

## Development of a Continuous Process To Make Casein Films

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A semicontinuous pilot plant process was developed to make films from calcium caseinate plasticized with glycerol. Calcium caseinate/glycerol solutions were wetted and spread on polyethylene or Mylar belts and were readily removed after forming films. A Meier rod was used to spread the solution onto the belt. Feed rates were 3.3 and 5 g/cm. The resulting films using the polyethylene belt had a tensile strength of 5 MPa and 30% elongation with thickness of 0.14 mm. When the glycerol concentration was adjusted, the process produced films with properties similar to those previously reported for a batch process. It is anticipated that this process will apply to other proteins.

**KEYWORDS:** Casein; dairy film; coating

### INTRODUCTION

The use of films or coatings made from proteins and other biobased materials, as an alternative to polymeric nonbiodegradable films, has been the subject of many review articles (1–6) and is a subject of continuing research. Films are usually formed as free-standing thin sheets, whereas coatings are sprayed or formed directly on the product. Films or coatings based on proteins are desirable because they are environmentally friendly, biodegradable, and, more importantly, offer a lucrative outlet for surplus agricultural materials, such as casein. Edible films or coatings may be used to inhibit migration between components in foods, to carry food ingredients, and to improve the mechanical integrity and handling characteristics of foods. They may also be used as mass transfer barriers between the food and the environment, controlling the transfer of moisture, oxygen, carbon dioxide, and aromas (7). Edible films and coatings are not a replacement for synthetic packaging materials for prolonged storage of food but are to be used to improve food quality, extend shelf life, and possibly improve the efficiency of packaging materials (1). In nonfood applications, dairy protein films have potential for use in products such as diapers and cleaning and dusting wipes. Caseins are phosphoproteins comprising ~80% of the proteins in milk (8).

Casein exists in micelles, colloidal complexes of protein and salts, and function biologically to deliver protein, Ca, and phosphorus to the neonate (9). The micelles consist of submicelles, bound by soluble calcium phosphate and citrate. The micelle is a porous structure and has the ability to accommodate large amounts of water and, as such, is easily influenced by environmental processing conditions but resistant to temperature. The submicelles consist mostly of  $\alpha_{s1}$ -,  $\alpha_{s2}$ -, and  $\beta$ -caseins and the micelle-stabilizing  $\kappa$ -casein and associate by strong hydrophobic bonding (10, 11).

An extensive review of the properties of milk protein films cast from aqueous solutions of casein and caseinates, the component casein proteins, and NFDM is presented in McHugh and Krochta (12). Films were cast by pouring aqueous solutions of the protein into a plastic or glass Petri dish or onto any other flat surface (the substrate) that can contain the solution and allowing it to dry under constant relative humidity—usually at 50% relative humidity—and usually at room temperature. The films are sensitive to relative humidity, are water soluble, and are brittle and crack if a plasticizer such as glycerol (GLY) is not added to impart flexibility and elongation. GLY reduces intermolecular forces in films by inserting itself between the protein chains (13, 14). Electrostatic attraction between Ca and the hydroxyl groups of GLY is possible. GLY may also establish hydrogen bonding with amino acid residues of casein.

Tomasula et al. (15, 16) developed a continuous pilot plant process to produce casein with some of its micellar calcium intact and no added salts of neutralization. The process uses high-pressure CO<sub>2</sub> to reduce the pH to precipitate casein. When the pressure is removed, the CO<sub>2</sub> is vented and the pH returns to near the pH of milk. No salts of neutralization are present because there is no mineral acid present that needs to be neutralized.

The somewhat intact micellar casein presents the possibility of producing biodegradable polymer coatings with unique physical properties. Tomasula et al. (13) compared films made from high-pressure CO<sub>2</sub> with films made from calcium caseinate. CO<sub>2</sub>-casein films were barely soluble in water and were slightly stronger and stiffer than the corresponding calcium caseinate films. GLY was the plasticizer in all cases.

The films were made in Petri dishes and dried in ambient air (13). The “laboratory” Petri dish procedure took ~1–2 days to make films at room temperature. We want to develop a continuous process on a pilot plant scale and ultimately develop a commercially feasible continuous process to make continuous

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polymer coatings. Ideally, CO<sub>2</sub>-casein would be used in these studies, but it is still a research material available in limited quantity. However, calcium caseinate is commercially available and should provide a suitable surrogate for developing the process. After the process is developed using commercially available calcium caseinate, CO<sub>2</sub>-casein will be substituted into the process and evaluated.

There are various ways to produce films commercially. The simplest process is casting (17), which is used for free-standing films. The liquid solution is poured onto a smooth, flat surface to a controlled thickness. The film can also be prepared with a spreader. The spreader draws solution from a product reservoir with an adjustable gate to control thickness. The spreader is drawn over the substrate to deposit solution at the desired thickness. This is a batch process not conducive to continuous processing and is normally too costly. The solution can sometimes be extruded into a continuous film (18). For example, some films are produced by extruding a viscous aqueous suspension into a neutralizing coagulation bath, washing, plasticizing, and drying. This usually applies to highly concentrated, viscous solutions or melts at elevated temperatures. Sometimes, the best method is to dip rollers into the solution to pick up the solution (19). The film forms on the rolls and is then doctored off. For a dilute aqueous solution, such as studied in this research, the solution is usually spread onto a moving belt and passed through a dryer.

The critical issues in developing a continuous film process for casein are dryer parameters, a suitable belt, and the feed system. The general dryer conditions were previously established (20) during the determination of the drying curves. From the drying curve study, 34–35 °C was the maximum temperature that did not cause visible deterioration of the finished film. Forced warm air accelerated drying. The solution must spread evenly on the belt surface, and the surface must release the film after drying (18). There are various commonly available belt materials such as polyurethane, polyester, nylon, PVC, cotton, rubber, silicone, and polyethylene (22). Although vendors can supply general characteristics of the belt materials, actual selection is by trial and error.

The general dryer conditions were determined from earlier research. Kozempel et al. (20) determined the drying kinetics of calcium caseinate/GLY solutions. They showed that most of the drying time is devoted to constant-rate drying. Films start to form at 15% solids (wet basis). Therefore, it would appear to be economically advantageous to start with 15% solutions, but when this is done, the films usually have defects due to trapped air bubbles.

Surface tension is the tendency of liquids to minimize the surface area. It is "a measure of the inward forces that must be overcome to expand the surface area of a liquid" (21). It is characterized by the ability of an object denser than the liquid, such as a metal pin, to float on the liquid. Kozempel et al. (20) determined the surface tension of various solution concentrations from 4 to 15% solids. The surface tension is constant from 4% to near 10% solids and then rises rapidly above 10% solids. The increased surface tension prevents the removal of bubbles, resulting in defects. Hence, 10% was chosen as the appropriate starting concentration. At 35 °C drying temperature, the resultant films were slightly yellow and were progressively darker as the drying temperature increased. Because higher temperatures result in faster and more economical drying, a temperature of 34 °C was chosen for drying.

The objective of this study is to develop a continuous process for making calcium caseinate films. The methodology developed

in this study should extend to other protein- and carbohydrate-based films. The specific objectives of this study are to determine a suitable belt material and a suitable feed system and to integrate them into a continuous process based on the previously determined dryer parameters.

## EXPERIMENTAL PROCEDURES

Film solutions were 10% solids, wet basis. The solids were 70% calcium caseinate and 30% glycerol except for one experiment that was 40% glycerol. The raw materials were a commercial calcium caseinate, Alanate 310, NZMP (North America) Inc., and glycerol, 99.5% ACS reagent grade, Aldrich. The solvent was deionized water. The calcium caseinate and glycerol were thoroughly mixed in the water with a Braun handblender, MR310, Braun Inc., Lynnfield, MA, for  $\geq 2$  min. The solutions were refrigerated at least overnight to permit foam to disappear. In a fully continuous process, the foam would likely be actively removed, probably using vacuum.

The dried films were placed in a humidifier maintained at room temperature and 55% relative humidity (RH) for at least 1 day before testing. A Texture Analyzer model TA-XT2 tensile tester (Texture Technologies Corp., Scarsdale, NY) equipped with a 5 g load cell was used to measure tensile strength and elongation to break. Five replicates were run for each batch film sample using 5 mm wide specimens. The samples were cut 5 mm wide with a razor blade and a template. Continuous films were sampled at 15 cm intervals. Five replicates were run for each interval using 5 × 40 mm specimens. A gauge length of 25 mm and an extension rate of 0.2 mm/min were used. Samples were stored at 50% RH for at least 24 h before testing. A model 3 micrometer (B.C. Ames Co., Waltham, MA) was used to measure film thickness. Reported values are the mean of three measurements selected randomly over the face of the film sample. The precision of the thickness measurements was  $\pm 5\%$ .

Testing belt materials for suitability was subjective. The material had to meet two criteria: (1) if the solution spread (wetted) readily over the belt surface instead of beading up and (2) if the dry film was easily removed manually without excessive sticking or tearing, the material was considered to be acceptable.

**Batch.** Initially, small pieces of belt and other materials were screened for the tendency of the film solution to wet or spread uniformly on the following materials and for ease of removal after drying: (1) gloss polyethylene (PE) PE2 M65 C COS, Frank Callahan, Inc., Colmar, PA, 9 × 16 cm × 1.5 mm thick; (2) matte finish PE EM 10/2, Burrell Leder Beltech, Grand Rapids, MI, 8 × 15 cm × 2.25 mm thick; (3) polyurethane (PU) SP2C EM 8/2 Burrell Leder Beltech, 9 × 16 cm × 1.6 mm thick; (4) aluminum foil 0.017 mm thick; (5) a solid bar of aluminum; and (6) glass. All materials except the aluminum foil, aluminum bar, and glass were glued to a 15 mm thick board to keep them flat. Film solutions were pipetted onto the surfaces and permitted to form a uniform liquid coating. Preliminary samples on PE and PU were permitted to dry at room temperature. Ambient conditions were 21 °C with a relative humidity ranging from 20 to 50%.

After this initial screening for wetting and ease of release, two of the samples, gloss and matte PE, were cleaned and further tested by pipetting film solution onto the test surfaces and allowed to form a uniform liquid coating. They were placed in the continuous dryer (batch test) but with no movement (dryer drive off). The dry bulb temperature was 32–35 °C, and the relative humidity ranged from 12 to 23%.

A sample of Mylar [poly(ethylene terephthalate)] was received after the PE samples had been tested in the dryer. The Mylar belt was tested by drying film solutions at room temperature, 21 °C. Relative humidity was low, 7–20%. The film solutions were pipetted onto the surfaces and permitted to form a uniform liquid coating. The samples of aluminum foil and the aluminum bar were tested similarly to Mylar.

**Continuous.** The only continuous pilot plant dryer available was a microwave dryer (Cober Electronics Inc., Stamford, CT) with electrical and steam heat available. The dryer was 6.25 m long and 0.3 m wide: The dryer was not level from entrance to exit. The belt changed elevation by  $\sim 6$  mm at several points in the dryer. The first such point was  $\sim 100$  cm into the dryer. So, after  $\sim 90$  cm of the belt had been fed into the dryer (the nominal length of the PE belt), the belt drive

**Table 1.** Physical Properties of Batch Films Formed on Various Belt Materials

belt	tensile strength, <sup>a</sup>		% elongation at break <sup>a</sup>		thickness, <sup>a</sup>	
	MPa	SD		SD	mm	SD
PE gloss	1.83a	0.612	38.6a	22.269	0.255a	0.071
PE matte	2.97b	1.025	29.0a	23.740	0.169b	0.048
Mylar	2.30ab	0.419	29.6a	21.589	0.232a	0.057

<sup>a</sup> Within each column, means with no letter in common are significantly different ( $p \leq 0.05$ ).

was stopped and the film permitted to dry without traversing the length of the dryer. The microwave heating system was disconnected. Air flow was 19 L/s.

A 25.5 cm × 92 cm PE belt was glued to a 3 mm thick sheet of polycarbonate to present a flat, somewhat rigid surface to the film solution. The film solution was pumped with a Masterflex L/S model 77200-62 peristaltic pump (Cole-Parmer Instrument Co., Vernon Hills, IL) with Masterflex 06402-15 neoprene tubing. The liquid flow rate was 18.3 g/m. A Meier bar no. 500 spirally wound with 1.00 mm diameter wire (Testing Machines Inc., Islandia, NY) was used to distribute the film solution.

## RESULTS AND DISCUSSION

“Surface energy results from nonsymmetric bonding of the surface atoms/molecules” (23). The interaction of the surface energy of the belt and the surface tension of the solution determines the wetting ability of the belt/film solution combination, whereas the surface tension keeps the solution on the belt (23). The film solution must wet the belt surface and spread to a uniform coating. Additionally, it must remain on the belt until dry and be readily released.

If the surface energy of the belt material is too high or the surface tension of the solution too high, the solution will bead up and not form a film. If the surface energy of the belt material is too low or the surface tension of the solution too low, the solution will spread very thin, flow off the belt, and form a very thin film.

**Batch.** In the initial batch studies made with samples of various belt materials, the film solution wetted the aluminum foil and the aluminum bar and formed a uniform film. However, the dried film could not be removed without destroying it. As a comparison, the film solution beaded up on a glass surface and did not form a continuous film after drying but was easily removed. The polyurethane belt was very promising. The film solution wetted the belt and spread uniformly. The dry film was easily removed. However, on repeat applications with the same belt, the solution wetted the belt uniformly, but it became increasingly difficult to remove the finished film from the belt surface. According to the supplier of the belt material, there was no surface coating applied. Also, there was no visible residue on the belt either before or after cleaning. Apparently, the surface energy of the belt changed so the film could no longer be easily removed.

Two belt materials did work well—PE and Mylar. The PE came in two finishes, gloss and matte. **Table 1** lists the results of the tests with these three films. The tensile strength of the film formed on the matte finish PE was statistically significantly higher than that of the film formed on the gloss finish. There was no statistically significant difference in elongation. The thickness of the film formed on the matte finish PE was statistically significantly thinner than that of the film formed on either the gloss PE or the Mylar.

However, it became increasingly difficult with each trial to remove the finished film from the matte finish. After 11 repeat films, although somewhat difficult, the films were removable. The films had an opaque appearance because of numerous depressions corresponding to the matte finish. The gloss finish did not suffer from these shortcomings. The solutions easily wetted the surface, spread uniformly, and remained on the belt. The dry film was easily removed. With 52 repeat applications, the film was still easily removable. The film solution wetted the Mylar surface uniformly, and after 11 repeat applications the dry film was as easy to remove as with the gloss PE.

Even though the films formed on the matte PE belt were slightly superior in tensile strength, we chose the gloss PE belt to continue with the continuous process development because the films formed on the matte finish PE were opaque and much more difficult to remove. However, the matte belt might be the best choice depending on the end use. We chose the gloss PE belt over the Mylar because it performed as well as Mylar and it was conveniently available because we already had it. The Mylar belt material was too narrow and too thin for use in the continuous dryer and would have had to be purchased. We expect the Mylar would perform the same as PE.

**Continuous.** As stated earlier, we did not have a suitable continuous hot air belt dryer available in our pilot plant. The closest approximation was a continuous microwave dryer with electrically and steam-heated air fitted with a porous belt. We used this, minus the microwave heating, to simulate a continuous dryer.

In actual continuous dryer application, the belt material would be stretched tight to present a flat rigid surface rather than glued to polycarbonate sheets. Drying time was a minimum of 5.5 h. The belt speed needed to achieve this residence time in our small dryer was too slow to match a suitable solution feed rate to the belt.

A belt speed of 2.2 m/h and a feed rate of 18.3 g/m gave a fairly uniform film when dried. The spirally wound Meier rod tended to distribute solution preferentially to one side of the belt. Therefore, the solution was pumped  $\sim 10 \pm 2.5$  cm from the opposite side of the belt. To quickly establish a steady state feeding/distribution pattern on the belt, we wetted the Meier rod with feed solution. If the solution exceeded the width of the spiral windings on a side, it would be drawn off the belt by the unwound rod. To prevent this we occasionally had to elevate the rod slightly on the side to which the solution was drawn to properly distribute the solution. (When the proper belt loading was established, this was no longer a problem.) This prevented the solution from gaining contact with the unwound section of rod. If a solution did manage to go off the side, we stopped the solution feed and removed the Meier rod until fresh belt area was exposed. Then the process was begun again. The finished film did not dry and form a film within the remainder of a normal working day, so the dryer was turned off and the film permitted to finish overnight.

**Table 2** lists the physical properties of the films formed on gloss PE. The mean tensile strength and thickness, 3.9 MPa and 0.13 mm, compare favorably with the results reported by Tomasula (13), 1.9 MPa and 0.15 mm, respectively. However, the mean percent elongation at break (%EB) was much lower, 27 versus 71%. The standard deviation (SD) was quite high for all of the results, indicating the film was not uniform. This was obvious when we removed the film from the belt. The front or leading edge was thicker, presumably because the dryer was not perfectly level inside. **Figure 1** plots the film thickness starting at the leading edge. This confirms that the film became



**Table 2.** Physical Properties of Continuous Films Formed on Gloss PE at High Belt Loading<sup>a</sup>

run	<i>n</i> <sup>b</sup>	<i>m</i> <sup>c</sup>	tensile strength, MPa	SD	% elongation at break	SD	thickness, mm	SD	film width, cm
1	20	5	3.7	1.23	23.0	16.97	0.107	0.0302	11
2	7	2	4.9	1.14	19.6	24.60	0.071	0.0160	11
3	17	4	3.4	1.07	39.4	29.20	0.118	0.0436	24.1
4	25	6	3.4	1.35	17.0	15.52	0.143	0.0450	17.8
5	12	3	3.7	0.62	41.9	32.29	0.149	0.0381	19.1
6	21	6	3.0	1.16	32.2	24.95	0.148	0.0531	17.8
7	20	5	5.7	1.83	23.2	22.59	0.117	0.0498	19.7
mean	122		3.9	1.56	27.3	24.30	0.126	0.0480	17.2

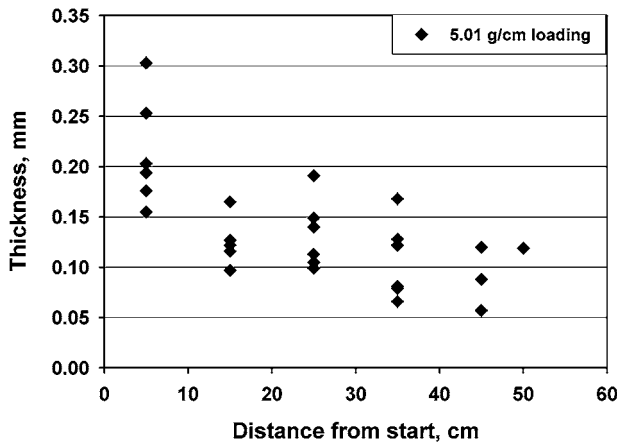
<sup>a</sup> Belt loading = 5.01 g/cm. <sup>b</sup> *n* = total number of samples = sample locations (*m*) × replicates. <sup>c</sup> *m* = number of sample locations on the film.

**Table 3.** Physical Properties of Continuous Films Formed on Gloss PE at Low Belt Loading<sup>a</sup>

run	<i>n</i> <sup>b</sup>	<i>m</i> <sup>c</sup>	tensile strength, MPa	SD	% elongation at break	SD	thickness, mm	SD	film width, cm
8	20	5	4.3	1.29	24.6	18.69	0.135	0.0400	20
9	8	2	5.5	1.15	16.1	24.01	0.081	0.1235	17
10	20	7	4.0	2.51	28.1	32.15	0.119	0.0781	20
11	18	6	5.2	2.23	21.3	21.46	0.113	0.0727	20
12	21	7	4.7	2.05	12.1	15.62	0.093	0.0429	22
13	16	6	3.4	1.05	33.3	36.17	0.115	0.0528	18
14	17	6	2.6	1.24	21.0	24.86	0.116	0.0725	20
mean	119		4.2	1.96	22.5	25.49	0.113	0.0608	19.6

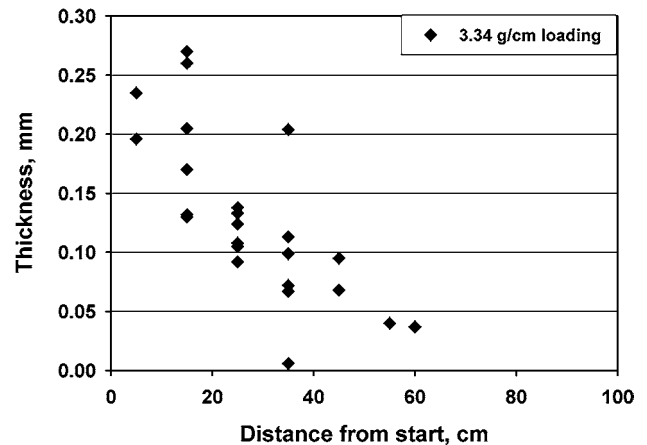
<sup>a</sup> Belt loading = 3.34 g/cm. <sup>b</sup> *n* = total number of samples = sample locations (*m*) × replicates. <sup>c</sup> *m* = number of sample locations on the film.

**Film Thickness vs. Distance**



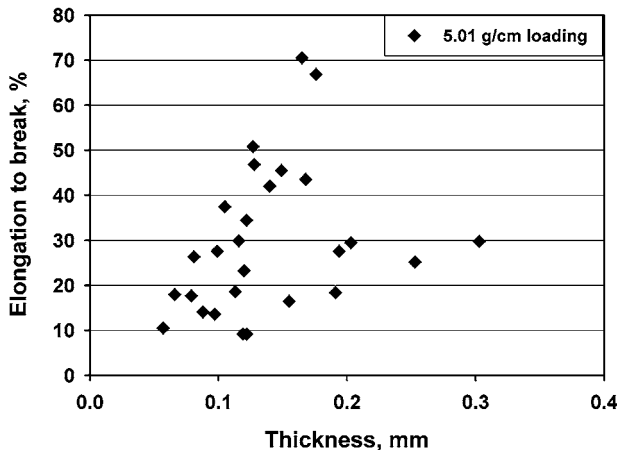
**Figure 1.** Film thickness versus distance into the dryer, belt loading = 5.01 g/cm.

**Film Thickness vs. Distance**



**Figure 3.** Film thickness versus distance into the dryer, belt loading = 3.34 g/cm.

**Elongation vs. Film Thickness**



**Figure 2.** Elongation versus film thickness, belt loading = 5.01 g/cm.

thinner along the length of the film, although it was relatively constant in the middle.

In addition, there were considerable defects such as bubbles in the film. We plotted the %EB versus the thickness in **Figure 2**. There was a trend for the %EB to increase with thickness, but, more importantly, there is considerable scatter in the data. The obvious conclusion is that this process can make a continuous film but the physical conditions must be improved to produce a uniform film.

We then increased the belt speed to 3.3 m/h and wetted the Meier rod with film feed solution. This made a big difference in the feed. In the previous experiments, the film solution would not always spread out uniformly and would sometimes go off the side of the belt. Under these improved conditions, the film solution spread immediately to the side but remained within the wire windings of the Meier rod. The belt loading was uniform. At this faster feed rate, the solution dried and the film formed in 5.5 h.

The results of these experiments are listed in **Table 3**. There was little difference in the mean values of thickness, tensile strength, and elongation due to belt loading. The mean thickness was statistically significantly ( $p \leq 0.05$ ) thinner with the increased belt speed and with the Meier rod wetted with feed solution. The film thickness was 0.113 versus 0.126 mm. However, there was no statistically significant difference in tensile strength, 4.2 versus 3.9 MPa, or %EB, 22.5 versus 27.3. The mean tensile strength and thickness, 4.2 MPa and 0.11 mm, were comparable to the results reported by Tomasula (13), 1.9 MPa and 0.15 mm, respectively. The mean elongation at break was much lower, 23 versus 71%.

Although the film feed was improved, the film was still not homogeneous. The leading edge was thicker than the end, and there were numerous bubbles. **Figure 3** plots the film thickness from the leading edge to the end of the film. The film became thinner along the length of the film. **Figure 4** plots the %EB versus film thickness. The data have a lot of scatter as in the previous experiments, but there is still a trend toward higher %EB values with thickness.

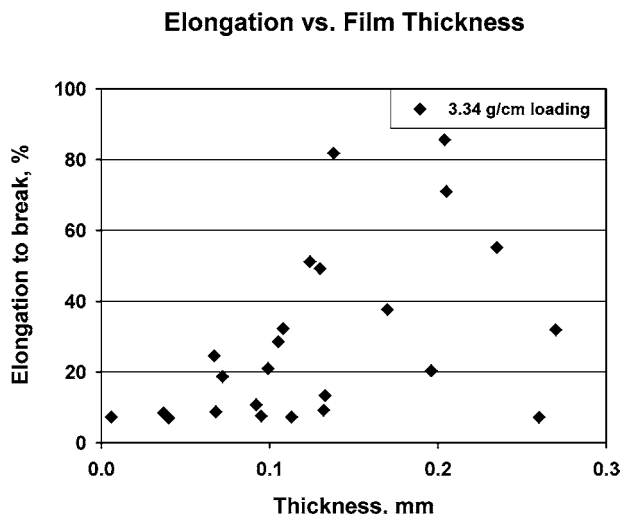


Figure 4. Elongation versus film thickness, belt loading = 3.34 g/cm.

This process produced a continuous film, but the quality was compromised by defects, namely, bubbles and ripples and uneven thickness. The uneven thickness was due to the unlevel dryer surface. This will be corrected for continued study by modifying the dryer. The source of the bubbles and ripples was probably due to the air flow and/or elevated temperature. To test this theory, we ran the feed and dryer as before but with no heat and only natural convection air flow. The belt loading was 3.34 g/cm. The film took several days to dry. There were no ripples and no apparent bubbles. The TS was 6.6 (1.738 SD), %EB was 16.1 (15.976 SD), and the thickness was 0.079 mm (0.0189 SD).

We repeated this experiment at the higher belt loading, 5.01 g/cm. There was little difference. The TS was 5.63 (1.904 SD), %EB was 20.6 (24.639 SD), and the thickness was 0.111 mm (0.0297 SD). There were no ripples and no apparent bubbles.

Next, we ran the feed and dryer with heat, 34–35 °C, and only natural convection air flow at the higher belt loading, 5.01 g/cm. The films formed overnight with the dryer left on. There were no ripples and no apparent bubbles. This indicates the air flow caused the ripples and bubbles to form. The TS was 5.0 (2.416 SD), %EB was 30.3 (24.732 SD), and the thickness was 0.136 mm (0.0101 SD). There was no statistically significant difference in physical properties at the higher temperature compared to the previous two experiments run at room temperature.

The mean tensile strength and thickness, 5.0 MPa and 0.136 mm, respectively, compare favorably with the results reported by Tomasula et al. (13), 1.9 MPa and 0.13 mm. However, the mean percent elongation at break (%EB) was much lower, 30 versus 71%. For whatever reason, the TS was higher and the %EB was lower. These two physical properties have an inverse relationship. Therefore, we used a higher concentration of GLY (40% GLY and 60% calcium caseinate) in water at the higher belt loading, 34–35 °C, and 35–40% RH. There was no noticeable difference in drying. The films formed overnight with the dryer left on. The mean TS of four experiments was 1.1 (0.700 SD), %EB was 59.4 (26.126 SD), and the thickness was 0.150 mm (0.0660 SD). These results are much closer to those reported by Tomasula et al. (13) at 30% GLY.

In this study the TS ranged from 2 to 5 MPa and the SD was 0.3–2.4. The low value for SD was for the Petri dish films. There was a big difference in the %EB and previously published data (20). The films formed in the Petri dish (20) were 70% EB, whereas the films formed on the PE or Mylar belt material

ranged from 22 to 39%. The film thickness was 0.17–0.26 when the films were formed statically, either in a Petri dish or on belt material. The thickness was less, 0.11–0.15 mm, in the continuous dryer. This was probably a function of the belt loading, which will be studied in continuing research.

There was a difference between the films formed in Petri dishes and on belts. It is especially obvious with %EB. Film solutions formed a meniscus on the outer edge of Petri dishes. The films formed and became clear beginning in the center. The last to dry and form a film was on the outer radius in the meniscus. The film solution on the belts wetted the belt and spread out. These films dried and formed films starting at the outer edges or sides. Typical film widths were ~20 cm on the belts. Petri dishes were ~9 cm in diameter.

Liquid solutions generally evaporate from the edge, and the solution diffuses to the evaporating edge. As a consequence, solids migrate to the edge. The result is a “coffee ring” type spot or residue (24). This might be happening in these experiments. The calcium caseinate and GLY diffuse to the edge as the solution dries and the film forms. The film would then not be uniform across the width. This could account for the high standard deviation in the films formed on belts. The SD is high for films formed on belts whether statically or in the continuous dryer. In the Petri dishes, the radius is much smaller and the evaporating surface appears to be in the center, not at the edge. Edge effects will be studied in continuing work. It might be necessary to include raised sides on the belts to force the films to form from the center out.

**Conclusions.** Caseinate films can be made in a continuous, albeit slow, process. PE or Mylar belt material permits caseinate solution to properly wet the belt and release easily after drying. The dryer conditions previously determined are suitable, and a Meier rod effectively spreads the solution.

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