J. Pharm. Pharmacol. 1986, 38: 414–419
Received October 11, 1985

Mechanical properties of some pigmented and unpigmented aqueous-based film coating formulations applied to aspirin tablets*

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The Brinell hardness and Young's modulus of pigmented and unpigmented films of hydroxypropyl methylcellulose alone, and in combination with either polyethylene glycol 400 (plasticizer) or polyvinyl alcohol, which were applied to aspirin tablets, have been measured. Generally hardness and modulus data showed similar trends. The hardness and modulus of hydroxypropyl methylcellulose fell in the presence of polyethylene glycol 400 as a result of its plasticizing action. On the other hand, the hardness and modulus of the film former rose slightly when polyvinyl alcohol was initially incorporated, probably due to the crystalline phase of the additive, and then decreased when the level of the additive was further raised. Hardness and modulus were higher in films pigmented with talc than in those containing titanium dioxide because of the plate-like shape of talc and its greater interaction with the polymer systems. Some correlation was found between the Young's moduli of the applied films and those of the corresponding free films, with the moduli of the latter two 2–5 times greater. Ageing at 37 °C and 75% r.h. was found to cause a decrease in the mechanical properties of the unplasticized film coating systems probably as a result of decreased molecular order and enhanced polymer chain mobility.

One of the main requirements of a pharmaceutical film coating is that it should provide adequate protection to the solid dosage form. Various studies (e.g. Stern 1976; Rowe 1976a; Fell et al 1979) indicate that the crushing strength of tablets is usually increased when they are film-coated. Solid dosage forms such as tablets and granules, are susceptible to various physical hazards such as impact, abrasion and friction. They may also experience substantial volume expansion in conditions of high temperature and humidity and this could lead to considerable strains on the applied film coating.

The capacity of a film coating to afford physical protection depends, to a large extent, on its mechanical characteristics. Increased magnitude of parameters such as tensile strength and Young's modulus do not necessarily indicate enhanced physical protection of a solid dosage form by the film coating. For example, although pigmentation generally increases elastic modulus, the incidence of edge splitting of film-coated tablets has been reported to increase with a decrease in the tensile strength: Young's modulus ratio (Rowe 1983). In other words, the incidence of edge splitting increased because the Young's modulus rose relative to the tensile strength of the coating. Rowe (1983) further observed that a decrease in the Young's modulus of unpigmented films generally resulted in a lower incidence of bridging of intagliations or monograms. The inference, therefore, is that Young's modulus plays a key role in determining the protective capacity of a film coating.

Unlike free films, parameters such as tensile strength, toughness and elongation of film coatings applied to tablets are difficult to measure. Nevertheless, indentation techniques offer a way of measuring the mechanical properties of applied (in-situ) film coatings such as hardness and Young's modulus. In the present study, the hardness and Young's modulus of pigmented (with talc or titanium dioxide) and unpigmented aqueous-based film coating formulations applied to aspirin tablets have been assessed. The Young's modulus data are correlated with similar data for free films and the effect of ageing on the mechanical properties of the applied film coatings are also examined.

MATERIALS AND METHODS

Three polymer systems were used: hydroxypropyl methylcellulose (Pharmacoat 606, Shin-Etsu Chem. Co. Ltd, Japan) alone, and hydroxypropyl methylcellulose blended with either 88% hydrolysed poly-

* Communicated in part at the British Pharmaceutical Conference, Leeds, September 1985.

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vinyl alcohol (Poval PA-5, Shin-Etsu, Japan) or polyethylene glycol 400 (BDH Chem. Ltd, Poole, UK). Talc (talc 4053, Richard Baker Harrison Ltd, Ilford, UK) and titanium dioxide (Bayertitan RC-K-20, Bayer (UK) Ltd) were the pigments or fillers used. The specific surface areas of these pigments (based on nitrogen adsorption measurements) are 1·81 and 8·42 $m^2 g^{-1}$, respectively. The titanium dioxide was rutile, surface-treated with alumina and an organic material, and water-repellent. For the pigmented formulations, the levels of the polymer additives (i.e. polyvinyl alcohol and polyethylene glycol 400) were 20 and 10 wt%, respectively. The dispersion of the pigments in the aqueous film formulations, the manufacture of 13 mm diameter flat-faced aspirin tablets and the application of the coating formulations to the tablets have been described previously (Okhamafe & York 1985a).

An indentation apparatus, similar in design to that proposed by White & Aulton (1980), and incorporating a displacement transducer model 1304 (Penny & Giles Potentiometers Ltd, Christchurch, UK) was employed for indentation measurements. The depth of penetration of a 4 mm diameter sapphire indenter in the film coating of a tablet when a 10 g load was applied for 60 s and the recovery of the film coating 60 s after removal of the load were monitored on a chart recorder. Measurements were made at four different points on one surface of a tablet and five coated tablets were assessed in each test.

Effect of ageing

The effect of ageing at 37 °C and 75% r.h. on indentation properties was examined by repeating the indentation tests on selected film coating formulations after ageing periods of 1, 3 and 5 months. The samples were equilibrated at 20 °C and 37% r.h. for one week after each ageing period before measurements were made. As a control, the indentation properties of similar film coating systems were also assessed following storage in tightly closed bottles at 20 °C for 5 months.

RESULTS AND DISCUSSION

Brinell hardness number (H_B) and Young's modulus (E) were calculated from the following simplified relationships (Baer et al 1961; Aulton 1982):

$$H_B = \frac{W}{\pi D_1 d_1} \quad (1)$$

$$E = \frac{W}{3d_2(d_1 D_1)^{\frac{1}{2}}} \quad (2)$$

where W is the indentation load applied, D_1 is the diameter of the indenter, d_1 the depth of penetration of the indenter in the film coating, and d_2 the recovery distance after the removal of the load. The mean values of the results obtained are presented in Figs 1–6, and Tables 1 and 2. Coefficient of variation of data was in the range of 19–30% which is comparable to the 20–25% reported by Ridgway et al (1970) and Rowe (1976b) who attributed the higher scatter of data to the inherent variability of the film coating rather than the measuring technique employed.

Generally, the hardness and Young's modulus data showed identical trends (see Figs 1–4). This is expected since both are measures of the stiffness or rigidity of the molecular segments of the films. Moreover, Van Krevelen (1976) has shown that the two parameters are directly related.

As Fig. 1 indicates, addition of the plasticizer (polyethylene glycol 400) to hydroxypropyl methylcellulose led to a decrease in film hardness and modulus. The enhancement of the segmental mobility of hydroxypropyl methylcellulose by the plasti-

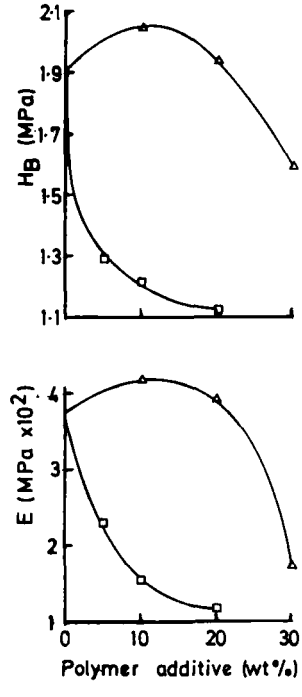

FIG. 1. Brinell hardness (H_B) and Young's modulus (E) of hydroxypropyl methylcellulose coatings containing either polyvinyl alochol (△) or polyethylene glycol 400 (□) at 20 °C and 37% r.h.

cizer resulted in a softer and more flexible film. On the other hand, the hardness and Young's modulus of the hydroxypropyl methylcellulose–polyvinyl alcohol blend were largely unchanged at low levels of the polymer additive (polyvinyl alcohol). However, when the concentration of the polymer additive exceeded 20 wt %, a downward trend was observed for both parameters. It may be that at low polyvinyl alcohol content, the presence of a crystalline phase of the polymer additive (see Okhamafe & York 1985b), which is a region of high molecular order and chain stiffness, prevented a fall in hardness and modulus. As the polyvinyl alcohol level was further raised, the plasticizing action of its amorphous phase became dominant over the chain stiffness effect of the crystalline phase.

The data for the pigmented systems (Figs 2–4) indicate that titanium dioxide did not enhance hardness and Young's modulus. For hydroxypropyl methylcellulose films, the parameters were initially lowered when titanium dioxide was incorporated but showed a small upward trend when filler level rose

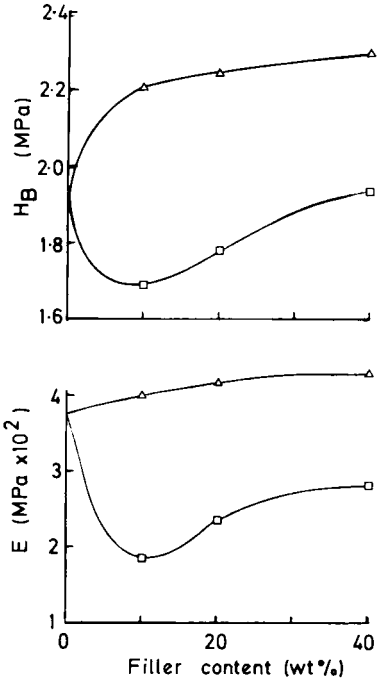

FIG. 2. Brinell hardness (H_B) and Young's modulus (E) of hydroxypropyl methylcellulose film coating pigmented with either talc (△) or titanium dioxide (□) at 20 °C and 37% r.h.

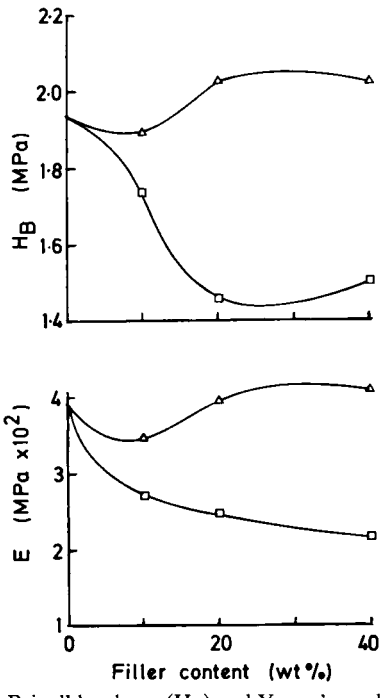

FIG. 3. Brinell hardness (H_B) and Young's modulus (E) of hydroxypropyl methylcellulose/polyvinyl alcohol film coating blend pigmented with either talc (△) or titanium dioxide (□) at 20 °C and 37% r.h.

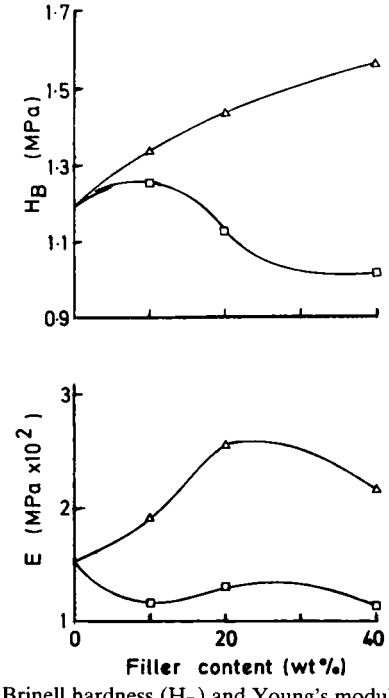

FIG. 4. Brinell hardness (H_B) and Young's modulus (E) of hydroxypropyl methylcellulose/polyethylene glycol 400 film coating blend pigmented with either talc (△) or titanium dioxide (□) at 20 °C and 37% r.h.

above 10 wt %. The hardness and moduli of the plasticized formulation and hydroxypropyl methylcellulose–polyvinyl alcohol blend were either unchanged or reduced in the presence of titanium dioxide. On the other hand, talc generally improved the hardness and modulus of plasticized and unplasticized hydroxypropyl methylcellulose films while exerting little influence on the system containing polyvinyl alcohol.

The two main categories of filler effects on mechanical properties, as discussed previously (Okhamafe & York 1984a), are: (i) hydrodynamic and (ii) reinforcing. The shape factor (i.e. particle length : width ratio, a hydrodynamic parameter) of talc, which is plate-like, is greater than that of spherical titanium dioxide particles. Talc should, therefore, more effectively hinder polymer mobility than titanium dioxide. Based on the acid-base concept (see Okhamafe & York 1984b), the reinforcing effect (the degree of pigment-polymer interaction) of talc is also greater than that of titanium dioxide. Thus, talc is clearly the more powerful hardness and modulus enhancer and this is supported by the results obtained.

Talc, with a hardness of 1·0 on the Moh scale (Patton 1973) is generally regarded as a soft pigment. Rutile titanium dioxide is considerably harder at 6–7 on the Moh scale (Kampfer 1973). It is apparent from the generally lower hardness of titanium dioxide-filled films that the hardness of the fillers did not influence the mechanical properties of the film coatings. This also implies that the indentation hardness and modulus measured were those of the polymer matrix, and that the pigments did not physically interfere with the measurements.

Correlation of applied films with free films

Table 1 shows the Young's moduli of the film coating formulations applied to aspirin tablets and those of corresponding free films. The data for the free films were taken from earlier works (Okhamafe & York 1983, 1984a, 1985c). The results generally indicate that the Young's moduli of free films were 2–5 times higher than those of equivalent applied films. A number of reasons can be advanced to explain this difference. Firstly, the measurement techniques employed—indentation and stress-strain—are different in principle. Secondly, different procedures were used to prepare the films. The free films were cast by a pouring technique while the applied films were obtained by a spraying method. An applied film may therefore be less coherent since it probably consists of contiguous multilayers of polymeric material. In addition, internal stresses may be higher in applied films as a result of the volume expansion of the tablet core, and the constant impact and abrasion of the tablets during a coating operation. A further reason might be that the drying rate is higher for applied films. The drying rate could influence the morphology and molecular features (such as crystallinity and glass transition) of the films as well as the magnitude and distribution of internal stresses.

Table 1. Comparison of the Young's moduli (E) of film coatings applied to tablets with those of equivalent free films (HMPC = hydroxypropyl methylcellulose; PVA = polyvinyl alcohol; PEG 400 = polyethylene glycol 400; TiO_2 = titanium dioxide).

		$E \times 10^2$ (MPa)	
Film coating		Applied film	Free film
HPMC alone		3·74	10·40
HPMC + PVA	10 wt%	4·17	8·99
	20 wt%	3·92	8·73
HPMC + PEG 400	10 wt%	1·53	7·04
	20 wt%	1·16	3·91
HPMC + talc	10 wt%	3·99	11·47
	20 wt%	4·17	12·77
	40 wt%	4·28	16·78
HPMC + TiO_2	10 wt%	1·85	11·12
	20 wt%	2·36	11·35
	40 wt%	2·81	11·61
HPMC/PVA + talc	10 wt%	3·48	8·71
	20 wt%	3·94	8·85
	40 wt%	4·11	9·75
HPMC/PVA + TiO_2	10 wt%	2·70	7·93
	20 wt%	2·47	8·45
	40 wt%	2·15	9·24
HPMC/PEG 400 + talc	10 wt%	1·91	4·99
	20 wt%	2·55	5·16
	40 wt%	2·14	5·56
HPMC/PEG 400 + TiO_2	10 wt%	1·17	3·77
	20 wt%	1·30	4·07
	40 wt%	1·14	5·31

Nevertheless, the modulus trends of both free and applied films were, in most cases, similar, with the exception of the polymer blends pigmented with titanium dioxide where, in contrast with the corresponding free films, the moduli of the applied films were not enhanced by increasing pigment content.

Effect of ageing

Figs 5 and 6 illustrate the effect of ageing at 37 °C and 75% r.h. on the hardness and Young's modulus, respectively, of some of the applied film coating formulations. Since the coated tablets were always equilibrated for one week at 20 °C and 37% r.h. before measurements were made, it is considered unlikely that the moisture taken up by the hydrophilic coatings at the ageing humidity would still be retained at the time of measurement. Therefore, the probability that any reductions in the measured

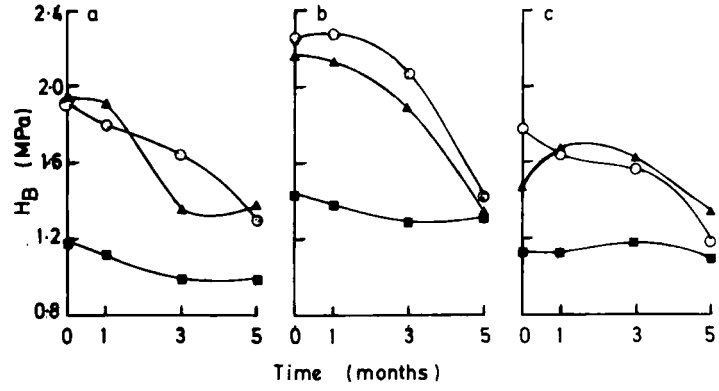

FIG. 5. Effect of ageing at 37 °C and 75% r.h. on the Brinell hardness (H_B) of film coatings of hydroxypropyl methylcellulose (HPMC) alone (○), HPMC/polyvinyl alcohol (▲) and HPMC/polyethylene glycol 400 (■). (*Note*: (a) = unfilled coatings; (b) = talc-filled coatings; (c) = titanium dioxide-filled coatings.)

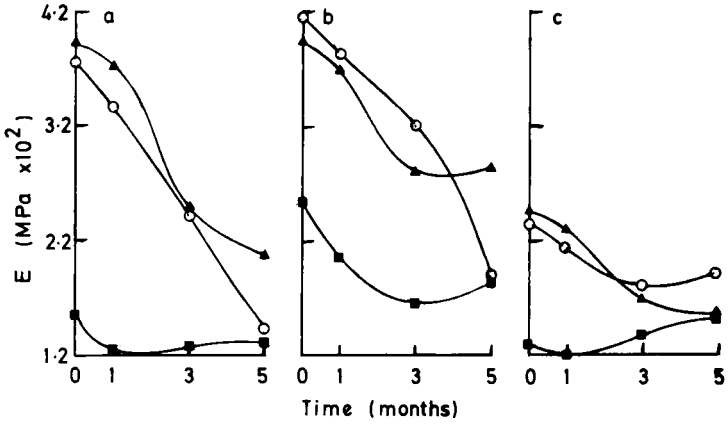

FIG. 6. Effect of ageing at 37 °C and 75% r.h. on the Young's modulus (E) of film coatings of hydroxypropyl methylcellulose (HPMC) alone (○), HPMC/polyvinyl alcohol (▲) and HPMC/polyethylene glycol 400 (■). (*Note*: (a) = unfilled coatings; (b) = talc-filled coatings; (c) = titanium dioxide-filled coatings.)

mechanical properties are due to the plasticizing effect of the water adsorbed during ageing is ruled out.

With the exception of the plasticized system (i.e. hydroxypropyl methylcellulose–polyethylene glycol 400), the hardness and moduli of the other filled and unfilled formulations decreased on ageing. The mechanical properties of the plasticized films (filled and unfilled) were generally unchanged except for the modulus of the system loaded with talc which appeared to have shown a small fall. An explanation of these observations probably lies in the effect of ageing on the molecular stability of the polymer matrices. The ageing conditions (37 °C and 75% r.h.) would be expected to enhance polymer chain mobility by a combination of thermal effects and the swelling action of water. This would then lead to a fall in molecular order. As previously reported (Okhamafe & York 1985b), both hydroxypropyl methylcellulose alone and its combination with polyvinyl alcohol exhibited some levels of crystallinity. It would appear from the present study that the crystallinity probably declined gradually during the ageing process as a result of a decrease in molecular order. The fall in hardness and modulus may be attributed to this phenomenon since there is a direct relationship between the degree of crystallinity and mechanical properties. The largely unchanged hardness and moduli of the plasticized films on ageing support this hypothesis because the films are non-crystalline, flexible and of a low molecular order (Okhamafe & York 1985b). Hence comparable molecular events leading to a decrease in mechanical properties could not have occurred in the plasticized systems.

An additional consideration, in view of the physi-

Table 2. Brinell hardness (H_B) and Young's moduli (E) of some film coatings applied to aspirin tablets before and after storage for 5 months in tightly closed bottles at 20 °C (coefficient of variation in parentheses).

		Initial		5 months later	
Film coating		H_B (MPa)	E × 10^2 (MPa)	H_B (MPa)	E × 10^2 (MPa)
HPMC		1·91 (26·3%)	3·74	1·73 (28·6%)	3·12
HPMC + 20 wt% PVA		1·94 (24·0%)	3·92	1·98 (21·7%)	4·90
HPMC + 20 wt% PEG 400		1·19 (29·9%)	1·53	1·88 (19·4%)	2·29
HPMC + talc	20 wt%	2·25 (21·4%)	4·17	2·20 (25·6%)	4·40
HPMC/PVA + talc	20 wt%	2·16 (23·5%)	3·94	2·14 (20·6%)	4·59
HPMC/PEG 400 + talc	20 wt%	1·44 (20·9%)	2·55	1·85 (23·2%)	2·33
HPMC + TiO_2	20 wt%	1·78 (21·5%)	2·36	1·28 (21·3%)	2·20
HPMC/PVA + TiO_2	20 wt%	1·46 (24·5%)	2·47	2·22 (29·2%)	3·93
HPMC/PEG + TiO_2	20 wt%	1·13 (20·0%)	1·30	2·15 (19·5%)	4·69

cochemical nature of the tablet core, is the sublimation tendency of salicylic acid. It was observed during an unrelated preliminary study that some salicylic acid appeared as needle-like crystals on the film coats of aspirin tablets aged under conditions more drastic than those employed in the present work. Aulton et al (1983) have reported that salicylic acid decreased the tensile strength and increased the elongation of ethyl cellulose film. This implies that salicylic acid has a plasticizing activity in ethyl cellulose which, like the main film-former used in this work, is a cellulose ether. Although sublimed salicylic acid was not visually observed on the coated aspirin tablets used in the present ageing tests, it seems possible that some of the salicylic acid produced as a result of aspirin degradation could have sublimed within the tablet film coating and thus exert a plasticizing effect. This plasticization would lower the hardness and moduli of the unplasticized films. The mechanical properties of the film coatings containing polyethylene glycol 400 (a plasticizer) would not be significantly affected by the presence of salicylic acid since they are already adequately plasticized.

Table 2 lists the Brinell hardness and Young's moduli of the tablet film coating systems before and after storage in a tightly closed bottle at 20 °C for five months. Over the storage period, the measured mechanical properties of all the film coating formulations appeared not to have decreased. In a few cases, e.g. the plasticized film containing titanium dioxide as well as the unpigmented film, hardness and modulus actually increased. Taken together with the ageing data in Figs 5 and 6, these findings emphasize the importance of the effect of storage environment on the mechanical properties of tablet film coating formulations.

Acknowledgement

The authors are grateful to ICI plc and Dr R. C. Rowe in particular for the use of tablet film coating facilities.

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