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The adhesion characteristics of some pigmented and unpigmented aqueous-based film coatings applied to aspirin tablets*

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The adhesion of pigmented (with talc and titanium dioxide) and unpigmented aqueous-based films, derived from hydroxypropyl methylcellulose, to aspirin tablets and the effect of ageing on the measured adhesion have been assessed. The adhesion of hydroxypropyl methycellulose film attained maximum values at polyethylene glycol 400 and polyvinyl alcohol levels of 10 and 20 wt%, respectively. Above these concentrations, adhesion decreased. For solid loaded films it is proposed that the effect of pigments on film adhesion is dependent on the balance between their influence on the internal stress of the film coating and the strength of the film-tablet interface. Adhesion was enhanced when a pigment increased the strength of the interface faster than it increased internal stress, and vice versa. A simple relation between the measured adhesion and the incidence of edge splitting of film-coated tablets was not observed. Generally, film adhesion fell when the tablets were aged at 37 °C and 75% r.h. as a result of swelling-induced stresses in the film and at the film tablet interface. The effect of ageing on the adhesion of the system plasticized with polyethylene glycol 400 was eased when the film was pigmented. Adhesion was largely unaffected with film-coated tablets stored in tightly closed bottles at 20 °C for five months.

Good adhesion between a film coating and the tablet to which it is applied is desirable in film coating practice for several reasons. First, complete loss of adhesion could severely hinder the ability of the film coat to afford mechanical protection to the tablet. Second, there is the possibility that adhesion loss could lead to accumulation of substantial quantities of moisture at the film-tablet interface, a development that may have adverse effect on the active contents of tablets. In addition, adhesion studies have also provided useful information on internal stress in films and film coating defects such as edge splitting, cracking and bridging of intagliations (Sato 1980; Rowe & Forse 1981; Rowe 1982).

Mittal (1980, 1982) has differentiated between 'fundamental adhesion'and 'practical adhesion' by defining practical adhesion as the summation of all the intermolecular interactions between the film and the substrate (i.e. fundamental adhesion) and other factors, which though not interfacial in nature, do affect adhesion, e.g. stresses in the films, type of rupture and the adhesion measurement technique. A similar distinction, which referred to 'intrinsic adhesion' and 'measured adhesion' has been made by Rowe (1981a). Thus practical or measured adhesion is the actual force required to detach a film from a unit area of the substrate. Sato (1980) has indicated that the presence of high internal stress in a film coating usually lowers the measured adhesion. If the mode of film detachment from the tablet is adhesive (rather than cohesive) and other variables such as measurement conditions are constant, the measured adhesion will depend on the balance between intrinsic (or fundamental) adhesion and the magnitude of internal stress in the film coat.

In the present paper, the adhesion process in pigmented (with talc and titanium dioxide) and unpigmented aqueous-based films, derived from hydroxypropyl methylcellulose and applied to aspirin tablets, is considered in detail. The effect of ageing on adhesion, and the presumed link between measured adhesion and the incidence of edge splitting of film-coated tablets are also examined.

MATERIALS AND METHODS

Hydroxypropyl methylcellulose USP (Pharmacoat 606) and polyvinyl alcohol (Poval PA-5)—with a degree of hydrolysis of 88%—were manufactured by Shin-Etsu Chem. Co. Ltd, Japan. The plasticizer, polyethylene glycol 400 was obtained from BDH Chemicals Ltd, Poole, UK. The talc used (talc 4053, Richard Baker Harrison Ltd, Ilford, UK) had a specific surface area of 1·81 $m^2 g^{-1}$ (based on nitrogen adsorption measurements) while the titanium dioxide (Bayertitan RC-K-20, Bayer (UK) Ltd) was rutile, surface-treated with alumina and an organic

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material, water repellant and had a specific surface area of 8·42 $m^2 g^{-1}$. Granular aspirin (Asagran, Monsanto, UK) together with microcrystalline cellulose (Avicel PH-105, FMC Corp.) and potato starch (BDH Chemicals Ltd) were the tablet constituents.

Three main polymer systems were evaluated: hydroxypropyl methylcellulose alone, and in combination with either polyvinyl alcohol or polyethylene glycol 400. For the pigmented films, the level of polymer additives for polyvinyl alcohol and polyethylene glycol 400 was 20 wt% and 10 wt% of hydroxypropyl methylcellulose, respectively. The pigments were dispersed in aqueous formulations of the polymer systems using a mixer-emulsifier (Silverson Machines Ltd, London) for 30 min.

Flat-faced aspirin tablets, 13 mm diameter, containing 5 wt% each of microcrystalline cellulose and potato starch were directly compressed at 122 MPa in an instrumented single punch tablet machine (type E-2, Manesty Machines Ltd, Liverpool) and coated with 10% w/v aqueous film formulations (pigmented and unpigmented) in a 15·1 cm Wurster column to produce 3·2 wt% of film coating per tablet. The tablets were stored for one month at 20 °C, 37% r.h., and film-tablet adhesive force measured using a technique and an adhesion tester similar to that reported by Fisher & Rowe (1976). Ten tablets were used in each test and coefficients of variation, ranging from 4–20%, are similar to those previously reported (Fisher & Rowe 1976; Fung & Parrott 1980).

Effect of ageing

The effect of ageing on adhesion was determined for tablets coated with film formulations containing 20 wt% of talc or titanium dioxide and equivalent pigment-free formulations. The coated tablets were aged over five months in a desiccator at 37 °C, 75% r.h. Before adhesion measurement, the tablets were equilibrated at 20 °C and 37% r.h. to eliminate moisture. As a control, similar tablets were stored in tightly closed bottles at 20 °C and their adhesion measured after five months.

RESULTS AND DISCUSSION

In all cases adhesive rather than cohesive failure occurred, as indicated by the absence of traces of the tablet substrate on the detached films. Adhesion was calculated as the force per unit area required to detach the film coat from the flat face of the tablet.

The mean adhesion values are shown in Figs 1–3 and Tables 1 and 2. Fig. 1 indicates that the adhesion of hydroxypropyl methylcellulose film to aspirin tablets reached a peak and then declined when the concentration of either polyethylene glycol 400 and polyvinyl alcohol was increased. Adhesion maxima occurred at 10 and 20 wt% of the polymer additives, respectively. The data for the pigmented systems (Fig. 2) showed different trends and seemed dependent on both the polymer matrix and pigment type.

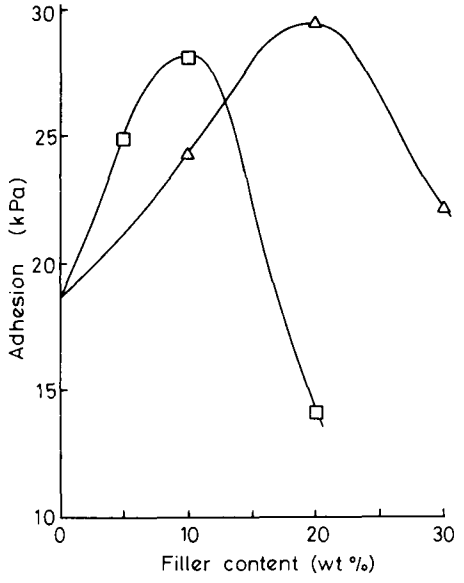

FIG. 1. Adhesion of HPMC film coats containing either polyvinyl alcohol (△) or polyethylene glycol 400 (□) to aspirin tablets at 20 °C and 37% r.h.

The factors that influence the adhesion of film coatings to a tablet and the precise contribution of each factor are often difficult to assess. However, Rowe (1981a) has proposed that film-tablet adhesion would be influenced by two major factors: (a) intrinsic adhesion or the strength of the interfacial bond between the film and tablet, and (b) the internal stress of the film coat. The stress crack resistance (toughness index) and Young's modulus data of free films corresponding to the applied film coatings of the present work were reported in a recent study (Okhamafe & York 1984, 1985a). These data are listed in Table 1 along with the corresponding adhesion and incidence of edge splitting data.

The initial rise in the measured adhesion of the unpigmented coating formulations corresponds to a lowering of the Young's modulus of the free films. Rowe (1981b) has proposed that a direct relationship can be found between reduction in internal stress and lowering of Young's modulus. Apparently the addition of polyvinyl alcohol and polyethylene glycol 400 to the film former initially increased adhesion because the polymer additives, with varying degrees

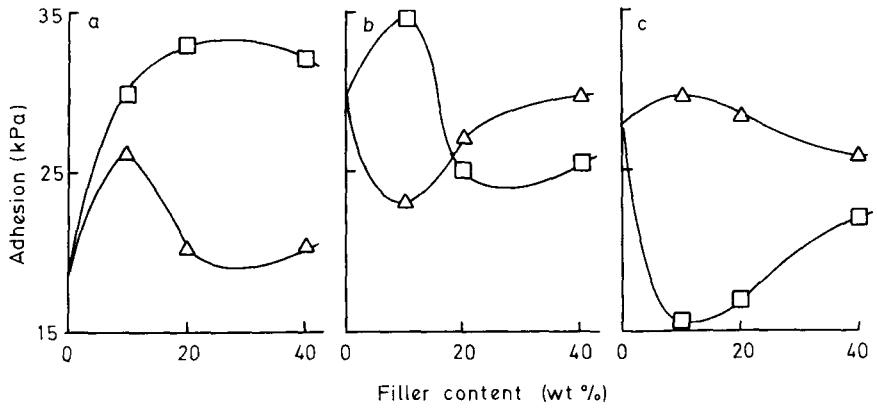

FIG. 2. Adhesion of film coatings of (a) HPMC, (b) HPMC/polyvinyl alcohol and (c) HPMC/polyethylene glycol 400 containing either talc (△) or titanium dioxide (□) to aspirin tablets at 20 °C and 37% r.h.

of plasticizing activity, decreased the internal stress of the film coatings. However, as the levels of the additives were further raised, adhesion soon reached a maximum value and then began to decrease as a result of the presence of increasing amounts of the polymer additives at the film-tablet interface. A previous report (Okhamafe & York 1984) shows that the polymer additives are acidic while the film former is neutral. Since aspirin is acidic, application of the acid-based (electron donor-acceptor) concept (Fowkes 1982) predicts poor interactions between the polymer additives and the tablet substrate. Thus the polymer additives, while reducing the internal stress of the film coatings, and therefore promoting adhesion, also seemed to have increased stress at the film-tablet interface as a result of its poor interaction with the tablet core. The latter effect is dominant at high concentrations of the polymer additives and hence the fall in the measured adhesion.

For the pigmented systems, there was no definitive relationship between the measured adhesion and Young's modulus. Although pigmentation generally enhanced the modulus, the adhesion of plasticized (with PEG 400) and unplasticized hydroxypropyl methylcellulose films increased as the content of titanium dioxide was raised. The film blend containing polyvinyl alcohol and talc showed a similar trend while the other pigmented systems exhibited reduced adhesion with increased pigment level.

The toughness index, which fell with increase in pigment concentration, could indicate the trend of internal stress since stress concentration at the pigment-polymer interface rises with increased pigment content. Thus pigmentation might at first be thought to be detrimental to measured adhesion but this ignores the possibility that pigment-polymer interaction could have a strengthening effect on the film-tablet adhesion bonding. Pigment-polymer interaction generally stiffens the polymer segments at the pigment-polymer interface and according to Kwei (1965), this stiffening phenomenon can be transmitted as far down the polymer network as 1500 Å from the pigment-polymer border. It is conceivable that if some of the pigment particles in the polymer system are within 1500 Å (or any similarly established distance) of the film-tablet interface, the polymer matrix, as well as the film-tablet interface, could be stiffened and thereby strengthened. This is believed to have occurred in the pigmented systems herein examined.

Thus the pigments incorporated in an applied film coat can exert two opposing effects on adhesion: one decreases adhesion by increasing internal stress and the other increases adhesion by strengthening the film-tablet interface. The measured adhesion depends on the balance of these two effects. It is possible, on this basis, to provide explanations of the adhesion data for the pigmented films. The adhesion of hydroxypropyl methylcellulose film was initially increased in the presence of talc because of a stronger film-tablet interface and a smaller increase in the internal stress of the film, but above 10 wt% of the pigment, the internal stress factor began to dominate and adhesion fell. In pigmented hydroxypropyl methylcellulose/polyvinyl alcohol films, the internal stress factor was initially dominant because the presence of some polyvinyl alcohol at the film-tablet interface hinders the strengthening of the interface. Increased talc content subsequently leads to a faster rise in the strength of the film-tablet interface than in the internal stress of the film. A reverse situation applies in the presence of titanium

dioxide. Where observed, the higher adhesion of titanium dioxide-filled films, when compared with equivalent films loaded with talc, could be attributed to the lower internal stress of the former as indicated by the toughness index data in Table 1. A combination of the ability of a plasticized film to accommodate the effect of stress better than the unplasticized film, and the hindrance by the polymer additive of the strengthening effect of pigment-polymer interaction on the film-tablet interface may account for the unchanged adhesion of talc-filled films containing polyethylene glycol 400.

Table 1. Relationship between stress crack resistance (toughness index TI) and Young's modulus (YM) of free films, and the measured adhesion and the incidence of edge splitting of equivalent film coats applied to aspirin tablets. HPMC = hydroxypropyl methycellulose; PVA = polyvinyl alcohol; PEG400 = polyethylene glycol 400, TiO_2 = titanium dioxide.

	wt%	TI (kJ m^{-3} × 10^3)	YM (MPA) × 10^2	Adhesion (kPa)	Edge splitting (%)
HPMC alone		8·14	10·40	18·6	12
HPMC + PVA	10	8·89	8·99	24·22	0
	20	7·56	8·73	29·4	0
	30	—	—	22·1	0
HPMC + PEG 400	5	—	—	24.9	0
	10	5.36	7·04	28·1	0
	2`	5.64	3·91	14.1	0
HPMC + talc	10	3·12	11·47	26·0	0
	20	2·75	12·77	20·2	5
	40	0·98	16·78	20·3	8
HPMC + TiO_2	10	3·68	11·12	29·8	14
	20	2·25	11·35	32·9	32
	40	1·54	11·61	32·1	83
HPMC/PVA + talc	10	3·01	8·71		0
	20	2·35	8·85	27·0	3
	40	0·77	9·75	29·5	8
HPMC/PVA + TiO_2	10	4·46	7·93	34·4	6
	20	2·32	8·45	24·9	19
	40	1·57	9·24	25·4	62
HPMC/PEG 400 + talc					
	10	2·50	4·99	29·4	0
	20	1·90	5·16	28·3	6
	40	0·71	5·56	25·8	4
HPMC/PEG 400 + TiO_2					
	10	4·63	3·77	15·6	29
	20	1·34	4·07	16·8	36
	40	1·19	5·31	22·1	89

Rowe (1982) has suggested that an inverse relationship could be found beween the measured adhesion and the incidence of edge splitting. The data in Table 1 show that the hypothesis held up well for the pigmented systems containing polyethylene glycol 400 but not for the pigmented films containing hydroxypropyl methylcellulose alone and those containing polyvinyl alcohol in addition. The exact relationship between edge splitting and measured adhesion is likely to be complex, and a number of factors such as the magnitude of the internal stress, the substrate type and the conditions under which the film coating was applied, may be involved.

Effect of ageing

The effect of ageing at 37 °C and 75% r.h. on the film-coated tablets, illustrated in Fig. 3, was generally a reduction in measured adhesion, and this behaviour was exhibited by both pigmented and unpigmented films. Although Walker (1967) obtained a similar result for paint-metal adhesion which he attributed to the formation of a water-sensitive layer in the film close to its boundary with the metal, the films employed in the present study, unlike the paint films, are hydrophilic. In addition, the aspirin tablet surface is less hydrophobic than the metal substrate. Therefore the processes leading to adhesion loss will not be exactly the same.

The data obtained (Fig. 3) can be discussed as before, in terms of the internal stress of the coatings and the strength of the film-tablet interface. Since the stored film-coated tablets were always equilibrated at 20 °C and 37% r.h. for one week before measurements were made, the presence of the water taken up during ageing in the film coat or at the film-tablet interface is considered unlikely. In a few films, notably unpigmented and talc-pigmented hydroxypropyl methylcellulose/polyvinyl alcohol films as well as talc-filled hydroxypropyl methylcellulose film, adhesion increased after one month of ageing but fell sharply when the ageing period was further extended. It is unlikely, in these cases, that the strength of the film-tablet interface increased under the ageing conditions. The probability is that the level of the internal stress initially fell following a decrease in molecular order and a rise in segmental mobility, in view of the crystallinity (high molecular order and rigidity) of the polymer matrices which has previously been reported (Okhamafe & York

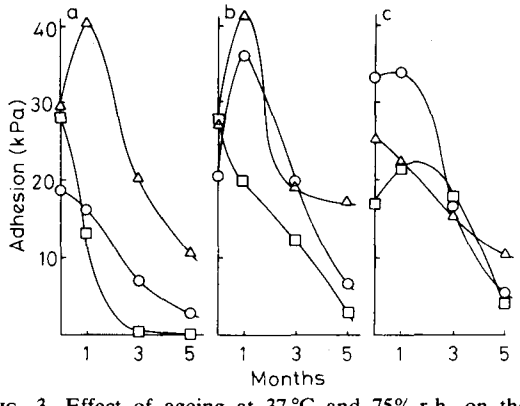

FIG. 3. Effect of ageing at 37 °C and 75% r.h. on the adhesion of HPMC (○), HPMC/polyvinyl alcohol (△) and HPMC/polyethylene glycol 400 (□) film coats to aspirin tablets (Note: (a) = unpigmented films; (b) = talc-filled films; (c) = titanium dioxide-filled films).

1985b). However, prolonged ageing leads to even greater molecular motions and re-orientation as the polymer matrix absorbs moisture and heat energy, and expands or swells in the process. This results in the creation of new stresses in all the film coatings (including the plasticized systems) as well as at the film-tablet interface, a development that could not be completely reversed by pre-test equilibration at 20 °C and 37% r.h. The combination of increased stress in the film coat and at the film-tablet interface (thus weakening interfacial bonding) is the cause of the loss in adhesion. The data in Fig. 3 also indicate that the unpigmented film containing polyethylene glycol 400 virtually lost all adhesion by the third month but the effect of ageing was noticeably eased when the film was pigmented with either talc or titanium dioxide. The effect of pigmentation could be explained in terms of the stiffening of the film-tablet interface by the pigments, and the lower swelling-induced stresses as a result of volume restraint by the pigment particles and the insolubility of the pigments.

Table 2. Adhesion of some film coats applied to aspirin tablets before and after storage for 5 months in tightly closed bottles at 20 °C (coefficient of variation in brackets).

Film coat		Adhesion (kPa) Initial	5 months later
HPMC		18·6 (13·0%)	20·8 (15·9%)
HPMC + 20 wt% PVA		29·4 (7·4%)	31·7 (15·1%)
HPMC + 20 wt% PEG 400		28·1 (18·1%)	25·8 (16·2%)
HPMC + talc	20 wt%	20·2 (11·6%)	23·4 (15·6%)
HPMC/PVA + talc	20 wt%	27·0 (16·3%)	25·7 (18·7%)
HPMC/PEG 400 + talc	20 wt%	28·3 (9·6%)	25·2 (18·4%)
HPMC + TiO_2	20 wt%	32·9 (12·5%)	21·1 (13·5%)
HPMC/PVA + TiO_2	20 wt%	24·9 (19·0%)	19·2 (14·0%)
HPMC/PEG 400 + TiO_2	20 wt%	16·8 (16·9%)	28·8 (17·5%)

As Table 2 shows, the measured adhesion of most of the film coats did not change significantly (*t*-test at the 95% confidence level) after they had been stored in tightly closed bottles for five months. The exceptions were unplasticized hydroxypropyl methylcellulose containing titanium dioxide (decreased adhesion) and the plasticized film loaded with titanium dioxide (increased adhesion). With the data in Fig. 3, these results highlight the need to provide suitable storage conditions for film-coated tablets.

CONCLUSIONS

Maxima in measured adhesion of hydroxypropyl methylcellulose film to aspirin tablets were obtained at 10 wt% polyethylene glycol 400 and 20 wt% polyvinyl alcohol contents.

The effect of talc and titanium dioxide on film adhesion was attributed to the balance between their influence on the internal stress of the film coat and the strength of the film-tablet interface. Adhesion decreased when a pigment increased the internal stress of the film faster than it increased the strength of the film-tablet interface and vice versa.

A simple relation between the measured adhesion and the incidence of edge splitting of film-coated tablets was not observed.

Whilst adhesion was largely unaffected when the film-coated tablets were stored in tightly closed bottles at 20 °C for five months, adhesion generally decreased when they were aged at 37 °C and 75% r.h. The unpigmented film containing polyethylene glycol 400 virtually lost all adhesion after ageing for three months but adhesion was less adversely affected by ageing when the film was pigmented with either talc or titanium dioxide.

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