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Dry and wet strengths of polymeric films prepared from an aqueous colloidal polymer dispersion, Eudragit RS30D

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Summary

The mechanical properties of polymeric films are traditionally evaluated on dry cast or sprayed films. In this study, films were cast from an aqueous colloidal polymer dispersion [poly(ethylacrylate methylmethacrylate-trimethylammonioethylmethacrylate chloride) copolymer, Eudragit RS30D]. The mechanical properties were measured not only in the dry but also in the wet state by a puncture test, wherein a puncturing probe was driven through the dry or wet film. The puncture strength and elongation at puncture were measured as a function of crosshead speed, film thickness, type and concentration of the plasticizer (triethyl citrate or acetyl tributyl citrate), and storage humidity. The puncture strength decreased and the elongation increased with increasing plasticizer concentration with dry films. An HPLC assay was used to determine the amount of plasticizer in the polymer films before and after exposure to an aqueous medium in order to correlate the plasticizer content in the films with the resultant mechanical properties. Triethyl citrate, a water-soluble plasticizer, leached from the polymeric films resulting in significant changes in the mechanical properties when compared to dry or wet films prepared with the non-leaching, water-insoluble acetyl tributyl citrate. The mechanical properties of the films were affected by the storage humidity.

Introduction

Water-insoluble polymers are applied as coatings to solid dosage forms either from organic polymer solutions or preferably, because of the absence of organic solvents, from aqueous colloidal polymer dispersions (latexes or pseudolatexes). The permeability and mechanical properties of such coatings are often determined on free polymer films prepared by casting or spraying techniques. The mechanical properties, including tensile strength and elongation at break, energy to break, and various moduli, are traditionally evaluated by stress-strain tensile tests (Allen et al., 1972; Fell et al., 1979; Aulton and Abdul-Razzak, 1981; Okhamafe and York, 1983). The effect of type and concentration of plasticizer (Entwistle and Rowe, 1979; Aulton and Abdul-Razzak, 1981), solid inclusions (Aulton and Abdul-Razzak, 1984; Okhamafe and York, 1984, 1985), method of film preparation (Vemba et al., 1980), or storage conditions (Aulton and Abdul-

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Razzak, 1981; Ononokpono and Spring, 1988) on the mechanical properties have been widely studied by stress-strain techniques.

The tensile tests are generally performed on dry films in order to characterize the mechanical properties of the coatings during coating and further processing (e.g., compression of coated beads) and storage. However, the mechanical properties of the dry films are not good predictors of how the coated dosage form will perform when in contact with aqueous fluids. With regard to the practical performance of the coated dosage form, it is of interest to know how water may affect the mechanical properties. During dissolution studies or under in vivo conditions, aqueous fluids will diffuse across the polymeric coating into the solid dosage form; the films will be in a wet or hydrated state. The mechanical properties of wet coatings may be completely different to those of dry films because of hydration of the polymer and/or possible leaching of the plasticizer. With water-soluble drugs and excipients, significant osmotic pressures could develop within the coated dosage form (Iyer et al., 1990; Ozturk et al., 1990), thus potentially causing ruptures in the wet films and subsequent loss of protective or sustained release properties.

The objective of this study was therefore to evaluate the mechanical properties of not only dry but also wet films which had been exposed to dissolution media. A puncture test was used to measure the puncture strength and the % elongation at puncture (Bourne, 1979; Dao, 1983; Radebaugh et al., 1988). Polymeric films of Eudragit RS30D, a commercially available acrylic pseudolatex widely used in the coating of solid dosage forms, were investigated.

Materials and Methods

Materials

The following chemicals were obtained from commercial suppliers and used as received: triethyl citrate (TEC; Citroflex-2), acetyl tributyl citrate (ATBC; Citroflex A-4) (Morflex Chemical Co., Greensboro, NC), Eudragit RS30D [poly (ethylacrylate methylmethacrylate-trimethylammonioethylmethacrylate chloride) copolymers with a ratio of 1:2:0.1; Röhm Pharma, Darmstadt, Germany], methyl alcohol (HPLC grade, Mallinckrodt Specialty Chemicals Co., Paris, KY).

Methods

Preparation of the polymeric films

The polymeric films were prepared by casting the plasticized colloidal polymer dispersion, Eudragit RS30D, (plasticization time prior to casting, 5 h) on a teflon protective overlay (Cole-Parmer Instrument Co., Chicago, IL) mounted on a glass plate (area of casting, 9.5×13.5 cm^2; standard formulation: casting volume, 40 ml; plasticizer content based on polymer, 20% w/w; total solids content, 5 g; approximate film thickness, 300 μm). The low viscosity of the polymer dispersions obviated the need of a casting knife. The film thickness was varied by adjusting the amount of polymer solids in the pseudolatex while keeping the casting volume constant. The films were oven-dried for 48 h at 40°C and 30% relative humidity. The dried films were peeled from the teflon surface, cut into 4×4 cm^2 test sections, and stored at 22°C and 54% relative humidity for 48 h prior to puncture tests or exposure to the aqueous medium. The film thickness of dry films was determined in five places using a micrometer (Paul N. Gardner Co., Inc., Pompano Beach, FL).

Preparation of the wet films

The test films were confined individually in bags (7×7 cm^2, made from a 40-mesh plastic screen with three sides sewn-closed) in order to prevent folding of the polymeric films during exposure to the aqueous medium. The bags were then placed horizontally at the bottom of medium-filled vessels (USP XXII rotating paddle method; 500 ml 0.1 M NaCl, 37°C, 25 rpm, $n = 3$). An exposure time of 24 h was selected based on gastrointestinal transit time and film thickness.

Determination of the mechanical properties of wet and dry films

The puncture test was performed on an Instron (Model 4201, Instron Corp., Canton, MA, 1

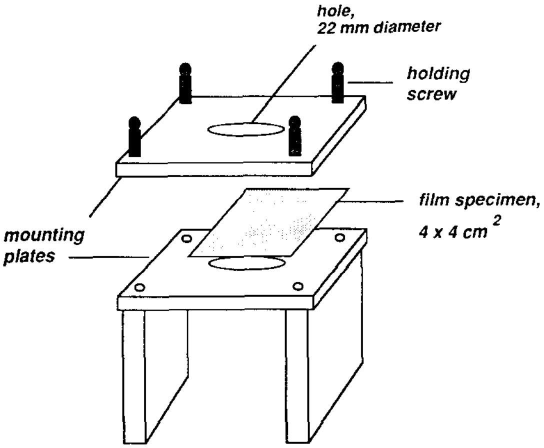

Fig. 1. Schematic diagram of the film holder used in the puncture test device.

kN load detecting transducer). The custom-designed film holder is schematically illustrated in Fig. 1 and was similar to those previously described (Steger, 1979; Bourne, 1982; Radebaugh et al., 1988). Dry or wet film specimens were positioned in the film holder between the two mounting plates followed by tightening of the holding screws to prevent slippage of the films. The wet films were carefully blotted to remove water from the film surface prior to the mounting. The puncture probe (length, 50 mm; diameter, 5 mm; hemispherical probe end), which was attached to the driving load cell, was then driven downward through the center of the mounted film (diameter of the opening of the film holder, 22 mm) at a crosshead speed of 10 mm/min to record load vs displacement data at room temperature. The load (kg) and displacement (mm) at film rupture were converted to puncture strength (MPa) and % elongation at rupture (puncture strength $= F/A_{cs}$, where F is the load required for puncture and A_{cs} denotes the cross-sectional area of the edge of the dry film located in the path of the cylindrical opening of the film holder; % elongation $= [\{(R^2 + D^2)^{1/2} - R\}/R] \times 100$, where R is the radius of the film exposed in the cylindrical hole of the film holder and D represents the displacement of the probe from point of contact to point of puncture). The conversion of peak load to the puncture strength provided the normalization of the data for differences in film thickness (Radebaugh et al., 1988). It was impossible to obtain accurate values for the thickness of the wet films because of irregular swelling characteristics and film surfaces and weak films. The thickness of the dry films was used in the calculations. The punctured wet films were then oven-dried at 40°C for 24 h in order to determine the water and residual plasticizer content.

The following variables were investigated: crosshead speed, 2.5, 5, 10, 15, and 20 mm/min; thickness of Eudragit RS30D films, 100–800 μm with 200–300 μm increments; type of plasticizer, triethyl citrate and acetyl tributyl citrate; plasticizer concentration, 10–40% w/w based on polymer solids, 5 or 10% increments; storage humidities, 0, 20, 54, 75, and 97% relative humidity.

Determination of plasticizers by HPLC

A previously developed HPLC assay was used for the analysis of the plasticizers present in the films before and after exposure to the aqueous medium (Bodmeier and Paeratakul, 1991). The chromatographic system consisted of a solvent delivery module (LC-9A), a UV spectrophotometric detector (SPD-6A), an automatic sample injector (SIL-9A), an integrator (Chromatopac CR601) (Shimadzu, Kyoto, Japan), and an analytical column (Beckman-Ultrasphere, C-18, 5 μm particle size, 25 cm $\times$ 4.6 cm i.d.). The mobile phases consisted of methanol:double distilled water mixtures (70:30 vol.% for TEC (I); 90:10 vol.% for ATBC (II)).

The polymeric films (500–700 mg) were accurately weighed and dissolved in methanol (14 ml for (I); 18 ml for (II)), followed by the addition of water (6 ml for (I); 2 ml for (II)) to precipitate the polymer, and ultracentrifugation (45000 rpm, 30 min; Beckman Ultracentrifuge L5-50). The supernatant was diluted with the mobile phase prior to injection. All film samples were stored in an oven at 40°C for 16 h prior to extraction for plasticizer content in order to evaporate residual moisture in the films. The loss of plasticizers at this drying temperature/time was negligible ($<$ 0.5%).

The residual plasticizer content in the wet films was determined after drying of the films.

The amount of plasticizer leached into the aqueous medium and the residual plasticizer content in the films matched the original plasticizer content within 2–5%.

Scanning electron microscopy (SEM)

The dried films were coated for 70 s under an argon atmosphere with gold-palladium (Pelco Model 3 Sputter Coater) and then observed with a scanning electron microscope (Jeol JSM 35C) to examine the surface morphology before and after exposure to the aqueous medium.

Storage of films at different relative humidities

The prepared films were cut and stored in desiccators containing a desiccant or saturated solutions of different salts for maintaining relative humidities at 0% (silica gel), 20% ($MgBr_2$), 54% ($Mg(NO_3)_2$), 75% (NaCl), and 97% (K_2SO_4), respectively, at 22°C (Nyqvist, 1983). The relative humidity in each chamber was determined with a hygrometer (Airguide humidity indicator, Airguide Instrument Co., Chicago, IL). The mechanical properties and moisture uptake or loss (determined by weighing the films before and after storage) of the dry film samples were determined after storage for 7 days.

Results and Discussion

This paper discusses the effect of the exposure of polymeric films to an aqueous medium on the stress-strain behavior. The polymeric films were prepared by casting and drying an aqueous colloidal polymer dispersion, Eudragit RS30D, a pseudolatex widely used in the coating of solid dosage forms. The mechanical properties, including the load and displacement at rupture, of dry and wet films were measured by a puncture test whereby a puncturing probe was attached to the driving load cell of an Instron and driven through the polymeric film. The device measured the load as a function of probe displacement; the load and elongation at puncture were then converted to puncture strength and % elongation, which were the parameters used to characterize the mechanical properties of the films.

One testing variable affecting the results of the puncture test was the rate at which the puncturing probe was driven through the film (Fig. 2). The puncture strength increased and the elongation at puncture decreased with increasing crosshead speed (varied from 2.5 to 20 mm/min). As a result of the increasing puncture strength and decreasing elongation, the modulus at punc-

TABLE 1

Effect of film thickness on the mechanical properties of dry and wet Eudragit RS30D-20% TEC films (S.D. in parentheses; n = 3)

Dry film thickness (μm)	Peak load (kg)	Displacement (mm)	Puncture strength (MPa)	Elongation (%)	Actual TEC content (% w/w)
Dry films					
127 (4)	0.45 (0.01)	23.2 (0.8)	1.58 (0.05)	133.6 (6.9)	19.97 (0.81)
294 (15)	1.04 (0.02)	24.6 (0.7)	1.59 (0.01)	145.3 (6.2)	20.17 (0.23)
553 (20)	1.96 (0.04)	25.8 (0.4)	1.58 (0.03)	155.2 (3.5)	20.01 (0.51)
826 (39)	2.98 (0.12)	26.3 (1.0)	1.61 (0.06)	159.0 (8.8)	20.10 (0.47)
Wet films					
127 (4) [a]	0.07 (0.02)	3.6 (0.7)	0.10 (0.06)	5.3 (1.6)	6.62 (0.05) [b]
294 (15) [a]	0.29 (0.02)	8.3 (0.7)	0.44 (0.03)	25.1 (4.0)	10.83 (0.09) [b]
553 (20) [a]	0.67 (0.03)	11.7 (0.8)	0.54 (0.03)	46.2 (5.6)	14.32 (0.27) [b]
826 (39) [a]	1.16 (0.06)	14.5 (1.1)	0.63 (0.03)	65.7 (7.9)	17.53 (0.31) [b]

[a] Thickness of dry films.

[b] Determined after drying of wet films.

ture (slope at puncture, data not shown), calculated from the ratio of load to displacement at puncture, increased with increasing crosshead speed. The films had less time to relax to the applied stress with increasing crosshead speed. These results were consistent with data obtained from tensile tests (Bartenev and Zuyev, 1968; Nielsen, 1974; Shah, 1984); the tensile strength and modulus increased, while the elongation decreased with increasing strain rate. In this study, a speed of 10 mm/min was selected for all further experiments.

The effect of film thickness on the mechanical properties of dry and wet Eudragit RS30D films (plasticized with 20% triethyl citrate) is shown in Table 1. With dry films, the values of peak load increased with increasing film thickness whereas the displacement stayed fairly constant. Similar relationship between film thickness and the load at break have been reported with films of cellulose derivatives (Fell et al., 1979). After normalizing the load at puncture with respect to film thickness, the resulting puncture strength of dry films was almost independent of film thickness. This could indicate the presence of similar microstructures in films of varying thicknesses. The puncture strength and % elongation of wet films were significantly lower when compared to those of dry films; the puncture strength and elongation of wet films increased with increasing film thickness. This could be explained by the more pronounced leaching of triethyl citrate, a water-soluble plasticizer, from thinner films during exposure to the aqueous medium. The actual concentration of triethyl citrate after drying of the wet films increased with increasing film thickness.

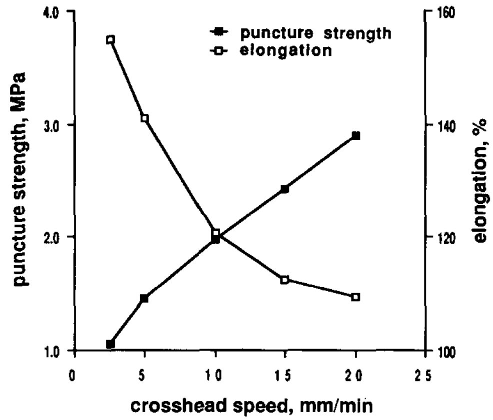

Fig. 2. Effect of crosshead speed on the puncture strength and % elongation at puncture of dry Eudragit RS30D-triethyl citrate films (TEC concentration, 20% w/w of polymer; film thickness, 323 μm).

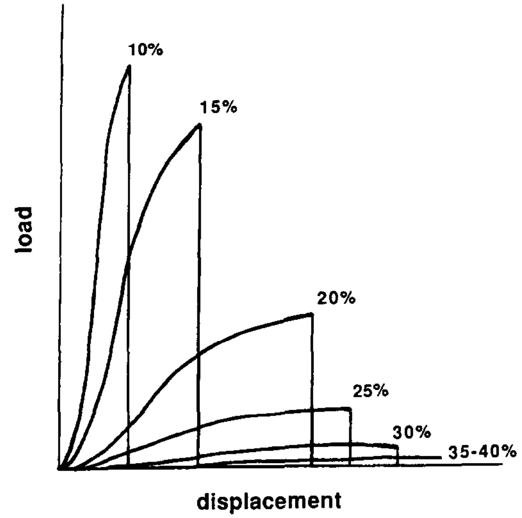

Fig. 3. Load vs displacement profiles of dry Eudragit RS30D-triethyl citrate films (plasticizer concentration, 10–40% w/w of polymer).

Triethyl citrate (TEC, water-soluble) and acetyl tributyl citrate (ATBC, water-insoluble) were evaluated as plasticizers in order to study the effect of leaching or permanence of the plasticizer during exposure to aqueous media on the mechanical properties of wet films. The concentration of the plasticizer was varied between 10 and 40% w/w based on the polymer. Films could be cast from polymeric dispersion containing less than 10% plasticizer, however, the resultant films were very brittle. This plasticization level was insufficient for film formation because of incomplete coalescence or fusion of the colloidal polymer particles. Typical plots of load vs displacement for dry Eudragit RS30D films plasticized with different amounts of TEC (10–40% w/w based on polymer solids) are shown in Fig. 3. The load increased with increasing displacement; the resulting curves had a sigmoidal shape. Initially, the slope increased up to an inflection point followed by a decrease in slope thereafter. The proportionally smaller increase in load at higher displacements could probably be explained with the relaxation of the polymeric film to the applied stress during testing. Films plasticized with low levels of plasticizer (10–15% w/w) were either broken into many small pieces or were star-cracked upon puncture. Polymeric films plasticized with 20–30% w/w plasticizer had a circular hole in the middle of the film specimen after the test. At high TEC concentration (35–40% w/w), the films were so flexible that they did not rupture at the maximum possible elongation.

The puncture strength and elongation of dry and wet Eudragit RS30D films plasticized with TEC or ATBC are shown in Fig. 4. The puncture strength of dry films plasticized with TEC decreased and the elongation increased with increasing TEC concentration (Fig. 4A). The addition of increasing amounts of plasticizer resulted in softer and more flexible films. Plasticization generally causes a decrease in the intermolecular forces along the polymer chains resulting in a decrease in the glass transition temperature, tensile strength, and reduction in the brittleness of polymeric materials (Sears and Darby, 1982). At a TEC level of 10% w/w, the polymeric films were hard and brittle as shown by a high puncture strength and a low elongation. Eudragit RS30D films plasticized with TEC contents higher than 25% w/w became tacky due to excess amount of the water-soluble plasticizer. Polymeric films plasticized with 35 and 40% TEC were extremely tacky and did not break at the end of test, the point at which displacement corresponded to an elongation of approx. 365%.

Although dry films with TEC concentrations above 20% w/w were quite flexible, the respective wet polymeric films were soft and weak as shown by low values of puncture strength and % elongation (Fig. 4B). The weakening of the films

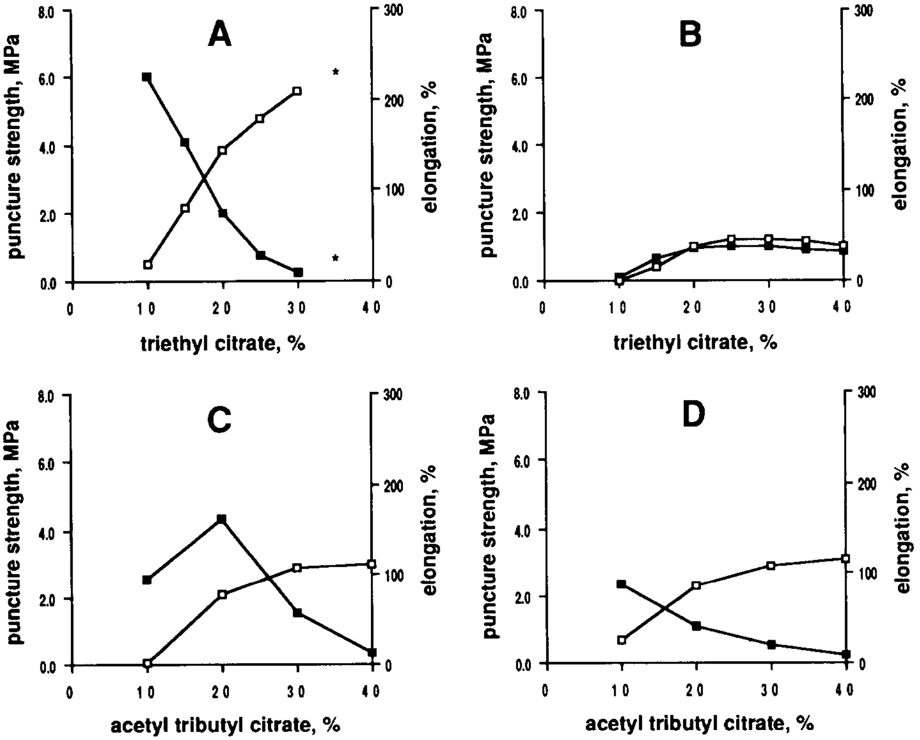

Fig. 4. Effect of initial plasticizer concentration on puncture strength and % elongation at puncture of (A) dry and (B) wet Eudragit RS30D-triethyl citrate films; and (C) dry and (D) wet Eudragit RS30D-acetyl tributyl citrate films (film thickness, 254–314 μm): (□) % elongation; (■) puncture strength. (∗) Films plasticized with 35 and 40% TEC did not rupture.

TABLE 2

Triethyl citrate (TEC) concentration in Eudragit RS30D films before and after exposure to 0.1 M NaCl solution

Theoretical TEC concentration (%)	Actual TEC concentration (%)		TEC remaining (% of original TEC)
	Before	After	
10	9.90	6.78	68.48
15	14.97	9.33	62.12
20	19.95	11.23	56.29
25	24.08	12.16	50.50
30	29.10	12.13	41.68
35	34.09	11.84	34.70
40	39.05	10.83	27.73

upon exposure to aqueous media could be explained with the hydration or water uptake of the polymeric films and the leaching of the water-soluble plasticizer, TEC. Table 2 shows the amount of TEC in the films before and after exposure to the aqueous medium. Approx. 10% w/w plasticizer remained in films having original plasticizer concentrations above 10% w/w. The percent of TEC remaining in the film based on the original TEC concentration decreased sharply with increasing TEC concentration. Incorporation of a water-soluble, leachable plasticizer such as TEC caused a significant increase in the porosity of the polymeric films due to the leaching of the water-soluble component. Scanning electron micrographs of Eudragit RS30D films containing 30 and 40% TEC after exposure to the aqueous medium (original film surfaces were smooth) revealed a very porous structure as a result of TEC leaching while Eudragit RS30D films containing 20% TEC had a smooth surface (Fig. 5A–C). The pore size (generally < 10 μm) increased with increasing TEC concentration. In a previous study with drug-containing pseudolatex-cast films, the release of propranolol HCl from Eudragit RS30D plasticized with TEC concentrations in excess of 30% w/w was found to be very rapid and this rapid drug release was attributed to leaching of

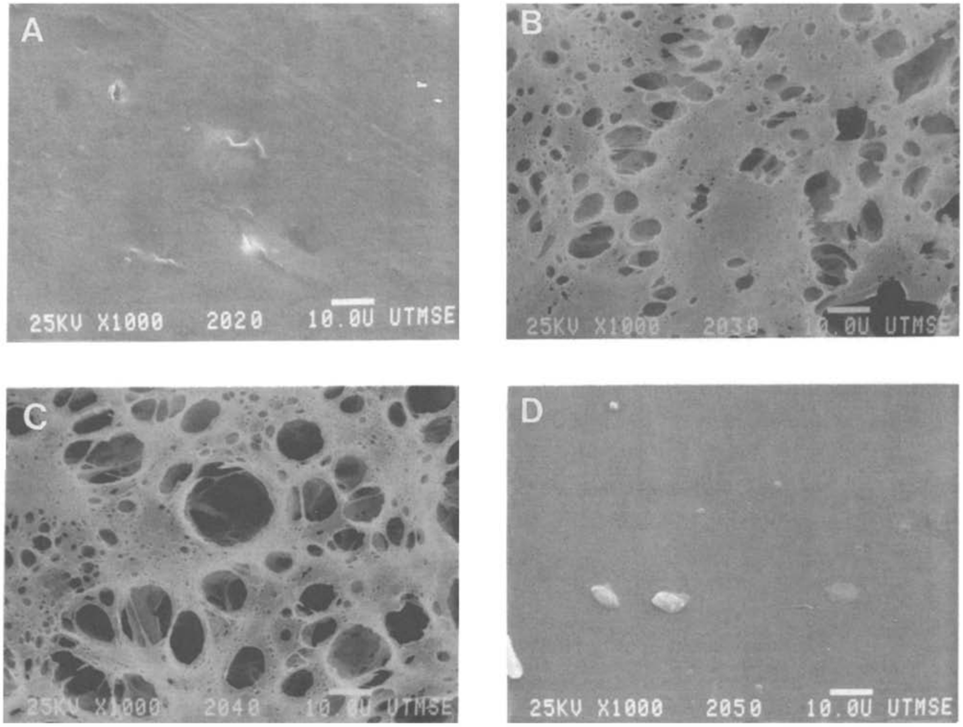

Fig. 5. Scanning electron micrographs (×1000) of the surfaces of Eudragit RS30D films plasticized with (A) 20%, (B) 30%, (C) 40% triethyl citrate and (D) 40% w/w acetyl tributyl citrate after exposure to 0.1 M NaCl solution.

the water-soluble plasticizer from the polymeric films (Bodmeier and Paeratakul, 1990).

Polymeric films of Eudragit RS30D plasticized with ATBC, a water-insoluble plasticizer, possessed some similarities in their mechanical properties in both dry and wet states (Fig. 4C and D). The elongation values were comparable for both dry and wet films. The wet Eudragit RS30D-ATBC films were also significantly more flexible than the corresponding wet Eudragit RS30D-TEC films. Polymeric Eudragit RS30D coatings plasticized with ATBC should therefore be able to yield to increasing osmotic pressures developing within the coated dosage form upon contact with dissolution or biological fluids without rupturing, while coatings plasticized with TEC, which have very low elongation values, may rupture and lose their protective function. The percent of ATBC remaining in the films was 100.0 ± 3.7% in all cases. These results confirmed the complete retention of the water-insoluble plasticizer in the films and thus its contribution to the mechanical stability of the polymeric films in the wet state. The lower puncture strength of the wet films, an observation also made with Eudragit RS30D-TEC films, could be attributed to the hydration of the films. The water uptake of all films varied between 35 and 50% (determined gravimetrically and based on the polymer weight). Except for the ATBC concentation of 10% w/w, the puncture strength of dry films decreased with increasing ATBC concentration. Ten percent ATBC was not sufficient to cause complete film formation of the colloidal polymer dispersion, the extremely brittle films were characterized by a low puncture strength and almost no elongation at rupture.

After exposure to the aqueous medium, the films were air-dried. Interestingly, Eudragit RS30D-TEC films were opaque, wrinkled or rolled-up, and no longer flexible, while Eudragit RS30D-ATBC, although being opaque in the wet state, became clear and flexible after drying. Be-

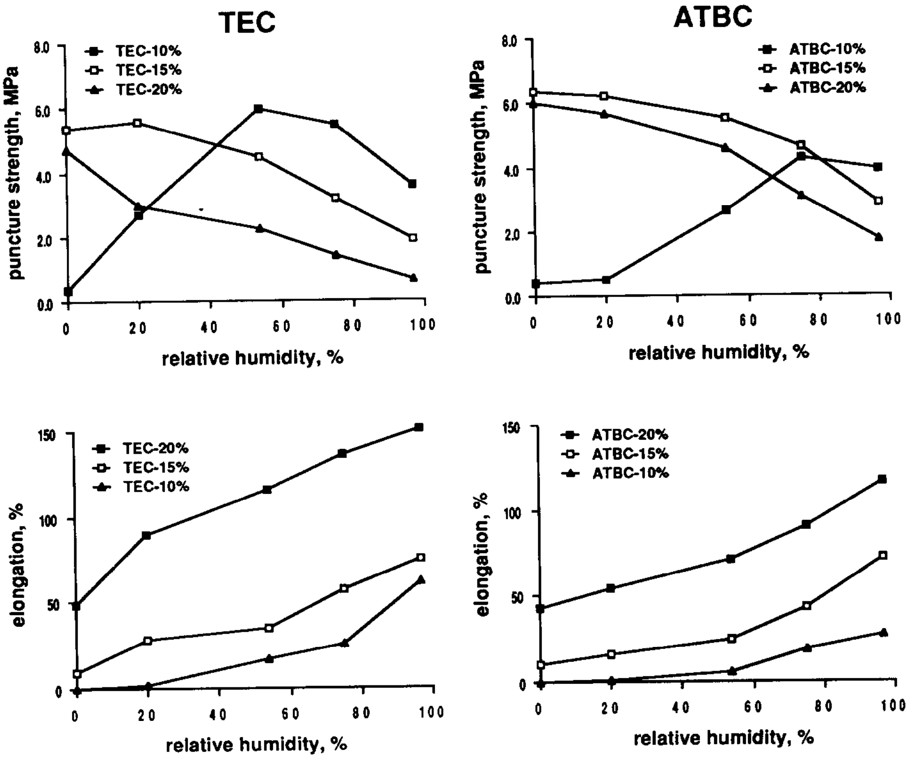

Fig. 6. Effect of relative humidity on the puncture strength and % elongation at puncture of Eudragit RS30D-triethyl citrate or -acetyl tributyl citrate films containing 10, 15, or 20% w/w plasticizer (film thickness, 310 μm).

TABLE 3

Moisture uptake or loss (%) during storage of Eudragit RS30D films at different relative humidities (7 days, 22°C)

Relative humidity (%)	Triethyl citrate (% w/w)			Acetyl tributyl citrate (% w/w)		
	10	15	20	10	15	20
0	−1.82	−1.63	−0.69	−0.57	−0.46	−0.43
20	−0.83	−0.82	0.79	−0.48	−0.42	0.35
54	0.44	0.58	0.96	0.32	0.35	0.91
75	1.97	1.98	2.81	1.60	1.54	2.22
97	16.74	12.98	12.63	15.95	12.60	11.45

cause of the leaching of TEC, the Eudragit RS30D-TEC films did not shrink to a large extent during drying; pores remained where water was present in the hydrated films thus explaining the formation of porous, opaque and brittle films. On the contrary, ATBC remained within the Eudragit RS30D-ATBC films; the films were flexible enough to shrink during drying resulting in non-porous, clear, and flexible films. The surface of Eudragit RS30D-40% ATBC after exposure to the aqueous medium was smooth and non-porous (Fig. 5D) when compared to films containing the same amount of TEC (Fig. 5C).

The mechanical properties were influenced not only by the type and concentration of the plasticizer but also by the storage conditions. The effect of storage humidity on the mechanical properties of dry Eudragit RS30D films plasticized with TEC and ATBC at different plasticizer levels (10, 15, and 20% w/w) is shown in Fig. 6. The % elongation increased with increasing storage humidity at all three plasticizer levels. At a low plasticizer concentration (10% w/w), the films stored at 0 and 20% relative humidity were very brittle and weak as shown by low values of both puncture strength and elongation. At a plasticizer level of 15 and 20%, the puncture strength and modulus at puncture (data not shown) decreased with increasing storage humidity with both plasticizers, reflecting the plasticizing effect of water. The moisture uptake of the films as a function of plasticizer concentration and storage humidity is shown in Table 3. Except for the storage at 97% relative humidity, the moisture uptake or loss, as expressed in percent weight gain, was less than 2%. Small changes in water content could therefore significantly affect the mechanical properties of these films.

Future studies will address the time-dependent changes in the mechanical properties of wet films, especially with regard to time-dependent concentrations of plasticizers and water in the film. In conclusion, it was shown that the mechanical properties of Eudragit RS30D films in the dry and wet states could be measured in a reproducible manner using a puncture test. The puncture strength and elongation of dry and wet films were significantly different and were influenced by the type and concentration of plasticizer. The mechanical properties of wet polymeric films could strongly affect the performance of a coated solid dosage form in dissolution or biological fluids; they should be taken into considerations when formulating sustained release coated dosage forms.

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