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Textile Research Journal 2010; 80; 263 originally published online Nov 20, 2009;

DOI: 10.1177/0040517508099914

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On the Kubelka–Munk Single-Constant/Two-Constant Theories

Abstract As the basic theory for color mixing in intensely light-scattering materials, the Kubelka–Munk single-constant/two-constant theories have played an important role in color science and technology. However, in practice there are some shortcomings. This paper checked the theories with a series of systematical experiments, data processing and analyzing, and has revealed several disagreements with the experimental evidence in the case of the additivity color-mixing law for turbid materials. Explanations on some of these problems are given, which are different from the conventional views.

Key words Kubelka–Munk theory, color-mixing law, single-constant, two-constant, new opinion

In 1931, Kubelka and Munk [1] proposed the first thorough mathematical treatment of a system simultaneously absorbing and scattering light, which led to what has since been known as the Kubelka–Munk (K-M) theory. Then in 1940, Duncan [2] made an assumption that the absorption coefficients and scattering coefficients for individual pigments in a mixture were additive, that is, in proportion to their respective concentrations. In 1942, Saunderson [3] adopted the same assumption in his analysis of color phenomena. This assumption has thus been used widely. Some call it “the Duncan formula” or “the Duncan’s additivity theorem” [4, 5], while others term it “the usual mixing law” [6, 7]. The K-M theory incorporated with the additivity assumption has gradually evolved into the so-called K-M single-constant/two-constant theories [8, 9], which have been playing an important role in color science and technology ever since [8–17].

However, the K-M single-constant/two-constant theories (K-M1C2C) are not without dispute in practical use. Some problems have long been reported and were generally regarded as restrictions of K-M theory [18–21]. This paper looks into the theory from a different view, and through systematical experiments, data processing and analyzing, several clear conflicts between the experiments and the K-M1C2C were revealed. Discussions of possible causes are provided.

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Theories

Kubelka–Munk Theory for Turbid Materials

For a detailed description of the K-M theory, one can refer to literature [1, 22–24]. Only the equations from the K-M theories used in this paper are listed below. According to the K-M theory, the relation of the absorption and scattering of incident light of a turbid material can be expressed as

$$\frac{K}{S} = \frac{(1 - R_\infty)^2}{2R_\infty} \quad (1)$$

and

$$\begin{aligned} K &= S(a - 1) \\ S &= \frac{1}{Xb} \ln \frac{1 - R/(a + b)}{T} \end{aligned} \quad (2)$$

where

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$$a = \frac{1 + R^2 - T^2}{2R}$$

$$b = \sqrt{a^2 - 1}$$

K is the K-M absorption coefficient (cm^{-1}) and S the K-M scattering coefficient (cm^{-1}); R_∞ is the reflectance at infinite thickness; T is the transmittance; and R is the reflectance with a perfectly black backing, referred to as R_0 in Kubelka [22]; X is the thickness of the turbid material.

Mixing Law for Colorants in Turbid Media

In order to predict the color of a mixture of pigments in turbid materials, it was assumed that the absorption coefficients and scattering coefficients for the individual pigments were additive, that is, in proportion to their respective concentrations [3, 4], that is

$$K = K_0 + C_1K_1' + C_2K_2' + \dots + C_nK_n'$$

$$S = S_0 + C_1S_1' + C_2S_2' + \dots + C_nS_n' \quad (3)$$

where K and S are the mixture's absorption and scattering coefficients; K_i' and S_i' are each colorant's unit absorptivity and scattering; C_i is the concentration of each colorant; K_0 and S_0 are the values for the substrate, added by Allen in 1980 [25] and since adopted.

K-M Single-constant/Two-constant Theory

If the assumption of additivity is valid, then

$$\frac{K}{S} = \frac{K_0 + C_1K_1' + C_2K_2' + \dots + C_nK_n'}{S_0 + C_1S_1' + C_2S_2' + \dots + C_nS_n'} = \frac{(1 - R_\infty)^2}{2R_\infty} \quad (4)$$

Thus, the K-M theory and Equation (4) build a bridge between the reflectance (R_∞) of a colored turbid material and the concentrations (C_i) of the colorants using the two constants, K_i' and S_i' of each colorant, hence the name K-M two-constant theory.

Further, researchers argued [8, 9] that in many of the color-matching problems, most of the scatter is introduced either by a white pigment, or by the substrate itself, that is, in the case of textiles, papers. In other words, the value of S_0 is dominantly large in comparison with the total scatter provided by the colorants, as described in Equation (5). In this case, Equation (4) can be reduced to Equation (6):

$$S = S_0 + C_1S_1' + C_2S_2' + \dots + C_nS_n' \approx S_0 \quad (5)$$

$$\frac{K}{S} = \frac{K_0 + C_1K_1' + C_2K_2' + \dots + C_nK_n'}{S}$$

$$= \left(\frac{K_0}{S}\right) + C_1\left(\frac{K_1'}{S}\right) + C_2\left(\frac{K_2'}{S}\right) + \dots + C_n\left(\frac{K_n'}{S}\right) \quad (6)$$

$$= \left(\frac{K}{S}\right)_0 + C_1\left(\frac{K}{S}\right)_1 + C_2\left(\frac{K}{S}\right)_2 + \dots + C_n\left(\frac{K}{S}\right)_n$$

$$= \frac{(1 - R_\infty)^2}{2R_\infty}$$

The K-M theory in Equation (6) is called the K-M single-constant theory, for only one constant, $(K/S)_i'$, is needed to relate R_∞ to C_i .

Obviously, since the basis for Equations (4) and (6) is Equation (3), the additivity assumption, our present research is to examine the validity of the assumption.

Experimental

Materials

CIBACRON C (Ciba Specialty Chemicals) reactive dye-stuffs were used in this study as the colorants, including Red C-2G, Yellow C-RG, and Blue C-R. Yet only the data gained from the Red C-2G are given below, for the other dyes showed similar results. Both sodium carbonate and sodium chloride were used as dyeing auxiliaries. A dull-white twill of intensely light-scattering fabric was chosen as the turbid substrate. The fabric was 0.50 mm thick with scattering transmittance/total transmittance over 99%, to which the K-M theory applies [23].

Dyeing

The white fabric was dyed by the dip-pad-steaming process shown in Figure 1 with the dyeing formula listed in Table 1 [26] and dye concentrations in Table 3.

Measurement

A Cary 500 integrating sphere spectrophotometer from Varian Technologies (Palo Alto, CA) was used to measure

Table 1 Dye formula for CIBACRON C.

Dye and auxiliary	Dosage ($\text{g} \cdot \text{liter}^{-1}$)		
CIBACRON C	< 10	10–30	> 30
Sodium carbonate	10	20	30
Sodium chloride	60	90	90

Table 2 Concentration of CIBACRON Red C-2G in standard solution

Sample	0#	1#	2#	3#	4#	5#	6#	7#
Concentration (%)	0	0.0217	0.0357	0.0706	0.1516	0.3035	0.4544	0.6060

Fabric—dipping in dye bath (at room temperature)—padding (at room temperature, with about 75% liquor retention)—steaming (at 102–104°C, for about 2.5 min)—cool water washing (bath ratio 25:1, twice)—hot washing (80°C, bath ratio 25:1, twice)—soap boiling (soap 3g/L, 93–95°C, bath ratio 25:1, about 15min)—hot washing—cool washing—drying

Figure 1 The dyeing process flow chart.

the transmittance T spectra, reflectance R spectra (one-layer fabric applied) and the infinite thickness reflectance R_∞ spectra (eight-layer fabrics applied) in the region of 280–800 nm with an 1-nm interval at room temperature.

The concentrations of dye on fabrics were obtained by the acid dissolution method [27]. The white fabric (0.5 g) was weighed, cut into fine pieces and dehydrated until the weight stabilized. Then, 0.1 g of the fabric pieces were weighed and totally dissolved with 10 ml 75% sulfuric acid in a beaker for about 1.5 hour. Next, the solution was diluted with 40 ml distilled water and poured into a 100 ml flask containing a dyestuff with known concentration. More distilled water was added and shaken to uniformity for testing. A total of eight flasks of such standard dye solutions with different dye concentrations were prepared as listed in Table 2. Taking the 0# solution (with zero dye) as the reference sample, the absorbance curves of the other seven samples were measured using a Cary 500 spectrophotometer. Its photometric accuracy was ± 0.003 at 1.0 Abs checked with wave filter NIST930, wavelength accuracy was ± 0.1 nm and wavelength setting reproducibility was less than 0.025 nm offset in ultraviolet and visual range.

Then, the dyed fabric solutions were prepared using a similar method as described above, but the white fabric was replaced with dyed fabrics R1 to R10 (see Table 3) and no more dye was added. Also taking the new 0# sample as the reference, the absorbance curves of the sample dissolved with dyed fabric R1 to R10 was measured accordingly using the Cary 500 spectrophotometer.

Results

Three CIBACRON C reactive dyestuffs, Red C-2G, Yellow C-RG and Blue C-R showed similar results. Thus in

Table 3 Concentration of CIBACRON Red C-2G in a dye bath and on fabric

Sample	Concentration of dye (weight percentage, %)	
	In dye bath (C_d)	On fabric (C_f)
R0	0	0
R1	0.02	0.17
R2	0.04	0.26
R3	0.08	0.28
R4	0.16	0.37
R5	0.30	0.55
R6	0.50	0.77
R7	1.00	1.02
R8	2.00	1.62
R9	4.00	2.89
R10	6.00	4.10

this paper, only the data gained from Red C-2G are given below. One can refer to the authors' other paper [28] for the data from Yellow C-RG.

Theoretical Concentration and Effective Concentration

The dyes on fabrics were calculated according to the Bouguer-Lambert-Beer law. Based on the data from standard solutions, the working curve was obtained by the least square method, as $A = 0.02374C$ (correlation coefficient $r = 0.9997$), where C is the dye concentration in Table 2 and A is the dye absorbency at absorption peak (509 nm). Then the concentrations C_f of the dyes on fabrics were solved by substituting their absorbency A values at absorption peak (509 nm) into $A = 0.02374C$. The results, together with the given concentration in the dye bath C_d , are shown in Table 3.

Analyzing the relationship of dye concentration in the dye bath (theoretical concentration, C_d) and the dye concentration on fabric (effective concentration, C_f) in this experiment by the least squares method, the two are shown to be highly correlated, with the correlation coefficient r being 0.9978 as seen in Figure 2. Therefore, according to common practice, the dye bath concentration instead of the dye concentration on fabric is used in our discussion.

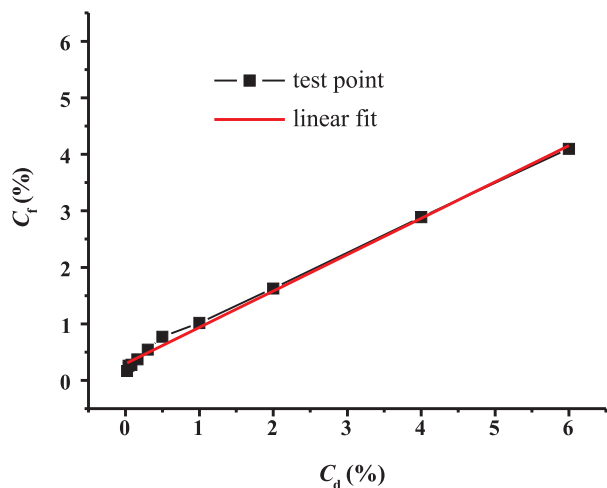


Figure 2 Relationship between concentrations of dyes in dye bath C_d and on fabric C_f .

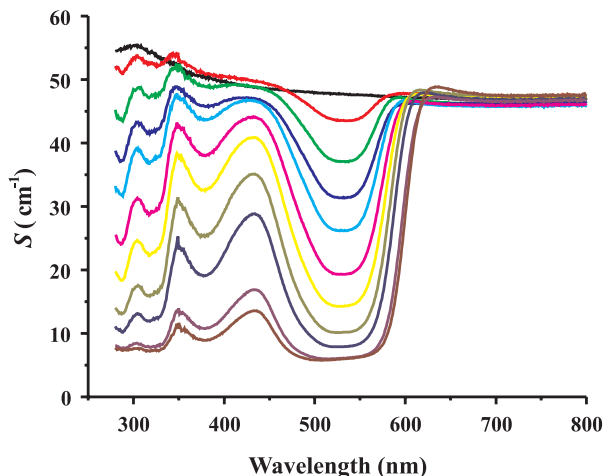


Figure 4 Comparison of scattering coefficient (S) curves for fabrics dyed with different concentrations of CIBACRON Red C-2G (fabric R0 top to R10 bottom).

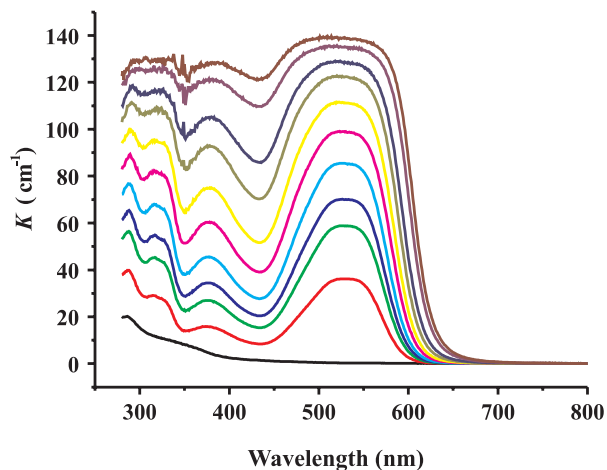


Figure 3 Comparison of absorption coefficient (K) curves for fabrics dyed with different concentrations of CIBACRON Red C-2G (fabric R10 top to R0 bottom).

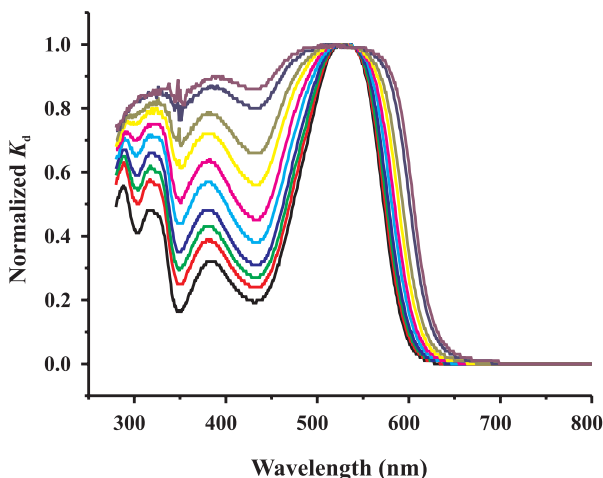


Figure 5 Comparison of normalized absorption coefficient (K_d) curves for fabrics dyed with different concentrations of CIBACRON Red C-2G (fabric R10 top to R1 bottom).

Scalability of K , S and K/S

The K_0 and S_0 values for the undyed fabric (R0) and the K and S values for the dyed fabrics (R1–R10) were calculated by Equation (2) using measured transmittance T and reflectance R values. The results are plotted in Figures 3 and 4. The K values of the dyed fabrics increase with the dye content C_d . For easy comparison, K_d values were calculated by subtracting K_0 from K , then normalized by their respective

maximal absorption peak values and the normalized K_d values were re-plotted in Figure 5.

It is clear that the normalized spectral curves of K_d are different and their absorption peaks broaden with the increasing C_d . Therefore, K_d (or K) is not scalable [15], that is, the dye contents influence not only the size or amplitude but also the shape of the curves. By the same reasoning, the scattering coefficient S is non-scalable as well.

Table 4 Relationship between S , K_d , $(K/S)_d$ and concentration C_d of CIBACRON Red C-2G

Item	Fitted equation	Correlation coefficient r
K_d - C_d	$K_d = 176.65 - 57.42 C_d^{-0.2328}$	0.9979
S - C_d	$S = 11.029 C_d^{-0.3857}$	0.9921
$(K/S)_d$ - C_d	$(K/S)_d = 13.364 C_d + 0.8093$	0.9965
	$(K/S)_d = 6.2528 \ln C_d + 11.697$	0.9928

The $(K/S)_0$ for the white fabrics (R0) and the K/S for the dyed fabrics were calculated by Equation (1) using the measured R_∞ . The $(K/S)_d$ values, calculated by subtracting $(K/S)_0$ from the corresponding K/S values, rise with the increasing C_d , as depicted in Figure 6. The $(K/S)_d$ values were then normalized by their respective maximal absorption peak values and the normalized $(K/S)_d$ values were re-plotted in Figure 7. It is seen that the normalized $(K/S)_d$ spectral curves can be treated as identical at low dye concentrations (below 2% for the dye C-2G; the value changes for the other dyes), that is, it can be assumed scalable at low concentration. Whereas, when the C_d rises to 4% and 6%, the peak widths broaden and the scalability is no longer valid.

Relationship Between K and S for Dyed Fabrics

The K and S of dyed fabrics are negatively correlated, as shown in Figures 3 and 4. Take the K values and S values at 525 nm absorption peak to compute their relation. The least squares method is used to fit the data and the linear correlation coefficient r is -0.9977 as seen in Figure 8.

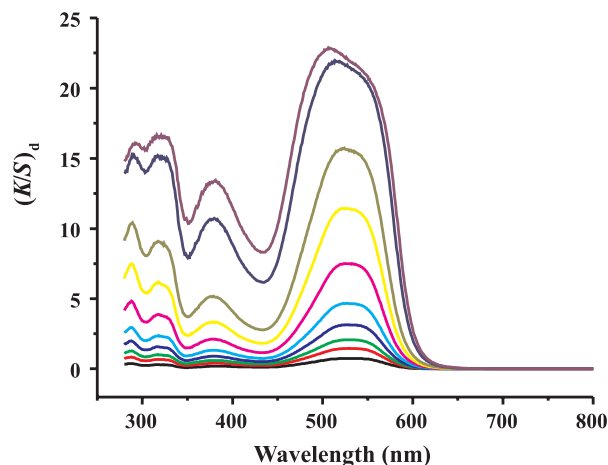
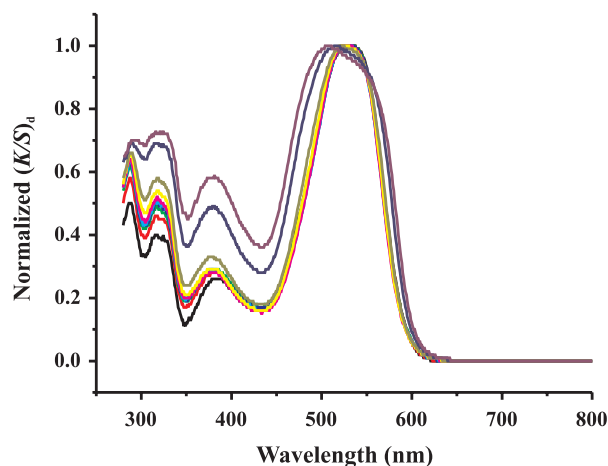
Relationships Between K , S , K/S and Concentration

Further analysis of the relations between the S values for the dyed fabric, K_d and $(K/S)_d$ values for dyes on fabrics at 525 nm with the dye concentration C_d , yields several results provided in Table 4 and Figures 9–11.

Discussion

Correlation between K and S

The K and S values of dyed fabric are highly but negatively correlated in colorants' absorption bands, that is, when dye concentration rises, K ascends whereas S descends as shown in Figures 3, 4, 8, 9, and 10. The explanation is as follows. The white fabric already has many scattering points (the sites which arouse light scattering). After being


Figure 6 Comparison of $(K/S)_d$ curves for fabrics dyed with different concentrations of CIBACRON Red C-2G (fabric R10 top to R1 bottom).

Figure 7 Comparison of normalized $(K/S)_d$ curves for fabrics dyed with different concentrations of CIBACRON Red C-2G (fabric R10 top to R1 bottom).

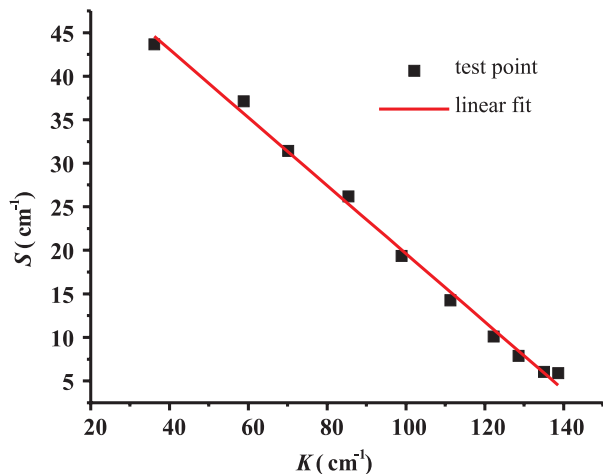


Figure 8 Relationship between K and S values of fabrics dyed with different concentrations of CIBACRON Red C-2G at a wavelength of 525 nm.

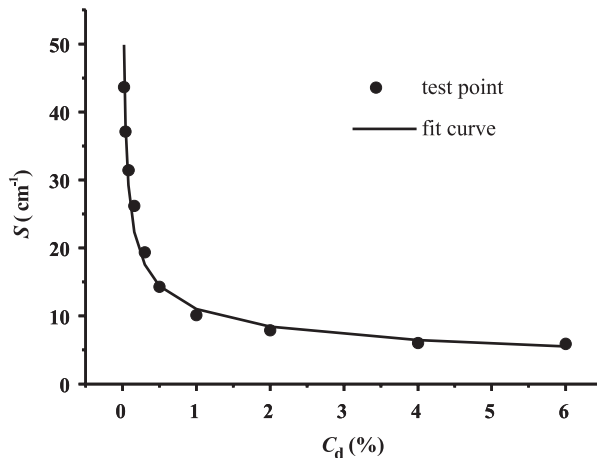


Figure 10 Relationship between S and C_d values of fabrics dyed with different concentrations of CIBACRON Red C-2G at a wavelength of 525 nm.

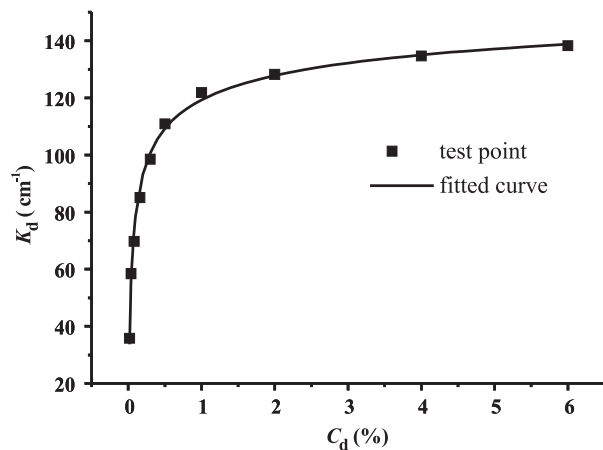


Figure 9 Relationship between K_d and C_d values of fabrics dyed with different concentrations of CIBACRON Red C-2G at a wavelength of 525 nm.

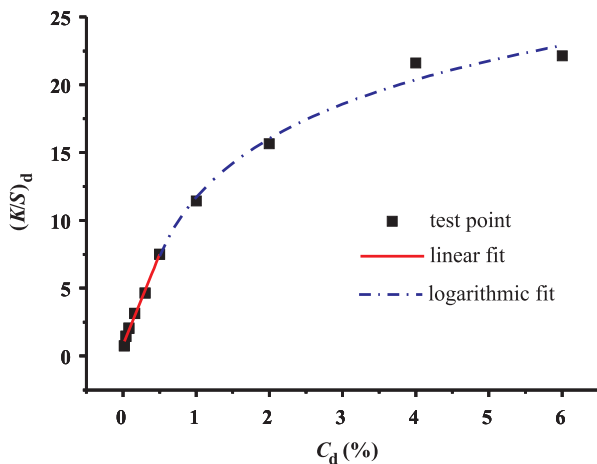


Figure 11 Relationship between $(K/S)_d$ and C_d values of fabrics dyed with different concentrations of CIBACRON Red C-2G at a wavelength of 525 nm.

dyed, many absorbing points corresponding to the absorption bands are added to the fabric. In most cases, the refractive indexes of dyes are close to those of fabric, so the scattering caused by dyes is small and can be ignored. As a result, with increasing dye concentration, the distances between the scattering points and absorbing points on the dyed fabric are below the limit of incoherence [29], that is, scattering and absorbing on dyed fabric become coherent. Hence, with the increase of C_d , K increases while S decreases.

In fact, the inverse correlation between K and S also exists in other colored turbid materials. The data (Table 5) extracted from Shah and Billmeyer's study [7] on the mixtures of pigments are just such examples. There is a reported interaction between K and S on papers [30]. Obviously, the mixing law, Equation (3), does not take into consideration the inverse correlation between K and S . The ever increasing of S with dye concentration indicated in Equation (3) does not fit well with the experimental results

Table 5 Correlation of K and S values of pigment mixtures (from Shah and Billmeyer [7])

Content of Saturn Yellow pigment (%)	0	0.346	0.517	0.688	0.854
K at 460 nm (cm^{-1})	1.1998	29.2279	42.9342	54.1214	74.6716
S at 460 nm (cm^{-1})	68.5498	59.6449	55.3315	51.9185	52.9752

shown in Figures 4 and 10 and Table 5. Consequently, assumption (5) is also incorrect.

Unscalability of K_d and S and their Nonlinear Relations with C_d

The spectra curves of normalized K_d broaden with C_d (Figure 5) so that K_d is not scalable or is non-linear to C_d . The scattering is hence unscalable and is non-linear to C_d too, due to its high correlation with the absorption. Thus Equation (3) is invalid.

Scalability of $(K/S)_d$ and its Relationship with C_d

At low dye concentration, $(K/S)_d$ can be approximated as scalable and linear to C_d as shown in Figures 7 and 11, that is because the broadened amounts in the K and S spectra would cancel each other. However, when C_d exceeds a certain value, this is no longer the case because the broadened values in K spectra are much greater than those in the S spectra, causing the in-scalability of $(K/S)_d$.

Further Remarks

If Equation (3) were valid, for mixtures of different colorants, one could establish enough simultaneous equations (no less than for the unknown parameters), through which the unit absorptivity K' and unit scattering S' for each colorant can be determined. However, this approach has long been recognized to be infeasible [15] because the calculated unit absorptivity K' and unit scattering S' of a colorant changes in different colorant mixtures. A recommendation has been made that K and S only appear as a ratio, that is, Equation (4) rather than (3) should be used, even though there are still problems in using Equation (4) to solve unknown parameters.

For example, the primary binary mixtures method is used to solve the K' and S' values for pigments. Walowitz et al. [18] stated that this method did not always yield reliable predictions, and frequently the K' and S' values produced using different binary mixtures did not agree very well with the predictions. The validity of this method further diminishes when several colorants were used in a mixture at high concentrations. Marcus and Welker's [19] study directly showed the influences of pigment volume concentration (PVC) on the K' and S' values for the pigments, for exam-

ple, S' of titanium dioxide at 600 nm decreased by 40.4% as the PVC increased from 4.30% to 19.94%. All these are in conflict with Equation (4), which regards K' and S' as colorant's constants.

For fabric dyeing, the K-M single-constant theory is usually used. According to Equation (6), $(K/S)_d$ should be linear to C . However, as shown in Figure 11 and known to all in the field, the relationship is inherently non-linear. Conventionally there are two explanations for this phenomenon in the textile industry. One attributes the behavior to the fact that the rate of dye uptake decreases as the fabric approaches the maximum amount of dye uptake, resulting in the non-linearity. In other words, as the dye bath concentration continues to increase, both the rate and amount of dye absorbed by the fabric, that is, the effective concentration, level off. However, our experimental results show that even in the range where dye uptake is proportional to dye bath concentration as exhibited in Figure 2, non-linearity still occurs in Figure 11. The other explanation attributes the phenomenon to the neglect of the surface reflection in the K-M model. In this study, the K-M K , S and K/S , with and without surface reflection correction, have been calculated and analyzed in parallel. Surface reflection correction has shown little effect on the results. Hence, surface reflection is conventionally ignored in practical color matching for fabric. The figures with surface correction are not presented in this paper. We will discuss the influence of surface reflection correction on the K-M parameters in a future paper.

Then, why does the single-constant K-M theory seem to be valid for fabric color modeling where the dye concentration is always low? Now, after data analysis in this paper, it is easily understood that the original explanation that scatter by colorants is negligible in comparison to that of the substrate is too simplistic, and neglects the internal connection between K and S . The reason that (K/S) is scalable [15] at low concentration and appears linear to C is that the errors in Equations (3) and (5) cancel each other in the ratio (K/S) . It's interesting that based on invalid assumptions in Equations (3) and (5), a more feasible Equation (6) was derived, albeit at low dye concentration.

As for the K-M single-constant/two-constant theory, because of the great works by those who developed the many practical methods to get more accurate values for K' and S' , the theory still has a role to play in the color field.

Conclusions

Taking dyed fabric as an example of colored turbid material, the K-M light absorption coefficients K , scattering coefficients S , and their ratio K/S , were investigated systematically through experiments. The results question the rationality of the additive color-mixing law for turbid materials from three aspects. First, both the K-M absorption coefficient K and scattering coefficient S are not scalable and non-linear to dye concentration. Secondly, the absorption coefficient K and the scattering coefficient S are demonstrated to be highly yet negatively correlated, a fact ignored when establishing the theories. Thirdly, in the absorption band, the scattering coefficients decrease with increasing dye concentration, questioning the linearity assumed.

This paper sees the K-M single-constant two-constant theory from a different point of view. Many problems it encounters in practical use are due to the irrationality of the additivity assumption, rather than the K-M turbid medium theory itself. Some explanations are given, which are different from the conventional ones, especially about why the single-constant K-M theory seems applicable to the fabric coloring process.

Acknowledgements

Hongying Yang wishes to acknowledge the support of the China Postdoctoral Science Foundation. Thanks also go to Ms Yulan Zhang and Ms Wei Zhang at the East China University for Science and Technology for allowing us to test and check our ideas on their Cary 500 spectrophotometer, and to Ciba Specialty Chemicals for providing the dye-stuffs and technique support.

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