ELSEVIER

International Journal of Pharmaceutics 227 (2001) 113–119

international journal of pharmaceutics

www.elsevier.com/locate/ijpharm

The influence of additives on the spreading coefficient and adhesion of a film coating formulation to a model tablet surface

Heena Khan [a], John T Fell [a,*], Graeme S Macleod [b]

[a] *School of Pharmacy and Pharmaceutical Sciences, University of Manchester, Manchester M13 9PL, UK*
[b] *Manesty, Evans Road, Speke, Liverpool, L24 9LQ, UK*

Received 6 February 2001; received in revised form 20 April 2001; accepted 21 April 2001

Abstract

The surface energies of film coating formulations based on hydroxypropyl methylcellulose and containing microcrystalline cellulose, lactose and Tween 20, respectively, have been assessed. The approach taken allowed the components of the surface energy, in terms of the Lifshitz–van der Waals and the acid-base contributions, to be determined. Spreading coefficients of these coating formulations were determined on a model tablet surface whose surface energy had been similarly characterised. The determined spreading coefficients were high and positive indicating that spreading and wetting would not be a controlling factor in the formation of an adequate film coat. The adhesion of the coats to the core was measured and showed that the inclusion of additives influenced the adhesion of the film. Maximum adhesion was obtained when microcrystalline cellulose was included in the coating formulation that presumably allowed a strong interaction with the same component in the tablet core. Adhesion was enhanced when the tablet cores were made at a higher compaction force. Atomising air pressure had little influence on the adhesion.

Keywords: Tablet coating; Adhesion; Surface energetics; Spreading coefficients

1. Introduction

Irrespective of the purpose for which a film coat is applied to a tablet core, one of the requirements for success is good adhesion of the coat to the tablet. This adhesion can be influenced by the properties of the coating formulation, as well as those of the tablet (Rowe, 1977). The prerequisite for good adhesion is the spreading of the atomised droplets over the surface of the tablet, and limited penetration of the coating solution into the pores of the tablet (Aulton, 1995). Both of these are controlled by the surface energetics of the tablet and the coating solution.

It is now recognised that interactions between solids and liquids, as in the case of spreading, can

* Corresponding author. Tel.: +44-161-275-2365; fax: +44-161-273-2396.
E-mail address: jfell@fs1.pa.man.ac.uk (J.T. Fell).

0378-5173/01/$ - see front matter
PII: S0378-5173(01)00789-X

be explained by analysing the surface energy in terms of the apolar and acid–base components of the total energy. The theory underlying this approach has been adequately explained in the literature (van Oss et al., 1988a,b). Spreading coefficients, determined in this manner, have proved useful in explaining such diverse interactions as adhesion to container walls (Buckton and Chandraia, 1993) and mucoadhesion (Rillosi and Buckton, 1995), and have been applied to the surface energies of ethylcellulose films (Oh and Luner, 1999).

Spreading coefficients determined in this way are essentially representative of the interactions between the components of the surface energies. Positive values indicate spontaneous spreading with higher values favouring the process. Coating formulations consist of a polymer and additives including pigments, such as titanium dioxide, plasticisers, such as polyethylene glycol, or materials such as lactose, microcrystalline cellulose or surfactants. The influence of these materials on the adhesion of the film to the tablet is variable, depending inter alia on material type and particle size (Felton and McGinity, 1997, 1999). The influence of different additives on the surface energetics of the coating formulations has not been examined.

The purpose of the work, reported in this paper, is to determine the spreading coefficients, using the acid–base approach, of hydroxypropyl methylcellulose solutions containing additives, on a model tablet surface. These values will then be related to the adhesion of the film to the tablet. Additionally, surface roughness and droplet size values are taken into account.

2. Materials and methods

2.1. Materials

The materials used in the preparation of the tablet cores were microcrystalline cellulose (Avicel PH 102, FMC, USA), lactose (Tablettose 80, Meggle, Germany), magnesium stearate (BDH, UK) and colloidal silicon dioxide (Aerosil 200, Degussa, Germany). The coating formulations consisted of hydroxypropyl methylcellulose (Pharmacoat 606, Shin-Etsu Chemical Co. Japan), polyethylene glycol 400 (BDH, UK), microcrystalline cellulose (PH 105, FMC, USA), lactose (Tablettose 80, Meggle, Germany) and Tween 20 (Sigma, UK). The formulations were prepared in glass distilled water. To determine the surface energy components of the materials, the liquids used were di-iodomethane (Sigma, UK), glycerol (BDH, UK) and glass distilled water. The solids were polytetrafluoroethylene and polyvinyl chloride (both RS Components, UK).

2.2. Tablet preparation

Tablets consisting of 75.2% microcrystalline cellulose (MCC), 24.2% lactose, 0.4% magnesium stearate and 0.2% colloidal silicon dioxide (all w/w) were manufactured using a rotary tablet machine (Unipress Diamond, Manesty, UK) equipped with flat, bevelled-edge punches, 10mm in diameter. Two batches of tablets with average breaking loads of 127 N and 191 N were produced. Each tablet so produced had an average weight of 360 mg.

2.3. Film coating of tablets

Four coating formulations consisting of a) 9% hydroxypropyl methylcellulose (HPMC), 1% PEG 400; b) 9% HPMC, 1% PEG 400, 2% MCC; c) 9% HPMC, 1% PEG 400, 2% lactose; d) 9% HPMC, 1% PEG 400, 0.5% Tween 20 have been used for coating studies. Two kilogram batches of both sets of tablets were coated in a perforated drum coater (AccelaCota 10, Manesty, UK), modified using a base plate to reduce working capacity, and fitted with a Manesty spray gun. The drum speed was kept at a constant 15 rpm, the spray rate was 14 g/min, the gun distance was 20 cm, and the fan air pressure was set at 0.4 bar. Two atomising air pressures were used (0.5 and 2.0 bar) to coat two batches for both sets of tablets. For each coating run, samples were coated to a theoretical weight gain of 4%.

2.4. Viscosity determination

The viscosities of the four coating formulations were measured at 20 °C using a Brookfield LV-8 viscometer (Brookfield, USA).

2.5. Droplet size determination

Droplet sizes for each of the coating formulations were measured at room temperature using a Malvern Spraytec Droplet Size Analyser (Malvern, UK). For each of the coating formulations, droplet sizes were measured for two atomising air pressures of 0.5 bar and 2.0 bar, at a constant spray rate of 14 g/min, gun distance of 20 cm, and fan air pressure 0.4 bar. Measurements were taken in triplicate.

2.6. Adhesion testing

The adhesion of the films to the tablet cores was tested using a specially designed tablet tester (Force Measurement Systems, Glasgow, UK). The instrument principally consists of a 125 N load cell capable of measuring the force required to remove the film coat perpendicular to the tablet surface. For each set of tablets, ten tablets were selected within 0.5% of the respective theoretical weight gain to minimise variation in film thickness. The film coating at the bevelled edge of the tablet under test was carefully removed using a sharp scalpel, taking care not to disturb the remaining film coat. The tablet was mounted horizontally into the lower stationary tablet holder. Double-sided adhesive tape (RS Components, UK) was affixed to the upper moving arm and lowered to the surface of the tablet at a constant speed. The upper moving arm was then raised at a constant speed of 10 mm/min. The force required and the deflection profile generated was recorded. A personal computer recorded the development of the force (N) and the displacement (mm) at the tablet–film interface during the pull test.

2.7. Surface roughness

Measurements of surface roughness were made using a PneumoSurtronic 3 (Taylor Hobson, UK). A cut of length of 0.80 mm (Lc) and an assessment length of 4.0 mm (Ln) were used. Measurements were taken across the diameter of ten tablets for each of the tablet batches.

2.8. Surface tension of the coating formulations

Surface tension values have been determined by the Wilhelmy plate method using a platinum plate suspended from a microbalance (Cahn Electrobalance, USA). The mean surface tension for each formulation was calculated from an average of three readings taken from each of three separately prepared formulations.

2.9. Contact angles and spreading coefficients

For the determination of the spreading coefficients, the test liquids used were distilled water, di-iodomethane and glycerol, the di-iodomethane being the apolar liquid.

Contact angles were measured using a sessile drop technique. The drops were viewed through a microscope fitted with a protractor eyepiece, the microscope and tablet holder being mounted on an optical bench. Mean contact angles were calculated from ten measurements taken within 10 s, using 20 μl of the liquid under test, dropped from a height of 5 mm. Calculation of the spreading coefficients was performed according to Good et al. (1991).

3. Results

The surface energies of the coating formulations are shown in Table 1. The approach used in determining the surface energies allows subdivision of the total energy into γ^{LW}, the contribution from Lifshitz–van der Waals component and γ^{AB}, the acid–base component. The latter is further subdivided into the Lewis acid (electron acceptor) parameter, γ^{+}, and the Lewis base (electron donating) parameter, γ^{-}. For all the coating formulations, the γ^{AB} component makes only a small contribution to the total surface energies. The surface energies of the tablets are shown in Table

2. The γ^{AB} component is relatively small though higher than that for the liquids. There is a change in the surface energy values as the compaction force used to make the tablets increases.

The contact angles exhibited by the coating formulations on the tablets are shown in Table 3, which also shows the spreading coefficients calculated using the approach of Good et al. (1991).

The inclusion of additives changes the contact angle of the coating formulation to a limited extent. The spreading coefficients are all high and positive, indicating effective spreading of the coating formulations on the surfaces of the tablets.

The adhesion of the film coats to the tablets, expressed as the maximum force required to remove the films, is shown in Table 4. The low variability between replicate measurements implies that the defects were not introduced during the removal of the film from the bevelled edge of the tablets. For HPMC, alone and with the inclusion of lactose and Tween, atomising air pressure has no significant effect on the adhesion of the film. Atomising air pressure does have a significant effect on the adhesion when MCC is included as an additive, and in all cases, compaction pressure significantly influences the measured adhesion. The physical properties of the coating solutions in terms of the viscosity and droplet size on spraying are given in Tables 5 and 6. The surface roughness values of the tablets are 2.7 μm ($\pm$ 0.10, $n = 10$) for those prepared at 127 N force and 1.74 μm($\pm$ 0.004, $n = 10$) for those prepared at 191 N.

Table 1
Surface energy components (mJ/m^2) of the coating formulations

Coating formulation	γ^{TOT}	γ^{LW}	γ^{AB}	γ^-	γ^+
HPMC	45.6	44.3	1.3	0.05	7.91
MCC	45.6	45.2	0.3	3.8E−3	7.20
Lactose	45.6	44.7	0.9	0.03	6.96
Tween	41.6	41.0	0.6	0.01	6.48

Table 2
Surface energy components of the tablets

Tablet breaking load (N)	γ^{TOT}	γ^{LW}	γ^{AB}	γ^-	γ^+
127	54.7	47.7	7.0	51.1	0.2
191	51.8	45.9	6.0	51.7	0.2

Table 3
Contact angles (degrees) and spreading coefficients (mJ/m^2) for the coating formulations on the tablets

Coating formulation	127 N		191N	
	Contact angle	SC	Contact angle	SC
HPMC	43 $\pm$ 0.48	41.2	46 $\pm$ 0.47	39.6
MCC	54 $\pm$ 0.34	40.2	54 $\pm$ 0.32	38.6
Lactose	50 $\pm$ 0.37	39.0	51 $\pm$ 0.52	37.4
Tween	53 $\pm$ 0.93	41.8	52 $\pm$ 0.47	40.2

SC = Spreading coefficient; contact angle = mean ($n = 10$, $\pm$ SD).

Table 4
Maximum adhesive strength of the film coats applied under different conditions

Tablet and coating conditions	Adhesion (kN/m^2)	Std. error of mean
HPMC 127N 0.5 AAP	4.09	±0.63
HPMC 127N 2.0 AAP	8.65	±2.04
HPMC 191N 0.5 AAP	115.53	±13.36
HPMC 191N 2.0 AAP	110.50	±12.42
LACTOSE 127N 0.5 AAP	12.10	±2.83
LACTOSE 127N 2.0 AAP	15.09	±2.20
LACTOSE 191N 0.5 AAP	69.48	±11.32
LACTOSE 191N 2.0 AAP	61.46	±12.10
MCC 127N 0.5 AAP	18.86	±4.87
MCC 127N 2.0 AAP	65.23	±13.83
MCC 191N 0.5 AAP	211.89	±25.62
MCC 191N 2.0 AAP	163.32	±26.25
TWEEN 127N 0.5 AAP	5.03	±1.10
TWEEN 127N 2.0 AAP	12.58	±0.94
TWEEN 191N 0.5 AAP	99.82	±17.29
TWEEN 191N 2.0 AAP	98.87	±9.12

AAP = atomising air pressure; WG = weight gain on coating.

Table 5
Viscosities of the coating formulations

Coating formulation	Viscosity (mPa•s)
HPMC	191
MCC	227
Lactose	217
Tween	197

4. Discussion

The determination of the surface energetics of powdered systems is complex. Each method has its drawbacks, particularly the sessile drop method on a compacted powder as used in the current study. Despite this, it is the appropriate method for this study, as determinations are made on the compact that is then subjected to the coating procedure. The change in surface energy with compaction pressure (Table 2) has been noted previously (Buckton and Newton, 1986) and may be related to a change in the surface characteristics of the material caused by the compaction pressure. Again, it is this surface that will interact with the coating fluid.

The surface energies of the coating formulations are determined from contact angle measurements against solid polyvinyl chloride where measurement using the sessile drop technique is not so contentious. The results (Table 1) show that the total surface energy of the coating formulations is essentially accounted for by the γ^{LW} component. The liquid surfaces are exhibiting monopolar behaviour due to saturation by HPMC and the inclusion of additives makes little difference to this. The tablet surfaces, on the other hand, exhibit a small contribution from the γ^{AB} component, displaying bipolar behaviour due to contributions from the microcrystalline cellulose and lactose.

The adhesion of the coat to the tablet will depend on a complex set of interacting factors related to the coating formulation, the tablet core and processing conditions. A primary requirement is that the coating formulation spreads completely over the surface of the tablet and adhesion will be enhanced if some penetration into the pores of the tablet takes place. Processing conditions aside, these effects will be controlled by the interaction

Table 6
Sauter mean diameters of sprays using different coating formulations and atomising air pressures

Coating formulation	AAP	D(3,2)[a] μm
HPMC	0.5	39.9
HPMC	2.0	15.8
Lactose	0.5	38.6
Lactose	2.0	14.6
MCC	0.5	30.4
MCC	2.0	12.9
Tween	0.5	34.9
Tween	2.0	13.9

[a] D(3,2) = Sauter mean diameter.

of the fluid with the tablet core, and the high, positive spreading coefficients determined for the current tablets and coating formulations indicate that complete wetting of the tablet cores will take place. The inclusion of additives to the coating formulations does not significantly change the spreading coefficients and thus, changes in adhesion are not due to alterations in wetting effects. The similarities in the viscosities of the fluids (Table 5) implies that this is not a controlling factor in spreading, although the evaporation of droplets during their passage from the spray nozzle to the tablet bed will inevitably change the liquid viscosity.

The inclusion of additives does, however, influence adhesion (Table 4), with the type of additive, atomising air pressure and compaction pressure all showing significant effects. The magnitude of the adhesion values are in keeping with those of other workers (Jordan et al., 2000; Lehtola et al., 1995). An increase in adhesion with an increase in compaction pressure has been demonstrated previously (Lehtola et al., 1995; Brown and Davies, 2000). One explanation is that although the surface area of the tablets compacted at lower compaction pressures will be higher, as witnessed by the surface roughness values obtained in this study, the effective surface area of contact is lower due to lack of penetration of the coating fluid into the 'valleys' on the surface of the tablet. This is unlikely in the current study, as the spreading coefficients indicate a high degree of interaction between the coating fluid and the tablet. A further possibility is that failure in the adhesion test occurs partially at the interparticulate contact areas of the powder particles in the tablet, rather than at the film–tablet interface, leading to a lower than expected value. A rough interface between the film and the tablet may also act as a focus for stress concentration, leading to failure at lower than expected values. It is noteworthy that the maximum adhesion is exhibited by the coating formulation containing MCC. The major component of the tablet core is MCC so there will be a high interaction between the coat and the core. Additionally, cellulose is bipolar (Oh and Luner, 1999) and will interact with the other main component of the tablet core, lactose. This higher adhesion exhibited with the inclusion of MCC will not necessarily be observed if the formulation of the tablet core is different to the one studied.

Changes in atomising air pressure will lead to changes in droplet size (Table 6) and this may influence surface coverage of the spray and rates of drying. Increasing atomising air pressure will also increase droplet velocity (Macleod, unpublished results) and this will increase momentum and assist in spreading and penetration on impact. Only in the case of the addition of MCC does atomising air pressure have an influence on adhesion, and then the results are not consistent with changing compaction pressure. For the other coating fluids, atomising air pressure does not significantly affect the measured adhesion, and this is in keeping with a liquid that spreads easily on the tablet core.

This study has shown that the inclusion of additives in a basic film coating formulation can markedly influence the adhesion of the film to the tablet core. By examining the surface energetics of both the liquid and solid components of the process, it can be concluded that, in the system under study, the interaction between the two, as quantified by the spreading coefficients, is sufficient such that wetting effects are not responsible for the observed changes in adhesion. When the film coating liquid dries, the additives are, in essence, concentrated, and interactions between them and the tablet will become more important.

Acknowledgements

We wish to thank Manesty and The School of Pharmacy and Pharmaceutical Sciences, University of Manchester for financial support to HK.

References

Aulton, M.E., 1995. Surface effects in film coating. In: Cole, G. (Ed.), Pharmaceutical Coating Technology. Taylor and Francis, London, pp. 118–151.

Brown, S.R., Davies, C.N., 2000. The effect of film-coat thickness and tablet hardness on film-coat adhesion. J. Pharm. Pharmacol. 52, 171 Supp.

Buckton, G., Chandraia, B., 1993. Consideration of adhesion to modified container walls, by use of surface energy and polarity data, and Lewis-acid Lewis-base interactions. Int. J. Pharm. 94, 223–229.

Buckton, G., Newton, J.M., 1986. Assessment of the wettability of powders by the use of compressed powder discs. Powder Technol. 46, 201–208.

Felton, L.A., McGinity, J.W., 1997. Influence of plasticizers on the adhesive properties of an acrylic resin coplymer to hydrophilic and hydrophobic tablet compacts. Int. J. Pharm. 154, 167–178.

Felton, L.A., McGinity, J.W., 1999. Adhesion of polymeric films to pharmaceutical solids. Eur. J. Pharm. Biopharm. 47, 3–14.

Good, R.J., Chaudhury, M.K., van Oss, C.J., 1991. In: Lee, L.H. (Ed.), Fundamentals of Adhesion. Plenum Press, New York, pp. 153–172.

Jordan, M.P., Easterbrook, M.G., Hogan, J.E., 2000. Quantitative Studies on the Film Coating of Tablets. Technical Department, Colorcon Limited (GB).

Lehtola, V.-M., Heinamaki, J.T., Nikupaavo, P., Yliruusi, J.K., 1995. Effect of some excipients and compression pressure on the adhesion of aqueous-based hydroxypropylmethylcellulose film coatings to tablet surface. Drug Dev. Ind. Pharm. 21 (12), 1365–1375.

Oh, E., Luner, P.E., 1999. Surface free energy of ethylcellulose films and the influence of plasticizers. Int. J. Pharm. 188, 203–219.

Rillosi, M., Buckton, G., 1995. Modelling mucoadhesion by use of surface energy terms obtained from the Lewis acid–base approach. Int. J. Pharm. 117, 75–84.

Buckton, G., 1995. Surface characterization: understanding sources of variability in the production and use of pharmaceuticals. 47, 265–275.

Rowe, R.C., 1977. The adhesion of film coatings to tablet surfaces — the effect of some direct compression excipients and lubricants. J. Pharm. Pharmacol. 33, 610–612.

van Oss, C.J., Chaudhury, M.K., Good, R.J., 1988a. Interfacial Lifshitz–van der Waals and polar interactions in macroscopic systems. Chem. Rev. 88, 927–941.

van Oss, C.J., Chaudhury, M.K., Good, R.J., 1988b. Additive and nonadditive surface tension components and the interpretation of contact angles. Langmuir 4, 884–891.