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Adhesion of film coatings to tablet surfaces — a theoretical approach based on solubility parameters

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Summary

Based on the assumption that tablet/film adhesion is due entirely to the summation of the interactions of the intermolecular bonding forces in a perfectly bonded system, it is possible to derive equations to predict both the interaction parameter and ideal butt adhesive strength using solubility parameters readily available in the literature. The trend predicted for the adhesion of hydroxypropyl cellulose to a series of low-energy polymer substrates is consistent with that shown by Johnson and Zografi (1986). The trends predicted for the adhesion of a series of cellulose derivatives to both microcrystalline cellulose and anhydrous lactose are consistent with those seen in practice.

Introduction

A major prerequisite for pharmaceutical film coatings is good adhesion to the tablet substrate. Previous studies in tablet/film adhesion (Wood and Harder, 1970; Nadkarni et al., 1975; Fisher and Rowe, 1976; Fung and Parrott, 1980; Porter, 1980; Okamafe and York, 1985) have tended to concentrate on the method of measurement and its applicability in the study of the effect of film-forming variables with only a few concentrating on substrate variables (Rowe, 1977, 1978). Generally the experimental work that has been reported has been inconclusive in demonstrating any quantitative relationship between the measured adhesion and the surface energetics of the system. However, in a recent study, Johnson and Zografi (1986), using a well-controlled butt adhesion test where the film is detached normal to the substrate surface, have demonstrated that, for a system involving hydroxypropyl cellulose cast or sprayed onto several polymer substrates, the measured adhesion extrapolated to zero thickness did reflect the surface energetics of the system. Unfortunately the equations used by these workers involved a knowledge of polar and non-polar components of the interfacial tension at the polymer film–vapour interface and the substrate–vapour interface and although these data were available for the polymer substrates studied they are not generally reported in the literature. In this paper a similar approach is proposed based on solubility parameters, on which more data are readily available (Barton, 1983).

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Theoretical considerations

In 1967 Gardon proposed an equation for calculating the ideal tensile (butt) adhesive strength ($\sigma_{\max}$) between two slabs of material (A and B) assumed to be separated by a plane of a thickness (λ_{AB}) corresponding to the equilibrium distance at zero potential energy:

$$\sigma_{\max} = \frac{1.03 W_{AB}}{\lambda_{AB}} \tag{1}$$

where W_{AB} is the work of adhesion. The equation is directly analogous to that used by Johnson and Zografi (1986). This equation may be rewritten in terms of the solubility parameters ${}^{A}\delta$ and ${}^{B}\delta$ (measured in units of MPa$^{1/2}$) of the two materials thus obviating the need for calculating W_{AB} (Gardon, 1967):

$$\sigma_{\max} = 0.25\phi\,{}^{A}\delta\,{}^{B}\delta \tag{2}$$

where ϕ is the so called interaction parameter (Girifalco and Good, 1957) for which Wu (1973) derived the equation:

$$\phi = 2\left[\frac{{}^{A}x_{d}\cdot{}^{B}x_{d}}{{}^{A}x_{d}\cdot g_1 + {}^{B}x_{d}\cdot g_2} + \frac{{}^{A}x_{p}\cdot{}^{B}x_{p}}{{}^{A}x_{p}\cdot g_1 + {}^{B}x_{p}\cdot g_2}\right] \tag{3}$$

where x_d and x_p are the fractional non-polarity and polarity of each material and g_1 and g_2 are constants defined by the ratios of their surface energies, γ, i.e.,

$$g_1 = \frac{{}^{A}\gamma}{{}^{B}\gamma} \quad \text{and} \quad g_2 = \frac{{}^{B}\gamma}{{}^{A}\gamma} \tag{4}$$

These constants may be equally determined from solubility parameters using the relationship (Gardon 1967)

$$\gamma = \frac{\delta^2 V^{1/3}}{K} \tag{5}$$

where V is the molar volume and K is a constant. Thus from equations (4) and (5)

$$g_1 = \frac{{}^{A}\delta^2 \cdot {}^{A}V^{1/3}}{{}^{B}\delta^2 \cdot {}^{B}V^{1/3}} \tag{6}$$

and

$$g_2 = \frac{1}{g_1} = \frac{{}^{B}\delta^2 \cdot {}^{B}V^{1/3}}{{}^{A}\delta^2 \cdot {}^{A}V^{1/3}} \tag{7}$$

Although Wu (1973) also defined x_p and x_d in terms of the polar and non-polar components of the surface energy, these may also be calculated from the dispersion or non-polar component of Hansen's 3-dimensional solubility parameter (Hansen, 1967), i.e.,

$${}^{A}x_{d} = \left[\frac{{}^{A}\delta_{d}}{{}^{A}\delta}\right]^2 \tag{8}$$

and

$${}^{A}x_{p} = 1 - \left[\frac{{}^{A}\delta_{d}}{{}^{A}\delta}\right]^2 \tag{9}$$

It should be noted that although Hansen defined his partial solubility parameters by the equation:

$$\delta^2 = \delta_d^2 + \delta_p^2 + \delta_h^2 \tag{10}$$

where δ_p and δ_h are the polar (dipole/dipole interaction) and hydrogen bonding components respectively, x_p cannot be calculated using δ_p alone since this would neglect any additive effect of hydrogen bonding.

Discussion

Values for the Hansen partial solubility parameter of a range of materials of interest in film coating including those polymers used as substrates by Johnson and Zografi (1986) are shown in Table 1. Unfortunately no data exist for hydroxypropyl cellulose other than its solubility parameter (24.9 MPa$^{1/2}$; Sakellariou et al., 1986)

TABLE 1

Molar volumes, Hansen solubility parameters and fractional polarities of various polymers and substrates

Material	Molar volume V ($cm^3 \cdot mol^{-1}$)	Solubility parameter				Fractional polarity	
		δ	δ_d	δ_p	δ_h	x_p	x_p
		($MPa^{1/2}$)				(Eqn. 9)	(Surface tension data)
Polymethylmethacrylate [1]	85.6	22.6	18.6	10.5	7.5	0.32	0.27 [4], 0.28 [5]
Polyethylene terephthalate [1]	144.5	22.1	19.0	8.1	6.0	0.26	0.26 [4]
Polyethylene [1]	32.8	17.6	17.6	0	0	0	0.09 [4]
Polyvinylchloride [1]	45.1	21.4	18.2	7.5	8.3	0.28	0.15 [5]
Polyvinyl acetate [1]	72.4	25.6	20.9	11.3	9.6	0.33	0.33 [5]
Cellulose acetate [1]	860.5	25.1	18.6	12.7	11.0	0.45	
Ethyl cellulose [1]	749.6	20.6	16.7	2.9	11.7	0.34	
Microcrystalline cellulose [2]	216.0	39.3	19.4	12.7	31.3	0.76	
Anhydrous lactose [3]	236.8	39.9	19.6	26.2	23.1	0.76	

[1] Data ex Barton (1983).
[2] Data ex Phuoc et al. (1987).
[3] Date ex Phuoc et al. (1986)
[4] Data ex Johnson and Zografi (1986).
[5] Data ex Wu (1973).

and molar volume (559.0 cm^3 mol^{-1}; calculated). However, if it is assumed that the fractional polarity for this polymer calculated from the surface tension data of Johnson and Zografi (1986) of 0.4 is correct (a valid assumption considering the good agreement between the fractional polarity values calculated using Eqn. 9 and those given in the literature) then it is possible to calculate ideal butt adhesive strengths for the same systems as used by Johnson and Zografi (1986). Table 2 shows values for the interaction parameter and ideal butt adhesive strength for these systems. As expected the calculated values of the ideal butt adhesive strength are orders of magnitude greater than the measured adhesion but the trend predicted, i.e. polyethylene terephthalate > polymethylmethacrylate > polyethylene, is as measured by Johnson and Zografi (1986).

The value of the interaction parameter is important since it provides information on the possible mode of failure of the system, i.e. interfacial or

TABLE 2

Correlation of the interaction parameter (ϕ) and the estimated ideal butt adhesive strength (σ_{max}) with measured adhesion data for the hydroxypropyl cellulose film/polymer substrate combinations used by Johnson and Zografi (1986)

Polymer	ϕ	σ_{max} (MPa)	Measured adhesion (kPa)	
			Cast film	Sprayed film
Polyethylene terephthalate	0.78	107.3	451.1	–
Polymethylmethacrylate	0.74	104.1	402.1	470.7
Polyethylene	0.36	39.4	313.8	294.2

TABLE 3

Predicted interaction parameters (ϕ) and ideal butt adhesive strengths (σ_{max}) for various polymer film/substrate combinations

Substrate	Film	ϕ	σ_{max} (MPa)
Microcrystalline cellulose	cellulose acetate	0.82	202.2
Microcrystalline cellulose	hydroxypropyl cellulose	0.74	181.0
Microcrystalline cellulose	ethyl cellulose	0.64	129.5
Anhydrous lactose	cellulose acetate	0.80	200.3
Anhydrous lactose	hydroxypropyl cellulose	0.72	178.8
Anhydrous lactose	ethyl cellulose	0.62	126.2

cohesive within the weaker component. If the interaction parameter is close to unity then interfacial failure in a perfectly bonded system will not be possible because the interfacial bond strength would be greater than the tensile strength of the weaker component. If the interaction parameter is significantly lower than unity, as in the 3 systems in Table 2, interfacial failure will always occur even in perfectly bonded systems. Johnson and Zografi (1986) reported that visual inspection which had been previously validated by a colorimetric detection test confirmed interfacial failure with these 3 systems.

Application of the equations to systems involving microcrystalline cellulose and anhydrous lactose coated with the cellulose derivatives, hydroxypropyl cellulose, ethyl cellulose and cellulose acetate yield some interesting trends (Table 3). In all cases the microcrystalline cellulose yielded higher interaction parameters and higher ideal butt adhesive strengths. This trend is one that has been shown to occur in real systems (Rowe, 1977, 1978). In addition, the trend of cellulose acetate > ethyl cellulose has been found for a variety of substrates in our laboratories.

While, at best, the equations derived above can do nothing more than predict trends in film/tablet adhesion, the data presented clearly illustrate the potential of this kind of approach. Data on the solubility parameters of excipients used both in tablet formulation (Phuoc et al., 1986, 1987) and in film coating (Matsuura et al., 1976; Rowe, 1986; Sakellariou et al., 1986) are now becoming more available and further data can be easily calculated from group molar attraction constants readily available in the literature (Barton, 1983).

References

Barton, A.F.M., *Handbook of Solubility Parameters and other Cohesion Parameters*, CRC, Baton Rouge, FL, 1983.

Fisher, D.G. and Rowe, R.C., The adhesion of film coatings to tablet surfaces—Instrumentation and preliminary evaluation. *J. Pharm. Pharmacol.*, 28 (1976) 886–889.

Fung, R.M. and Parrott, E.L., Measurement of film-coating adhesiveness. *J. Pharm. Sci.*, 69 (1980) 439–441.

Gardon, J.L., Variables and interpretation of some destructive cohesion and adhesion tests. In Patrick, R.L. (Ed), *Treatise on Adhesion and Adhesives, Vol. 1*, M. Dekker, New York, 1967, pp. 269–324.

Girifalco, L.A. and Good, R.J., A theory for the estimation of surface and interfacial energies. I Derivation and application to interfacial tension. *J. Phys. Chem.*, 61 (1957) 904–909.

Hansen, C.M., The three dimensional solubility parameter–key to paint component affinities. I. Solvent, plasticizers, polymers and resins. *J. Paint Technol.*, 39 (1967) 104–117.

Johnson, B.A. and Zografi, G., Adhesion of hydroxypropyl cellulose films onto low energy solid substrates. *J. Pharm. Sci.*, 75 (1986) 529–533.

Matsuura, T., Blais, P. and Sourirajan, S., Polar and nonpolar parameters for polymeric reverse osmosis membrane materials from liquid chromatographic data. *J. Appl. Polym. Sci.*, 20 (1976) 1515–1531.

Nadkarni, P.D., Kildsig, K.O., Kramer, P.A. and Banker, G.S., Effect of surface roughness and coating solvent on film adhesion to tablets. *J. Pharm. Sci.*, 64 (1975) 1554–1557.

Okamafe, A.O. and York, P., The adhesion characteristics of some pigmented and unpigmented aqueous-based film coatings applied to aspirin tablets. *J. Pharm. Pharmacol.*, 37 (1985) 849–853.

Phuoc, N.H., Phan Tan Luu, R., Munafo, A., Ruelle, P., Hô Nam-Tran, Buchmann, M. and Kesselring, U.W., Determination of partial solubility parameters of lactose by gas-solid chromatography. *J. Pharm. Sci.*, 75 (1986) 68–72.

Phuoc, N.H., Hô Nam-Tran, Buchmann, M. and Kesselring, U.W., Experimentally optimized determination of the partial and total cohesion parameters of an insoluble polymer (microcrystalline cellulose) by gas-solid chromatography. *Int. J. Pharm.*, 34 (1987) 217–223.

Porter, S.C., The effect of additives on the properties of an aqueous film coating. *Pharm. Tech.*, 4 (1980) 67–75.

Rowe, R.C., The adhesion of film coatings to tablet surfaces — the effect of some direct compression excipients and lubricants. *J. Pharm. Pharmacol.*, 29 (1977) 723–726.

Rowe, R.C., The measurement of the adhesion of film coatings to tablet surfaces – the effect of tablet porosity, surface roughness and film thickness. *J. Pharm. Pharmacol.*, 30 (1987) 669–672.

Rowe, R.C., The prediction of compatibility/incompatibility in blends of ethyl cellulose with hydroxypropyl methyl cellulose or hydroxypropyl cellulose using 2-dimensional solubility parameter maps. *J. Pharm. Pharmacol.*, 38 (1986) 214–215.

Sakellariou, P., Rowe, R.C. and White, E.F.T., The solubility parameters of some cellulose derivatives and polyethylene glycol used in tablet film coating. *Int. J. Pharm.*, 31 (1986) 175–177.

Wood, J.A. and Harder, S.W., The adhesion of film coatings to the surfaces of compressed tablets. *Can. J. Pharm. Sci.*, 5 (1970) 18–23.

Wu, S., Polar and nonpolar interactions in adhesion. *J. Adhesion*, 5 (1973) 39–55.