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Determination of the critical pigment volume concentrations of pigmented film coating formulations using gloss measurement

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Summary

Critical pigment volume concentrations for a variety of pigments and opacifiers dispersed in a hydroxypropyl methylcellulose film have been determined using gloss measurement. The values vary between 7 and 12% for the iron oxide pigments, 13.5 and 15% for titanium dioxide and 12 and 35% for talcs of varying particle size. The results are comparable with those determined using other methods and are consistent with the known theories of pigment dispersion and packing.

Introduction

Additives in the form of pigments (eg. aluminium lakes of water soluble dyes), opacifiers (eg. titanium dioxide) and various inorganic materials (eg. iron oxides and talc) are often included in film coating formulations (Pickard and Rees, 1974; Porter, 1980; Rowe 1983). As the proportion of a solid additive in a polymer film is increased, the amount of polymer required to completely surround the particles in the dry film increases until at a critical concentration there is just sufficient polymer to fill all the interparticle voids. This concentration, known as the critical pigment volume concentration (CPVC), is of interest to the formulator since it is the point at which marked changes occur in the mechanical properties, appearance and permeability of the dry film (Asbeck and van Loo 1949).

The critical pigment volume concentration is characteristic of a specific additive/polymer combination for well-defined dispersion conditions and hence its theoretical determination is virtually impossible. Although Asbeck and van Loo (1949) proposed an independent experimental method based on filtering a coating formulation through a fine filter and calculating the void volume from the volumes of the filter cake and the pigment particles, the majority of the generally used techniques are based on the measurement of the discontinuity of a specific property of the dry film with increasing pigment volume concentration. In this paper gloss measurement is the specific property studied since, although the gloss of a film decreases with increasing pigment volume concentration, at the critical pigment volume concentration it either tends to remain constant or, in some cases, increases (Guillaume, 1969).

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Materials and Methods

All the additives were obtained from D.F. Anstead Ltd. (Billericay, Essex, U.K.) and used as received (Table 1). Dispersions in a 5% w/v aqueous-methanol solution of hydroxypropyl methylcellulose (Pharmacoat 606, Shin-Etsu Chemical Co., Tokyo, Japan) were prepared using a small laboratory ball mill and films were prepared by casting onto a treated siliconised glass substrate bounded by a nylon ring 10 cm in diameter. The films were dried on a level surface overnight in a hot oven at 60 ° C and stored in a desiccator until required.

Gloss measurements were made on each film (stuck to pieces of black card to prevent curling and wrinkling) by determining the maximum light specularly reflected at 60 ° C using a photodetector coupled to a signal processor (model D48-7 glossmeter, Hunter Associates Laboratory Inc., VA, U.S.A.). The instrument was calibrated using standard ceramic glass tiles of known gloss (Rowe, 1985).

Results and Discussion

Some typical results are shown in Fig. 1. It can be seen that for both the titanium dioxide and the yellow lake pigment the measured gloss decreases with increasing pigment volume concentration until at concentrations in excess of 12% there is no further decrease. However, for the black iron oxide there is a decrease in the measured gloss at low pigment volume concentrations with an increase at high pigment volume concentrations. If the data for the black iron oxide is plotted on a semi-logarithmic scale as suggested by Guillaume (1969) then the critical pigment volume concentration can be determined at the intersection of the two straight lines as shown in Fig. 2. Accrued results using both methods of analysis are shown in Table 2.

It can be seen from the accrued results that whereas the iron oxides all have relatively low critical pigment volume concentrations, the values for the titanium dioxide, the yellow lake pigment

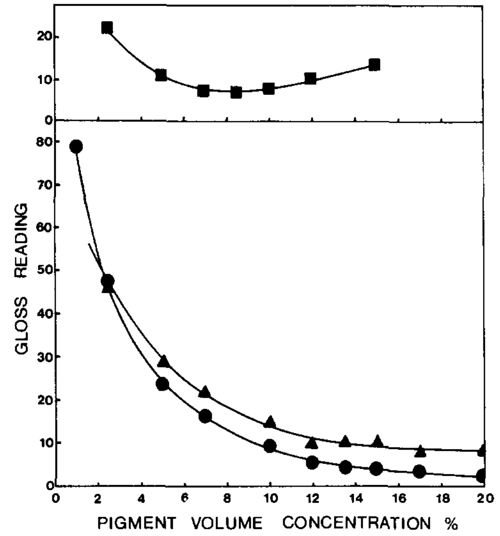

Fig. 1. Measured gloss vs pigment volume concentration for (▲), titanium oxide; (●), yellow 6 lake; (■), black iron oxide.

TABLE 1

Pigment properties

Pigment	Particle shape	Surface area * $m^2 g^{-1}$
Black iron oxide	cubical	3.84
Red iron oxide	spherical	7.56
Yellow iron oxide	acicular	13.07
Titanium dioxide	rounded	7.60
FD&C yellow 6 lake	irregular	93.10
Talc A	lamellar	2.99
Talc B	Lamellar	14.33

* Determined by B.E.T. nitrogen adsorption.

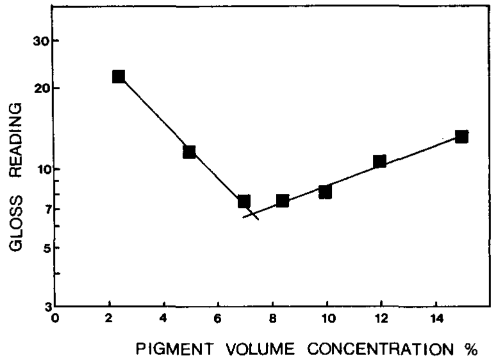

Fig. 2. Log (measured gloss) vs pigment volume concentration for black iron oxide.

TABLE 2

Critical pigment volume concentrations

Pigment	CPVC (% v/v)
Black iron oxide	7.0– 8.5
Red iron oxide	8.5–10.0
Yellow iron oxide	10.0–12.0
Titanium dioxide	13.5–15.0
Yellow 6 lake	12.0–13.5
Talc A	12.0–15.0
Talc B	25.0–35.0

and the talcs are very much higher. The values for the iron oxides appear to be related to the shape and surface area of the pigment with black iron oxide with its cubic shape and low surface area exhibiting the lowest value and yellow iron oxide with its acicular shape and high surface area exhibiting the highest value. It is interesting to note that the critical pigment volume concentration for the titanium dioxide is some 50% higher than that for the red iron oxide despite the fact that these materials have a similar particle shape and surface area.

The values recorded in this study are comparable with those determined by other workers using a variety of techniques for similar film formulations based on hydroxypropyl methylcellulose. Of interest are the values determined for titanium dioxide from permeability measurements, 10.5–13.5% (Porter 1980); from diffusion coefficient measurements, 12.5–13.5% (Okamafe and York, 1984a) and from tensile mechanical testing, 9.7–12.7% (Okamafe and York, 1984b). Unfortunately the data on talc-filled film is ambiguous with no transitions in permeability/diffusion properties being reported. However, Okamafe and York (1984b) did show transitions in the tensile mechanical properties of hydroxypropyl methylcellulose films at talc concentrations of 8.9–14.1% and 11.9–18.8% for large and small talc particles with surface areas of 1.34 $m^2\ g^{-1}$ and 1.81 $m^2\ g^{-1}$ respectively. These values are comparable with those determined for the large-sized talc used in this study.

As stated previously, the critical pigment volume concentration is a characteristic of a specific additive/polymer combination for well-defined dispersion conditions and hence will depend on the nature of the polymer and any interaction between it and the pigment (eg. adsorption) and the packing characteristics of the pigment. Adsorption of polymers from solution onto pigment surfaces is a well known phenomena and is often used to control flocculation of pigments in paints. Unfortunately, there is little literature data regarding the adsorption/interaction for the pigments and polymers used in tablet film coating. However, for titanium dioxide there is direct evidence of both adsorption from solution (Brodnyan, 1968) and an interaction in the dry polymer film (Okamafe and York, 1984c) and it is this effect that is thought to be the reason for the relatively high critical pigment volume concentration of this pigment compared with that for red iron oxide.

The differences seen in the critical pigment volume concentrations for the iron oxides is thought to be due to their shape and packing characteristics. The black iron oxide presumably aggregates and traps more polymer leaving less polymer available for film formation. On the other hand, the acicular particles of yellow iron oxide would form aggregates with an open structure immobilising less polymer. The strength of the aggregate will also be important as in the case of the yellow lake pigment. Aluminium lakes are known to be highly aggregated with high aggregate strength difficult to break down except with high-energy dispersion systems, Despite their high surface area (Table 1) there will be little external surface available to the polymer and a reasonably high critical pigment volume concentration can be achieved. Talc is not prone to aggregation and forms a parallel planar structure within the film resulting in high critical pigment volume concentrations. The differences between the values for the two talcs remains a matter for conjecture.

In conclusion the results show that gloss measurement can provide a rapid, simple method of determining the critical pigment volume concentrations of pigmented film coating formulations. The results are comparable with those determined using other more laborious methods and are consistent with the known theories of pigment dispersion and packing.

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