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Quantitative opacity measurements on tablet film coatings containing titanium dioxide

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Summary

The opacity of tablet film coatings consisting of hydroxypropyl methylcellulose containing titanium dioxide has been analyzed quantitatively with particular emphasis on the theoretical and practical significance of the various methods for evaluating the relative opacity of different film formulations. By taking reflectance measurements with both black and white backing tiles at various film thicknesses it is possible, by the application of the Fell relationship, to estimate the approximate conditions (e.g. pigment concentration and film thickness) required to produce opaque films. However, if a quantitative assessment of opacity is required, the application of the Kubelka-Munk equation provides data not only of a more fundamental nature but also more precise. The scattering coefficients calculated for the various film coating formulations compare favourably with those for the same pigment in paint formulations.

Introduction

The opacity of tablet film coatings is of a particular importance in the case of drug substances which are either light-sensitive or exhibit interbatch colour variation and which are normally administered in a tablet formulation. The opacity of a film coating can be assessed relatively easily by means of a contrast ratio defined as the ratio of the measured reflectance of the film placed on a black substrate to the

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measured reflectance of the film placed on a white substrate (Rowe, 1984). However, although the results are consistent with the known theories of light scattering and absorption, comparisons between different film formulations can only be made at constant film thickness— an important variable in the development of film opacity. In this report the effect of film thickness on the opacity of hydroxypropyl methylcellulose films containing titanium dioxide is quantitatively analyzed with particular emphasis on the theoretical and practical significance of the various methods for evaluating the relative opacity of different film formulations.

Materials and Methods

All reflectance measurements were made on films removed from the entire face of a 11.1 mm diameter flat-faced tablet (care being taken to remove any adhering substrate) using a Hunterlab 4-filter, tristimulus colorimeter (Model D25A, Hunter Associates Laboratory, VA, U.S.A.) fitted with a 6.5 mm diameter viewing aperture. Contrast ratios were determined by dividing the reflectance value with the black backing tile (R_B) by the reflectance value with a white backing tile (R_w) and expressing the result as a percentage. The Y tristimulus values of the black and white backing tiles were kept constant at 0.02% and 85.0%, respectively. Ten measurements were made on each formulation at each film thickness and the means and standard deviations calculated. Film thicknesses were measured using a micrometer.

Film coating was carried out in a 24-inch Accela-Cota (Manesty Machines, Liverpool) using an airborne spray system at an application rate of 50 $ml \cdot min^{-1}$ and air inlet temperature of 60 °C. The film formulation consisted of a 5% w/v solution of hydroxypropyl methylcellulose (Pharmacoat 606, Shin-Etsu Chemicals, Japan) containing polyethylene glycol 300 (20% w/w based on polymer) as plasticizer and anatase titanium dioxide (9.5%, 17.3% and 29.5% on a dry weight basis) as pigment.

Results and Discussion

The effect of film thickness on the measured contrast ratios of the 3 film formulations is shown in Fig. 1. In all cases the contrast ratio increases with film thickness although the relationship is exponential rather than linear. It is this exponential relationship that can be exploited in the Fell relationship to provide information on the ability of a formulation to produce an opaque film.

The Fell relationship

The Fell relationship, first introduced and recommended by Sawyer (1941), states that a plot of the logarithm of the contrast ratio against the reciprocal of the film thickness yields a straight line and that this straight line may be extrapolated to determine the film thickness necessary for 'complete hiding' defined in the paint industry as a film having a contrast ratio of 98%. While this definition of complete hiding appears to be a good compromise in the case of paint films applied to conceal the undersurface, such a definition may not be satisfactory in the case of tablet film coatings applied to protect the drug in the tablet core from light degradation. Therefore, in applying the Fell relationship to tablet film coating formulations, the extrapolation has to be extended to a contrast ratio of 100%.

Applying these concepts to the 3 film coating formulations in this work yields the results shown in Table 1. It can be seen that in all cases there is a good correlation between the logarithm of the contrast ratio and the reciprocal of the film thickness ($P < 0.001$). Extrapolation of the line to predict film thicknesses at contrast ratios of 98 and 99% appears satisfactory but there is an anomalous result at a contrast ratio of 100% for the film containing the highest concentration of titanium dioxide. This may well be due to the inaccuracies in the measurement technique being exaggerated on extrapolation to this extreme, especially since this formulation had the shallowest

TABLE 1

RESULTS ON APPLYING THE FELL RELATIONSHIP TO THE 3 COATING FORMULATIONS

	Titanium dioxide concentration % w/w		
	9.5	17.3	29.5
Correlation coefficient	0.9905	0.9950	0.9985
Film thickness (μm) for contrast ratios:			
98%	252	122	92
99%	342	151	131
100%	526	198	227

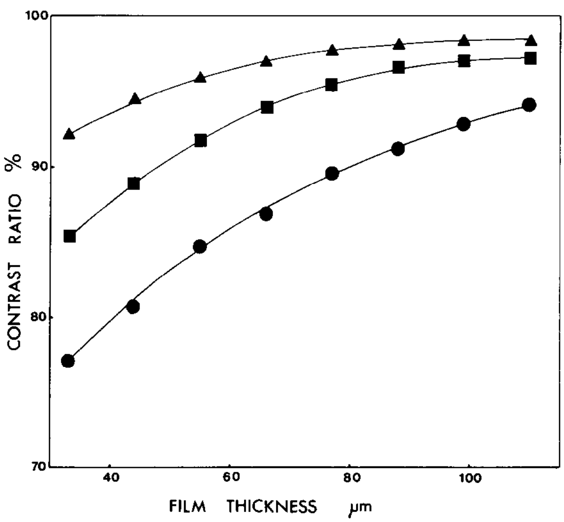

Fig. 1. The effect of film thickness and pigment concentration on the contrast ratios of hydroxypropyl methylcellulose films containing titanium dioxide: ●, 9.5% w/w; ■, 17.3% w/w; ▲, 29.5% w/w.

gradient. Such inaccuracies have been discussed in detail by Switzer (1955) who recommended the use of defined ranges of both the contrast ratio and film thickness. Mitton (1973) has since recommended that all values for contrast ratios of below 83% be disregarded in the analysis.

It can be seen, therefore, that, although the Fell relationship is relatively simple and easy to apply and has an advantage in that it can be used to compare film formulations at a constant contrast ratio rather than at a constant thickness as used previously (Rowe, 1984), it lacks precision. A further objection to the method is that it does not take into account the reflectivity (R_∞) of the film defined as the reflectance of a film so thick that any increase in thickness has no further effect. This is an important fundamental property of a film as is fully recognized in the Kubelka-Munk theory.

Kubelka-Munk theory

In contrast to the well-known Mie theory (Mie, 1908) which can be used to analyze the scattering and absorption event for each single spherical particle within a film well separated from all other particles, the Kubelka-Munk theory (Kubelka and Munk, 1931) can be used to analyze the reflectance of light at a differential layer within a film without any specification of the size, shape or separation of particles within that layer. In the derivation of their theory Kubelka and Munk assumed diffuse light within a film which is infinite (optically) in area, of finite thickness and optically fixed to the substrate. Two constants are needed to explain the optical characterization of the film, viz. S, the scattering coefficient, a measure of the light scattered by the film and K, the absorption coefficient, a measure of the light absorbed. S, K and R_∞ (expressed as a fraction) are related by the simple equation:

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

In the context of opacity measurement the equation has since been extended (Kubelka, 1948):

$$Sx = \frac{1}{b} \coth^{-1}\left(\frac{1 - aR_B}{bR_B} \right) \tag{2}$$

where

$$a = 1 + \frac{K}{S} = \tfrac{1}{2}\left(\frac{1}{R_\infty} + R_\infty \right) \tag{3}$$

and

$$b = \frac{1}{S}(2KS + K^2)^{1/2} = \tfrac{1}{2}\left(\frac{1}{R_\infty} - R_\infty \right) \tag{4}$$

In these equations x is the thickness of the film and R_B the measured reflectance (expressed as a fraction) of the film with a black substrate. The product of the scattering coefficient, S, and the film thickness, x, is usually referred to as the scattering power of a film.

Eqn. 2 can be solved by either computation or by the use of published tables and graphs (Judd and Wyszecki, 1963; Mitton and Jacobsen, 1963; Mitton, 1970). However, in the case of white opaque films as used on this work, where $R_\infty \rightarrow 100\%$ and $S_x < \infty$, Eqn. 2 can be simplified to:

$$S_x = \frac{R_B}{1 - R_B} \tag{6}$$

Hence, it can be seen that it should be possible to calculate both the scattering power and scattering coefficient of a film coating formulation simply from the measurement of its reflectance with a black substrate. Data on the film coating formulation containing 9.5% w/w titanium dioxide (Table 2) show very good agreement between the scattering powers and scattering coefficients calculated using

TABLE 2

DATA ON THE FILM COATING FORMULATION CONTAINING 9.5% w/w TITANIUM DOXIDE

Film thickness (μm)	R_B (%)	Scattering * power	Scattering * coefficient (mm^{-1})	Scattering ** power	Scattering ** coefficient (mm^{-1})
33	67.73	2.10	63.64	2.10	63.64
44	72.02	2.57	58.41	2.75	62.50
55	76.27	3.21	58.36	3.25	59.09
66	78.40	3.63	55.00	3.75	56.82
77	81.43	4.39	57.01	4.60	59.74
88	83.30	5.15	58.52	5.40	61.36
99	84.63	5.51	55.66	5.66	57.72
110	86.58	6.45	58.63	6.58	59.82

* Data calculated using Eqn. 6

** Data calculated using Eqn. 2 where $R_\infty = 95\%$.

TABLE 3

SCATTERING COEFFICIENTS FOR THE 3 FILM COATING FORMULATIONS

Titanium dioxide concentration		Scattering coefficients (mm^{-1})	
% w/w	% v/v	Calculated from Eqn. 2	Calculated from Eqn. 6
9.5	3.4	60.09 ± 2.32	58.15 ± 2.62
17.3	6.5	106.65 ± 4.74	103.91 ± 4.91
29.5	12.2	173.48 ± 7.31	168.07 ± 6.48

Eqn. 6 and those using the more general Eqn. 2 as extrapolated from the graphs and tables given by Mitton and Jacobsen (1963) and Mitton (1970). An advantage of this method is that, by using the data from each film thickness, it is possible to calculate the means and standard deviations of the scattering coefficients for each film coating formulation (Table 3). These data are comparable with that reported for anatase titanium dioxide dispersed in acrylic lacquer films (Mitton, 1973)—Fig. 2. This is to be expected because, although the S value in the Kubelka-Munk theory applies to the total system, in practice polymeric film formers contribute little or no scattering to the system and therefore the S value may be considered as due entirely to the pigment alone.

The scattering coefficient as defined above, although being due entirely to the pigment, makes no allowance for the number of pigment particles present within the film and gives no indication of the scattering efficiency of individual pigment particles. This criticism can be countered by calculating the scattering coefficient not in terms of reciprocal length but in terms of an area per unit weight of pigment. This can be done by multiplying the scattering power of a film coating by the area covered per unit weight of the dry film and dividing by the weight fraction of the pigment. Calculations on the 3 film coating formulations containing 9.5%, 17.3% and 29.5% w/w titanium dioxide yield values of 593.3 ± 26.7, 527.9 ± 23.7 and 430.1 ± 16.8 $mm^2 \cdot mg^{-1}$, respectively. These results are again comparable with those reported in the paint literature for anatase titanium dioxide (Mitton, 1973). It is interesting to note that the trend in these values is the reverse of that shown in Table 3, i.e. each pigment particle in the films containing the higher concentrations of

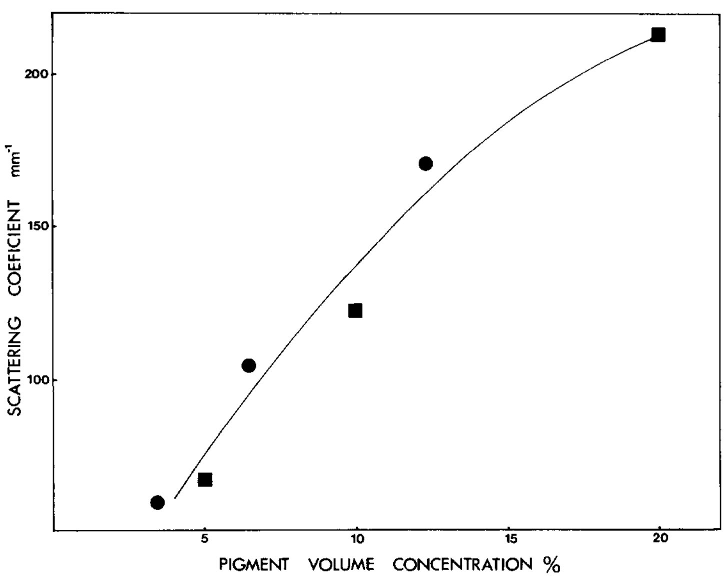

Fig. 2. A comparison of the scattering coefficients for anatase titanium dioxide dispersed in tablet film coatings (this study, ●) and acrylic lacquer paint films (data ex. Mitton (1973), ■).

titanium dioxide no longer scatters light as efficiently as that in the film containing the lowest concentration of titanium dioxide. This is due to a crowding or shadowing effect due to the closeness of packing of the pigment particles. It is known in the paint literature (Mitton, 1973) that such an effect starts at pigment volume concentrations as low as 10% v/v.

Conclusion

The Fell relationship is of use as a simple and practical method for estimating the approximate conditions, e.g. pigment concentration and film thickness, required to produce an opaque film. However, if a quantitative assessment of film coating opacity is required, the application of the Kubelka-Munk equations provides data not only of a more fundamental nature but also superior in reliability due to the ability to use measurements without range restriction and the ability to obtain data from measurements at each film thickness and hence provide a statistical estimate on the precision of the values determined for the scattering coefficient. The data produced from the anatase titanium dioxide used in tablet film coatings compare favourably with that for the same pigment in paint formulations.

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