

Research papers

Influence of processing variables on the properties of free films prepared from aqueous polymeric dispersions by a spray technique

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Abstract

A novel method for the preparation of free films from aqueous polymeric dispersions by a spray technique is reported. The apparatus included a spray gun, rotary drum and a temperature controlling system. The influence of spray rate and processing temperature on the mechanical properties of free films prepared from aqueous dispersions of Eudragit® L 30D-55, and L 100-55 (methacrylic acid-ethyl acrylate copolymer), Shin-Etsu AQOAT® (hydroxypropyl methylcellulose acetate succinate), Aquateric® (cellulose acetate phthalate) and Aquacoat® (ethyl cellulose), plasticized with triethyl citrate, was investigated. The processing temperature was monitored using a telemetric system. Reproducible free films were obtained from the five polymers using this apparatus. The tensile strength and elongation of films of the two Eudragit® latex dispersions, having a minimum film formation temperature (MFT) less than 20°C, were not influenced by spray rate or processing temperature between 30 and 40°C. The mechanical values of free films from Shin-Etsu AQOAT® were significantly decreased at a slower spray rate, but processing temperature did not affect film properties. This polymeric dispersion contained larger particles than the acrylic dispersions and the free films had a low MFT. The Aquateric® dispersion, having a high MFT, contained larger particles than the acrylic latexes and produced films at high spray rates and slow drying conditions. The drying temperature significantly influenced the elongation properties of the films. The mean tensile strength of free films from Aquacoat®, a high-MFT latex, was slightly higher at higher processing temperature, but this was not significant. The spray rate did not alter the mechanical properties of films prepared from this pseudolatex.

Keywords: Eudragit; Hydroxypropyl methylcellulose acetate succinate; Cellulose acetate phthalate; Ethyl cellulose; Aqueous polymeric dispersions; Minimum film formation temperature; Free films

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1. Introduction

The successful introduction of a wide variety of aqueous-based polymeric solutions and dispersions for film coating of pharmaceutical dosage forms, has stimulated a great deal of research activity by pharmaceutical scientists. For environmental and economic reasons, the aqueous coating of oral solid dosage forms has rapidly replaced solvent-based film coating operations. The aqueous-based polymers that are currently available on the market include the acrylic resins, cellulosic derivatives, and a vinyl polymer. For gastric-soluble film coating, water-soluble polymers such as hydroxypropyl methylcellulose are used as the coating material. Since polymers for enteric coating or sustained-release formulations are practically insoluble in water, they are usually applied as aqueous dispersions. The mechanism of film formation from an aqueous polymeric dispersion is more complex compared to that from an aqueous or organic solution, since the polymeric particles dispersed in the water must coalesce to form a continuous film. Several researchers have reported on the mechanisms of film formation and have proposed models for film formation from aqueous polymeric dispersions (Eckersley and Rudin, 1990; Roulstone et al., 1991; Chevalier et al., 1992; Winnik and Wang, 1992). An important factor for film formation is the driving force that causes the coalescence of polymeric particles which results from water evaporation or capillary force. Since coalescence occurs only above a certain temperature, i.e., minimum film formation temperature, temperature and water evaporation are considered to be major factors which affect the film properties of coating materials.

Commercial aqueous polymeric latex dispersions are prepared by emulsion polymerization while pseudolatexes are prepared by emulsification of polymeric solutions. Pseudolatexes may also be made from suspensions of spray-dried or mechanically milled solid polymeric particles. Properties of free films prepared from aqueous polymeric dispersions have received little attention in the literature and this is probably due to the difficulties that researchers have experienced in preparing free films with reproducible properties.

Cast methods have been widely used for the preparation of polymeric films from organic or aqueous solution. For aqueous latex and pseudolatex dispersions, however, polymeric particles will tend to settle during the drying process in the cast method, which will lead to uneven film formation. Free films prepared by a spray method provide a better simulation of the coating process. The comparison of mechanical properties of free films between cast and spray techniques was recently reported and the spray technique was found to provide more reproducible results for each polymer (Obara and McGinity, 1994).

During the aqueous film coating of pharmaceutical dosage forms, several parameters including process temperature and spray rate must be controlled. These parameters will influence the product performance and need to be optimized for each polymeric system. This is also true for free film preparation using a spray technique. It was therefore of interest to investigate the processing parameters that influence the film formation of several commercial polymeric dispersions. The objectives of the present study were initially to develop a novel method for preparing uniform free films of aqueous polymeric dispersions using a spray technique, and then to investigate the influence of processing variables on the physical-mechanical properties of the free films. Five commercially available acrylic and cellulosic polymers for enteric and sustained-release coating were employed in this study.

2. Materials and methods

2.1. Materials

The following materials were used as film-forming agents: methacrylic acid-ethyl acrylate copolymer aqueous dispersion (Eudragit® L 30D-55, Röhm GmbH, Darmstadt, Germany), methacrylic acid-ethyl acrylate copolymer redispersible powder (Eudragit® L 100-55, Röhm GmbH, Darmstadt, Germany), ethyl cellulose aqueous dispersion (Aquacoat®, type ECD-30, FMC Corp., Newark, DE), cellulose acetate phthalate powder (Aquateric®, type CD-901, FMC Corp.,

Newark, DE), and hydroxypropyl methylcellulose acetate succinate powder (Shin-Etsu AQOAT®, type AS-MF, Shin-Etsu Chemical/Biddle Sawyer Corp. New York, NY). Triethyl citrate was obtained from Morflex, Inc., Greensboro, NC. Diethyl phthalate and dibutyl sebacate were obtained from Eastman Kodak Co., Rochester, NY.

2.2. Preparation of aqueous polymeric dispersions

The aqueous dispersions of Eudragit® L 30D-55, Shin-Etsu AQOAT®, Aquateric®, and Aquacoat® were prepared as described previously (Obara and McGinity, 1994). The dispersion of Eudragit® L 100-55 was prepared by re-dispersion of the polymeric powder in water followed by the addition of a 1 N NaOH solution. The amount of NaOH was 1.3% of the polymer (Lehmann, 1986).

2.3. Minimum film formation temperature (MFT)

The measurement of the MFT was based on ASTM D2354-91 with minor modifications. The apparatus consisted of an MFT bar (main board: 17' × 5' × 3/8', legs: 5' × 5' × 3/8'), fabricated from stainless steel, an oil bath, air supply system, and a Plexiglas cover. Borders were made on the main board by attaching plastic bars so that the dispersions could be poured. A temperature gradient was generated by putting one side of the bar into an oil bath. The other side was exposed to room temperature (23°C). The temperature of the oil bath was set so that the bar temperature of the oil side was approximately 45°C. The flow rate of drying air was 20 l/min. After the temperature gradient reached equilibrium, the dispersions were poured into the molds on the bar and covered. The amount of dispersion to be poured was varied such that the final film thickness was approximately 150 μm. The dispersions were dried in approximately 5 h under this condition. The MFT was determined as the minimum temperature where the film was clear and free of cracks.

2.4. Preparation of free films

The apparatus for film preparation is shown in Fig. 1. It was modified from the original design

that was reported previously (Obara and McGinity, 1994). The apparatus consisted of a spray gun with a vertical driving system, a rotary drum (3' high, 3' diameter), a chamber, and a temperature controlling system. Heated air was supplied into the chamber, and the flow rate of the inlet air was measured by an anemometer (Model 491 FM, Kurz Instruments). A radiotelemetric system was applied to measure the temperature of the drum surface. This system included a small radio transmitter installed inside the drum and a receiver connected to a meter (Macay, 1970). A thermistor connected to the transmitter was attached to the center of the drum surface. The thermistor sensed the temperature, and a signal that was modulated by an audio tone was transmitted. The audio tone was proportional to the temperature at the surface of the drum. The signal was received through an aluminum antenna, which was placed beside the chamber, and the temperature was read by the meter.

The free films were prepared by spraying the dispersions onto a Teflon® sheet attached to the rotating drum. The inlet temperature was controlled to maintain a desired drum surface temperature. The inlet air flow was approximately 500 l/min. The distance between the spray nozzle and the drum surface was 20 cm. The spray rate was controlled by a programmable timing controller. After the spray cycle was completed, the film was dried under the same air flow condition for 10 min, and the Teflon® sheet was removed carefully from the drum. The sheet was then put into a 100% relative humidity chamber to soften the film. The film was peeled off the Teflon® sheet and cut into rectangular specimens of 1.3 × 8 cm. The film specimens were then cured in an oven at 60°C for 24 h. They were then stored in a 50% relative humidity chamber at 23°C until the mechanical tests were conducted.

2.5. Mechanical tests

Tensile strength and percent elongation of film specimens were measured using an Instron Model 4201, based on the ASTM D882-75d method. The details were described in the previous report (Obara and McGinity, 1994).

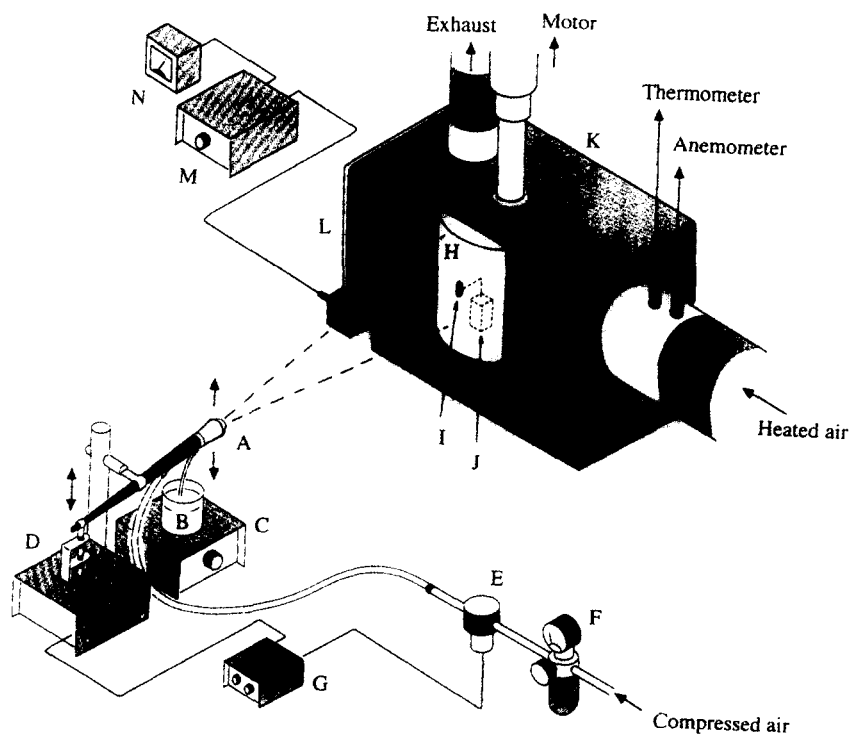


Fig. 1. Spray apparatus for the preparation of free films. A, spray gun; B, sample beaker; C, magnetic stirrer; D, vertical driver; E, solenoid valve; F, regulator; G, programmable timing controller, H, rotary drum; I, thermistor; J, transmitter; K, chamber; L, antenna; M, receiver; N, temperature meter.

3. Results and discussion

3.1. Minimum film formation temperature of aqueous dispersions

Prior to the preparation of free films by the spray method, it was necessary to determine the minimum film formation temperature (MFT) of each dispersion to select the appropriate temperature for the spray operation. While the MFT has been studied on Eudragit® polymers (Lehmann, 1989) and Aquacoat® (Lippold et al., 1990), the MFT values for Shin-Etsu AQOAT® and Aquateric® have not been reported.

Table 1 shows the results of the MFT measurements. Since the Eudragit® L 30D-55, L 100-55,

and Shin-Etsu AQOAT® dispersions, plasticized with TEC (20, 20 and 28%, respectively) formed clear films at room temperature, the MFT was less than 23°C. The MFT of the Aquateric® dispersion, plasticized with DEP (25, 35 and 45%), was found to be 40, 38 and 36°C, respectively. Below these temperatures, no coalescence or film formation was observed and only a layer of powder remained. The water-soluble plasticizer, TEC, resulted in an MFT that was 2–4°C lower than the DEP. The Aquacoat® dispersions, plasticized with DBS (15, 25 and 30%), gave intact clear films above 34, 28, and 27°C, respectively. These data are in a good agreement with Lippold et al (1990). As reported by these researchers, films that formed at temperatures be-

low the MFT exhibited many cracks although the layer itself was clear, which indicated, that some coalescence still occurred at a temperature less than the MFT. TEC exhibited a MFT that was 2°C lower than for Aquacoat® with DBS.

The water-insoluble plasticizers, DEP and DBS, have been the recommended plasticizers for Aquateric® and Aquacoat®, respectively. The results in Table 1 suggest that the water soluble plasticizer, TEC, would be an ideal plasticizer for these polymers, based on the MFT data. Another advantage of the water soluble plasticizers is that the distribution of plasticizer into a polymeric dispersion rapidly reaches equilibrium. The water-insoluble plasticizers are often emulsified into the dispersion and the equilibration time can sometimes be quite lengthy. However, the utilization of water-soluble plasticizers in enteric or sustained-release film coatings would also need to be investigated to ensure that the product performs in the desired manner. Based on our current results, TEC was used as a plasticizer for all polymers used in this study due to the ease of film preparation and consistency in film properties.

Table 1
Minimum film formation temperature of aqueous polymeric dispersions

Polymer	Plasticizer ^a	%	MFT ^b (°C)
Eudragit® L 30D-55	TEC	20	<23
Eudragit® L 100-55	TEC	20	<23
Shin-Etsu AQOAT®	TEC	28	<23
Aquateric®	TEC	25	36 ± 2
		35	35 ± 2
		45	34 ± 2
	DEP	25	40 ± 2
		35	38 ± 1
		45	36 ± 2
Aquacoat®	TEC	15	32 ± 2
		25	30 ± 2
		30	<23
	DBS	15	34 ± 2
		25	28 ± 2
		30	27 ± 1

^aTEC, triethyl citrate; DEP, diethyl phthalate; DBS, dibutyl sebacate.

^bMean ± S.D. of three experiments.

3.2. Film preparation by the spray apparatus

Since the temperature of film formation is an important factor for aqueous polymeric dispersions, the methods to control the process temperature must be carefully controlled and maintained in order to obtain reproducible free films. When the drum in Fig. 1 was rotated during the spraying process, it was difficult to directly measure the drum surface temperature with a conventional thermometer. A thermometer when fixed close to the drum surface did not indicate an accurate surface temperature. A small wireless temperature measurement device was found to solve this problem. Due to its small size, it could be readily installed inside the drum. The rotating motion of the measuring point enabled a more accurate determination of the temperature of the drum surface in spite of the temperature gradient which might occur from the heated air that was supplied from one side of the chamber. The wireless telemetric system used in this study was similar to that which has been widely used in animal studies (Macay, 1970). This device was found to be stable in spite of its simplicity. The surface temperature during the spray process was pulsating, since the intermittent spray cycle was used to prepare the films. The range of this temperature fluctuation was within ±1.5°C. By monitoring the surface temperature of the drum, reproducible free films could be prepared for each polymer studied in this investigation.

An additional modification to the apparatus that was reported earlier was the drive system which was attached to the spray gun to oscillate it vertically. This modification enabled the preparation of free films with more uniform thickness throughout the sprayed area and enhanced the recovery of products, compared to the previous apparatus (Obara and McGinity, 1994).

3.3. Influence of processing variables on the free film properties

Lippold et al. (1989) reported that optimal film formation was obtained when Aquacoat®-coated microcapsules were heat-treated at 10°C above the MFT. In our experiments, the surface temper-

Table 2
Formulation of aqueous dispersions for free film preparation

Polymer	Solid content (%)	Plasticizer ^a content (% of polymer)
Eudragit® L 30D-55	10	20
Eudragit® L 100-55	10	20
Shin-Etsu ACOAT®	5	28
Aquateric®	10	35
Aquacoat®	10	23

^aTriethyl citrate was used for all polymers.

ature during the spray process was also set to a temperature at least 10°C above the MFT for each of the high-MFT polymeric dispersions. The curing temperature was set at 60°C for all polymers since this temperature was more than 10°C above the MFT for each polymeric dispersion.

The formulations of the aqueous dispersions used for the preparation of free films are shown in Table 2. For polymeric dispersions having low minimum film formation temperatures, such as Eudragit® L 30D-55, L 100-55, and Shin-Etsu ACOAT®, two different temperatures, 30 and 40°C were compared. The influence of spray rate was also investigated, and two spray rates were compared at the same surface temperature. The spray rate was another important parameter since it will impact the moisture content of the films. The moisture content during the spraying process would influence film properties since one of the driving forces for coalescence is provided by water evaporation. The high spray rate in this study kept the film surface wet during the spraying process, whereas the surface was maintained dry with the lower spray rate. Care was taken at the higher spray rate, however, to avoid overwetting of the surface, which would cause droplets to

appear on the surface of the film and an uneven film to form.

The results in Table 3, demonstrate the tensile strength and elongation of the free films of Eudragit® L 30D-55, prepared under different temperature conditions and spray rates. Since films formed from this polymeric dispersion had a MFT less than room temperature, it was anticipated that changing the temperature would not significantly affect the film properties. As shown in Table 3, both tensile strength and elongation were not significantly changed by temperature or spray rate. Table 4 shows the results for the Eudragit® L 100-55 dispersion. The results were similar to those obtained from the Eudragit® L 30D-55 dispersion with respect to the mechanical properties which were not significantly altered by changing variables. It was observed, however, that both the tensile strength and the percent elongation were significantly lower than those of Eudragit® L 30D-55 films, although the monomer structure and its content in these two polymers are the same. Eudragit® L 100-55 is a re-dispersible fine powder with a particle size that is larger than that of the Eudragit® L 30D-55 latex dispersion. It also requires the addition of alkali

Table 3
Effect of processing variables on mechanical properties of free films prepared from Eudragit® L 30D-55 aqueous dispersion (TEC 20%)

Processing condition		Mechanical properties ^a	
Spray rate (g min)	Surface temperature (°C)	Tensile strength (MPa)	Elongation (%)
2.5	40	25.4 ± 5.0	2.3 ± 0.6
1.0	40	23.2 ± 5.3	2.0 ± 0.9
0.8	30	24.5 ± 7.0	2.5 ± 1.2

Table 3

Effect of processing variables on mechanical properties of free films prepared from Eudragit® L 30D-55 aqueous dispersion (TEC 20%)

Processing condition		Mechanical properties ^a	
Spray rate (g/min)	Surface temperature (°C)	Tensile strength (MPa)	Elongation (%)
2.5	40	25.4 ± 5.0	2.3 ± 0.6
1.0	40	23.2 ± 5.3	2.0 ± 0.9
0.8	30	24.5 ± 7.0	2.5 ± 1.2

^aMean ± S.D. of five experiments.

Table 4

Effect of processing variables on mechanical properties of free films prepared from Eudragit® L 100-55 aqueous dispersion (TEC 20%)

Processing condition		Mechanical properties ^a	
Spray rate (g/min)	Surface temperature (°C)	Tensile strength (MPa)	Elongation(%)
2.5	40	15.6 ± 3.0	1.0 ± 0.2
1.0	40	15.8 ± 3.0	0.8 ± 0.2
0.8	30	14.5 ± 2.7	0.9 ± 0.2

^aMean ± S.D. of five experiments.

Table 5

Effect of processing variables on mechanical properties of free films prepared from Shin-Etsu AQOAT® aqueous dispersion (TEC 28%)

Processing condition		Mechanical properties ^a	
Spray rate (g/min)	Surface temperature (°C)	Tensile strength (MPa)	Elongation (%)
2.5	40	16.2 ± 0.7	2.9 ± 0.2
1.0	40	4.8 ± 0.8 ^b	1.3 ± 0.2 ^b
1.0	30	15.9 ± 0.9	2.7 ± 0.5

^aMean ± S.D. of five experiments.

^bSignificantly different from value at the higher spray rate ($P < 0.01$).

to stabilize the dispersion. It is hypothesized that the differences in the physical-mechanical properties of films from these two dispersions were due to the difference in the nature of the dispersion. The results clearly demonstrated, however, that reproducible free films from both Eudragit® L 30D-55 and Eudragit® L 100-55 dispersions prepared by a spray method could be obtained since the surface temperature and the spray rate did not affect the properties of the film.

The parameter settings and results for the Shin-Etsu AQOAT® dispersion are seen in Table 5.

The surface temperature did not affect the film properties due to the lower MFT of the plasticized polymer. The temperature was not a significant factor, between 30 and 40°C provided the sprayed film had sufficient moisture. At 30°C, the surface was kept sufficiently moist at a spray rate of 0.8 g/min. At 40°C, the lower spray rate resulted in brittle films of significantly lower tensile strength and elongation. It was possible that spray-dried particles were trapped in the films prepared from low spray rates, resulting in an incomplete coalescence of the polymer. These re-

sults suggested that a specific moisture content was required for complete film formation from this polymeric dispersion. The differences in moisture requirements between the Shin-Etsu ACOAT® and the Eudragit® polymers could be due to the size of the polymeric particles. The particles in the Shin-Etsu ACOAT® dispersion are solid with a mean particle size of approximately 5 μm (Nagai et al., 1989), while the Eudragit® L 30D-55 dispersion included latex particles with a mean particle size of 0.2 μm . A higher moisture content would be required for the complete coalescence of the harder and larger particles. The mean particle size of redispersed Eudragit® L 100-55 is larger than L 30D-55, but the particles of the L 100-55 are softened by the addition of alkali and the presence of the plasticizer.

For polymers with a high MFT such as Aquateric® and Aquacoat®, the spray temperature was shown to be a significant parameter to control compared to polymers having a low MFT. Table 6 shows the influence of processing conditions on the film properties from the Aquateric® dispersion. Maintaining the surface temperature at 50°C failed to result in film formation even though a higher temperature than its MFT was employed. A powdered aggregate resulted without any coalescence as reported previously (Obara and McGinity, 1994). At this temperature and spray rate, the moisture content during the spraying process was insufficient to achieve complete coalescence. Higher spray rates to increase the mois-

ture content of sprayed layers would therefore be required to achieve coalescence. Since it was difficult to maintain a surface temperature at 50°C with higher spray rates, the spray rate was increased and a lower surface temperature (45°C) was employed. Under this condition, films were not formed during the spraying step, but formed in the drying step. However, drying in the chamber under heated air often resulted in many small cracks in the films probably due to the rapid drying conditions. Intact films were obtained when the drying process was carried out in an oven, by transferring the sprayed sheet from the chamber to the oven immediately after the spraying step. For this polymer, it was observed that the film properties were significantly influenced by drying temperature. At a drying temperature of 45°C, the films had many small cracks. Intact films were obtained when the drying temperature was 50 or 60°C. These results suggested that the temperature of 10°C above the MFT was not sufficient for this dispersion to form films by the spray method and both a high moisture content and a temperature at least 15°C higher than the MFT were required to obtain a continuous film from this dispersion. The percent elongation of the Aquateric® films prepared by drying at 60°C was significantly higher than for films which were dried at 50°C. This suggested that the coalescence at the temperature of 15°C above the MFT was still not the optimized condition. Further studies are in progress to explain this phenomenon.

Table 6

Effect of processing variables on mechanical properties of free films prepared from Aquateric® aqueous dispersion (TEC 35 %)

Processing condition			Mechanical properties ^a	
Spray rate (g/min)	Surface temperature (°C)	Drying temperature ^b (°C)	Tensile strength (MPa)	Elongation (%)
2.5	50	60	Film did not form	
3.8	45	45	Film with cracks	
3.8	45	50	7.3 ± 1.5	3.8 ± 1.0
3.8	45	60	6.7 ± 1.4	9.2 ± 2.0 ^c

^aMean ± S.D. of five experiments.

^bOven temperature. As reproducible films were not obtained by rapid drying using the present apparatus, the sprayed sheet was removed from the drum immediately after spraying and transferred to an oven to dry slowly for 1 hour.

^cSignificantly different from the value at the lower drying temperature ($P < 0.01$).

Table 7
Effect of processing variables on mechanical properties of free films prepared from Aquacoat® aqueous dispersion (TEC 23 %)

Processing condition		Mechanical properties ^a	
Spray rate (g/min)	Surface temperature (°C)	Tensile strength (MPa)	Elongation (%)
2.5	50	8.5 ± 1.5	2.9 ± 0.2
2.5	40	7.5 ± 1.0	3.0 ± 1.0
1.0	50	8.3 ± 1.7	2.9 ± 1.2
0.8	30	Film with cracks	

^aMean ± S.D. of five experiments.

Table 7 shows the influence of processing conditions on the properties of films prepared from the Aquacoat® dispersion. Due to the high MFT of this dispersion, a significant influence of processing temperature on film properties was anticipated. At a surface temperature of 30°C, many small cracks were evident in the films. These findings were in agreement with the results from the MFT measurement. At 40 and 50°C, intact films were obtained. The difference in tensile strength of films prepared at these two temperature conditions was not significant. The effect of drying temperature on the physical properties of Aquacoat® films using a cast method has been reported (Arwidsson et al., 1992). The increase in drying temperature enhanced the tensile strength of the films. In this study, the difference of the surface temperature between 40 and 50°C was not significant, but the mean tensile strength at 40°C was lower than at 50°C. The spray rate did not influence the properties of films prepared at 50°C. This was expected since the size of dispersed particles is similar to that of the Eudragit® dispersions.

4. Conclusions

In summary, free films from aqueous polymeric dispersions of enteric and sustained-release formulations were prepared with good reproducibility using a novel apparatus. The telemetric temperature measurement system was demonstrated to be an effective system to monitor the film formation temperature on a rotating drum ap-

paratus. Temperature was found to influence the film properties especially for high MFT polymeric dispersions, such as Aquacoat® and Aquateric®. The spray rate was also an important parameter influencing the film properties for dispersions with large solid particles, such as Aquateric® and Shin-Etsu AQOAT®. The mechanical properties of free films prepared from Eudragit® L 30D-55 and Eudragit® L 100-55 were found to be independent of the processing variables that were examined in this investigation.

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