The flockings were labeled blue, white, tan, brown, red brown, and red.

For the purpose of determining the optical absorption and scattering coefficients, the opacifiers and the mineral earth pigments were separately incorporated into 30 g of medical-grade elastomer (Silastic MDX4-4210 silicone elastomer, Lot HH-062180, Dow Corning Corp., Medical Products Div., Midland, MI 48640) in concentrations of 0.1, 0.05, and 0.02 wt%. The flockings were separately incorporated into the silicone base at concentrations of 2, 1, and 0.5 wt%. To the colored silicone base was then added 10 wt% curing agent (MDX4-4210 curing agent, Lot HH-033211, Dow Corning Corp., Medical Products Div., Midland, MI 48640). The mix was then placed under reduced atmospheric pressure in order to eliminate incorporated air bubbles, and was poured into an aluminum mold which produced specimens 60 mm by 120 mm, with a thickness within 0.25 mm of 1.25 mm. The mold and elastomer were returned to reduced pressure and cured at 65°C for eight hours. Two specimens of each concentration of each colorant and two specimens of unpigmented elastomer were fabricated, making a total of 128 specimens for optical absorption and scattering determination.

In addition, thicker specimens were formed for each colorant at the greatest concentration given above. These specimens were formed in an identical manner, except that 60 g of elastomer base was used, and the molds produced specimens 40

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mm in diameter and 40 mm in height. Twenty-one specimens were formed for the determination of reflectivity, which is the reflectance at infinite optical thickness.

Titanium oxide and black dry mineral earth colorants were chosen for the fabrication of the dark and light backings to be used for optical coefficient determinations. One percent by weight of each of these two colorants was used separately to make backings as described above for the thick reflectance specimens.

The optical coefficients were determined by the methods fully described by Woolsey *et al.* (1984) and by Johnston *et al.* (1986), with one exception: No custom-made sample port plate was used, since the elastomeric specimens were of sufficient size to be held within the standard sample port. Reflectance measurements were made with illumination beam condensers in place, such that any reflected light scattered toward the edge of the sample would be included in the measurement. To summarize the determination method, the reflectance spectrum of each thin specimen was measured with the specimen in optical contact with each of the two backings, and the Kubelka-Munk absorption coefficient, K, and scattering coefficient, S, were calculated by the solution of two conditions of the Kubelka-Munk theory. For each concentration of each colorant and for the unpigmented material, two determinations of the optical coefficient were obtained and averaged at every wavelength.

The linear relationship between the optical coefficients of each colorant and the colorant concentration were analyzed by linear regression analysis at wavelengths of 450, 500, and 650 nm. The optical coefficients of the unpigmented elastomer were also included.

The reflectivity was calculated by the equation of Kubelka (1948):

$$Rinf = [1 + K/S + (K^2/S^2 + 2K/S)^{1/2}]^{-1}$$

where K and S are the average optical absorption and scattering coefficients, respectively. This equation was corrected for interfacial reflections (Duntley, 1942; Saunderson, 1942) using the value of internal reflection proposed by Mudgett and Richards (1973). The calculated color parameters were determined by the method of C.I.E. (1976, 1985). The observed reflectivity for the 21 thick specimens was measured on a diffuse reflectance spectrophotometer (Beckman Acta-C3 UV-visible spectrophotometer with ASPH-U integrating sphere, Beckman Instruments, Inc., Irvine, CA 92664). The observed color parameters were determined from the average of two measurements of reflectivity. The color difference between the calculated and observed color parameters was then determined within the (L*, a*, b*)-uniform color space (C.I.E., 1976). Statistical analysis of the color differences was performed for those mineral earth and flocking colorants which had identical color names.

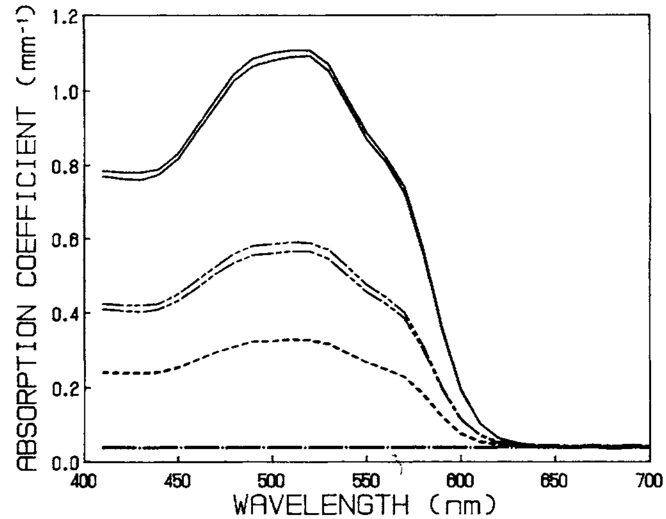

Fig. 1 — Spectra of optical absorption coefficients for maxillofacial elastomer with red flocking in concentrations of 2.0 wt% (solid line), 1.0 wt% (long-short-short dashed line), 0.5 wt% (dashed line), and 0.0 wt% (dash-dot line).

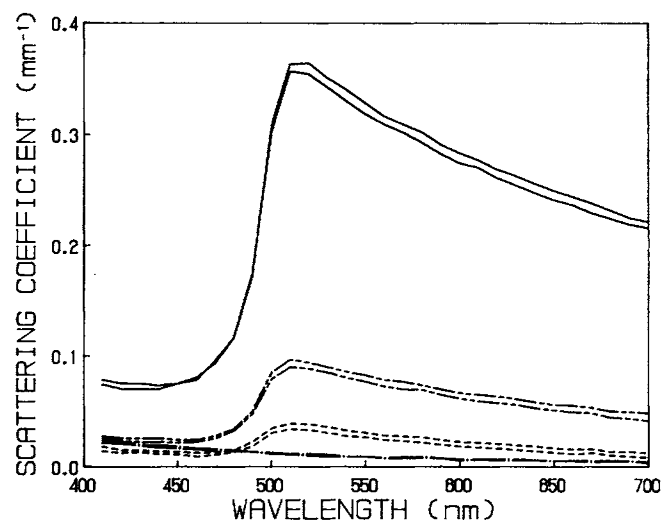

Fig. 2 — Spectra of optical scattering coefficients for maxillofacial elastomer with yellow dry mineral earth pigment in concentrations of 0.10 wt% (solid), 0.05 wt% (long-short-short dashed line), 0.02 wt% (dashed line), and 0.00 wt% (dash-dot line).

Results.

In general, the absorption spectra of the chromatic colorants exhibited absorption maxima at the wavelengths corresponding to the opposite color of each colorant studied. For example, Fig. 1 displays the spectral optical absorption coefficients of the unpigmented elastomer and of the elastomer with the three concentrations of red flockings. Fig. 2 similarly displays the spectral scattering coefficients of yellow dry mineral earth pigment.

The results of all linear regression analyses of the optical absorption (K) and scattering (S) coefficients are presented in Tables 1 and 2, respectively. The intercept is the regression estimate of each coefficient for unpigmented elastomer, and the slope represents the increase in coefficient per increase in concentration in wt%. The average optical absorption coefficients for unpigmented elastomer were 0.039, 0.038, and 0.038 mm^{-1} at the wavelengths of 450, 500, and 650 nm, respectively. The average optical scattering coefficients for unpigmented elastomer were 0.018, 0.009, and 0.005 mm^{-1} at the wavelengths of 450, 500, and 650 nm, respectively. The standard error is that of the regression for each coefficient at the average concentration studied. Since there were 126 regression analyses, only those with a statistical probability of 0.0003 or less were considered statistically significant. Figs. 3 through 6 present the values of selected optical coefficients and show the positions of the linear regression lines relative to the observed optical coefficients.

Fig. 7 presents the calculated and observed reflectivities for 0.1 wt% dark buff dry mineral earth colorant. Table 3 gives the color differences between calculated and observed color

TABLE 1
LINEAR REGRESSION ANALYSES OF OPTICAL ABSORPTION COEFFICIENT (K) VERSUS COLORANT CONCENTRATION

Colorant	Wavelength (nm)	Intercept (per mm)	Slope (per mm)	Standard Error	P
	450	0.033	−20	0.0077	0.0337
Titanium Dioxide	550	0.033	−26	0.0063	0.0045
	650	0.034	−31	0.0049	0.0005
	450	0.040	−16	0.0019	0.0001
Tin (IV) Oxide	550	0.040	−17	0.0024	0.0003
	650	0.041	−15	0.0025	0.0007
	450	0.040	−22	0.0021	<0.0001
Zinc Oxide	550	0.041	−20	0.0025	0.0001
	650	0.041	−18	0.0029	0.0005
	450	0.039	5.5	0.0022	0.0369
Kaolin	550	0.038	5.3	0.0019	0.0240
	650	0.039	4.2	0.0018	0.0498
	450	0.038	2.2	0.0019	0.2647
Zinc Stearate	550	0.038	1.3	0.0016	0.4112
	650	0.039	0.7	0.0015	0.6600
	450	0.038	2.4	0.0019	0.2330
Talc	550	0.038	3.0	0.0016	0.0584
	650	0.038	2.0	0.0013	0.1466
	450	0.053	462	0.0146	<0.0001
Red Artskin	550	0.050	442	0.0123	<0.0001
	650	0.032	−12	0.0014	0.0001
	450	0.051	361	0.0114	<0.0001
Med. Brown Artskin	550	0.045	289	0.0064	<0.0001
	650	0.040	82	0.0016	<0.0001
	450	0.042	1154	0.0073	<0.0001
Black Artskin	550	0.039	1174	0.0068	<0.0001
	650	0.036	1219	0.0058	<0.0001
	450	0.039	−23	0.0035	0.0004
White Artskin	550	0.040	−20	0.0026	0.0002
	650	0.041	−16	0.0027	0.0007
	450	0.041	288	0.0046	<0.0001
Light Brown Artskin	550	0.039	123	0.0020	<0.0001
	650	0.041	6.7	0.0025	0.0300
	450	0.036	470	0.0045	<0.0001
Dark Buff Artskin	550	0.036	71	0.0023	<0.0001
	650	0.040	−22	0.0012	<0.0001
	450	0.041	12	0.0026	0.0031
Blue Artskin	550	0.038	238	0.0038	<0.0001
	650	0.038	186	0.0027	<0.0001
	450	0.034	291	0.0066	<0.0001
Red Brown Artskin	550	0.033	243	0.0066	<0.0001
	650	0.034	77	0.0042	<0.0001
	450	0.026	1240	0.0651	<0.0001
Yellow Artskin	550	0.032	−23	0.0063	0.0091
	650	0.033	−23	0.0055	0.0012
	450	0.050	32	0.0113	<0.0001
Blue Flocking	550	0.056	49	0.0184	<0.0001
	650	0.053	43	0.0152	<0.0001
	450	0.034	0.25	0.0054	0.3774
White Flocking	550	0.034	0.23	0.0040	0.2783
	650	0.035	0.24	0.0036	0.2089
	450	0.049	11	0.0098	<0.0001
Tan Flocking	550	0.043	5.8	0.0045	<0.0001
	650	0.042	2.3	0.0032	<0.0001
	450	0.058	28	0.0199	<0.0001
Brown Flocking	550	0.050	21	0.0130	<0.0001
	650	0.043	10	0.0052	<0.0001
	450	0.056	40	0.0195	<0.0001
Red Brown Flocking	550	0.051	34	0.0141	<0.0001
	650	0.042	16	0.0048	<0.0001
	450	0.048	39	0.0108	<0.0001
Red Flocking	550	0.047	42	0.0112	0.0002
	650	0.041	0.03	0.0026	0.8331

TABLE 2
LINEAR REGRESSION ANALYSES OF OPTICAL SCATTERING COEFFICIENT (S) VERSUS COLORANT CONCENTRATION

Colorant	Wavelength (nm)	Intercept (per mm)	Slope (per mm)	Standard Error	P
	450	−0.159	610	0.0314	<0.0001
Titanium Dioxide	550	−0.020	504	0.0274	<0.0001
	650	0.021	417	0.0238	<0.0001
	450	0.016	74	0.0032	<0.0001
Tin (IV) Oxide	550	0.007	55	0.0026	<0.0001
	650	0.004	44	0.0019	<0.0001
	450	0.015	98	0.0033	<0.0001
Zinc Oxide	550	0.007	63	0.0023	<0.0001
	650	0.004	47	0.0017	<0.0001
	450	0.019	4.7	0.0009	0.0012
Kaolin	550	0.010	2.5	0.0006	0.0063
	650	0.006	2.5	0.0007	0.0087
	450	0.018	2.9	0.0009	0.0158
Zinc Stearate	550	0.010	1.6	0.0008	0.0737
	650	0.005	2.7	0.0005	0.0012
	450	0.017	2.6	0.0012	0.0556
Talc	550	0.009	1.6	0.0003	0.0022
	650	0.005	2.0	0.0004	0.0023
	450	0.019	8.2	0.0018	0.0028
Red Artskin	550	0.010	11	0.0014	0.0002
	650	0.002	99	0.0039	<0.0001
	450	0.018	6.4	0.0008	0.0002
Med. Brown Artskin	550	0.010	16	0.0006	<0.0001
	650	0.004	49	0.0016	<0.0001
	450	0.018	11.1	0.0013	0.0001
Black Artskin	550	0.009	8.2	0.0009	0.0001
	650	0.006	9.2	0.0013	0.0003
	450	0.012	119	0.0056	<0.0001
White Artskin	550	0.006	66	0.0032	<0.0001
	650	0.003	46	0.0020	<0.0001
	450	0.016	16	0.0016	<0.0001
Light Brown Artskin	550	0.007	34	0.0023	<0.0001
	650	0.003	42	0.0026	<0.0001
	450	0.011	113	0.0076	<0.0001
Dark Buff Artskin	550	−0.002	172	0.0112	<0.0001
	650	−0.051	159	0.0103	<0.0001
	450	0.016	21	0.0017	<0.0001
Blue Artskin	550	0.008	7.0	0.0010	0.0003
	650	0.004	12	0.0009	<0.0001
	450	0.019	5.6	0.0013	0.0043
Red Brown Artskin	550	0.010	8.8	0.0012	0.0002
	650	0.005	33	0.0010	<0.0001
	450	0.006	61	0.0119	0.0016
Yellow Artskin	550	−0.026	320	0.0416	0.0002
	650	−0.025	246	0.0346	0.0003
	450	0.020	0.72	0.0025	0.0009
Blue Flocking	550	0.011	0.02	0.0024	0.8619
	650	0.007	0.07	0.0020	0.4727
	450	0.018	1.7	0.0016	<0.0001
White Flocking	550	0.010	1.2	0.0010	<0.0001
	650	0.006	1.0	0.0010	<0.0001
	450	0.017	0.58	0.0007	<0.0001
Tan Flocking	550	0.009	0.49	0.0006	<0.0001
	650	0.005	0.47	0.0004	<0.0001
	450	0.018	0.34	0.0013	0.0020
Brown Flocking	550	0.010	0.27	0.0009	0.0008
	650	0.005	0.43	0.0007	<0.0001
	450	0.018	0.85	0.0011	<0.0001
Red Brown Flocking	550	0.010	0.52	0.0015	0.0003
	650	0.005	0.84	0.0009	<0.0001
	450	0.018	0.60	0.0009	<0.0001
Red Flocking	550	0.010	0.28	0.0007	0.0002
	650	0.004	1.20	0.0010	<0.0001

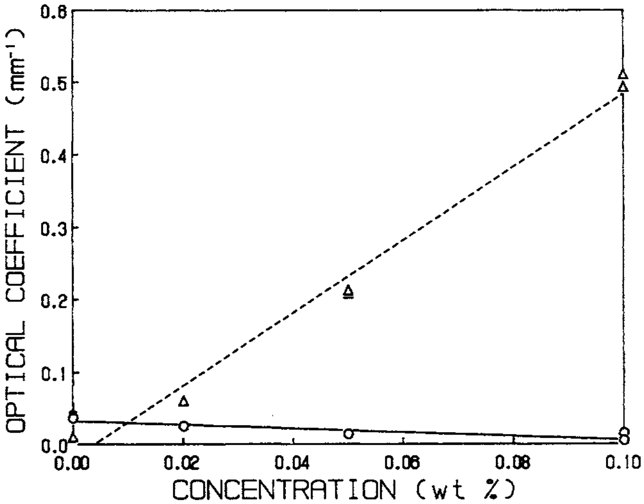

Fig. 3 — Scatter diagrams and regression lines for Kubelka-Munk absorption (open circles and solid line) and scattering (open triangles and dashed line) coefficients *vs*. concentration at 550 nm for titanium dioxide.

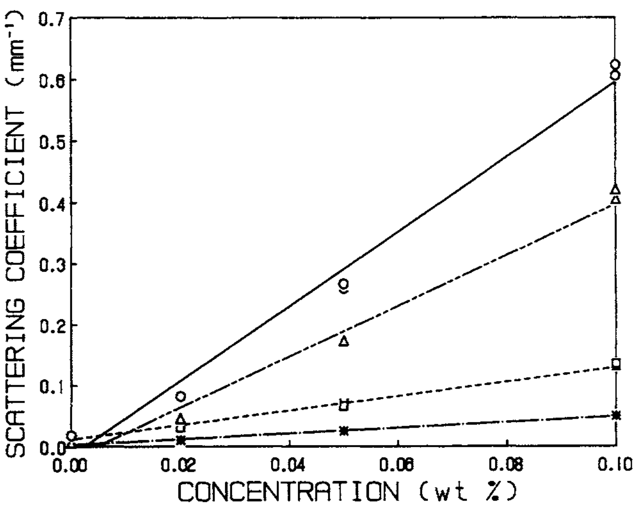

Fig. 4 — Scatter diagrams and regression lines for Kubelka-Munk scattering coefficients *vs*. concentration at 450 and 650 nm for titanium dioxide (450 nm: open circles and solid line; 650 nm: open triangles and long-short-short dashed line) and white dry mineral earth pigment (450 nm, open squares and dashed line; 650 nm, * and dash-dot line).

parameters at infinite optical thickness for all colorants studied. These color differences for the mineral earth and flocking colorants of similar color labels are presented in Table 4 with the paired comparison analysis of variance.

Discussion.

The low error associated with each regression of the optical coefficients upon colorant concentration indicates the consistency with which the Kubelka-Munk theory derives absorption and scattering coefficients relative to the concentrations of many types of colorants within maxillofacial elastomer. Wherever a statistically significant regression was not found, that optical coefficient was not expected to change with concentration for that pigment; *e.g.*, the absorption coefficient for any opacifier may remain at a relatively low value regardless of colorant concentration. The generally consistent values of the intercepts for each optical coefficient indicate the general lack of optical interaction between the maxillofacial elastomer and the color-

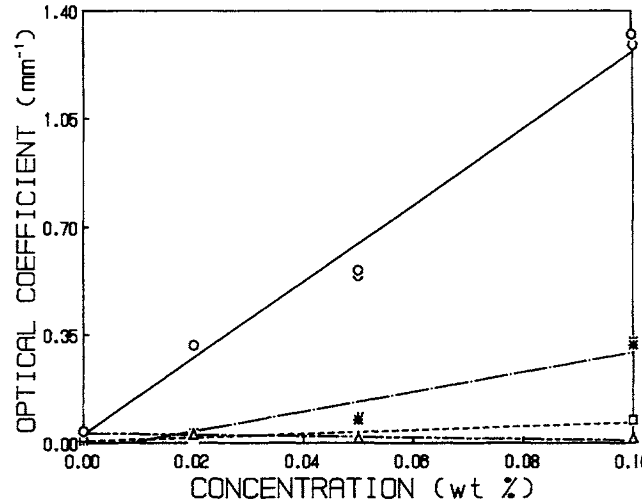

Fig. 5 — Scatter diagrams and regression lines for Kubelka-Munk absorption (K) and scattering (S) coefficients *vs*. concentration at 450 nm (K, open circles and solid line; S, open squares and dashed line) and 550 nm (K, open triangles and long-short-short dashed line; S, * and dash-dot line) for yellow dry mineral earth pigment.

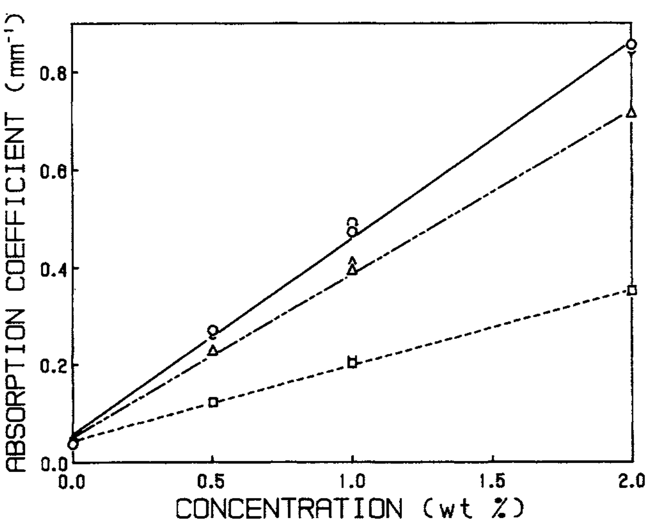

Fig. 6 — Scatter diagrams and regression lines for Kubelka-Munk absorption coefficients *vs*. concentration at 450 (open circles and solid line), 550 (open triangles and long-short-short dashed line), and 650 nm (open squares and dashed line) for red brown flockings.

ants studied, although specific exceptions are noted, *e.g.*, K of red and yellow Artskin and brown flocking (Table 1), S of titanium dioxide, dark buff, and yellow Artskin (Table 2). Further study of the size of any porosity in the unpigmented elastomer and the particle size of the colorants is recommended.

The high regression slope for titanium dioxide indicates the superior scattering ability of this colorant. Since a high scattering coefficient is related to high lightness and opacity, titanium dioxide, of all the colorants studied, had the greatest ability to lighten and opacify.

However, the limited general agreement of the observed color with that calculated by the Kubelka-Munk theory indicates that this theory may not always be used to predict the color of pigmented maxillofacial elastomer within a tolerance of even two perceivable color difference units. Despite limitations of this magnitude, the Kubelka-Munk theory has been

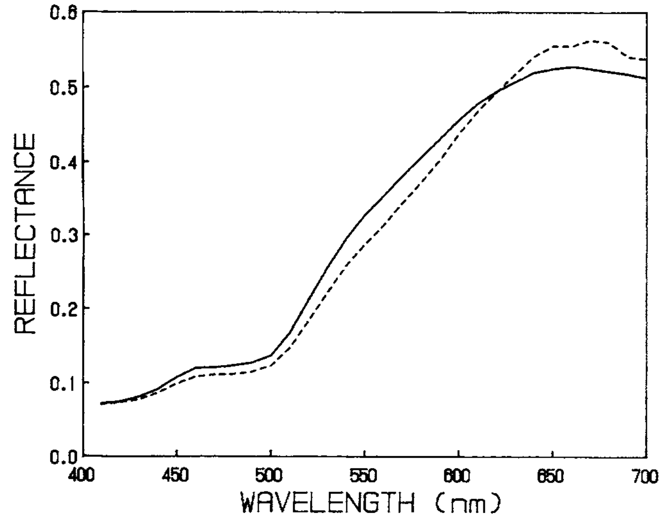

Fig. 7 — Spectra of calculated (dashed line) and observed (solid line) reflectivities for dark buff dry mineral earth pigment at 0.10 wt%.

TABLE 3
COLOR DIFFERENCE BETWEEN OBSERVED AND CALCULATED COLOR PARAMETERS AT INFINITE OPTICAL THICKNESS

Delta E(L*,a*,b*)	Pigment Identification
2.72	0.1% titanium dioxide
2.65	0.1% tin (IV) oxide
1.41	0.1% zinc oxide
5.31	0.1% kaolin
2.68	0.1% zinc stearate
3.02	0.1% talc
2.96	Average of six opacifiers
2.59	0.1% red Artskin
4.30	0.1% med brown Artskin
0.11	0.1% black Artskin
1.21	0.1% white Artskin
4.98	0.1% light brown Artskin
4.38	0.1% dark buff Artskin
4.74	0.1% blue Artskin
3.62	0.1% red brown Artskin
5.28	0.1% yellow Artskin
3.47	Average of nine mineral earth colorants
0.82	2% blue flocking
1.84	2% white flocking
3.05	2% tan flocking
1.09	2% brown flocking
1.16	2% red brown flocking
1.67	2% red flocking
1.60	Average of six flocking colorants

promoted as the basis for colorant formulation, since perfect formulations are impossible, and since considerable improvement can be obtained over trial-and-error methods (Johnston, 1973; Kuehni, 1975).

This study also indicates (Table 3) that, on the average, the color predicted by the Kubelka-Munk theory was closer to the observed color of a flocking than to that of a mineral earth pigment labeled with the same color. This greater agreement for the fibrous colorant may be due to the more complete elimination of collimated light within the mass of the fiber-filled elastomer, since collimated light is not directly accounted for in the Kubelka-Munk theory. The application of this theory seems based on the assumption that incident collimated illumination is totally and immediately transformed to diffuse light upon entering the turbid medium. The greater volume of the fibrous particles appears to permit most of the incident collimated light to be either scattered or absorbed.

TABLE 4
MEANS AND STATISTICAL ANALYSIS OF COLOR DIFFERENCES AT INFINITE OPTICAL THICKNESS FOR MINERAL EARTH AND FLOCKING COLORANTS WITH LIKE COLOR LABELING

Average Color Difference

	Colorant Type	
Labeled Color	Mineral Earth	Flocking
Blue	4.74	0.82
White	1.21	1.84
Red	2.59	1.67
Light brown or Tan	4.98	3.05
Medium brown	4.30	1.09
Red brown	3.62	1.16
Average	3.57	1.60

Analysis of Variance

Source of Variation	d.f.	M.S.	F-ratio	Probability
Pigment type	1	11.638	8.649	<0.05
Labeled color	5	1.385	1.029	>0.25
Residual (error)	5	1.346		

d.f. = degrees of freedom.
M.S. = mean square.

These results support a general lack of perfect agreement of the Kubelka-Munk theory with observed colors, but the relative ease of application of this two-flux reflectance theory is of great importance. Techniques are available for corrections based on a trial coloration and subsequent re-formulation which give negligible color differences between re-formulation and the target color (Kuehni, 1975). The general magnitude of the color differences between the theoretical and observed values supports the further study of this theory for mixtures of colorants within maxillofacial elastomer.

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