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The adhesion of film coatings to tablet surfaces—a problem of stress distribution

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An important prerequisite of a film coating is that it should adhere strongly to the tablet core. Unfortunately only limited research has been carried out on the measurement of the adhesion of film coatings to tablet surfaces. The first method used was a peel test (Wood & Harder 1970; Nadkarni et al 1975) in which a strip of the film coating was peeled from the tablet surface using a tensile tester. To overcome the many deficiencies of the peel test, an instrument was designed to remove a known area of film coating normal to the film/tablet interface (Fisher & Rowe 1976). This method has since been used extensively to study the various factors affecting the adhesion of film coatings to tablet surfaces (Rowe 1977, 1978; Porter 1980; Fung & Parrott 1980). The results of these studies have generally been interpreted in terms of the physicochemical interaction between the film-coating polymers and tablet excipients. Recently, however, work on the bridging of the intagliations (Rowe & Forse 1980, 1981)—a practical consequence of poor film/tablet adhesion and thought to be due to the presence of high internal stress in the film coating—has highlighted the need to reappraise the results based on the stress distribution at the film/tablet interface. This has been undertaken in this report.

A film coating applied to a tablet surface is under the influence of two stresses (Fig. 1): one due to the forces of adhesion holding it to the substrate (A), and one inherent in the film (P) parallel to the tablet surface—the sum of the stress due to shrinkage of the film on evaporation of the solvent and the thermal stress due to differences in thermal expansion of the film coating and tablet substrate during changes in temperature arising out of the coating process. If the substrate was flexible, P would cause it to bend (Chow et al 1976) but since it is non-flexible this stress will cause a reaction R at the interface acting in the opposite direction to A. The relationships between P, R and t—the thickness of the film coating—are not fully understood but it is known that R will increase as both P and t increase

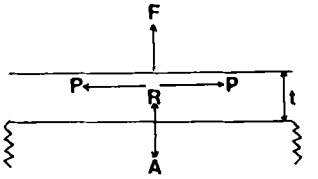

Fig. 1. A schematic diagram showing the stress distribution at the film/tablet interface.

(Meissner & Baldauf 1951). It can be seen that F, the so called measured adhesion, is given by the equation:

$$F = A - R \quad (1)$$

and, if R exceeds A, then loss of adhesion occurs resulting in bridging of the intagliations. (R has been incorrectly referred to as the residual internal stress previously Rowe & Forse 1980.)

P can be calculated by reference to Croll (1979) and Sato (1980) as recently suggested by Rowe (1981) in the context of film cracking.

$$P = \frac{E}{1 - \upsilon}\left[\frac{\phi_s - \phi_r}{3(1 - \phi_r)} + \frac{\Delta\alpha\Delta t}{1 + \upsilon}\right] \quad (2)$$

Where E is the Young's modulus of the film coating, υ is the Poisson's ratio, ϕ_s is the volume fraction of the solvent at the solidification point, i.e. where the coating solution first behaves as a solid rather than a viscous liquid. ϕ_r is the volume fraction of the solvent remaining in the dry film at ambient conditions. $\Delta\alpha$ is the difference between the thermal expansion coefficients of the film coating α_c and the tablet substrate α_s. Δt is the difference between the glass transition temperature of the film coating Tg and the ambient temperature T.

It is pertinent, therefore, to reappraise the published film/tablet adhesion results in terms of the factors affecting R and P, i.e. the substrate formulation, the addition of plasticizers and pigments to the film coating, the solvents used, the thickness of the film applied, and the conditions under which measurements were made.

Substrate formulation

Variations in the adhesion results for a constant film formulation applied to a variety substrates have generally been interpreted in terms of the surface polarity and roughness of the substrate (Rowe 1977). However, from equation 2 it can be seen that if substrates have a different thermal expansion coefficient $\Delta\alpha$ will vary. If $\Delta\alpha$ is large then P will be large and the measured adhesion F will be small. Data recently compiled on the expansion coefficients of a variety of materials representative of the excipients commonly used in tablet formulation does show a wide variability (Rowe 1980) and hence this factor should certainly be taken into account when substrates are compared.

Addition of plasticizers

The results on the addition of plasticizers on the adhesion of hydroxypropyl methylcellulose films to tablet substrates would appear to be variable. Fisher & Rowe (1976) found a slight but statistically insignificant decrease in the adhesion on the addition of glycerol and propylene glycol but Porter (1980) found a slight increase with the same plasticizers and a slight decrease with the polyethylene glycols. These results have been interpreted in terms of the molecular association between the plasticizer and the polymer and the polymer and the tablet excipients. Plasticizers, however, cause a decrease in P by decreasing both the Young's modulus, E, and the glass transition temperature, Tg, of the film coating. This will cause a decrease in R and hence an increase in the measured adhesion. This increase will be dependent on the concentration of the plasticizer and on its degree of interaction with the polymer since plasticizers with a high degree of interaction will decrease the glass transition temperature to a greater extent than those with a poor interaction (Entwistle & Rowe 1979). Further support for this mechanism can be obtained from the work on bridging of the intagliations (Rowe & Forse 1981) which showed that the incidence of bridging was decreased on the addition of plasticizers, the magnitude of the decrease being dependent on the degree of plasticizer/polymer interaction measured by the lowering of the glass transition temperature of the film.

Addition of pigments

The addition of pigments to film coatings is usually detrimental to the adhesion (Fisher & Rowe 1976) although recent work by Porter (1980) did not show any significant effect even on the addition of high concentrations of pigments. The generally held view is that embedded pigments will interfere with the intermolecular bonding at the film/tablet substrate. Extensive work in the field of organic coatings (Sato 1980) has shown that the addition of any pigments or additives to films causes an increase in the internal stress P by causing an increase in both the Young's modulus and glass transition temperature of the film. The effect, however, is not only dependent on the concentration of the pigment but also on the particle morphology since pigments with flaky particles have less of an effect than pigments with irregular particles due to the former arranging themselves parallel to the film/substrate interface resulting in less stress being developed (Ioune 1943). Pigments should, therefore, cause a decrease in the measured adhesion, the decrease being dependent on the particle shape. Some evidence of this can be seen in a comparison of the measured adhesion of a constant film formulation (hydroxypropyl methylcellulose—Pharmacoat 606—Shinetsu Chemical Co) containing a constant concentration of 25% w/w pigment (based on polymer)—Table 1. In this case the adhesion is highest with talc and yellow iron oxide and lowest with FD & C Yellow 5 aluminium lake.

Solvent

The effect of solvents on the adhesion of a film coating to a tablet substrate has only been studied in detail using the

Table 1. The effect of pigments in the measured adhesion of hydroxypropyl methycellulose film to a placebo tablet substrate (pigment concentration 25% w/w based on polymer)—measured using the method described by Fisher & Rowe (1976).

Pigment	Particle shape	Measured adhesion *kPa*
Non pigmented	—	28·5
FD & C Yellow 5 aluminium lake	Irregular	22·5
Black iron oxide	Cubic	24·6
Red iron oxide	Spherical	26·0
Yellow iron oxide	Acicular	28·1
Talc	Flaky	29·5

peel test by Nadkarni et al (1975) who showed that the adhesion was best when solvents with solubility parameters close to that of the polymer were used. In this case ϕ_s will be small since polymer solutions prepared using good solvents will solidify at lower dilutions than those prepared using poorer solvents resulting in the internal stress P being minimal.

Film thickness

The relationship between P and t, the thickness of the film coating and hence F, the measured adhesion, has already been studied (Rowe 1978) confirming the mechanism given above. An extension of the work into the study of bridging of the intagliations gave results also in agreement with this mechanism (Rowe & Forse 1980). This factor also offers the opportunity of obtaining a true value for A—the intrinsic adhesion at the interface—since by extrapolating measured adhesion values to zero thickness, P and hence R can be effectively eliminated.

Conditions of measurement

The value of keeping the conditions of measurement constant during testing can now be seen since any change in both the ambient temperature and the humidity will affect the value of P by affecting Δt and ϕ_r. The latter will, of course, be especially important for hygroscopic polymers since in this case ϕ_r will increase with increasing humidity resulting in a decrease in both P and R and hence an increase in the measured adhesion. Evidence for this can be obtained from the work of Fung & Parrott (1980) who showed an increase in the measured adhesions of a variety of film-coated tablets on increasing the relative humidity.

Conclusions

The model based on a modified stress distribution at the film/tablet interface which provides for the presence of internal stresses within the film is able to account for the trends in the results obtained from both adhesion testing and experiments on the bridging of intagliations. It should, therefore, be considered in parallel with the current models of simple intermolecular association when attempting to explain results from such studies. Unfortunately it is one further variable to be considered before fundamental studies into the strength of the adhesive bond at the film/tablet interface can be undertaken.

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Inability of degraded carrageenan fractions to induce inflammatory bowel ulceration in the guinea-pig

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Ulcerative disease of the large bowel has been induced readily in several animal species (viz. guinea-pigs, rabbits, rhesus monkeys) by the oral administration of degraded carrageenan, a sulphated polysaccharide derived from the red seaweed, *Eucheuma spinosum* (Marcus & Watt 1969; Benitz et al 1973; Abraham et al 1974). Since the degraded form of carrageenan is difficult and expensive to obtain, Watt et al (1979) described in this journal a method for the hydrolysis of a native form of *Eucheuma spinosum* carrageenan, leading to the more ulcerogenic degraded product.

Using the same method of degradation as these workers, we investigated the actions of a variety of carrageenan fractions on the guinea-pig bowel, with the purpose of developing a reliable model for the assessment of anticolitic drugs. The carrageenan fractions were obtained from Sigma Chemical Co. Ltd (London, U.K.), a source not des-

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