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Studies of the mechanical properties of free films prepared using an ethylcellulose pseudolatex coating system

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Abstract

Free films produced using a commercially available ethylcellulose (EC) pseudolatex coating system (Aquacoat®) were prepared by a spraying method. The effect of either of 10 different plasticizers representing three chemical classes (citrate esters, di-acid esters and fatty acids/alcohols) on free film mechanical properties was determined using an Instron tensile testing apparatus. The plasticizers were each added to the EC pseudolatex formula at levels of 25, 30 or 35% (w/w), respectively. Increasing the amount of plasticizer led to an increase in free film elongation and a decrease in modulus and stress. Differences among the individual plasticizers and classes of plasticizers evaluated were correlated with their molecular structure and a rank order could be identified. Selected films containing 30% plasticizer were evaluated under conditions of elevated temperature and humidity for storage times of up to 1 month. The results indicated that plasticizer type and amount and elevated storage temperature and humidity influence free film mechanical behavior, particularly free film elongation and toughness. These changes probably arise from further gradual coalescence of EC pseudolatex particles.

Key words: Ethylcellulose; Mechanical testing; Pseudolatex; Aqueous coating; Plasticizer; Curing; Polymer film

1. Introduction

Ethylcellulose (EC) represents one of a small number of approved polymers available for pharmaceutical applications. Traditionally, ethylcellulose has been applied using solvent-based systems; however, the advent of latex technology in the pharmaceutical industry has provided a means for applying the polymer using an aqueous vehicle. Thus, problems associated with solvent-based systems such as flammability/explosion hazards, toxicity and environmental contamination can be eliminated using latex systems. Two types of EC pseudolatices are commercially available. Pseudolatices differ from true latex systems in their method of preparation; both are aqueous dispersions of polymer in water (Chang et al., 1987; Ghebre-Sellassie et al., 1988).

The most notable difference between solvent-based and latex/pseudolatex coating systems lies in the mechanism of film formation. Such differ-

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ences influence the release profile through films which are applied to drug loaded spheres (Iyer et al., 1990). The formation of film coatings from a polymer solution involves spraying of the coating solution onto a suitable substrate followed by the wetting and deformation of the sprayed droplets and entanglement of the polymer chains. As the solvent evaporates, the polymer gels and, with further loss of water, a continuous film is produced. In contrast, the formation of films from latex systems involves a somewhat different series of events. Like solution-based coatings, latex systems are applied by spraying methods. However, if film formation is to occur, the dispersed polymer spheres with an average size of 0.171 μm (Nakagami et al., 1991) must experience coalescence. The polymer chains are not solvated so additional energy is required to cause entanglement. This process involves overcoming particle-particle repulsion due to electrostatic forces and particle rigidity. Capillary forces resulting from the high surface tension produced as water between latex spheres evaporates provide much of the energy required for film formation (Bindschaedler et al., 1983).

However, while capillary forces provide a portion of the energy to cause coalescence, formation of films from Aquacoat®, one type of EC pseudolatex, is not possible without the addition of plasticizers. Plasticizers are added to an Aquacoat® formulation in order to reduce the glass transition temperature, T_g. Thus, if the coating formulation is applied at temperatures above T_g, coalescence will be facilitated. Following the application of the coating dispersion, an additional processing step commonly referred to as 'curing' is also required, since coalescence is not necessarily completed during the coating process. Curing involves exposing the coated material to temperatures above T_g for a period of time such that the process of coalescence may be completed. Careful selection of the type and amount of plasticizer as well as the processing temperatures ensures a uniform, reproducibly coated product.

Despite the importance of the plasticizer and processing temperatures to the formation of films from latex systems, few studies have addressed these factors from a fundamental perspective. The mechanical evaluation of free films provides such a fundamental means for qualitatively evaluating the nature of the interactions between a coating polymer and various additives including plasticizers (Allen et al., 1972; Entwistle and Rowe, 1979; Okhamfe and York, 1983, 1984, 1986; Jenquin et al., 1992). While numerous studies of this type may be cited for films prepared from polymer solution-based systems, limited studies are available for the pseudolatex systems. By exposing free films to various temperatures following preparation, it may also be possible to study the curing process.

Thus, the principle objectives of this work were (1) to study the effects of plasticizer type and amount on the mechanical behavior of free films in order to gain insight into the nature of the polymer/plasticizer interactions and (2) to evaluate the effects of storage on free film mechanical behavior as a means for studying the curing process. These results should be useful in formulating films for optimum performance in pharmaceutical applications.

Table 1
Plasticizers used for the preparation of free films

Branched esters	Di-acid esters	Fatty acids/alcohols
Triethyl citrate (TEC) [a]	dibutyl sebacate (DBS) [c]	oleic acid (OLAC) [e]
Acetyl triethyl citrate (ATEC) [a]	dimethyl sebacate (DMS) [c]	oleyl alcohol (OALC) [b]
Tributyl citrate (TBC) [a]	dibutyl adipate (DBA) [d]	
Acetyl tributyl citrate (ATBC) [a]		
Triacetin (TRI) [b]		

[a] Morflex, Greensboro, NC; [b] Eastman Kodak, Kingsport, TN; [c] Sigma, St. Louis, MO; [4] Aldrich, Milwaukee, WI; [e] Fisher Scientific, Fair Lawn, NJ.

2. Materials and methods

2.1. Chemicals

All chemicals were used as received. Aquacoat® was obtained as a gift from FMC (Newark, DE). The plasticizers used are listed in Table 1.

2.2. Composition of films

Films were prepared using the 10 different plasticizers at three levels. Appropriate levels of plasticizer were determined during preliminary studies to include 25, 30, and 35% (w/w) plasticizer evaluated as a percentage of the ethylcellulose weight in Aquacoat® (27% ethylcellulose). Thus, for each film prepared, 250 g of Aquacoat® were used and the amount of plasticizer added was 16.9 g (25%), 20.2 g (30%), or 23.6 g (35%).

Additional films were prepared to study the effect of curing time and temperature. Only four different plasticizers (DBS, TBC, ATBC, and OALC) at 30% (w/w) were evaluated in this part of the study. The free film samples were stored on teflon tape which was adhered to a glass plate. The following storage conditions were employed: 45°C, ambient relative humidity (RH); 45°C, 75% RH; 60°C, ambient RH; and 80°C, ambient RH. 'Ambient' refers to the RH of the rooms in which the ovens were housed. This humidity translates to a nearly 0% level in each of the sample ovens in which humidity was considered to be ambient. Samples were removed at the following times: 0, 3 days and 1, 2, 3, and 4 weeks and were maintained at ambient room conditions (22°C, 55% RH) prior to mechanical testing.

2.3. Apparatus for the preparation of films

The apparatus used for the preparation of free films consisted of a horizontally mounted rotating turntable (Harbaugh Machine, Youngwood, PA) with a diameter of 25 cm and adjustable speed, a spray nozzle (Glatt Air Techniques, Ramsey, NJ) and a source of warm drying air.

2.4. Film preparation process

Each film formula was prepared by adding the plasticizer to Aquacoat® and stirring for 1 h using a propeller mixer at a fixed speed. The coating formula was fed through the binary nozzle at a fixed flow rate (12 ml/min) and atomizing air pressure was fixed at 0.7 bar. The nozzle was held manually at a height of approx. 15 cm above the surface of the turntable. Films were sprayed onto three strips of teflon tape (5 cm × 24 cm) which were adhered to a glass plate that was ultimately placed onto the leveled turntable surface. After the application of each layer, warm drying air at 70°C was applied until the layer appeared to be dry. The source of the warm air was held approx. 1–3 cm above the film surface. The temperature of the air was measured at the source so actual drying air temperature at the film surface was somewhat lower due to evaporative cooling. For each trial, 25 coating layers were applied. Following the application of the final coat, the films were placed in an oven at 45°C for 1 h. The films were then removed from the teflon surface and were cut into uniform pieces of 2.5 cm × 4 cm. Film thickness was determined using a Starrett thickness gauge (Model 1015 MAZ, Athol, MA). Sample weight was also determined to provide an additional test of sample uniformity. Those samples used in the storage study were placed in a standard oven (Fisher Scientific, Fair Lawn, NJ) or a controlled humidity oven (Model PH-2G, Tabai, Japan) as described above. The changes in the weights and thickness of these samples were determined after they were removed from their respective chambers.

2.5. Mechanical evaluation of free films

Mechanical evaluation of free films was performed using a tensile testing apparatus (Model 4206, Instron Corp., Canton, MA) which was interfaced to a computer. Because of the brittle nature of the films, samples were tabbed using masking tape prior to placement into the sample grips. The cross-head speed was set at 1 mm/min. Stress at failure (N/mm^2), strain at failure (%)

and modulus (N/mm^2) were calculated from the raw data following transformation into a spreadsheet program and conversion to SI units. Stress at failure (N/mm^2) was calculated as the maximum load divided by the cross-sectional area of the sample prior to stretching (stress = force/ area = N/mm^2). It was assumed that, since the samples did not experience large percent elongation, the cross-sectional area did not change significantly. Strain at failure (%) was calculated as the change in the length at the time of sample failure of the films divided by the original length (L_0) expressed as a percent $\{(\Delta L/L_0) \times 100\%\}$. Young's modulus was calculated as the slope of the elastic (linear) portion of the stress-strain profile. Five samples were used to calculate a mean value for the study of plasticizer type and amount. Three samples were evaluated to determine a mean for the study of the effect of storage.

2.6. *Thermal analysis of films*

Thermal behavior of films was evaluated using a differential scanning calorimeter (Model 120, PL Thermal Sciences, Sunbury-on-Thames, U.K.). Covered samples, approx. 18 mg, were tested under a N_2 atmosphere at a heating rate of 10°C/min. The temperature range evaluated was −20 to 80°C.

3. Results and discussion

The samples produced using the spraying method were macroscopically homogeneous with respect to their content, thickness and weight. Homogeneity of content was assessed by visual observation of the films. No regions appeared where the translucency of the films was variable so it was assumed that a macroscopic separation of phases during the film formation process did not occur. Nevertheless, while no gross phase separation was visible, the films produced from the pseudolatex were not transparent, unlike those typically produced using solvent-based systems. This translucency may result because of the presence of pseudolatex emulsifying agents (i.e., sodium lauryl sulfate, cetyl alcohol) which migrate in the film structure forming minuscule 'islets' (Bindschaedler et al., 1987a, 1989).

Attempts to produce free films by casting methods did not succeed as phase separation occurred. The casting procedure was performed at 60°C. The slow process of evaporation during this casting process led to a breakdown of the dispersion so that EC particles become flocculated and hence separated from the aqueous phase. Chainey et al. (1985) employed a 'flash casting' method which involved the spraying of latexes of various polymers onto a hotplate and found this method to be superior for free film

Table 2
Mean weight (g) and thickness (mm) for free films produced using an EC latex containing 25, 30 or 35% (w/w) plasticizer (n = 5) (the percent relative standard deviation appears in parentheses next to the mean values)

Plasticizer	% plasticizer (w/w)					
	Weight (g)			Thickness (mm)		
	25	30	35	25	30	35
DBS	0.4335 (3.10)	0.4755 (4.16)	0.5004(1.78)	0.44 (1.9)	0.48(4.2)	0.50(1.8)
DBA	0.4169(11.9)	0.5121 (7.87)	0.3774(4.16)	0.41(10)	0.52(8.7)	0.38(5.2)
DMS	0.5061 (2.89)	0.3375 (2.52)	0.5828(2.78)	0.53 (3.4)	0.33(2.5)	0.57(4.0)
TEC	0.4574 (4.65)	0.5357 (2.86)	0.4837(3.98)	0.45 (2.2)	0.53(2.8)	0.47(4.0)
ATEC	0.4213 (4.04)	0.4881(10.3)	0.5594(8.49)	0.43 (5.9)	0.50(9.3)	0.54(7.6)
TBC	0.6508 (2.67)	0.5134 (5.48)	0.4734(6.64)	0.61 (5.9)	0.51(5.5)	0.47(5.5)
ATBC	0.5146(10.0)	0.5210 (9.54)	0.4515(3.55)	0.54 (7.5)	0.52(8.6)	0.45(3.3)
TRI	0.5116 (3.74)	0.4376 (3.10)	0.4481(0.95)	0.53 (3.4)	0.42(2.7)	0.45(2.0)
OALC	0.5430 (6.44)	0.4542 (3.42)	0.5446(2.20)	0.55 (4.0)	0.47(3.5)	0.56(2.2)
OLAC	0.3597 (3.50)	0.4386 (2.86)	0.5491(3.01)	0.39 (4.2)	0.44(3.1)	0.53(3.0)

preparation compared with casting onto other substrates.

The results for the mean thickness and weight of the free films produced using the spray apparatus are presented in Table 2. The films produced were fairly uniform with respect to thickness and weight. Mean film weight varied from 0.3375 to 0.6508 g. Mean film thickness ranged from 0.38 to 0.61 mm. The percent relative standard deviation ($n = 5$) was always less than 12% and was usually below 8%.

3.1. Effect of plasticizer type and amount on free film mechanical properties

Film coating systems which produce tough films with high mechanical strength and elongation are the best suited for tablet coating (Munden et al., 1963). Alternatively, softer (lower modulus, higher elongation) films are recommended for use in transdermal systems (Lin et al., 1991).

A typical stress-strain profile of Aquacoat® films in which DBS was employed as the plasticizer is shown in Fig. 1. Increasing the amount of plasticizer leads to a reduction of the maximum stress and an increase in the strain at rupture values. Additionally, the area under the stress-strain curve increases, indicating that these films become tougher (higher energy to break) when larger amounts of plasticizer are employed.

Comparing the values obtained for the stress, strain and modulus of all the films evaluated (Fig. 2), one sees similar trends with plasticizer content. Increasing the amount of plasticizer from 25 to 35% leads to a reduction of free film modulus and stress values while strain at rupture values increase. Similar trends were obtained by Lin et al. (1991) for Eudragit E100 films. Increasing plasticizer content from 0 to 5.5% led to an increase in percent elongation and a reduction of strength. Inconsistencies in these trends were evident such as the results for TBC, ATBC and TRI. These disparities may be explained by considering that the quality of the films produced using these plasticizers at the lowest levels was poor.

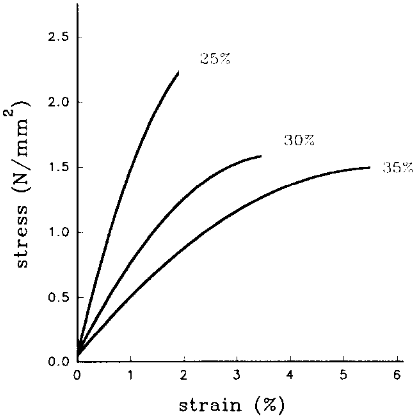

Fig. 1. Stress-strain isotherms for free films containing different amounts of DBS.

Fig. 2 also illustrates the relative effectiveness of plasticizers and even classes of plasticizers to influence free film mechanical behavior. If maximum stress for the free films is considered, the values which were obtained for the di-acid esters and for the fatty acid/alcohol classes were generally lower than those obtained for the citrate esters and triacetin. This observation may be correlated with molecular structure as both the di-acid esters and the fatty acids/alcohols are long-chain molecules while the citrate esters and triacetin are branched.

Within a given chemical class, additional generalizations are possible which, again, imply different types of interactions between the plasticizers and EC. For example, when considering modulus, a parameter characteristic of sample stiffness: higher modulus values are associated with films having greater stiffness while lower modulus values represent softer films. For the free films prepared using plasticizers from the class including the citrate esters or triacetin, the lowest modulus values were obtained for free films prepared using TEC or TBC. These two plasticizers have free hydroxyl groups while ATEC, ATBC and triacetin are completely esterified molecules.

A note should be added concerning plasticizer molecular weight. While it appears that TEC produces even softer films than TBC, this is not the case when considering the plasticizers' content on a molar basis. An inverse relationship exists between the molecular weight of these plasticizers and their modulus values. Thus, by

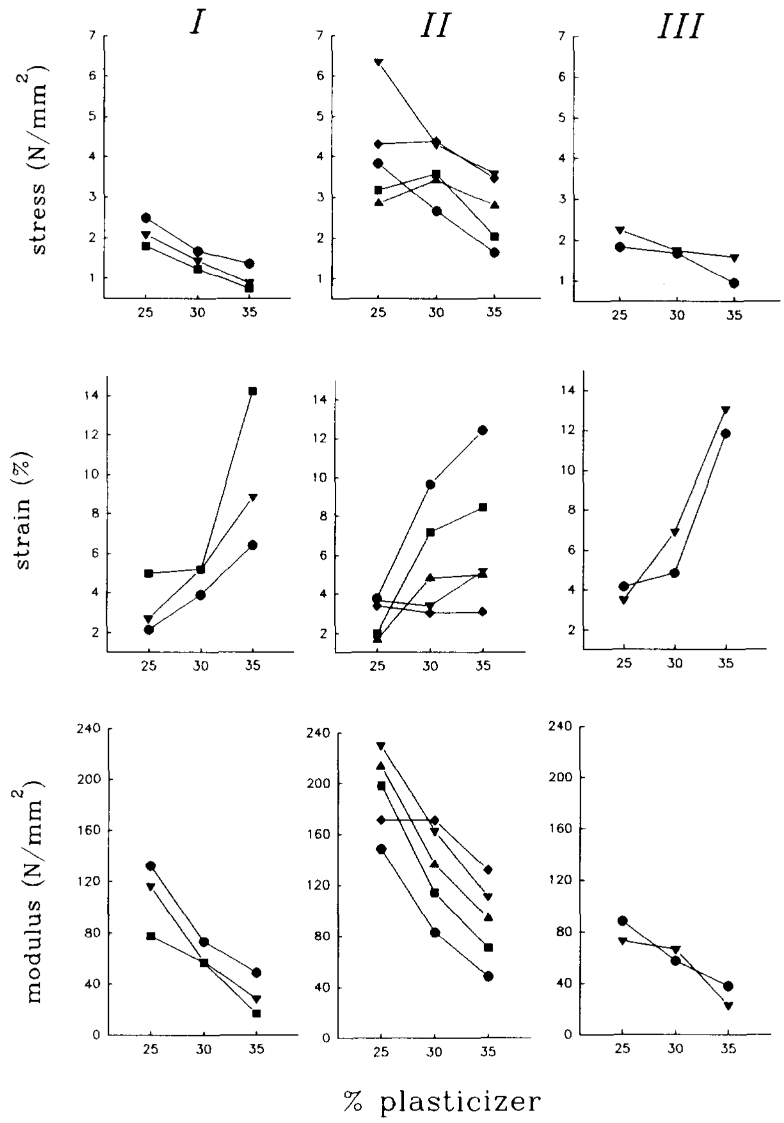

Fig. 2. Effect of plasticizer type and amount on free film stress (N/mm^2), strain (%) and modulus (N/mm^2). I: (●) DBS, (▼) DBA, (■) DMS; II: (●) TEC, (▼) ATEC, (■) TBC, (▲) ATBC, (◆) TRI; III: (●) OALC, (▼) OLAC.

dividing the modulus by the molecular weight of the plasticizer, a fairly consistent value (the 'adjusted modulus') is obtained. Adjusted modulus values for films containing 35% plasticizer are presented in Table 3.

3.2. *Effect of storage on free film mechanical properties*

The effect of storage on the change in the weight of free films following storage is illustrated in Fig. 3. Weight change is dramatic at 80°C for all of the plasticizers studied. Permanence is generally considered an important criterion for the selection of a plasticizer to be used in a film coating formula. From this perspective, OALC would be considered the poorest plasticizer among the four evaluated. When compared to the other plasticizers, films prepared with OALC experienced the largest change in weight when stored at 60°C. At 80°C, following storage

Table 3
Molecular weight (g/mol), mean modulus (N/mm^2) and adjusted modulus for free films containing 35% w/w plasticizer

Plasticizer	Mol. wt (g/mol)	Modulus (N/mm^2)	Adjusted modulus
TEC	276.3	48.37	0.18
TBC	360.4	70.62	0.20
ATEC	318.3	110.87	0.35
ATBC	402.5	94.16	0.23
TRI	218.2	131.76	0.60
DBS	314.5	48.64	0.15
DBA	258.4	28.66	0.11
DMS	230.3	17.09	0.07
OLAC	282.5	23.19	0.08
OALC	268.5	38.04	0.14

times greater than 1 week, the OALC films had deteriorated to the extent that they were so brittle that they broke apart when attempts were made to remove them from the teflon surface. Nevertheless, for some latex coating systems, incorporation of large amounts of plasticizers followed by the evaporative loss of the plasticizer led to the formation of strong films (Bindschaedler et al., 1987b). For other latex systems plasticizers may be added to merely act as 'coalescing aids' and evaporate from the system with time following film formation (Wang and Winnik, 1990).

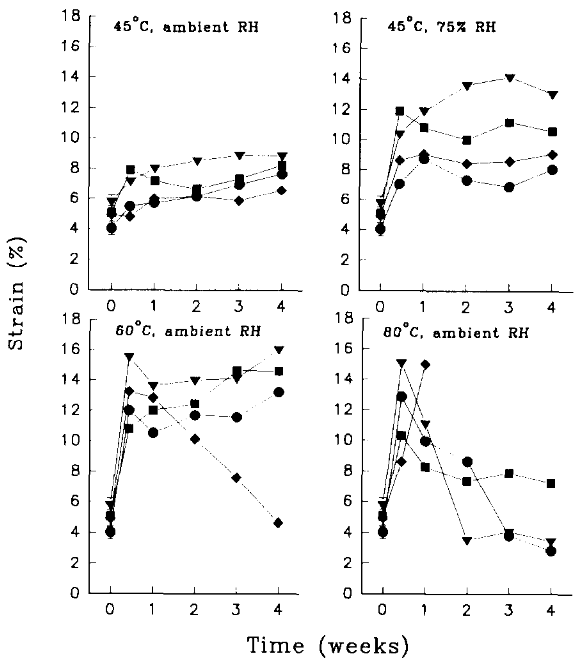

Fig. 4. Effect of storage on the strain (%) for free films: (●) DBS, (▼), TBC, (■) ATBC, (◆) OALC.

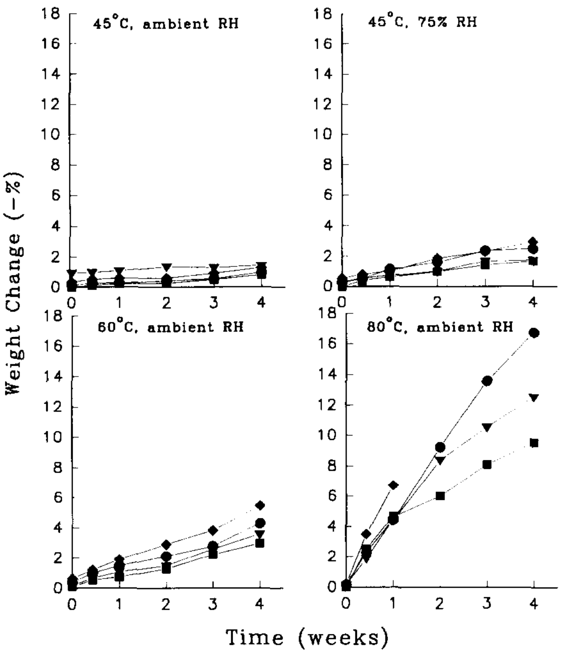

Fig. 3. Effect of storage on the weight change (−%) of free films: (●) DBS, (▼), TBC, (■) ATBC, (◆) OALC.

Fig. 4–6 represent the influence of storage under four different conditions on the strain, modulus and stress for free films. Considering the films stored at 45°C (ambient humidity), 45°C (75% relative humidity) and 60°C (ambient humidity), several trends may be noted. Storage at higher temperatures and under high humidity conditions is associated with increased in film elongation. Modulus is inversely related to strain at rupture. Stress does not change or changes only slightly following storage. Of all the tensile properties studied, strain at rupture changed more dramatically than the modulus or the stress.

Changes in the free film properties which resulted following storage at 80°C are markedly different from the results for films stored under the other conditions. The differences may be attributed to the evaporative loss of plasticizer (Fig. 3). For DBS, TBC and ATBC, this loss is associated with a reduction of strain and an increase in the modulus and stress values. For OALC, the trends were generally the opposite. Strain at rupture values increased while stress and modulus decreased. The opposite trend for OALC may be attributed to a greater degree of coalescence for these films while the trends observed for DBS, TBC and ATBC correlated with the behavior observed in the first part of this study: a decrease in the amount of plasticizer leads to an increase in free film stress and modulus and a decrease in the % elongation.

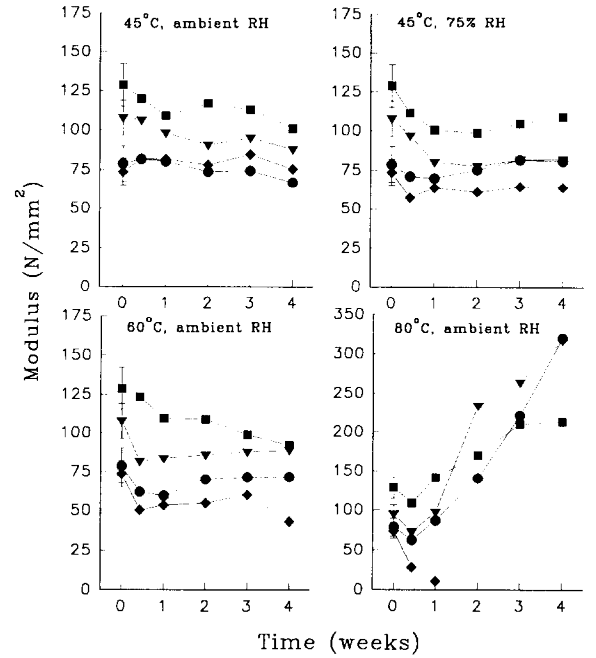

Fig. 6. Effect of storage on the modulus (N/mm²) for free films: (●) DBS, (▼), TBC, (■) ATBC, (◆) OALC.

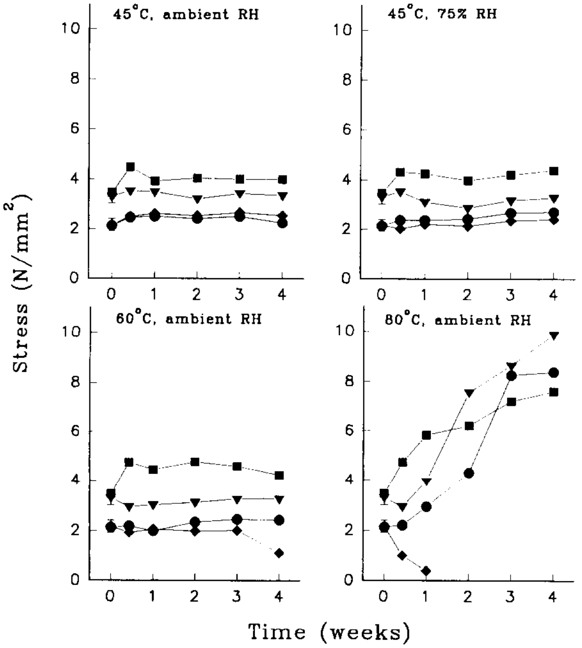

Fig. 5. Effect of storage on the stress (N/mm²) for free films: (●) DBS, (▼), TBC, (■) ATBC, (◆) OALC.

An example of a typical DSC profile for a film prepared in which TBC (30%) was incorporated as the plasticizer is presented in Fig. 7. This film was not exposed to the extended storage conditions described but was used in the evaluation of plasticizer type and amount. Characteristic of films prepared using any of the plasticizers evaluated are two transitions. These transitions do not appear to be glass transitions which are characterized by endothermic baseline shifts. Furthermore, dramatic transitions like these would not be expected for films prepared using ethylcellulose as glass transitions, determined using DSC, are subtle for this polymer (Van Bommel et al., 1989). Instead, they appear more as an endothermic phase transition. The transition between 50 and 60°C likely represents the melting of cetyl alcohol (m.p. 54–56°C). By storing films at lower temperatures, only partial coalescence is effected; more than one phase is still present so coalescence only occurs in this phase. At higher temperatures, 60 or 80°C, the films exist more uniformly so more extensive coalescence is possible. Hence, a greater change in free film mechanical properties results.

For films stored under conditions of elevated humidity, changes in free film mechanical properties were intermediate to those observed for storage under ambient conditions at 45 and 60°C. Changes in free films of HPMC, another cellulose ether, which occur following high humidity

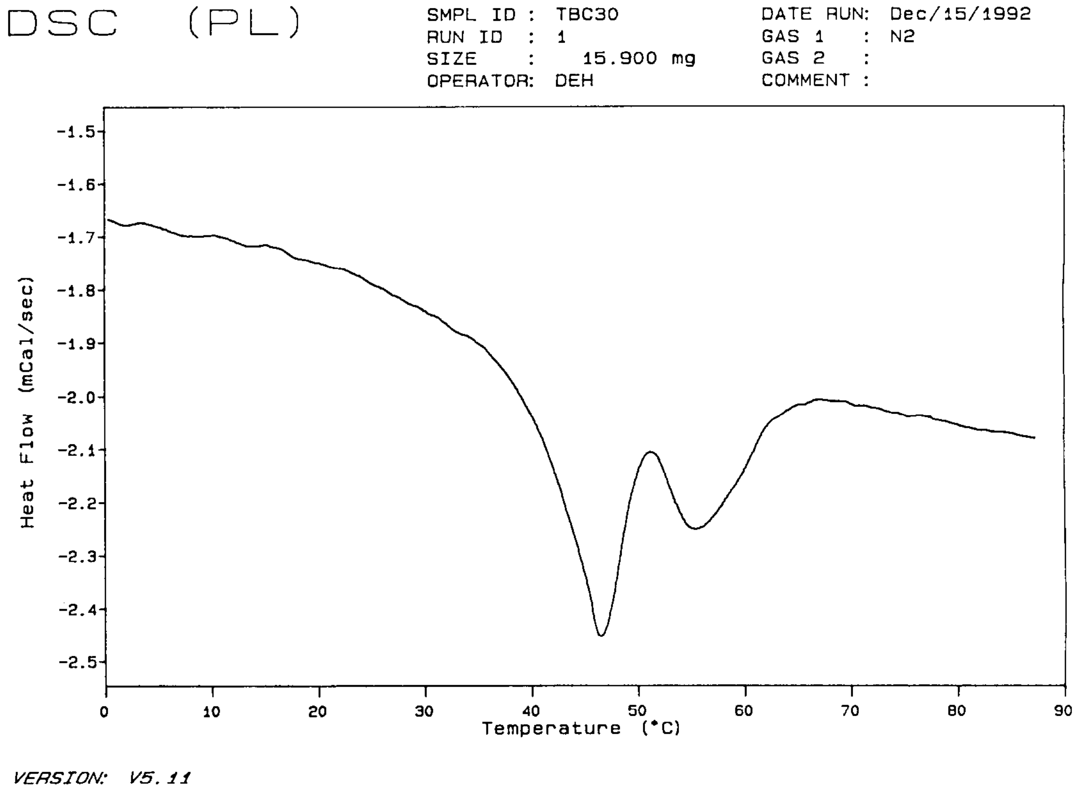

Fig. 7. DSC thermogram for a free film prepared using Aquacoat® with 30% TBC as the plasticizer.

storage were found by Aulton et al. (1981) and were explained by consideration of the competition of water for 'active sites' during storage. Water competes for bonding sites (i.e., free hydroxyl groups), thus reducing the number of polymer-polymer contacts (active centers). The water acts as a plasticizer. Thus, polymer chains are more free for random movement and greater coalescence is possible.

A comparison of the stress-strain profile of films prepared using TBC (30%) as the plasticizer following storage for 1 week under the various conditions (Fig. 8) indicates that the films toughen (greater area under the curve) with increased storage temperature or when samples stored at 45°C are exposed to elevated humidity. The effect at 45°C is slight and is likely insignificant. As stated earlier, tougher films are preferred as coat-

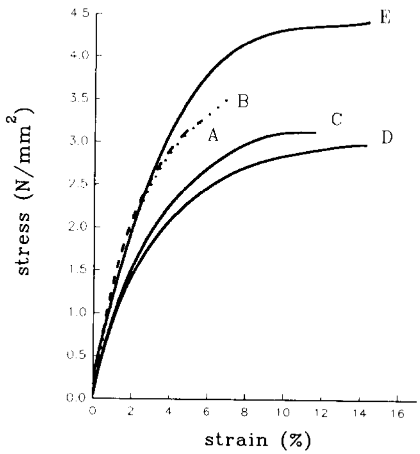

Fig. 8. Stress-strain isotherms for free films containing TBC (30%) following storage for 1 week under various conditions: (A) control, ambient storage; (B) 45°C, ambient RH; (C) 45°C, 75% RH; (D) 60°C, ambient RH; (E) 80°C, ambient RH.

ing materials. However, if exposure becomes too long the loss of plasticizer leads to a reduction of film elongation and toughness. An optimum curing time is implied by these findings.

While these studies imply that coalescence is taking place, it is not possible to definitively conclude that coalescence has been completed. An increase in the degree of coalescence implies that pseudolatex polymer chains have diffused completely across the particle-particle interface. However, more sophisticated techniques are necessary in order to confirm that coalescence has been completed. For example, Yoo et al. (1990) have correlated the interfacial diffusion process, using small-angle neutron scattering (SANS), with an increase in tensile strength of films produced from curing of sintered polystyrene latex particles.

4. Conclusions

Differences in the nature of plasticizer-polymer interactions may be deduced from mechanical studies of free films containing plasticizers of various chemical classes. A different type of molecular interaction is operative when either a long-chain or branched-type molecule is incorporated into the coating formula. Differences within a given class also reveal that a rank order may be obtained.

When free films are stored under various conditions, changes in free film mechanical parameters, particularly percent elongation, are indicative that further gradual coalescence has taken place. While curing at 45°C was associated with an improvement in free film mechanical properties, greater change is effected at higher temperatures (60°C) or following high humidity storage (45°C, 75% RH). While storage at 80°C leads to an improvement in free film mechanical properties following periods of short-term storage, the possible loss of plasticizer with time from films stored under these conditions, had detrimental effects.

Future work will involve preparation of coated beads containing drug is in progress and attempts will be made to correlate the characteristics of free and applied films prepared using Aquacoat®.

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6. References

Allen, D.J., DeMarco, J.D. and Kwan, K.C., Free films: I. Apparatus and preliminary evaluation. *J. Pharm. Sci.*, 61 (1972) 106–109.

Aulton, M.E. and Abdul-Razzak, M.H., The mechanical properties of hydroxypropylmethylcellulose films derived from aqueous systems: I. The influence of plasticizers. *Drug Dev. Ind. Pharm.*, 7 (1981) 649–668.

Bindschaedler, C., Gurny, R. and Doelker, E., Influence of emulsifiers on film formation from cellulose acetate latexes. Experimental study of phase separation phenomena due to sodium dodecyl sulfate: I. *J. Appl. Polym. Sci.*, 34 (1987a) 2631–2647.

Bindschaedler, C., Gurny, R. and Doelker, E., Influence of emulsifiers on film formation from cellulose acetate latexes. Modeling approach to the fate of emulsifiers in highly plasticizers films. II. *J. Appl. Polym. Sci.*, 37 (1989) 173–182.

Bindschaedler, C., Gurny, R. and Doelker, E., Mechanically strong films produced from cellulose acetate latexes. *J. Pharm. Pharmacol.*, 39 (1987b) 335–338.

Bindschaedler, C., Gurny, R. and Doelker, E., Theoretical concepts regarding the formation of films from aqueous microdispersions and application to coating. *Labo-Pharma-Probl. Tech.*, 31 (1983) 389–394.

Chainey, M. and Wilkinson, M.C., Preparation of polymer latex films by a flash casting technique. *J. Appl. Polym. Sci.*, 30 (1985) 4273–4285.

Chang, R., Hsiao, C.H. and Robinson, J.R., A review of aqueous coating techniques and preliminary data on release from a theophylline product. *Pharm. Technol.*, 11 (1987) 56–68.

Entwistle, C.A. and Rowe, R.C., Plasticization of cellulose ethers used in the film coating of tablets. *J. Pharm. Pharmacol.*, 31 (1979) 269–272.

Ghebre-Sellassie, I, Iyer, U., Kubert, D. and Fawzi, M.B., Characterization of a new water-based coating for modified-release preparations. *Pharm. Technol.*, 12 (1988) 96–106.

Iyer, U., Hong, W., Das, N. and Ghebre-Sellassie, I., Comparative evaluation of three organic solvent and dispersion-based ethylcellulose coating formulations. *Pharm. Technol.*, 14 (1990) 68–86.

Jenquin, M.R., Sarabia, R.E., Liebowitz, S.M. and McGinity, J.W., Relationship of film properties to drug release from monolithic films containing adjuvants. *J. Pharm. Sci.*, 81 (1992) 983–989.

Lin, S., Lee, C. and Lin, Y., The effect of plasticizers on compatibility, mechanical properties and adhesion strength of drug free Eudragit E films. *Pharm. Res.*, 8 (1991) 1137–1143.

Munden, B.J., DeKay, H.G. and Banker, G.S., Evaluation of polymeric materials: I. *J. Pharm. Sci.*, 53 (1964) 395–401.

Nakagami, H., Keshikawa, T., Matsumura, M. and Tsukamoto, H., Application of aqueous dispersions of water-insoluble polymers for tablet and granule coatings. *Chem. Pharm. Bull.*, 39 (1991) 1837–1842.

Okhamfe, A.O. and York, P., Analysis of the permeation and mechanical characteristics of some aqueous-based film coating systems. *J. Pharm. Pharmacol.*, 35 (1983) 409–415.

Okhamfe, A.O. and York, P.Y., Effect of solids-polymer interactions on the properties of some aqueous-based tablet film coating formulations: II Mechanical characteristics. *Int. J. Pharm.*, 22 (1984) 273–281.

Okhamfe, A.O. and York, P., Mechanical properties of some pigmented and unpigmented aqueous-based film coating formulations applied to aspirin tablets. *J. Pharm. Pharmacol.*, 38 (1986) 414–419.

Van Bommel, E.M.G., Fokkens, J.G. and Crommelin, D.J.A., Effects of additives on the physico-chemical properties of sprayed ethylcellulose films. *Acta Pharm. Technol.*, 35 (1989) 232–237.

Wang, Y. and Winnik, M.A., Effect of coalescing aid on polymer diffusion in latex films. *Macromolecules*, 23 (1990) 4731–4732.

Yoo, J.N., Sperling, L.H., Glinka, C.J. and Klein, A., Characterization of film formation from polystyrene latex particles via SANS: 1. Moderate molecular weight. *Macromolecules*, 23 (1990) 3962–3967.