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The opacity of tablet film coatings

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The opacity of tablet film coatings containing a variety of pigments and fillers has been assessed using a contrast ratio defined as the ratio of the measured reflectance of the incident light when the film is placed on a black substrate to the measured reflectance of the incident light when the film is placed on a white substrate. Films pigmented with calcium carbonate, calcium sulphate or talc had very low contrast ratios and only the inclusion of titanium dioxide imparted opacity. Films pigmented with the coloured lake pigments had contrast ratios dependent on both the parent dye and the dye concentration with the contrast ratios decreasing blue > red > orange > yellow. Films pigmented with the synthetic iron oxides all had very high contrast ratios. The results are consistent with the known theories of light scattering and absorption and illustrate the potential of this accurate, rapid and simple technique in the optimization of film formulations during product development.

A clear film, containing no colourant, coated onto a tablet core allows light to be reflected from the core surface with little or no distortion. Hence any variations in the surface due to colour variation of the formulation, or degradation of light sensitive materials, can be easily seen. As pigment is added to the film, the core becomes partially obscured and the film is said to achieve a certain amount of opacity. Eventually, depending on the pigment, its concentration and on the film thickness, the core becomes totally obliterated and the film is said to be totally opaque. In the paint industry the degree of opacity of a film is assessed by means of a contrast ratio defined as the ratio of the measured reflectance of the incident light when the film is placed on a black substrate to the measured reflectance of the incident light when the film is placed on a white substrate (Mitton 1973). Since the opacity of tablet film coatings is of particular interest in the case of drug substances which are light sensitive or which exhibit interbatch colour variation, it was decided to evaluate the potential of the contrast ratio for use in measuring the opacity of tablet film coatings.

MATERIALS AND METHODS

The opacity of the various coating formulations were determined by first removing the films from the entire face of a 11·1 mm diameter flat-faced tablet, then carefully removing any adhering substrate, and finally measuring the reflectance of the films with both black and white backing tiles using a Hunterlab, four filter, tristimulus colorimeter (Model D25A, Hunter Associates Laboratory Inc., Virginia, USA) fitted with a 6·5 mm diam. viewing aperture. The contrast ratio was determined by dividing the measured reflectance with the black backing tile by the measured reflectance with the white backing tile and expressing the result as a percentage—in this case a zero reading represents complete transparency and a reading of 100 total opacity. Since the thickness of the film and the specification of the two backing tiles are variable and must be taken into account, both were kept constant during any one experiment, the reflectance values (i.e. the Y tristimulus values) of the black and white backing tiles being 0·02 and 85% respectively. The results reported are the means and standard deviations calculated from ten measurements.

Three variables were studied in three separate experiments, firstly the effect of pigment and filler type; secondly the effect of pigment concentration using titanium dioxide and FD&C Yellow 5 lake as representative pigments, and thirdly the effect of particle size of an inert filler using a white dolomite as a model pigment (Microdol, A/S Norwegian Talc, Norway—see Rowe 1981). All the tablets used were coated with a film formulation consisting of either a mixture of four parts of hydroxypropyl methylcellulose (Pharmacoat 606, Shin-Etsu Chemical Co., Japan) and one part of ethylcellulose (Grade N7, Hercules Inc., USA), or hydroxypropyl methylcellulose alone. Both formulations contained glycerol (20% w/w based on polymer) as plasticizer, and were applied dissolved in a dichloromethane–methanol (70:30% v/v) solvent mixture using either a 6 inch diameter Wurster column or 24 inch diameter AccelaCota (Manesty Machines Ltd. Liverpool). All the pigments and fillers, with the exception of those used to study the effect of particle size, were ball milled with part of the coating solution for 4 h before use.

RESULTS

The contrast ratio results shown in Table 1 and Figs 1 and 2 can be placed in perspective by comparing them with values obtained for white paper—90% for typing paper 80 g m^{-2} and 60% for air-mail paper. Bearing this in mind, it can be seen that films containing the fillers calcium carbonate, calcium sulphate and talc were all relatively transparent and only the inclusion of titanium dioxide imparted opacity (Table 1). In contrast, the films containing the coloured lake pigments had contrast values depending on both the parent dye and the dye concentration, the degree of opacity decreasing blue > red > orange > yellow. No effect of colour could be seen in films containing the synthetic iron oxides, all of which were very opaque. The higher contrast ratio found for the film containing yellow iron oxide relative to that found for the film containing titanium dioxide confirms data recently published by Nyquist et al (1982) which showed that better light protection could be obtained with films containing yellow iron oxide compared with films containing titanium dioxide.

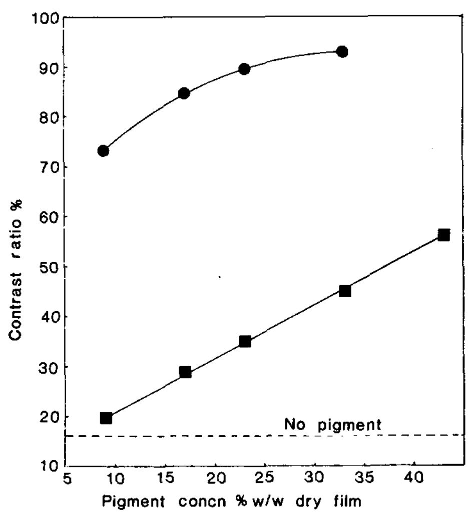

FIG. 1. The effect of pigment concentration (% w/w dry film) on the contrast ratio of tablet film coatings: ● Titanium dioxide; ■ FD&C Yellow 5 lake (25% dye content).

Increasing the pigment concentration of films containing both titanium dioxide and FD&C yellow 5 lake resulted in increased contrast ratios (Fig. 1) with an indication of a linear relationship in the case

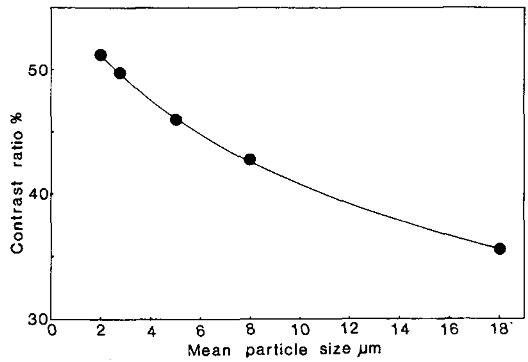

FIG. 2. The effect of the particle size of dolomite on the contrast ratio of tablet film coatings: pigment concentration 37·5% w/w; contrast ratio of non-pigmented film 26·0 ± 2·95.

of the latter. Increasing the particle size of the white dolomite resulted in a decrease in opacity (Fig. 2).

An interesting feature of all the results is the variation in the measured contrast ratio of the non-pigmented or filled films. This was found to be caused by differences in the method used for coating and the coating formulation (especially the polymer concentration), with higher polymer concentrations producing films with higher contrast ratios. However, since the formulation and process conditions were always kept constant during a particular experiment, it was felt that this effect could be effectively eliminated. Support for this stance may be obtained by considering the relative increases in contrast ratios found on the addition of 16% w/w FC&C Yellow 5 lake (25% dye content) in the two

Table 1. The effect of pigment and filler type on the opacity of tablet film coatings (pigment/filler concentration 16% w/w dry film).

Pigment/filler	Dye content	Contrast ratio %
No filler	—	33·3 ± 3·8
Calcium carbonate	—	46·7 ± 3·3
Calcium sulphate	—	46·8 ± 3·2
Talc	—	46·4 ± 2·2
Titanium dioxide	—	91·6 ± 1·2
Red iron oxide	—	99·5 ± 0·2
Yellow iron oxide	—	98·4 ± 0·8
Black iron oxide	—	99·6 ± 0·6
FD&C Blue 2 lake	13	97·5 ± 1·1
FD&C Blue 2 lake	30	99·5 ± 0·3
FD&C Red 3 lake	18	70·1 ± 3·3
FD&C Red 3 lake	39	81·3 ± 4·1
FD&C Yellow 5 lake	16	62·9 ± 3·5
FD&C Yellow 5 lake	25	65·2 ± 3·8
FD&C Yellow 5 lake	37	66·7 ± 3·3
FD&C Yellow 6 lake	17	73·2 ± 3·6
FD&C Yellow 6 lake	39	78·1 ± 3·1

experiments shown in Table 1 and Fig. 1. In both cases the relative increase in the contrast ratio is the same.

DISCUSSION

The opacity of a pigmented film is dependent on: (a) light reflection at the air/film and film/pigment interfaces, (b) light absorption by the pigment, (c) light scattering, (d) light refraction.

The laws of reflection and refraction at plane boundaries are well established, but the laws of light scattering are complex and have, as yet, only been established for simple systems of spherical particles at low concentrations. Of interest here is the Mie theory (Mie 1908) for the scattering and absorption of monodispersed spheres. In order to apply the theory it is necessary to have knowledge of the complex index of refraction of the pigment relative to the surrounding film. This factor (m) is related to the refractive index of the pigment (n_p), the refractive index of the film (n_o) and the absorption index or extinction coefficient of the pigment (k) by the expression:

$$m = \frac{n_p}{n_o}(1 - ik) \quad (1)$$

where i is the square root of −1.

For white pigments where absorption in the visible region is negligible, k will be extremely small and m, and hence the opacity of the pigmented film, will be dependent solely on the refractive index of the pigment (n_p) relative to that of the film (n_o). There are many equations relating n_p and n_o. Of importance are the Fresnel equation (Cooper 1969):

$$R = \frac{(n_p - n_o)^2}{(n_p + n_o)^2} \quad (2)$$

Where R is the amount of light reflected at the film/pigment interface (i.e. the Fresnel reflectivity) and the Lorentz-Lorentz equation (Mitton 1973):

$$M = \left(\frac{n_p}{n_o}\right)^2 - 1 \bigg/ \left(\frac{n_p}{n_o}\right)^2 + 2 \quad (3)$$

where M is referred to as the Lorentz-Lorentz expression—an important factor since it has to be shown practically that there is an approximate rectilinear relationship between the relative opacity of paint films containing white pigments and M^2 (Mitton 1973). Provided n_o is approximately 1·5 (a reasonable value for most polymeric films—Rowe & Forse 1983) and that n_p ranges between 1·5 and 2·75 (a reasonable range for most white pigments—Rowe & Forse 1983), then M can be approximated by the expression:

$$M = 0{\cdot}4(n_p - n_o) \quad (4)$$

This simple expression serves to explain why the difference between the refractive indices of a pigment and a film has generally been used as a rough guide to film opacity.

In the practical application of tablet film coatings containing the white pigments, it can be seen that the results are in partial agreement with those obtained with paint films with values for M, M^2 and R for titanium dioxide being much higher than those for the fillers calcium carbonate, calcium sulphate and talc (Table 2). However, there is no direct relationship between the values and the contrast ratios (Table 1).

Table 2. Calculation of the Fresnel reflectivity, R, and the Lorentz-Lorentz expression, M, for white pigments dispersed in films of hydroxypropyl methylcellulose (refractive index 1·49).

Pigment	Refractive index*	R	M (from equation 3)	M^2 (from equation 3)	M (from equation 4)	M^2 (from equation 4)
Titanium dioxide	2·52	0·062	0·382	0·146	0·412	0·170
Calcium carbonate	1·58	0·0008	0·040	0·0016	0·036	0·0013
Calcium sulphate	1·59	0·0011	0·056	0·0032	0·040	0·0016
Talc	1·57	0·0007	0·036	0·0013	0·032	0·0010

* These are mean values of the refractive indices since all the pigments are optically anisotropic i.e. they possess different refractive indices along different axes (see Rowe & Forse 1983).

For pigmented films, which show considerable absorbance in the visible spectrum, the index of absorption, k, will have a finite value and hence m will be dependent on both n_p and k, the relative importance of each factor being dependent on the specific pigment. For instance, an ideal black pigment which, by definition, absorbs all light, will have a very high value of k and m and hence opacity will be high irrespective of the refractive index of the pigment. Unfortunately, unlike values for refractive indices, values for the index of absorption of pigments are not available in the literature. One reason for this is that values will vary widely depending on the wavelength of the incident light, e.g. a red pigment, which strongly absorbs blue light, will have a larger value of k when measured in blue light than when when measured in red light. Since the low wavelength light is predominantly scattered, pigments that absorb at the higher wavelengths (e.g. the blue pigments) will tend to produce more opaque films than those which absorb at the lower wavelengths (e.g the yellow pigments). This trend is illustrated by the contrast ratio figures for the coloured lake pigments but not for the coloured iron oxides. The reason for this apparent anomaly lies in the relative n_p and k values for these two groups of pigments. For the coloured lake pigments, n_p is ca 1·54 (Rowe & Forse 1983) while k is ca 0·5 (Brockes 1964) but for the iron oxide pigments n_p is in excess of 2·0 (Rowe & Forse 1983) while k is ca 0·05 (Brockes 1964). Therefore, for the coloured lakes the opacity will be more dependent on light absorption and the 'colour' of the pigment but for the iron oxide pigments the opacity will be more dependent on light scattering.

The Mie theory for absorption and scattering, as stated previously, is only relevant where pigment concentration is low. As the pigment concentration is increased and the pigment particles become more closely packed, the scattered light from one particle will begin to interfere with that from a neighbouring particle and the theory fails. However, as a general rule, it can be stated that the opacity of a film will increase with increasing pigment concentration as shown in Fig. 1.

It has been known in the paint industry for many years that the particle size of a pigment is a factor in determining the opacity of paint films. There are many empirical relationships available in the literature predicting the optimum particle size of a pigment, probably the most convenient being the one suggested by Mitton (1973):

$$d_{opt} = \frac{\lambda}{\sqrt{2}\, n_o M \pi} \quad (5)$$

where d_{opt} is the optimum particle diameter and λ is the wavelength of the incident light. If this is applied to the dolomite filler ($n_p = 1{\cdot}62$) used in Fig. 2, then it can be calculated for blue light (λ = 450 nm), green light (λ = 560 nm) and red light (λ = 590 nm); the optimum particle diameters will be 1·19, 1·48 and 1·56 μm respectively. These are slightly less than any of the sizes in Fig. 2 but sufficiently close to show the relevance of the equation for pigmented tablet film coatings.

The overall results show that the differences in the opacity of tablet film coatings can be accurately and reproducibly assessed by means of a simple contrast ratio. The measurements are relatively easy to make and the results obtained are consistent with the known theories of light scattering and absorption. However, it must be realized that the film thickness is an important variable and until the interdependence of the film thickness and contrast ratio has been established then all measurements will need to be made at constant thickness.

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