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The cracking of film coatings on film-coated tablets—a theoretical approach with practical implications

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A theoretical approach of film cracking is presented using equations which allow for the calculation of both the internal stress in a film coating due to shrinkage of the film on evaporation of the solvent and the thermal stress due to differences in the thermal expansion of the film coating and tablet substrate during changes in temperature arising out of the coating processes. The practical implications of this approach in film formulation are discussed with particular reference to the choice of tablet core formulation, the grade of polymer used, the type and concentration of both plasticizers and pigments and the solvent system used in the coating process.

Stresses are invariably created when a film is deposited on a tablet substrate. These are created by shrinkage of the film on evaporation of the solvent and by differences in the thermal expansion of the coating and the substrate. Stresses can manifest themselves either on the macroscopic scale as edge splitting and peeling and cracking (Rowe & Forse 1980) or on the microscopic scale as minute cracks which can be seen only at high magnification (Fig. 1). Both defects could well affect the gastric resistance of entero-soluble films or the dissolution rate of sustained release films although, in the case of water soluble films applied for reasons of identification and elegance, the visual defects are the most important. In this paper a theoretical approach of cracking is presented and its practical implications in film formulation discussed.

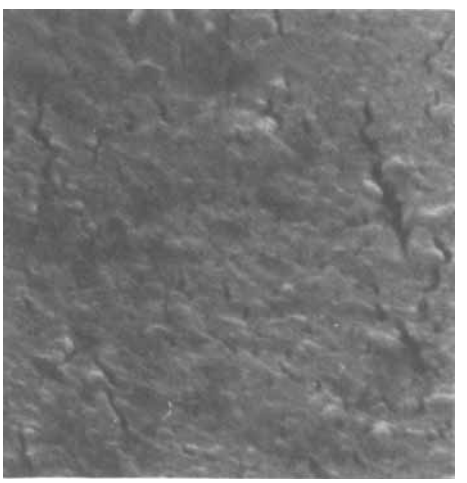

FIG. 1. A scanning electron photomicrograph of a surface of a placebo tablet based on lactose coated with a 35 μm thick film of hydroxypropyl methyl cellulose showing microscopic cracks in the film (× 18 000).

Theory

The total internal stress P in a film coating applied to a tablet substrate can be regarded as being composed of the sum of: (a) the internal stress due to shrinkage of the film on evaporation of the solvent P_S, (b) the thermal stress due to differences in the thermal expansion of the film coating and tablet substrate during changes in temperature arising out of the coating process P_T

$$\text{i.e. } P = P_S + P_T \quad .. \quad (1)$$

Both P_S and P_T can be calculated by reference to Croll (1979) and Sato (1980) respectively:

$$P_S = \frac{E}{1-\nu} \cdot \frac{\phi_s - \phi_r}{3(1-\phi_r)} \quad .. \quad .. \quad (2)$$

$$P_T = \frac{E}{1-\nu^2} \Delta\alpha \Delta t \quad .. \quad .. \quad (3)$$

where E is the Young's modulus of the film, ν is the Poisson's ratio of the film, 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 coefficient of the coating α_c and the tablet substrate α_s. Δt is the difference between the glass transition temperature of the coating Tg and the ambient temperature T.

It must be emphasized that both the equations 2 and 3 are concerned only with the equilibrium values of the two stresses. It is assumed that before solidification and/or above the glass transition temperature of the coating the polymer chains are mobile such that they can effectively minimize the stresses set up, but after solidification and/or below the glass transition temperature chain mobility is decreased and the stresses become 'frozen' in.

Generally cracking or splitting will occur when the internal stress approaches or exceeds the cohesive strength as indicated by the tensile strength, σ, of the film.

$$\text{i.e. } P \geqslant \sigma \quad .. \quad .. \quad (4)$$

Combining equations 2, 3 and 4. For cracking to occur

$$\frac{\sigma}{E} \leqslant \frac{1}{1-\nu}\left[\frac{\phi_s - \phi_r}{3(1-\phi_r)} + \frac{\Delta\alpha\,\Delta t}{1+\nu}\right] \quad (5)$$

Unfortunately accurate values for all the parameters in the equations are not available. However, it is possible to solve these equations using estimations based on previously published data. Consider, for example, a 35 μm thick film of hydroxypropyl methylcellulose containing 20% plasticizer applied to a tablet core consisting primarily of lactose and stored at ambient conditions (i.e. 25 °C and 50% relative humidity). For this example E and σ will be of the order $0{\cdot}9 \times 10^4$ MPa and 37 MPa respectively (Pickard 1979) with a Poisson's ratio of 0·35, α_s and α_c will be of the order of $2{\cdot}8 \times 10^{-4}$ and $3{\cdot}5 \times 10^{-4}$ respectively (Rowe 1980), Tg will be of the order of 129 °C (Entwistle & Rowe 1979) and ϕ_r will be of the order of 0·09 (calculated by reference to hydroscopity measurements on hydroxypropyl methyl cellulose). The main problem would appear to be in the identification of ϕ_s. Croll (1979) has suggested that it can be identified as the solvent concentration at which the glass transition temperature of the system corresponds to ambient temperature and has presented data on two polymer/solvent systems where $\phi_s = 0{\cdot}16$—$0{\cdot}17$ to show the validity of this approach. If it is assumed that ϕ_s will be 0·16 for the hydroxypropyl methyl cellulose/water system used in this example (a not unreasonable assumption since it is known that there is a marked inflection in the water uptake curve for this polymer at a similar concentration), then P_s and P_T can be calculated as 35·5 MPa and 7·4 MPa respectively. It is interesting to note that these values are reasonable in the light of quoted values for other organic coatings applied to a variety of substrates (Sato 1980), and that while individually they do not exceed the tensile strength of the film, when summed to obtain the total internal stress P, they just exceed it. In this example, therefore, cracking in the film could be a problem as confirmed by scanning electron photomicrography (Fig. 1).

Practical implications

If during product development cracking of the film occurs, the formulator has two courses of action; either to attack the root cause of the problem and to minimize the internal stresses or to accept the presence of these stresses and to formulate so as to minimize the incidence of the defect. These two actions are interrelated as will become apparent on discussing the various aspects of the formulation the formulator has under his control, i.e. the choice of the excipients used in the tablet core formulation, the type and concentration of the plasticizer, the solvent system and the addition of additives in the form of pigments, opacifiers etc.

Table core formulation

The only factor in the equations directly affected by the tablet core formulation is $\Delta\alpha$, the difference between the thermal expansion coefficients of the substrate and film coating. Data recently compiled on the expansion coefficients of some materials representative of the excipients used in both tablet core and film coating formulations have shown the wide variability likely to be encountered (Rowe 1980). Although these data cannot be regarded as directly comparable with that obtained from complete formulations it does enable trends to be predicted and compared with practical circumstances, e.g. if in the example given above the core formulation was changed to one based on magnesium carbonate where α_s is of the order of $0{\cdot}5 \times 10^{-4}$ then P_T will increase by some 4·3 times to a value of 31·8 MPa such that P would be nearly twice the value of the tensile strength of the film. In this case it is likely that catastrophic cracking of the film would occur as has been shown in practice for a similar formulation by Rowe & Forse (1980).

Polymer grade

In a recent study (Rowe & Forse 1980) it was shown that the incidence of edge splitting and peeling on a tablet coated with hydroxypropyl methyl cellulose

could be decreased on increasing the viscosity grade or molecular weight of the polymer. This result was interpreted in terms of the consequential increase in the tensile strength of the film. From examination of equation 5, where in this special case the right hand side of equation is a constant, it can be seen that the incidence of cracking will be related to the ratio σ/E, the higher this ratio the lower the incidence of cracking. This concept has been applied using recently available data on the tensile strength and Young's modulus of a variety of grades of hydroxypropyl methyl cellulose (Pickard 1979) and comparing the values of σ/E to the incidence of edge splitting and peeling seen with these grades (applied as 5% aqueous solutions) using the same tablet core and coating formulation as used by Rowe & Forse (1980). The results are summarized in Table 1. It can be seen that if the incidence of edge splitting was directly related to the tensile strength alone it would be predicted that the incidence of this defect would be highest for Pharmacoat 615. This is not the case, the ratio of tensile strength to Young's modulus gives a much better prediction.

Plasticizer type and concentration

Plasticizers are known to have a beneficial effect on the incidence of cracking on film coated tablets (data on hydroxypropyl methyl cellulose phthalate—Shinetsu Chemical Co., Japan), and yet are known to cause a decrease in the tensile strength of films (Greminger & Savage 1959). The equations given above provide an insight in these apparently contradictory observations.

Table 1. σ/E ratios and the incidence of edge splitting and peeling on a tablet coated with various grades of hydroxypropyl methyl cellulose.

Grade	Tensile strength* σ MPa	Young's modulus* E MPa	$\sigma/E \times 10^{-3}$	Incidence of edge splitting %
Pharmacoat 606	49·5	2040	2·43	6·5
Pharmacoat 615	48·6	1912	2·54	3·8
Methocel E15	51·2	1979	2·58	1·8
Methocel E50	66·4	2515	2·64	0

* Data from Pickard (1979).

Plasticizers act by interposing themselves between the polymer chains and interact with the forces holding the chains together thereby extending and softening the polymer matrix. They thus cause a decrease in both Young's modulus and the glass transition temperature of the film, effectively reducing the internal stress (eqns 2 and 3) and decreasing the incidence of cracking. The efficiency with which this occurs depends not only on the plasticizer concentration but also on its degree of interaction with the polymer—a plasticizer with a higher interaction will cause a greater lowering in the glass transition temperature than an equivalent concentration of one with a poor interaction (Entwistle & Rowe 1979). The addition of plasticizer also tends to cause an increase in the σ/E ratio relative to the amount of plasticizer added (Pickard 1979).

Solvent systems

The solvent system is a very important factor in influencing the magnitude of the P_R since it is the solvent which governs the concentration at which solidification occurs. In general it can be stated that polymer solutions gel at higher dilutions (i.e. ϕ_s increases) the poorer the solvent. In a poor solvent there is much more intermolecular association but in a good solvent the polymer chains are free to move and are independent of each other to a much later time during solvent evaporation. The mechanical properties of films are also dependent on the solvent system used, the poorer the solvent (as indicated by a lower intrinsic viscosity value) the lower the tensile strength and Young's modulus (Table 2) and the lower the ratio σ/E. This is probably the reason for the cracking observed during coating of tablets with hydroxypropyl methyl cellulose phthalate in various solvent mixtures con-

Table 2. The effect of solvent on the mechanical properties of films of hydroxypropyl methyl cellulose (data from Pickard 1979) and ethyl cellulose (data from Haas et al 1952).

Polymer	Solvent	Intrinsic viscosity $[\eta]$ dl g^{-1}	Young's modulus E MPa	Tensile strength σ MPa	Ratio $\times 10^{-3}$ σ/E
Hydroxypropyl methyl cellulose (Methocel E50)	Dichloromethane 80% w/w + Methanol 20% w/w	1·03	2126	55·3	2·60
" "	Water	1·23	3515	66·9	2·67
Ethyl cellulose (Grade N100)	Benzene	1·57	1868	59·0	3·16
	Chlorobenzene	1·52	1689	51·8	3·07

taining high proportions of isopropanol since it is known that this solvent is a very poor solvent for this polymer producing very poor quality films (Manufacturer's data—Shinetsu Chemical Co. Japan).

Pigments and additives

In general it can be stated that the addition of pigments or insoluble additives to films causes an increase in the Young's modulus of the film but a decrease in its tensile strength. It also causes an increase in the glass transition temperature resulting in an increase in both P_S and P_T (eqns 2 and 3). However, extensive work in the field of organic coatings has shown that the increase in the internal stress varies not only with the pigment or additive concentration but also with the type of material added. Of pharmaceutical interest is the work on benzyl cellulose (Ioune 1943) which showed that at constant pigment concentration the internal stress was highest in the formulation containing titanium dioxide compared with those containing either calcium carbonate or talc. In fact, in the case of talc, there was no increase in the internal stress with concentration. This effect is thought to be due to the differences in the particle morphology of the individual pigments since talc with its flake-like structure will arrange itself parallel to the substrate resulting in a restraint of volume shrinkage parallel to the plane of the coating.

It is also possible that differences in the thermal expansion coefficients of both the pigment or additive and the polymer will result in localized thermal stresses being set up around the pigment or additive particles. These stresses are extremely important since they can act as nuclei for crack propagation when a further external stress is imposed, e.g. as a result of the tablet striking a baffle in the coating pan.

Conclusion

The equations presented provide an insight into the mechanism of cracking of film coatings on coated tablets and the factors which affect it. The equations do not, however, take into account other stresses induced on storage e.g. molecular rearrangement—as has been shown to occur in cellulose acetate phthalate films (Delporte 1979)—shrinkage due to loss of plasticizer or swelling of the tablet core due to moisture uptake. These stresses will be dependent on storage conditions and hence are only likely to be able to be determined by experiment.

REFERENCES

Croll, S. G. (1979) J. Appl. Polym. Sci 23: 847–858

Delporte, J. P. (1979) Pharm. Ind. 41: 984–990

Entwistle, C. A., Rowe, R. C. (1979) J. Pharm. Pharmacol. 31: 269–272

Greminger, G. K., Savage, A. B. (1959) in: Whistler, R. L. (ed.) Industrial Gums—Polysaccharides and their Derivatives. Academic Press, pp 565–596

Haas, H. C., Farney, L., Vale, C. (1952) J. Colloid Sci. 1: 584–599

Ioune, Y. (1943) Kogyo Kagaku Zasshi 46: 784—taken from Sato (1980)

Pickard, J. F. (1979) Ph.D. thesis. Council for National Academic Awards

Rowe, R. C. (1980) J. Pharm. Pharmacol. 32: 851

Rowe, R. C., Forse, S. F. (1980) Ibid. 32: 583–584

Sato, K. (1980) Progr. Org. Coatings 8: 143–160