International Journal of Pharmaceutics, 14 (1983) 355–359
Elsevier Biomedical Press

Modulus enhancement in pigmented tablet film coating formulations

R.C. Rowe

ICI Pharmaceuticals Division, Alderley Park, Macclesfield, Cheshire SK10 2TG (U.K.)

(Received August 27th, 1982)
(Accepted September 14th, 1982)

Additives in the form of aluminium lakes of water-soluble dyes, opacifiers (e.g. titanium dioxide), various inorganic materials (e.g. iron oxides, calcium carbonate, talc and colloidal silica) and even active drug substances (e.g. caffeine and salicyclic acid) have been included in tablet and granule film coating formulations (Pickard and Rees, 1974; Friedman et al., 1979; Porter, 1980a; Rowe, 1981). The inclusion of such materials can affect the mechanical properties of the film coatings causing a decrease in their tensile strength and an increase in their modulus of elasticity thus affecting their performance in situ on the tablet surface (Rowe, 1982). If, as has been suggested by Toblosky (1971), filled or pigmented polymer films can be analyzed in an analogous fashion to suspensions, then modulus enhancement in such polymer films should be able to the expressed in terms of a modified Einstein equation:

$$E = E_0(1 + 2.5\phi + 14.1\phi^2) \quad (1)$$

where E is the modulus of the filled or pigmented film, E_0 the modulus of the unpigmented film, and ϕ is the volume fraction of the filler or pigment. The applicability of this equation in the analysis of modulus enhancement in pigmented tablet film coatings is discussed in this report using data generated by Abdul-Razzak (1980), Aulton (1981), Delporte (1981) and Porter (1980b).

It can be seen from Eqn. 1 that the pigment concentration is expressed as a volume fraction and hence, in order to comply with this criterion, helium density measurements were required on the pigments and film coating polymers used. Values obtained on representative samples using a helium pycnometer (Beckman model 930, Glenrothes, Scotland) are shown in Table 1. Fig. 1 shows modulus enhancement data for 3 standard aluminium lake pigments and titanium dioxide dispersed in hydroxypropyl methylcellulose films (data from Abdul-Razzak, 1980; Aulton, 1981) and for red iron oxide dispersed in a cellulose acetate phthalate film (data from Porter, 1980b). Results for titanium dioxide dispersed in a hydroxypropyl

TABLE 1

HELIUM DENSITIES OF PIGMENTS AND FILM COATING POLYMERS USED IN CALCULATIONS

	Helium density ($g \cdot cm^{-3}$)
Film coating polymers	
Hydroxypropyl methylcellulose (Methocel E5)	1.26
Cellulose acetate phthalate	1.37
Pigments	
Titanium dioxide	3.78
Red iron oxide	5.41
Erythrosine lake	2.08
Brilliant Blue FCF lake	1.85
Tartrazine lake	1.92

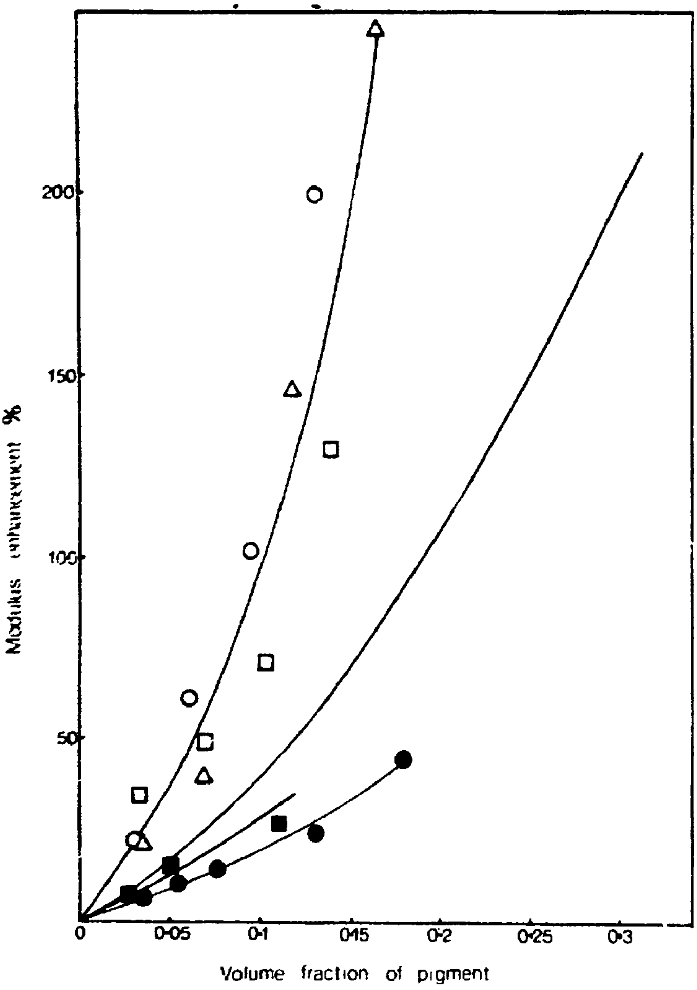

Fig. 1. The effect of pigments on the modulus enhancement of tablet film coating formulations: △, Brilliant Blue FCF lake; □, tartrazine lake; ○, erythrosine lake; ●, titanium dioxide—all materials dispersed in hydroxypropyl methylcellulose; ■, red iron oxide dispersed in cellulose acetate phthalate. The graph without symbols represents the values predicted using Eqn. 1.

methylcellulose film obtained using the data generated by Delporte (1980) were similar to those plotted in Fig. 1.

It can be seen that whereas the aluminium lakes in hydroxypropyl methylcellulose show large positive deviations from Eqn. 1 in that their modulus enhancement is much greater than predicted, titanium dioxide in the same film coating polymer shows a negative deviation in that the modulus enhancement is less than predicted. Red iron oxide in cellulose acetate phthalate, however, shows only a small negative deviation at higher pigment volume concentrations. In order to explain these deviations it is first necessary to consider the validity of the assumptions made in deriving Eqn. 1, i.e. firstly, that the filler or pigment particles are rigid, spherical and evenly dispersed throughout the polymer matrix, and secondly, that there is no interaction between the particles and the polymeric film former.

Particle shape

If it is assumed that the analogy between the modulus of filled or pigmented polymer films and the viscosity of colloidal suspensions is correct then an idea of the effect of irregularity of shape on modulus enhancement can be obtained by considering the equations derived for the effect of particle shape on the concentration dependence in the viscosity of colloidal suspensions. In this case it is known that any deviation from a spherical shape produces a positive deviation, the more irregular the shape the more the deviation (Frisch and Simha, 1956). The extent of the deviation that can be expected can be judged from the work of Simha (1950) in his equations for predicting the viscosity of suspensions containing dumbbell-shaped particles. If 2L is the length of the rod joining the two spheres each of radius, a, then by analogy to Eqn. 1 modulus enhancement in polymer films containing such particles should be able to be expressed by:

$$E = E_0\left(1 + \frac{3}{2}\frac{L^2}{a^2}\phi + \frac{87}{50}\frac{L^4}{a^4}\phi^2\right) \qquad (2)$$

It can be seen that for a dumbbell particle of L/A = 2 and at a volume concentration of 0.1, the predicted modulus enhancement should be 87.8% compared to 39.1% for a spherical particle at the same volume concentration. This solution is, of course, simplified since it assumes random orientation of the dumbbell. Any aggregation of particles will undoubtedly produce even larger deviations although alignment of asymmetric particles, provided they align in the direction of the application of stress during measurement, will tend to minimize this effect.

Unfortunately little basic work has been done on the shape characterization of the various fillers and pigments used in tablet film coating although qualitative descriptions are available for the majority (Table 2). These descriptions, although subjective, are consistent with the results in Fig. 1 in that the largest deviations occur with the largely irregular aluminium lake particles while little or no deviation occurs with the spherical red iron oxide particles. However, the negative deviation occurring with the largely rounded titanium dioxide particles is an anomaly and can only be explained on the basis of particle–polymer interaction.

TABLE 2

QUALITATIVE DESCRIPTIONS OF THE SHAPES OF PIGMENTS AND ADDITIVES USED IN TABLET FILM COATING

Pigment/additive	Qualitative description of shape
Titanium dioxide	rounded
Black iron oxide	cubic
Red iron oxide	spherical
Yellow iron oxide	acicular
Talc	flaky
Aluminium lakes	irregular

Particle-polymer interaction

Particle-polymer interactions can be of two types; hydrogen bonding or dipole-dipole interaction, resulting in a more swollen polymeric matrix with lower modulus, and, much less likely, covalent bonding, resulting in a cross-linked, rigid, polymeric matrix with higher modulus. It is likely that the interaction between titanium dioxide and hydroxypropyl methylcellulose is of the former type analogous to plasticization since it is already known that the material does interact with vinylchloride/vinylacetate copolymer lowering its glass transition temperature (Kumins and Roteman, 1963). It is interesting to note that calculations made on the data given by Delporte (1981) show that the negative deviation seen in Fig. 1 for titanium dioxide dispersed in hydroxypropyl methylcellulose is minimized on the addition of the plasticizers, propyleneglycol and polyethylene glycol 400, to the polymer. This implies that the interaction between the titanium dioxide and hydroxypropyl methylcellulose is through sites on the polymer which also interact with plasticizers, and that plasticizers can easily displace the titanium dioxide from these sites.

Although it must be realized that many more results are required before the analogy between the modulus of elasticity of pigmented or filled tablet film coatings and the viscosity of colloidal suspensions is proved conclusively and can be used in a predictive capacity, the data presented do show the wealth of information that can be accrued by a systematic analysis of simple modulus measurements.

Acknowledgement

The author wishes to thank Dr. M.E. Aulton for providing data from unpublished reports.

References

Abdul-Razzak, M.H., M. Phil. thesis, Council for National Academic Awards, 1980.
Aulton, M.E., private communication (1981).

Delporte, J.P., Influence de quelques additifs sur les propriétés de résistance méchanique de films isolés d'hydroxypropyl méthylcellulose de basse viscosité. J. Pharm. Belg., 36 (1981) 27–37.

Friedman, M., Donbrow, M. and Samuelov, Y., Placebo granules as cores for timed release drug delivery systems. J. Pharm. Pharmacol., 31 (1979) 396–399.

Frisch, H.L. and Simha, R., The viscosity of colloidal suspensions and macromolecular solutions. In Eirich, F.R. (Ed.), Rheology Theory and Application, Vol. 1, Academic Press, New York, 1956, pp. 525–614.

Kumins, C.A. and Roteman, J., Effect of solid polymer interaction on transition temperature and diffusion coefficients. J. Appl. Polym. Sci., Part A, 1 (1963) 527–540.

Pickard, J.F. and Rees, J.E., Film coating. 1. Formulation and process considerations. Manuf. Chem. Aerosol News, April (1974) pp. 19–22.

Porter, S.C., The effect of additives on the properties of an aqueous film coating. Pharm. Tech., March 1980a. 67–75.

Porter, S.C., PhD thesis, Council for National Academic Awards, 1980b.

Rowe, R.C., The effect of the particle size of an inert additive on the surface roughness of a film-coated tablet. J. Pharm. Phamacol., 33 (1981) 1–4.

Rowe, R.C., The effect of pigment type and concentration on the incidence of edge splitting on film-coated tablets. Pharm. Acta Helv., 57 (1982) 221–225.

Simha, R., The concentration dependence of viscosities of dilute solutions. J. Colloid Sci., 5 (1950) 386–392.

Toblosky, A.V., Viscoelastic properties of polymers. In Toblosky, A.V. and Mark H.F (Eds.), Polymer Science and Materials, Wiley Interscience, New York, 1971, pp. 207–229.