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Influence of plasticizers on the adhesive properties of an acrylic resin copolymer to hydrophilic and hydrophobic tablet compacts

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Abstract

The influence of plasticizers in film coating formulations on the adhesive properties of an acrylic resin copolymer was determined using the butt adhesion technique. Hydrophilic and hydrophobic plasticizing agents were incorporated into aqueous dispersions of Eudragit® L 30 D-55 and coated onto hydrophilic and hydrophobic tablet compacts. Using data obtained from a Chatillon digital force gauge attached to a motorized test stand, force-deflection profiles, similar to stress-strain curves generated in the tensile testing of free films, were constructed and the force of adhesion, elongation at adhesive failure, and adhesive toughness were determined. Plasticizer concentration and plasticizer type were found to influence the adhesive properties of the acrylic polymer. An increase in adhesive toughness was found when the concentration of triethyl citrate (TEC) in the coating formulation was increased from 20 to 30%, which was attributed to an increase in the elasticity of the film and a decrease in the internal stresses within the polymer. Films containing water soluble plasticizers were found to adhere more strongly to the tablet compacts than the water insoluble agents, due to more effective disruption of the intermolecular attractions between the polymer chains. Adhesion of the polymer to tablet compacts was found to be significantly influenced by the hydrophobicity of the tablet surface when the water soluble plasticizers were incorporated into the film coating, whereas no significant differences in the adhesive properties were found when the polymer was plasticized with water insoluble agents. Aging of the film-coated tablets resulted in a decrease in adhesive toughness, irrespective of the environmental storage condition. © 1997 Elsevier Science B.V.

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1. Introduction

Polymeric film coatings have been applied to pharmaceutical dosage forms for a variety of reasons including taste masking, as a moisture barrier, and as a method of controlling the release characteristics of drugs (Roy, 1994; Parker et al., 1974; Lehmann and Dreher, 1981). Many pharmaceutical polymers exhibit brittle properties and require the addition of a plasticizing agent to obtain an effective coating that is free of cracks, edging, or splitting. These plasticizers play a critical role in the performance of the film coating (Bodmeier and Paeratakul, 1994; Amighi and Möes, 1996). Plasticizers function by weakening the intermolecular attractions between the polymer chains, which generally results in a decrease in the tensile strength, a lowering of the glass transition temperature (Tg), and an increase in the elongation and flexibility of the films (Gutierrez-Rocca and McGinity, 1994). Use of a plasticizer has been found to be imperative when coating with polymeric materials, such as acrylic polymers, that are generally considered to be brittle in nature.

Good adhesion between a polymer and the surface of a tablet is a major prerequisite for the film coating of pharmaceutical dosage forms (Nadkarni et al., 1975; Rowe, 1977; Okhamafe and York, 1985). Loss of adhesion may lead to an accumulation of moisture at the film-tablet interface, significantly affecting the stability of drugs susceptible to degradation by hydrolytic mechanisms (Okhamafe and York, 1985). Loss of adhesion may also compromise the mechanical protection that the film-coating provides to the solid substrate (Stanley et al., 1981). In addition, experiments on adhesion may be useful to the pharmaceutical scientist during preformulation studies to investigate the relationship between tablet excipients and polymeric film coating formulations (Fung and Parrott, 1980).

Previous research on film-tablet adhesion has focused primarily on organic-based cellulosic films. Several studies have been published on the affects of solvents used in the coating formulation on polymer adhesion (Nadkarni et al., 1975; Wood and Harder, 1970). Fisher and Rowe (1976) showed that the force of compression used during tableting significantly influenced polymer adhesion. Excipients used in tablet formulations have also been found to affect film-tablet adhesion (Rowe, 1977; Lehtola et al., 1995; Felton and McGinity, 1996).

Film coating technology has shifted towards aqueous-based systems for environmental and economic reasons (Obara and McGinity, 1994). Few studies on the adhesive properties of aqueous polymeric dispersions have appeared in the pharmaceutical literature. In an earlier investigation involving aqueous dispersions of an acrylic polymer, it was reported that tablet hardness and tablet hydrophobicity significantly influenced polymer adhesion (Felton and McGinity, 1996). In the present study, hydrophilic and hydrophobic plasticizing agents were incorporated in aqueous dispersions of the acrylic polymer and coated onto hydrophilic and hydrophobic tablet compacts. The influence of plasticizer concentration and plasticizer type on the adhesive properties of the acrylic polymer was investigated. The affects of physical aging as a function of environmental storage condition on film-tablet adhesion were also studied.

2. Materials and methods

2.1. Materials

The aqueous dispersion of the enteric acrylic resin copolymer, Eudragit® L 30 D-55, was donated by Hüls America (Somerset, NJ). The plasticizers triethyl citrate (TEC), tributyl citrate (TBC), and dibutyl sebacate (DBS) were donated by Morflex (Greensboro, NC). The polyethylene glycol 6000 (PEG 6000) was obtained from Union Carbide (Houston, TX). Anhydrous lactose was purchased from Sheffield Products (Norwich, NY) and Capital City Products (Columbus, OH) supplied the hydrogenated castor oil under the trade name Sterotex® K. The magnesium stearate was purchased from Spectrum Chemical Mfg. (Gardena, CA) and the Cab-O-Sil® M-5P was donated by Cabot (Tuscola, IL). Scotch® double-coated tape 665 was supplied by 3M (St. Paul, MN).

2.2. Preparation of tablets

Tablet formulations to be coated with the acrylic polymer contained varying anhydrous lactose and up to 30% Sterotex K. All formulations contained 0.5% magnesium stearate as a lubricant and 0.5% Cab-O-Sil® M-5P as a glidant. Excipients were passed through a 40-mesh screen prior to compression. Tablets were manufactured using a Stokes B2, 16 station rotary tablet press (Stokes-Merrill, Bristol, PA). Flat-faced punches with a beveled edge were employed to compress the tablets. Due to differences in the densities and compactibilities of the excipients, the weight of the tablets was varied to maintain a constant surface area for all tablet formulations. All tablets had a diameter of 10.20 mm and a height of approximately 6.20 mm, irrespective of the formulation. Tablets were compressed to a hardness of 10 kg.

2.3. Coating of tablets

The acrylic coating suspensions were prepared by adding water to the Eudragit® L 30 D-55 dispersions to decrease the solids content to 20%. The 20 or 30% percent plasticizer which was incorporated into the film coating was based on the dry polymer weight. To ensure sufficient time for plasticization of the polymer, the aqueous dispersions were mixed with the water soluble plasticizers (TEC and PEG 6000) for at least 30 min and the water insoluble compounds (TBC and DBS) were mixed for 48 h prior to the initiation of coating (Felton et al., 1995).

Tablets were coated in a Vector Mini Hi-Coater Model HCT-20 (Freund, Tokyo, Japan). The bed temperature was held constant at 30°C, while the inlet temperature varied from 65 to 75°C. The spray rate of the polymeric dispersion was 2.0 g/min. The atomizing air pressure was 0.9 kg/cm^2. The rotational speed of the coating pan was set at 20 rpm. Sufficient polymer to achieve a 10% weight gain was applied and the thickness of the film coating was approximately 100 μm. After the coating process was completed, the coated tablets were stored at 40°C for 2 h to further promote coalescence of the polymeric film.

2.4. Determination of adhesive properties

Butt adhesion experiments were conducted using a Chatillon digital force gauge DFGS50 attached to a Chatillon TCD-200 motorized test stand (Chatillon Force Measurement, Greensboro, NC). A more detailed description of the apparatus has been published in previous reports (Felton et al., 1996; Wang et al., 1996). The film-coating at the beveled edge of the tablet was carefully removed using a scalpel. The tablet was affixed to the lower, stationary platen using double-sided adhesive tape. The force gauge, which was fitted with a 13 mm circular steel plate, served as the upper platen. Double-sided adhesive tape (Scotch® double-coated tape 665, 3M, St. Paul, MN) was placed on the top of the tablet. The adhesive tape was selected due to minimal interaction with the polymeric film. Other adhesives, such as cyanoacrylate esters or epoxy resins, may interact with the film coating and require longer contact times with the polymer. The upper platen was lowered to the surface of the tablet, as described in an earlier publication (Felton and McGinity, 1996). The upper platen was raised at a slow, constant rate of 2.5 mm/min. A personal computer (Leading Edge, Westborough, MA)

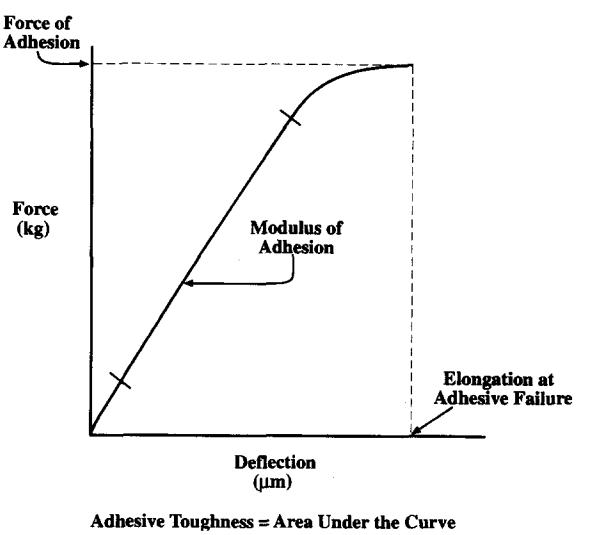

Fig. 1. Example of a force-deflection profile obtained from a butt adhesion experiment using a Chatillon digital force gauge attached to a motorized test stand.

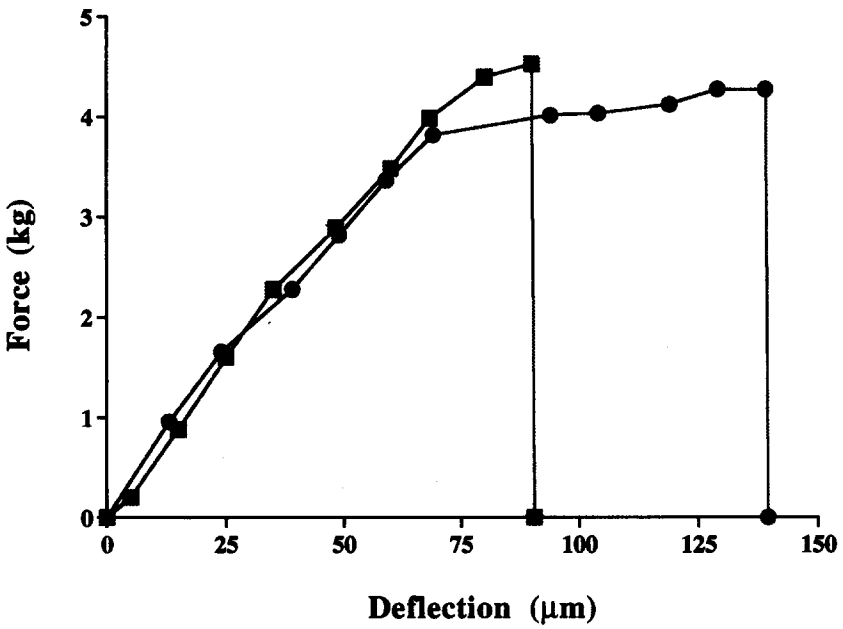

Fig. 2. Force-deflection profiles obtained from butt adhesion experiments as a function of plasticizer concentration (10% Eudragit® L 30 D-55, 15% wax tablet core). (■) 20% TEC and (●) 30% TEC.

recorded the force (kg) and the displacement (mm) at 0.01–0.02 mm intervals. Force-deflection profiles were constructed from the data. The force required to remove the film coating from the tablet, known as the adhesive force, and the elongation at adhesive failure, equivalent to elongation at break in tensile testing of free films, were determined. The slope of the force-deflection curve, referred to as the modulus of adhesion, and the area under the curve, known as the adhesive toughness, were calculated.

2.5. Contact angle measurements

A horizontal microscope (Zeiss, Germany) fitted with a protractor lens retical was used to determine contact angles between the tablets and the Eudragit® L 30 D-55 polymeric dispersion. Uncoated tablets were mounted on glass slides using a cyanoacrylate ester adhesive (Loctite, Rocky Hill, CT). A 2 ml glass pipette was used to deliver the plasticized aqueous dispersions onto the tablet surfaces. The contact angles were measured within 10 s. At least 15 measurements were made for each tablet and coating formulation.

2.6. Scanning electron microscopy

Transverse cross-sections of the film-coated tablets were mounted on brass stages and coated with gold-palladium for 60 s under an argon atmosphere using a Pelco Model 3 cold sputter module (TED Pella, Tustin, CA) in a high vacuum evaporator equipped with an omni-rotary stage. Scanning electron microscopy was performed using a Jeol Model 35 scanning electron microscope (Jeol, USA, Peabody, MA) at 25 kV.

2.7. Thermal analysis

The glass transition temperature (Tg) of the films of the coated tablets was determined using a modulated differential scanning calorimeter model DSC 2920 (TA Instruments, Houston, TX). The apparatus was calibrated using the melting transition of indium. To determine the Tg of the samples, the film from the coated tablets was carefully removed using a scalpel. Approximately 15 mg of the film was accurately weighed in aluminum pans. Thermal analysis was performed at a scan rate of 10°C per min from −10 to 130°C. The

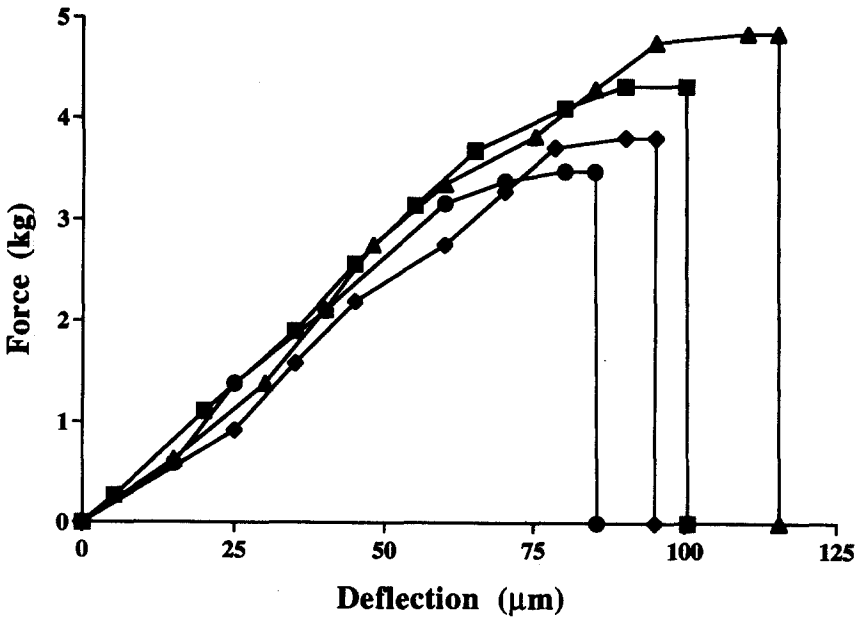

Fig. 3. Force-deflection profiles obtained from butt adhesion experiments as a function of plasticizer type. (20% plasticizer, 10% Eudragit® L 30 D-55, 15% wax tablet core). (▲) TEC, (■) PEG 6000, (◆) TBC and (●) DBS.

modulating signal was set at 0.32°C/min. No previous heating or quenching was performed on the samples. The Tg was calculated as the midpoint of the endothermic curve. Six samples were tested for each coating formulation.

3. Theoretical discussion

Previous researchers have used several adaptations of the butt adhesion technique and the peel test to determine the force required to separate a polymer from the surface of a substrate (Fung and Parrott, 1980; Wood and Harder, 1970; Fisher and Rowe, 1976; Johnson and Zografi, 1986). The equipment used in previous studies has suffered from a variety of deficiencies including the dependency of the peel angle on the elasticity of the film and the uniformity of adhesion, as well as inconsistent rates of deformation (Fisher and Rowe, 1976; Gardon, 1967). In the present study, modifications were made to a Chatillon digital force gauge attached to a motorized test stand to perform butt adhesion experiments, as described in an earlier publication (Felton and McGinity, 1996). An example of a force-deflection profile obtained from the Chatillon apparatus is shown in Fig. 1. This graph, similar to a stress-strain diagram commonly generated in the tensile testing of free films, permitted the visualization of the development of the force within the sample during the adhesion test. In addition to the force of adhesion, the elongation at adhesive failure, the modulus of adhesion, and the adhesive toughness of the film coating were determined in the present study. The elongation at adhesive failure is the distance the upper plate travelled up to the point of film separation. This term is analogous to the elongation at break obtained from tensile testing of free films and reflects the ductility of the polymeric film. The modulus of adhesion is the slope calculated from the linear portion of the force-deflection diagram and may be compared to the Young's modulus obtained from mechanical testing of free films. The adhesive toughness is equal to the work required to remove the film from the tablet surface and may be calculated from the area under the force-deflection profile.

Loss of adhesion has been reported as the result of an increase in the internal stresses within a

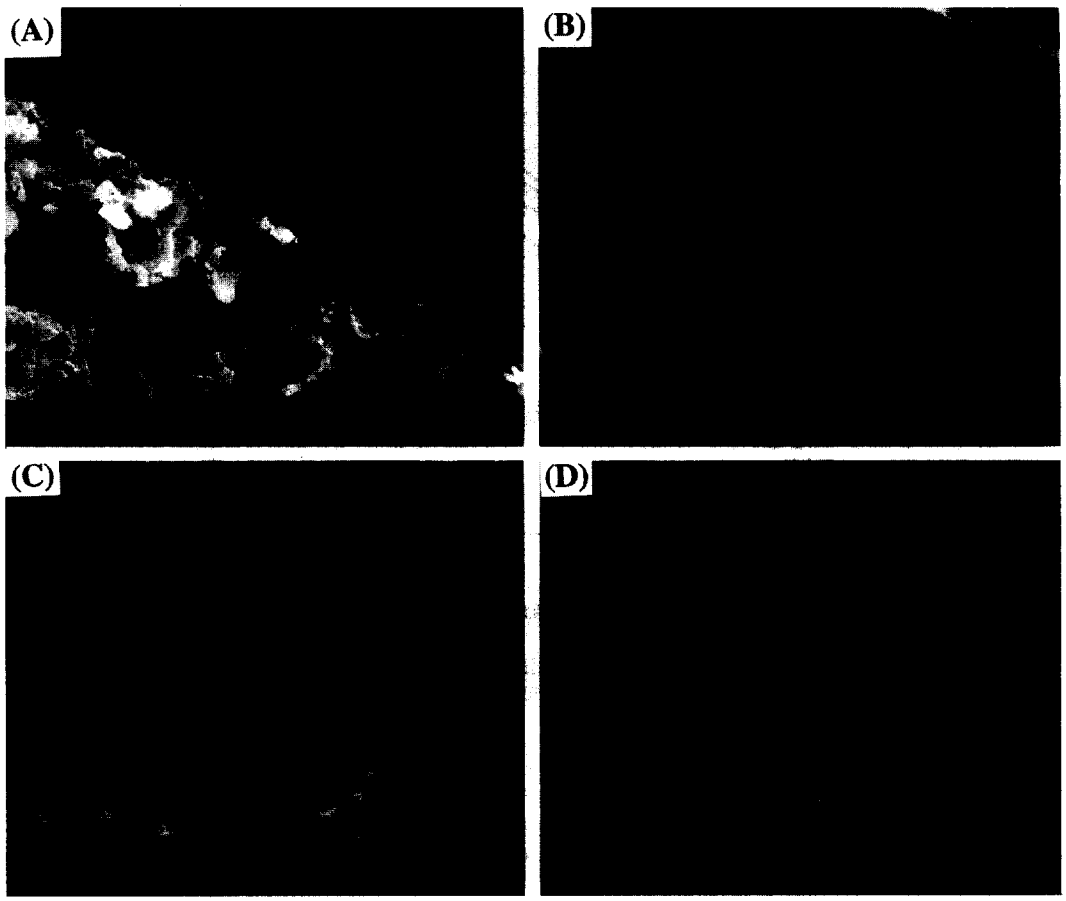

Fig. 4. Scanning electron micrographs of the film-tablet interface as a function of plasticizer type. (20% plasticizer, 10% Eudragit® L 30 D-55, 15% wax tablet core). (A) TEC, (B) PEG 6000, (C) TBC and (D) DBS.

polymeric film (Croll, 1979). When a polymeric solution or dispersion is applied to a substrate, an internal stress develops within the film (Rowe, 1983). The total stress within a film is the sum of all the stresses acting on the polymer, including the stress due to shrinkage of the film on evaporation of the solvent, thermal stress due to the difference in thermal expansion of the film and the substrate, and the volumetric stress due to the volume change when a substrate or polymer swells upon storage. Several researchers have developed equations to calculate the total stress within a film (Croll, 1979; Rowe, 1983; Sato, 1980). Eq. (1), recently developed by Okutgen and co-workers (Okutgen et al., 1995), includes contributions of volumetric changes of the tablet core in addition to the other well-established mechanisms. In Eq. (1): P is the total internal stress in the film, E is the elastic modulus of the film, v is the polymer's Poisson's ratio, Φs represents the volume fraction of the solvent at the solidification point of the film, Φr is the volume fraction of solvent remaining in the dry film at ambient conditions, $\Delta\alpha_{(cubic)}$ is the difference between the cubical coefficient of thermal expansion of the film coat and the substrate, ΔT represents the difference between the Tg of the polymer and the temperature of the film during manufacturing and storage; ΔV is the volumetric change of the tablet core and V denotes the original volume of the tablet core. While this equation has been derived for polymeric solutions, the theory is also applicable to polymeric

dispersions. From Eq. (1), the elasticity of the polymer is directly proportional to total stress within a film. Therefore, factors that decrease the elastic modulus should decrease the internal stress within the polymeric film, resulting in greater film-tablet adhesion. Since the addition of plasticizers to the film coating formulation has been reported to affect the elastic modulus of the film (Gutierrez-Rocca and McGinity, 1994), the influence of plasticizers on film-tablet adhesion was investigated in the present study.

$$P = \frac{E}{3(1-v)}\left[\frac{\Phi s - \Phi r}{1-\Phi r} + \Delta\alpha_{(cubic)}\,\Delta T + \frac{\Delta V}{V}\right] \quad (1)$$

4. Results and discussion

The affects of plasticizer concentration on the adhesive properties of the acrylic polymer are shown in Fig. 2. Increasing the concentration of TEC in the coating formulation from 20 to 30% caused a significant increase in the elongation at adhesive failure with only a slight, non-significant decrease in the force of adhesion. These results were attributed to an increase in the elasticity of the polymer and a lowering of the internal stresses within the film. Increasing the degree of plasticization of the polymer generally results in a decrease in the elastic modulus as the polymer becomes more flexible (Gutierrez-Rocca and McGinity, 1994). Since the elastic modulus is directly proportional to the total stress within the film, as seen in Eq. (1), stronger adhesion resulted. In addition, these finding demonstrate that the elongation at adhesive failure of the coating reflects the ductility of the polymer.

Table 1
Glass transition temperatures of Eudragit® L 30 D-55 containing 20% plasticizer ($n = 6$)

Plasticizer	Glass transition temperature °C (S.D.)
None	98.1 (0.4)
TEC	36.5 (1.1)
PEG 6000	38.6 (2.5)
TBC	51.2 (2.2)
DBS	62.0 (3.6)

Previous research conducted by Fisher and Rowe (1976) showed a slight, non-significant decrease in the force of adhesion of hydroxypropyl methylcellulose films when the plasticizer propylene glycol was increased from 10 to 20%. One may conclude from their study that the plasticizer concentration does not significantly affect the adhesive properties of the polymer. The data obtained in the present study, however, clearly demonstrates the increased work required to remove the film from the surface of the tablet when the concentration of the plasticizer was increased, as evidenced by the greater area under the curve or adhesive toughness. Furthermore, these findings show that the adhesive toughness in conjunction with the force of adhesion provides a more complete understanding of the mechanisms involved in the adhesive process.

Force-deflection diagrams obtained from the Chatillon butt adhesion experiments as a function of the plasticizer type are shown in Fig. 3. The force of adhesion, elongation at adhesive failure, and the adhesive toughness were greater for the acrylic films plasticized with the water soluble plasticizers, TEC and PEG 6000, whereas films containing the water insoluble plasticizers, TBC and DBS, exhibited lower adhesive strength. Scanning electron micrographs of the film-tablet interface support these findings, as shown in Fig. 4. When the water soluble plasticizers were incorporated in the film coating, few void spaces were observed, indicating good polymer adhesion to the substrate. Large voids between the film and the tablet surface were found when the water insoluble agents were used to plasticize the acrylic polymer, indicating poor adhesion. These results were attributed to the extent of polymer-plasticizer interactions and the effectiveness of the plasticizing agent in lowering the internal stresses within the film coating. The Tg of the polymer containing the various plasticizers are shown in Table 1. The TEC and PEG 6000 lowered the Tg to a greater extent than the water insoluble plasticizers, demonstrating that the water soluble plasticizers were more effective in disrupting the intermolecular attractions between the polymer chains. As the Tg was lowered, the polymeric film became more flexible and the elastic modulus of

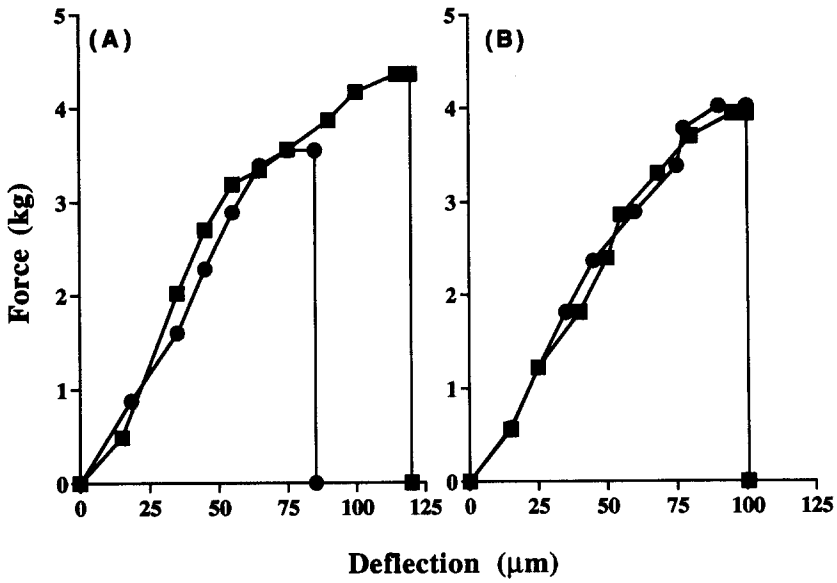

Fig. 5. Force-deflection profiles obtained from butt adhesion experiments as a function wax in the tablet core. (20% plasticizer, 10% Eudragit® L 30 D-55). (A) PEG 6000, (B) TBC, (■) 0% wax tablet core and (●) 30% wax tablet core.

the polymer decreased, resulting in lower internal stresses within the film and, thus, stronger adhesion. The results in the present study demonstrate a relationship between the mechanical, thermal, and adhesive properties of the acrylic polymer.

Previous research has shown that increasing tablet hydrophobicity caused a decrease in film-tablet adhesion when the acrylic polymer was plasticized with TEC, a water soluble agent (Felton and McGinity, 1996). In the present study, adhesion of the polymer plasticized with PEG 6000 was also found to be significantly influenced by the hydrophobicity of the tablet surface, as shown in the force-deflection profiles in Fig. 5a. When the amount of wax in the tablet core was increased from 0 to 30%, decreased adhesion of the polymer resulted. Interestingly, when TBC was incorporated into the film coating formulation, no significant differences in adhesion were found when the tablet hydrophobicity was increased, as shown in Fig. 5b. The adhesive properties of the films plasticized with DBS were also found to be unaffected by the hydrophobicity of the tablet. These results were attributed to the interfacial interactions between the film and the tablet surface.

To further investigate these results, droplets of the aqueous polymeric dispersions containing the various plasticizing agents were applied to the uncoated tablet cores and the contact angles of the coating suspensions were determined. Fig. 6 shows the contact angle of the acrylic dispersions as a function of the level of wax in the tablet. All coating formulations exhibited a significant increase in contact angle as the amount of wax in the tablet core was increased from 0 to 15%, irrespective of the plasticizing agent. When the wax content was increased from 15 to 30%, however, only the polymeric dispersions containing the water soluble plasticizers exhibited further increases in contact angles. Several researchers have demonstrated that contact angles of a polymeric solution were related to the surface free energy and that the wettability of the tablet surface influenced polymer adhesion (Wood and Harder, 1970; Harder et al., 1971; Huntsberger, 1967). Adhesion between a polymer and the surface of a tablet is due to the interactions of the intermolecular bonding forces. These forces consist primarily of hydrogen bonds, and, to a lesser extent, the weaker dipole-dipole and dipole-induced dipole interactions (Packham, 1992). As the

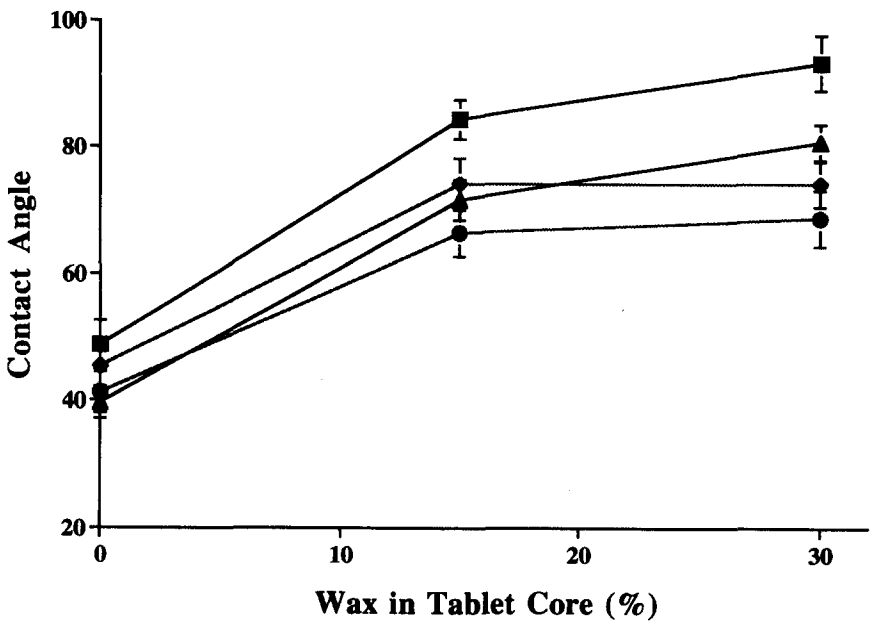

Fig. 6. Influence of plasticizer type on the contact angle of the aqueous acrylic dispersions as a function of wax in the tablet core. (▲) 20% TEC, (■) 20% PEG 6000, (◆) 20% TBC, (●) 20% DBS.

hydrophobicity of the tablet surface increased, fewer hydrogen bonds formed between the tablet and the polymeric films. For films containing the hydrophilic plasticizers TEC or PEG 6000, adhesion of the polymer was significantly decreased. With increased tablet hydrophobicity, a greater number of dipole-dipole interactions occurred between the tablet surface and the films containing the hydrophobic TBC or DBS, thus causing little change in the adhesive properties of the polymer.

The influence of storage conditions and physical aging of film-coated tablets and pellets is a major area of interest in the pharmaceutical sciences. Previous researchers have demonstrated the affects of storage conditions on the dissolution properties of tablets and on the mechanical properties of free films and film-coated solids (Felton et al., 1996; Amighi and Möes, 1989; Sinko et al., 1990; Gutierrez-Rocca and McGinity, 1993). The results in Fig. 7 depict the decrease in the adhesive properties of the acrylic resin as a function of storage condition. After 2 weeks of storage at 93% relative humidity and room temperature, dramatic decreases in the force of adhesion, elongation at adhesive failure, and adhesive toughness were found. These results are in agreement with Okhamafe and York (1985) who found a decrease in the force of adhesion of films derived from hydroxypropyl methylcellulose when coated tablets were stored at elevated humidity. The decreased adhesive properties were attributed to changes in the internal stresses within the film coating. While water molecules have been reported to plasticize the polymer (Hancock and Zografi, 1994; Gutierrez-Rocca and McGinity, 1993), the swelling of the film and tablet core as water diffuses through the coating during storage caused the formation of new stresses within the polymer and resulted in decreased adhesion.

Cohesive failure of the coated tablets occurred during the testing procedure when the tablets were stored at high humidity for periods of time greater than 2 weeks. Rather than the film being separated from the tablet surface, the tablet laminated during the adhesion experiments. The bonding between the film and tablet surface was greater than the bonding between the powdered particles within the tablet, thus the tablet was pulled apart. Cohesive failure of the coated tablets was due to a weakening of the tablet compacts by the sorption

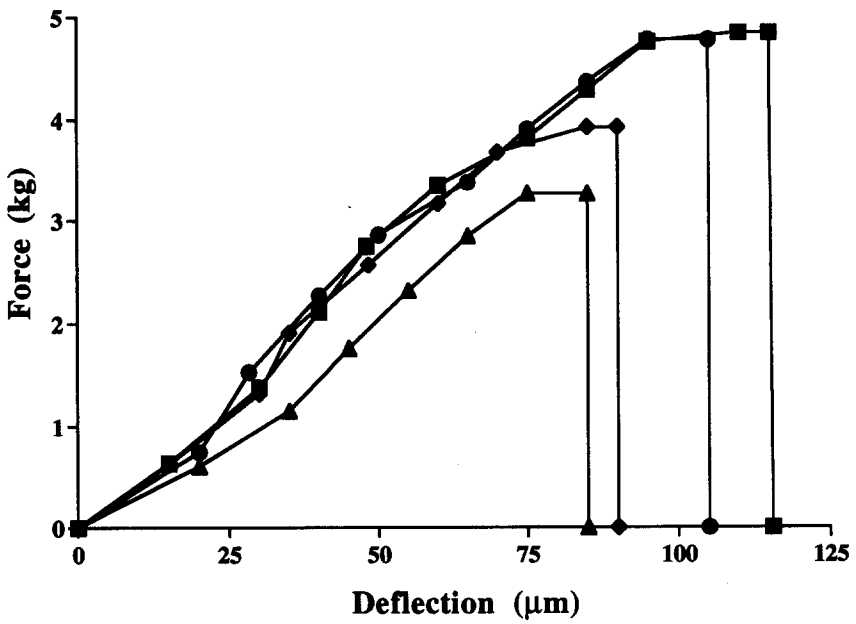

Fig. 7. Influence of aging on the force-deflection profiles obtained from butt adhesion experiments as a function of storage condition. (■) Initial, (▲) 2 weeks at 93% RH/RT, (◆) 3 months at 0% RH/RT and (●) 3 months at 40°C.

of moisture from the environment. This conclusion is supported by Riepma et al. (1992) who reported that the strength of the tablet compact decreased when lactose tablets adsorbed moisture from a humid environment.

The adhesive properties of the acrylic polymer were also found to decrease within 3 months of storage at 0% relative humidity and room temperature, as shown in Fig. 7. This decreased adhesion was again attributed to the internal stresses within the polymeric film. Previous reports in the literature have shown that storage of the acrylic polymer at low humidity causes the film to become brittle as the result of solvent or moisture loss (Gutierrez-Rocca and McGinity, 1993). In the present study, the decreased elongation at adhesive failure demonstrated that the polymer became less ductile and the elasticity of the film decreased, resulting in higher stresses within the film and decreased adhesive properties of the polymer.

The elongation at adhesive failure and the adhesive toughness of the acrylic film were found to decrease after 3 months of storage at 40°C, although no significant differences in the force of adhesion were noted, as depicted in Fig. 7. These results were attributed to changes in the mechanical properties of the film. At high temperatures, additional water within the film is driven off, accounting for the observed decreased elongation at adhesive failure and adhesive toughness. The temperature at which the coated tablets were stored was slightly higher than the Tg of the film. At temperatures above the Tg, polymer chains are mobile (Sinko et al., 1990) and can position themselves to minimize the internal stress in the polymer. Therefore, the decrease in the internal stresses in the polymer in combination with the decreased elasticity of the film from water evaporation resulted in minimal changes in the measured force of adhesion.

5. Conclusions

Plasticizers which interact to a greater extent with the polymer to disrupt polymer-polymer interactions were found to adhere more strongly to tablet compacts than agents that had limited plasticizing effects. The hydrophobicity of the tablet

surface significantly influenced adhesion when the polymer was plasticized with hydrophilic compounds whereas tablet hydrophobicity was not found to affect film adhesion when water insoluble plasticizers were incorporated in the coating formulation. Adhesion of the acrylic film to tablet compacts was generally found to decrease during storage. The present study demonstrated a relationship between the mechanical, thermal, and adhesive properties of an acrylic resin copolymer. The elongation at adhesive failure was found to reflect the ductility of the polymeric film. In addition, the present study showed that the adhesive toughness of the polymer in conjunction with the force of adhesion provided a more complete understanding of the mechanisms involved in the adhesion process.

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