Lecithin Gum Rheology and Processing Implications

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ABSTRACT: The rheology of canola, sunflower, and soybean lecithin gum was examined by studying samples of different moisture contents produced in a batch evaporator (70°C, 0.1 atm). Soybean lecithin was found to have the lowest viscosity, approximately 10 poise (100 s^{-1}, 70°C), compared to canola and sunflower lecithin with viscosities of approximately 90 and 90,000 poise, respectively. The high sunflower viscosity was attributed to the presence of long-chain waxes. Lecithin gum was shown to change from a Bingham (water continuous phase) to a pseudoplastic (oil continuous phase) type fluid as the moisture content of the lecithin gum decreased. The viscosity maxima occurred between 6.9 and 19.3% moisture content (100 s^{-1}), with the variation found to be related to the oil/water ratio of the system. Rheological results indicated that vertical scraped surface evaporator design could be optimized through the addition fluidizing of agents prior to the evaporator and/or increased heating at the evaporator outlet.

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KEY WORDS: Canola, lecithin gum, processing, rheology, soybean, sunflower.

Commercial lecithin has traditionally been derived from soybeans owing to its high quality and low cost. Alternative commercial sources of lecithin have emerged with the increased production of sunflower and canola/rapeseed. The qualities of lecithin derived from these "soft" varieties of oilseeds have often been seen as inferior to that of soybean in respect to color, taste, and consistency (1,2). However, with effective quality control measures "soft" seed lecithin of comparable quality to existing commercial soy lecithin can be produced (Schneider, M., personal communication, 1997).

The unique rheology of the lecithin-water system has lead to processing difficulties in the drying of lecithin. The rapid increase in viscosity that occurs when soybean lecithin gum contains 5–15% moisture (3) has further complicated thin film continuous drying. The rheological characteristics of soybean lecithin gum during drying are ill-defined in the literature. Even less is known about the rheological characteristics of sunflower and canola/rapeseed lecithin gum, apart from some processing difficulties noted in early attempts at production of rapeseed lecithin (4,5). This study focuses upon comparisons of lecithin gum rheology under steady state conditions and proceeds to derive practical engineering solutions to processing problems.

De-oiled lecithin, even in low concentrations in aqueous solutions, exhibits considerable viscoelastic and non-Newtonian behavior (6), to a degree not normally associated with low molecular weight polymer chains. This results from the formation of an electrostatic gel arising from the electrostatic nature of the polar portion of the lecithin molecule, the charged nature of the micelles, and the large dipole moment and hydrogen bonding capacity of water (7). The importance of water in the creation of an electrostatic gel was underlined through the confirmation that de-oiled lecithin in a hydrocarbon solvent exhibited Newtonian characteristics and the solutions followed a modified Einstein equation for suspensions of noninteracting spheres (7).

Lecithin gum approximates a lecithin/water/oil mixture in a ratio of about 35:50:15, depending upon degumming processing conditions. The phospholipid species of lecithin form an interface between the polar (water) and nonpolar (oil) components of the system, through bilayer and micellar structures.

MATERIALS AND METHODS

Materials. Canola (*Brassica napus*), sunflower (*Helianthus annus*), and soybean (*Glycine max*) lecithin gums were obtained from Cargill Oilseed processors, West Footscray, Victoria, Australia. The corresponding refined and deodorized vegetable oils were obtained from Meadow Lea Foods of the same location. Fatty acids of animal origins were supplied by Unichema, Port Melbourne, Victoria, Australia.

Lecithin gum drying. Samples (600 g) of lecithin gum were dried at a temperature of 70°C, under a vacuum of 0.1 atm, at an agitator speed of 60 rpm. A torque arm attached to the drive motor was used to detect relative changes in viscosity of the mixture (Fig.1). Sampling frequency was concentrated toward the period in which maximum torque was recorded, with a minimum of eight samples taken per run. Lecithin gum samples ($n = 27$) were put into sealed glass containers, and moisture and rheological measurements were performed within 2 h of the samples being taken. Fluidized lecithin samples ($n = 14$) were produced through the addition of oil and/or fatty acid to the lecithin gum. Fluidization achieved through the use of diavalent salts and acetylation was performed in accordance with the methodology described by Lantz (8) and Davis (9). Moisture was determined

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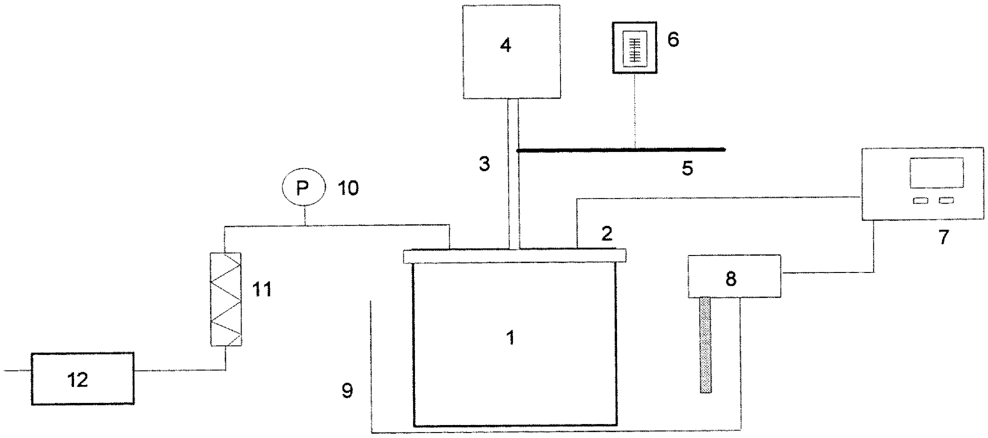

FIG. 1. Schematic diagram of batch drying of lecithin gum: 1, reaction vessel; 2, top flange; 3, agitator shaft; 4, drive motor; 5, torque arm; 6, weight scales; 7, temperature controller; 8, thermostat; 9, water bath; 10, pressure gauge; 11, condenser; 12, vacuum pump.

by placing duplicate samples in an oven at 105°C until constant weight was achieved. [This method produces apparent moisture levels which are 0.5–1% higher than the actual level (8)].

Rheological measurements. Analysis was performed at the Cooperative Research Centre (CRC) for Industrial Plant Biopolymers, functional testing laboratory. A Carri-Med Controlled Stress Rheometer CSL^2 (TA Instruments Ltd., Dorking, Surrey, England), was used with the cone and plate measurement system set at 2 cm, 2 degrees and 0.054 mm gap. Rheology measurements were performed at 70°C for hydrated and 25°C for fluidized samples.

To be able to measure the yield stress and viscosity of the samples, which varied from oil-like to paste-like in texture, a stress range from 10–4500 Pa was used. This was employed in 20 incremental steps on a log scale. The dynamic yield stress as defined by the Herschel-Bulkley equation was used in calculations by the rheometer's software. The instrument's measured yield stress was not used owing to sample slippage that occurred below a shear rate of 1.00×10^{-4} s^{-1}, rendering these results invalid. Sample slippage was attributed to hexane-insoluble matter in the samples. The viscosity of the hydrated and fluidized lecithin samples was determined from the shear stress vs. shear rate data, through the rheometer's software.

The viscosity curves produced by the regression model all had a correlation coefficient of above 0.97, with the majority exceeding 0.99. The shear rate range obtained for the lecithin samples varied from 1.00×10^{-4} to 3.30×10^{2} s^{-1}. The upper limit of the shear rate range was constrained by the 4500 Pa maximum operating shear stress of the rheometer. This necessitated the extrapolation of data to simulate the viscosity curve (Fig. 5) at commercial operating shear rates (10,000 s^{-1}), a practice used by commercial equipment designers.

RESULTS AND DISCUSSION

Rheology of hydrated lecithin. Flow curves of hydrated lecithin reveal the rheological complexity of these highly non-Newtonian mixtures. The flow curves (Fig. 2) reveal that lecithin gum initially exhibits Bingham flow characteristics, i.e.,

$$\sigma_{12} = A + B\dot{\gamma}\text{—Bingham equation} \qquad [1]$$

As the moisture levels decrease, so does the linearity of the curve, with the mixture becoming more accurately described by the Power Law equation. The Herschel-Bulkley equation, a hybrid of the Bingham and Power Law equations, best describes the fluid behavior of the hydrated lecithin (The Herschel-Bulkley equation is equal to the Power Law equation when $A = 0$).

$$\sigma_{12} = K\dot{\gamma}^{\eta}\text{—Herschel-Bulkley equation} \qquad [2]$$

This change in flow behavior can be explained by the dominating intermolecular forces changing from electrostatic forces at high moisture levels to van der Waals forces as the moisture levels decrease (7). The Bingham constant (A) may be seen as the yield stress required to overcome the electrostatic forces and enable fluid flow. The yield stress reached a maximum at between 9 and 19% moisture for the lecithin gum varieties analyzed (Fig. 3). The maximum yield stress will correspond to the minimum moisture level at which micelles and/or liquid crystalline domains maximize hydrogen bonding interaction. This moisture level represents the boundary between oil and water continuous-phase systems. Further removal of moisture beyond this point involves the removal of hydrogen bonding and a decreased surface area interface between the oil and water phases. Alternatively, the addition of water beyond this point adds to the layer of the water continuous phase and lowers resistance to flow. Both actions result in a lower yield stress.

Comparisons of lecithin gums. Although the sunflower and canola hydrated lecithin had yield stress maxima at similar moisture levels of 8.8 and 12.2%, respectively, the soybean hydrated lecithin peaked at a level of 19.3% moisture

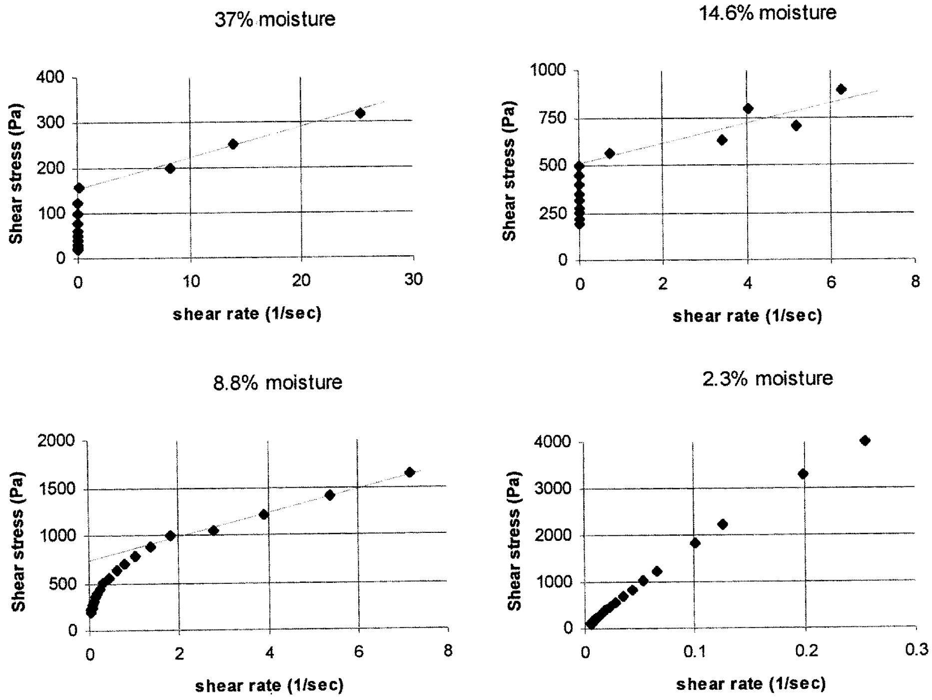

FIG. 2. Selective flow curves showing the rheological transition from a Bingham to a pseudoplastic type fluid as the moisture content of hydrated sunflower lecithin decreases.

(Fig. 3). This may be partly explained by the higher ratio of oil to lecithin [or lower ex-dryer Acetone Insolubles (AI) content] in this sample (Table 1). The increased oil level creates a more elliptical micelle (7), thus increasing the surface area available for hydrogen bonding. This theory was supported by the shift in the maximum yield stress to a higher moisture level on the addition of oil (and/or fatty acids) to the starting canola and sunflower lecithin gum (Fig. 4).

Commercial process. Drying of lecithin gum is performed commercially *via* either batch or continuous processing, with the latter technique dominating large-scale operations. Batch processing equipment consists of a reactor which is heated *via* a rotating coil heat exchanger. Although this drying method minimizes charring, it suffers from long drying periods of up to 8 h, and difficulties in consistently reducing the moisture levels to below 1% (Iwanyszcuk, R., private communication, 1997). These operating limitations can be directly attributed to the rheology characteristics within the evaporator. A combination of temperature and shear rate gradients across the reactor causes the viscosity of the mixture to vary significantly. It is conceivable that, at the same moisture level, the mixture may be in either a liquid or a near-solid state, depending on its position (and hence shear rate) in the reactor (Fig. 5). The resultant poor mixing (and hence poor heat exchange) contributes to both long reactor residence times and difficulties in achieving low moisture contents.

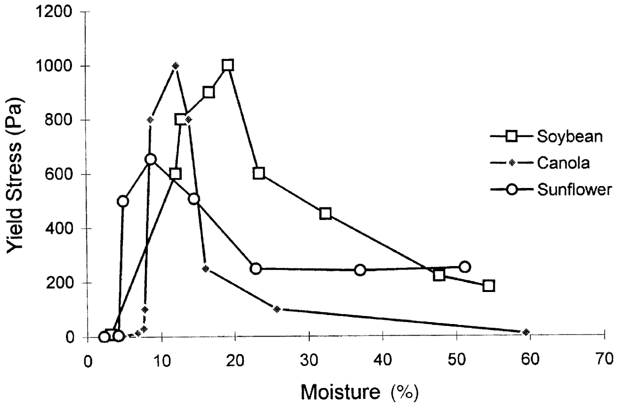

FIG. 3. Yield stress curves of soybean, canola, and sunflower hydrated lecithin.

The scraped surface evaporators used in the continuous processing have a film thickness of approximately 1–2 mm and an operating speed of approximately 600 rpm (5). As a commercial evaporator's circumference is at least 1 m, the operational shear rate will be in excess of 10,000 s^{-1}. The widespread usage of continuous thin film evaporators reflects the domination of lecithin production by the large-scale processors. However, while continuous processing is suited to steady-state production conditions, the maintenance of the steady state is not always possible. This is especially so with oilseed crushers which process multiple oilseed varieties. Unsteady-state startup conditions cause difficulties with vertical continuous evaporators, as lecithin gum viscosity changes with increasing shear rate (Fig. 5). As film thickness is directly proportional to shear rate within the evaporator, equipment maintenance is also critical. Worn blades have caused

TABLE 1
Characteristics of Fluidized Lecithin ($\dot{\gamma}$ = 1 s^{-1})

	Viscosity (poise at 25°C)	Acetone insolubles (%)
Commercial standard	200 max	62% min
Canola #1:ex-dryer	20000[a]	76
+ 3.5% Fatty acids	15000	73
+ 5% Fatty acids	6600	72
+ 5% Oil	4600	72
+ 5% Oil + 3.5% fatty acids	920	70
+ 10% Oil + 3.5% fatty acids	240	67
Canola #2: ex-dryer	1500	69
+ $MgSO_4$	690	69
Acetylated	920	69
Sunflower: ex-dryer	30000[a]	78
+ 10% Oil	6300	71
+ 20% Oil	230	65
Soybean: ex-dryer	5970	72
+ 3.5% Fatty acids	70	70
+ $MgSO_4$	64	72
Acetylated	120	72

[a] Viscosity measured at a shear rate of 0.1 s^{-1}.

severe processing difficulties and led to early design modifications which included a change from hinged to fixed blades (4) and the use of holes in the blades (5). In more recent times, the use of Ni/Cr plated cylinders with fixed stainless steel blades have been recommended to reduce maintenance costs (Veerabhadrappa, S., personal communication, 1997).

Processing implications. Hydrated canola and soybean lecithin viscosities at these shear rate levels tend to approach an asymptote at around 0.1 centipoise (cP), with marginally higher levels where electrostatic forces peak (Fig. 5). Conversely, below 5% moisture content sunflower lecithin solutions experience a rapid rise in viscosity due to the presence of sunflower waxes. This would suggest that under these processing conditions, an agitated thin film evaporator would not be suitable.

The commercial sunflower lecithin plant in Martfu, Hungary, has had minimal drying problems because of the lower levels of sunflower waxes present in their lecithin, an elevated operating temperature of 95°C at 100 mbar operating pressure (Kovari, K., personal communication, 1997) and the use of a horizontal scraped surface evaporator. This is more robust than a vertical evaporator, as mechanical rather than gravitational forces drive the flow of lecithin in the horizontal scraped surface evaporator (11).

On an operational level, processability and production output goals can have precedence over quality concerns (Veerabhadrappa, S., personal communication, 1997). However, through better control over lecithin rheology, production and quality objectives can both be achieved.

Effect of process temperature. The operating conditions at the Martfu plant are typical for continuous oilseed lecithin processing. Temperatures approaching or exceeding 100°C aid in increasing evaporator output and lowering fatty acid hydroperoxides [measured by the peroxide value (10)]. In contrast, reduced processing temperatures are desirable to avoid deterioration of organoleptic properties. In addition to phospholipids being easily hydrolyzed and oxidized, they are prone to react with decomposition products of fatty acid hydroperoxides and reducing sugars. The production of what are known as melanophospholipids, pigments not occurring in seeds, has been reported in the drying of rapeseed gum at elevated temperatures (5). Rapeseed/canola lecithin is especially prone to thermal degradation owing to the presence of reactive low molecular weight components: glucosinolates, phytates, and phenolics.

One method to improve fluid flow without compromising product quality would be to confine process temperature increases to around the evaporator outlet. This region has been responsible for process blockages because of elevated fluid viscosity resulting from heat losses from the outlet pump (12). Furthermore, whereas the viscosity profile of the lecithin gum is relatively constant at the operational shear rate, it may change dramatically in the low shear rate zone of the evaporator outlet (Fig. 6), especially if excessive residual moisture is present. This study suggests that the reported in-

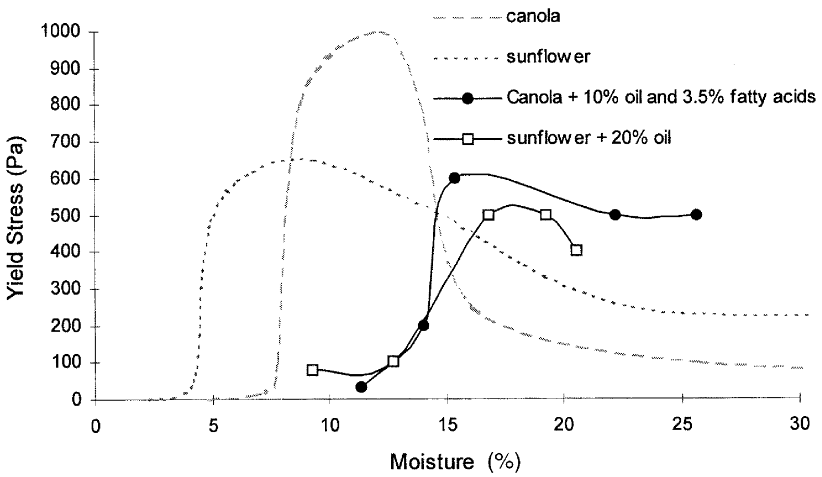

FIG. 4. Yield stress curves of canola and sunflower with viscosity-reducing additives.

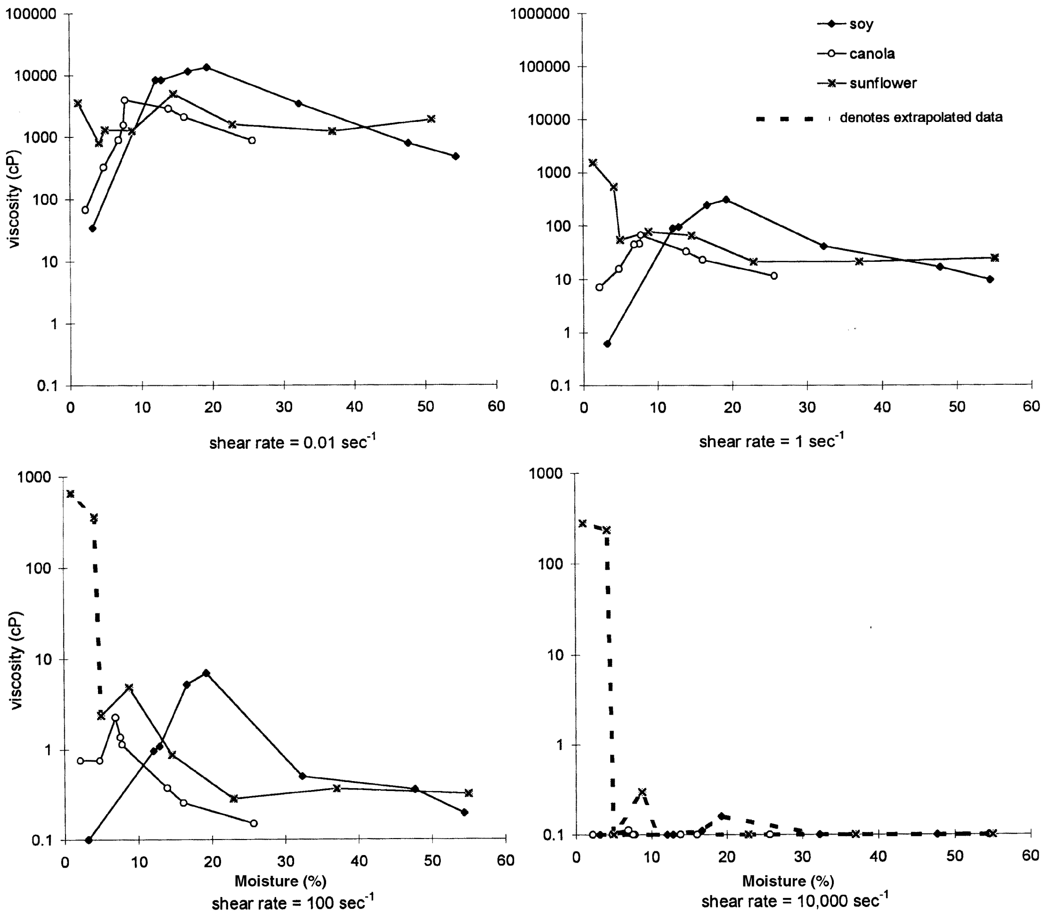

FIG. 5. Viscosity curves of lecithin sludge at 70°C.

ternal lecithin film breakage is not directly due to a rapid viscosity change of lecithin as previously postulated (3), as under the operating shear rate there is no rapid viscosity change. It is more probable that it results from fluid blockages at the evaporator outlet, which cause misalignment of the rotor shaft and hence propagate the process shock upstream. The resultant vibration of the shaft would cause rapid variations in the lecithin film thickness (and hence apparent viscosity) and thus induce the film breakage. It follows that the elevation of process temperatures to improve product flow should be centered around the outlet region. Although not a standard feature, some evaporator models have the option to extend the jacketed heating to cover the outlet zone. Alternatively, the use of lagging and a heat trace could serve a similar purpose. A temperature gradient stemming from the evaporator outlet may also be considered, because the low moisture content of the lecithin in this region is less prone to thermal degradation. An added benefit would be increasing the evaporation rate of residual amounts of moisture, which are reported to be approximately 2–3 times lower than for the evaporation of the bulk moisture (13). These figures were supported by the evaporation rate of the laboratory-scale batch evaporator, in which evaporation rates declined significantly once the moisture level had dropped below 4%.

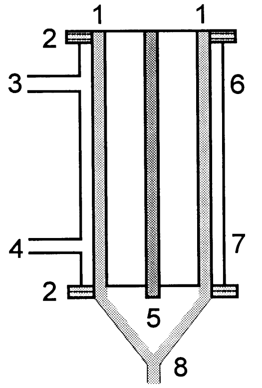

FIG. 6. Schematic diagram of vertical scraped surface evaporator: 1, lecithin thin film; 2, flange; 3, water/steam inlet; 4, water/steam outlet; 5, rotor shaft; 6, agitated zone entry—high shear, high moisture zone; 7, agitated zone exit—high shear, low moisture zone; 8, evaporator outlet —low shear, low moisture zone.

Effect of adding oil. The difficulties associated with sunflower lecithin drying may be avoided by lowering the equivalent dry-basis AI value through increasing the oil content of the wet gum. This may be achieved through the adjustment of degumming conditions (10) or the addition of oil prior to entering the evaporator. The lowering of the initial wet gum AI (dry basis) from 78 to 65% resulted in the dried lecithin viscosity at 70°C dropping from above 100 cP to reach an asymptote to 0.1 cP at the operational shear rate (i.e. 10,000 s^{-1}). The adjustment of AI levels in the lecithin gum before the drying operation has the advantages of eliminating postdrying unit operations and improving the lecithin gum rheology in the drying stage. An increased oil content ensures that the electrostatic-induced viscosity peak occurs at a higher moisture level and at a greater distance from the dryer outlet in a vertical scraped surface evaporator and thus ensures a greater robustness to process shocks. The mild steam-deodorizing action of the vertical evaporator also aids in the removal of crude oil volatile components which adversely affect product taste and odor.

Future processing directions. The processing problems experienced in removing residue moisture have led to the need for either extended drying periods (Iwanyszczuk, R., personal communication, 1997) (batch) or elevated drying temperatures (continuous) (5). The emergence of microwave technology in the chemical and food industries may offer some advantages compared to traditional evaporation techniques. Microwave drying has a number of advantages, including shorter drying times, smaller temperature gradients, and the ability to achieve low moisture levels (13). The production of chemically modified lecithin products is also likely to be enhanced by microwave-assisted organic reactions. In addition, bacterial growth would be minimized owing to the lower cycle times and aseptic environment induced by microwave radiation. This technology is likely to appeal to small producers who manufacture specialty lecithin products, whether by oilseed variety, chemical modification, or both.

Standardization of lecithin rheology. The viscosity of crude lecithin is commonly adjusted to below 200 poise at 25°C to improve its material handling properties. Fluidized lecithin is made by further dilution of the mixture with vegetable oil and/or fatty acids, with the constraints of competing acidity and purity requirements. The addition of divalent salts (7) and chemical modification through acetylation (8) have also been used to control lecithin viscosity.

The viscosity of dried lecithin fluctuates between different batches according to seasonal and varietal compositional characteristics. However, the results (Table 1) indicated that soybean lecithin tended to have a lower viscosity than sunflower or canola lecithin, although all were likely to satisfy the commercial viscosity specifications when diluted down to an AI content of 62%. The European Community specifications (Guideline 78/664/EWG) for lecithin (E322) specify that lecithin shall have greater than or equal to 60% AI, although commercial specifications are more commonly greater than or equal to 62%. The inferior consistency of sunflower and canola lecithin may be partly attributed to their "soft" seed characteristics, which lead to generally higher hexane insoluble levels in the form of fine seed particulates (Schneider, M., personal communication, 1997). In small lecithin production facilities, dilution of lecithin with vegetable oil has been the preferred method of controlling viscosity, with individual supply agreements allowing lower AI levels for bulk handling requirements (Iwanyszczuk, personal communication, 1997).

The rheology of oilseed lecithin gum is dependent upon both the oilseed variety and the composition of the lecithin gum. As opposed to batch evaporation, the high shear rates of the vertical scraped surface evaporator ensure that the lecithin gum experiences minimal viscosity variation under steady-state conditions. This study indicates that the low shear rate zone at the vertical scraped surface evaporator's outlet is most susceptible to flow blockages, especially if residual moisture in the lecithin is not minimized. Processing difficulties are more likely to be experienced in the processing of canola and sunflower lecithin, which have a relatively higher viscosity compared with soy lecithin. These processing difficulties may be avoided by a number of means, including: (i) implementation of external heating to the evaporator outlet zone, (ii) addition of fluidity-controlling additives to lecithin gum; (iii) increased evaporation temperature; and (iv) use of a horizontal scraped surface evaporator.

The latter two options are least preferred because an increased evaporator temperature is detrimental to lecithin quality and the use of a horizontal scraped surface evaporator increases capital costs.

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