Shear and Longitudinal Ultrasonic Measurements of Solid Fat Dispersions

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ABSTRACT: Dispersions of coating fat in corn oil (2.5–12.5 wt%) were prepared following two different protocols: Type A dispersions had an average crystal size of 30–36 µm, whereas type B dispersions were less than 1 µm. In both dispersions the fat crystals were aggregated into larger structures (up to 80 µm). The longitudinal ultrasonic properties (i.e., velocity, attenuation, and reflectance) were linearly related to the solid fat content, but only attenuation was sensitive to the different microstructures. The velocity and reflectance measurements were modeled using the Urick equation. Shear ultrasonic reflectance and oscillatory viscometry were used to measure the dynamic viscosity of all dispersions. According to both methods, type B samples were always more viscous than type A at a similar solids content. The correlation between the two techniques was good ($r^2 > 0.99$), but the numeric agreement was different for both systems.

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The microscopic properties of a fat crystal network (e.g., polymorphism, crystal size, and aggregation state), as well as the total amount of solids, play an important role in the texture of many food products. The importance of these parameters has prompted researchers to design methods to measure and therefore control crystal formation in semisolid dispersions. Among these, low-intensity ultrasound has demonstrated great potential. Low-powered ultrasound is ideal for sensing applications, as the material oscillations that constitute the wave are dependent on, but nondestructive of, the bonds of the material through which it passes (1). The material vibrations may occur either in the direction of sound propagation (longitudinal ultrasound) or normal to it (shear ultrasound).

Shear wave propagation is related to the shear modulus of the material; thus, it offers great potential for viscosity measurements. Unfortunately, food products are not capable of supporting shear sound propagation; therefore, only a surface technique such as ultrasonic reflectance can be implemented for measuring viscosity (1). Some authors have applied shear reflectance to characterize Newtonian and non-Newtonian fluids (2,3). Furthermore, the application of shear resonance has been extended to food operations such as coagulation of milk for yogurt manufacture (4) and, combined with longitudinal velocity, for study of the effect of water content in bread dough (5,6).

Although shear waves have not been applied to characterize semisolid fat dispersions, the application of longitudinal waves has found success. Several authors in the past have implemented longitudinal velocity to measure the solid fat content (SFC) of fat/oil mixtures such as purified lipids or pig adipose tissue dispersed in liquid oils (7,8). In a previous work, we showed that longitudinal ultrasonic reflectance could be used to give a good measurement of the SFC of coating fat and cocoa butter dispersions (9). Interestingly, the ultrasonic reflectance depended on the type of fat as well as the SFC, although the reasons for the difference were unclear and possibly included other aspects of fat structure such as crystal size, polymorphic form, or state of aggregation. However, the two fat systems used in that work were very different in crystal size and polymorphic form; thus, they did not allow a systematic understanding of the relationship with ultrasonic waves.

In the present work, dilute dispersions of the same type of fat (confectionery coating fat, CCF) with different crystal microstructures were prepared to test the ability of longitudinal ultrasound to characterize fat crystals. Shear ultrasonic reflectance was also used to measure the rheological properties of the dispersions.

MATERIALS AND METHODS

Sample preparation. CCF (CLSP870 nonlauric cocoa butter replacer "high *trans*"; Van den Bergh Food Ingredients, Joliet, IL) was dispersed in corn oil. Coating fat is typically a nonlauric, nonstabilizing fat with a stable polymorphic form that does not require a tempering procedure (10). The coating fat dispersions (2.5, 5, 7.5, 10, and 12.5 wt%) were held at 50°C for at least 30 min to ensure complete melting. The dispersions were then tempered in two different ways:

(i) Sample A: The dispersions were transferred from a water bath at 50°C to another at 0°C and equilibrated at this temperature for 6 h, followed by another 6 h at 10°C.

(ii) Sample B: A 5 wt% CCF dispersion at 50°C was flash-cooled (in liquid nitrogen) to approximately −150°C and aged for 7 min to allow complete crystallization. According to Garti *et al.* (11), the flash-cooling procedure allows the formation of submicron crystals in the liquid oil phase. The dispersion was then tempered at 0°C for 6 h, followed by 6 h at 10°C. After the tempering cycle, this dispersion was used as a seeding material. Coating fat dispersions (2.5, 5, 7.5, 10, and 12.5

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wt%) were transferred from a water bath at 50°C to another at 10°C. As soon as the samples reached the final temperature, 1 wt% seeding solution was added and gently stirred. The samples were immediately transferred into a water bath at 0°C and then tempered for 6 h, followed by another 6 h at 10°C.

Sample characterization. The SFC of the tempered fat samples was measured in triplicate by pulsed NMR (Minispec mq20; Bruker, The Woodlands, TX). The polymorphic form of coating fat crystals was identified by X-ray diffractometry with a CN2005 Miniflex XRD (Rigaku, Tokyo, Japan; Cu Kα, 30 kV, 10 mA) equipped with a temperature control device. The density of the dispersions was measured by using a vibrating tube densitometer (DE51 Density Meter; Mettler Toledo, Columbus, OH) accurate to ±10 μg cm^{-3}. All measurements were conducted at 10°C.

The dynamic viscosity of the CCF dispersions was measured using a controlled-stress rheometer RS 2000 (Rheometrics, Piscataway, NJ) operating with parallel-plate geometry (20 mm diameter, 1 mm separation). An oscillatory frequency sweep test was also conducted, in which the angular frequency was increased from 0.5 to 12 rad s^{-1} with an applied stress of 1 Pa.

The coating fat samples were dispersed in corn oil, and their size distribution was measured by static light scattering (LA-920; Horiba, Irvine, CA) (12). Tempered dispersions of coating fat (samples A and B) were imaged with a 40× lens using a polarized light microscope (BX40; Olympus, Melville, NY) equipped with a digital video camera (Power HAD DXC-970MD 3CCD; Sony, Tokyo, Japan). Slides were prepared by quickly transferring the tempered samples with a preheated Pasteur pipette onto a preheated glass slide. The slide was maintained at 10°C during the entire experiment on a hot/cold stage. The images were processed using Image Pro Plus software (Media Cybernetics, Carlsbad, CA).

Ultrasonic measurements. A modified pulse-echo technique with a 2.25 MHz broadband transducer (V606; Panametrics, Waltham, MA) was used to measure the velocity, attenuation, and reflection coefficient of all dispersions simultaneously (13,14). In shear mode, only the reflectance coefficient was measured by using a 10-MHz center frequency broadband shear transducer (Panametrics V221BA). Because the magnitude of the generated pulse could not be relied on as a constant on a day-to-day basis, the measured reflectance from both transducers was normalized to a similar measurement made against the calibration material (corn oil), i.e., normalized reflectance. In both cases, the transducers were excited with pulses from a Panametrics 500 PR signal generator, and a digital storage oscilloscope (9310c; LeCroy, Chestnut Ridge, NY) was used to capture the data; the signals were averaged over 200 pulses to reduce signal noise. A complete measurement took approximately 5 s to complete.

RESULTS AND DISCUSSION

Semicrystalline fat dispersions were prepared from the same starting ingredients by two tempering procedures. The density of both these samples increased linearly with the solids content (Fig. 1) and there was no difference in density between the two samples at the same solids level. The unique dependency of density on SFC suggests that the fat crystals were in the same polymorphic form despite their very different thermal histories. X-ray diffraction patterns of the more concentrated (12.5%) dispersions of each preparation (data not reported) confirmed they were of the same polymorphic form, i.e., β_2' (15).

The microstructures of the two dispersions were characterized by light scattering and by polarized light microscopy (Fig. 2). Sample A contained spherical crystalline structures (d = 30–36 μm) typically in 70- to 80-μm clumps. Sample B contained many smaller crystals approaching the resolution of the microscope (d < 1 μm). These crystals were also highly aggregated, and the aggregates were approximately 20–45 μm. Static light scattering of these samples also was used to estimate crystal sizes; typical distributions are shown in Figure 3. Sample A had a bimodal size distribution with a small peak (less than 5% of the total volume fraction) with a median value of 0.3 μm and the remainder of the crystals as a second peak with a median diameter of 36 μm. Sample B had a trimodal distribution with the fine peak (0.5 μm diameter) corresponding to single crystals and the two larger peaks (15 and 53 μm) from the presence of two populations of aggregates. Although light scattering is not an ideal method to estimate the size of nonspherical aggregates, there seemed to be a good agreement with the observations from the microscopy study, with the fine peak probably corresponding to isolated crystals and the coarse to aggregates. Clearly, dispersions as complex as these cannot be easily or meaningfully described in terms of a few length dimensions, but the goal of our sample preparation was to produce two chemically similar lipid dispersions with a range of SFC and widely differing microstructures, and this goal was achieved.

Over the frequency range measurable in longitudinal mode (0.5–3.5 MHz), there was no useful frequency dependence in

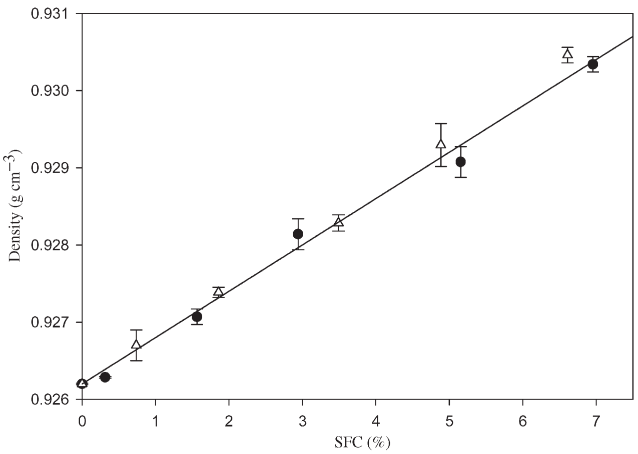

FIG. 1. Density of confectionery coating fat (CCF) in corn oil dispersions at 10°C. Samples A (●) and B (△). SFC, solid fat content. Error bars represent SD.

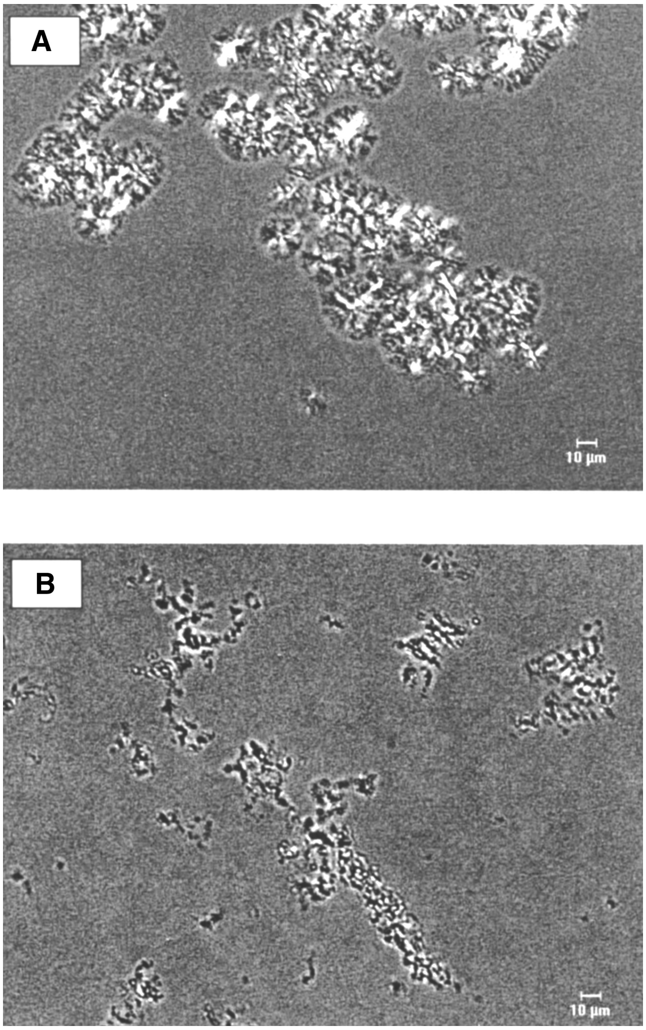

FIG. 2. Polarized light micrographs of CCF dispersion type A (A) and type B (B). Scale bars = 10 µm. For abbreviation see Figure 1.

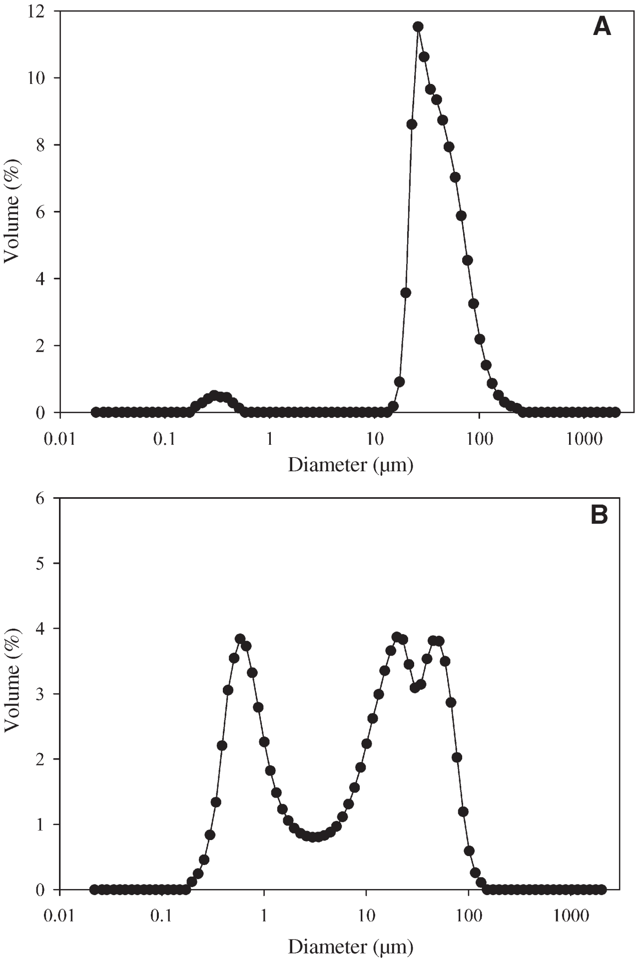

FIG. 3. Particle size distribution of CCF dispersion type A (A) and type B (B) at 10°C. For abbreviation see Figure 1.

the ultrasonic data (not reported); therefore, the group values were used in subsequent calculations. The ultrasonic velocity, attenuation, and reflectance coefficient changed linearly with SFC (Fig. 4). The changes in ultrasonic velocity with SFC were similar to those reported elsewhere (7,8). The percentage change in measured signal with an increase in SFC decreased in the order attenuation > velocity > reflectance. An increase in SFC from 2 to 3% would correspond to an increase of 13 N pm^{-1} in attenuation, to an increase of 4 ms^{-1} in speed of sound, and to a decrease of 0.01 in normalized reflectance, whereas the minimum resolvable differences in attenuation, velocity, and reflectance were approximately 1 N pm^{-1}, 1 ms^{-1}, and 0.001, respectively. There were slight intersample differences in the velocity data; although significant, these were not large. The relative differences persisted in the attenuation data and were larger, but they were not seen in the reflectance measurements. Other workers have seen differences in velocity in more concentrated dispersions not related to changes in SFC (16). Although it is possible that some of these differences were due to polymorphic transitions, it is also possible that differences in the crystal–crystal interactions, which become more significant at higher solids loadings, may have led to some structural sensitivity of ultrasonic velocity in more concentrated systems. Similarly, measurements at different frequencies could reveal structural differences at length/time scales inaccessible to these measurements.

The velocity data were modeled using the well-established Urick equation (7):

$$\frac{1}{c^2} = \sum_{j=1}^{n} \phi_j \rho_j \sum_{j=1}^{n} \phi_j \kappa_j \qquad [1]$$

where c is the ultrasonic velocity in the media and ρ_j, κ_j, and ϕ_j are, respectively, the density, adiabatic compressibility ($\kappa = 1/c^2\rho$), and volume fraction of phase j. In this specific system, two phases were present: Solid fat crystals were dispersed in a continuous phase of liquid corn oil with some liquid CCF. The speed of sound and density of liquid corn oil and solid CCF were measured [CCF velocity 2000 ± 10 m s^{-1} and density 1022 kg m^{-3} (the published density of tristearin was used as representative of a solid fat; see Ref. 7); corn oil velocity 1500 ± 1 m s^{-1} and density 926 kg m^{-3}], allowing the ultrasonic velocity in any mixture to be calculated.

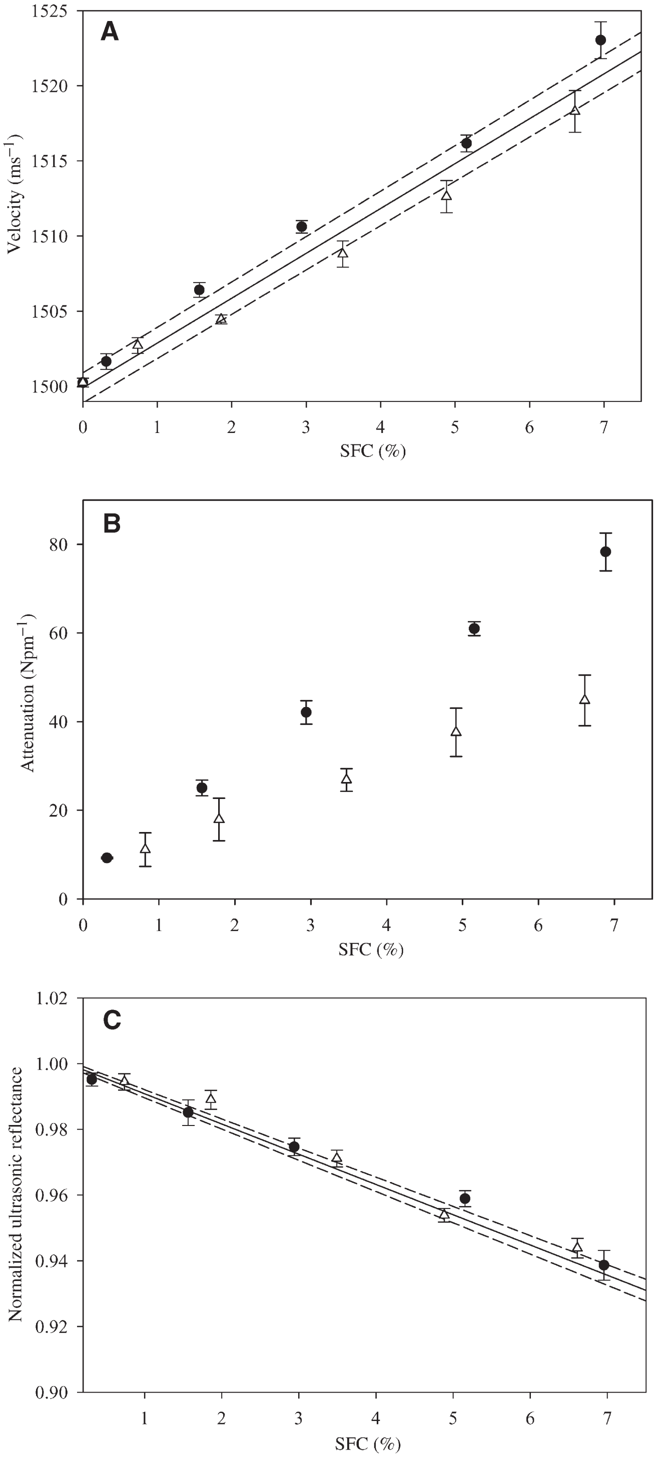

FIG. 4. Ultrasonic velocity (A), attenuation (B), and normalized ultrasonic reflectance (C) of CCF in corn oil dispersions tempered to 10°C for samples A (●) and B (△) as a function of SFC (by NMR). The solid line was generated from the Urick equation using the mean velocity and density values of corn oil and CCF, whereas the dashed lines were calculated using the extreme (±SD) of the constants used. For abbreviations see Figure 1.

The solid lines in Figure 4A correspond to the Urick calculations based on the mean and upper and lower limits (i.e., ±SD) of the physical constants used. The good agreement between the experimental and theoretical data confirms the value of the Urick equation for predicting the SFC in semicrystalline fats. Furthermore, the spread of data in the theoretical calculations encompasses the velocity data from both fat systems. We concluded that, under these conditions, ultrasonic velocity is not usefully sensitive to crystal microstructure.

The Urick equation was modified to model the ultrasonic reflectance data. The reflectance coefficient (*R*) is related to the acoustic properties of the sample through its impedance (*Z*):

$$R = \frac{I_r}{I_i} = \frac{\left(Z_d - Z_s\right)^2}{\left(Z_d + Z_d\right)^2} \qquad [2]$$

where I_r and I_i are the intensities of the reflected and incident sound waves, respectively, and the subscripts *s* and *d* refer to the sample and delay lines. Acoustic impedance is a complex parameter constituting a real and imaginary part ($Z = \rho c + i\alpha$, where $i = \sqrt{-1}$ and α is the attenuation coefficient). The complex impedance was first calculated from the Urick velocity, measured density, and measured attenuation data of all dispersions as follows (17):

$$Z^* = \frac{\omega\rho c}{\left(\omega^2 + c^2\alpha^2\right)} - \frac{i\omega\rho c^2\alpha}{\left(\omega^2 + c^2\alpha^2\right)} \qquad [3]$$

where ω is the angular frequency ($= 2\pi f$) and f is the frequency. The normalized reflectance could then be calculated from Equation 3. Based on the measured data for 12.5 wt% dispersions, the complex acoustic impedance and the complex normalized reflectance were only 0.04 and 0.01% different, respectively, from the real values (at lower concentrations the differences were much smaller). We concluded that the imaginary part of acoustic impedance can be considered negligible and that it is possible to simplify the impedance to $Z = \rho c$. This simplification was used to calculate the normalized reflection coefficient from the Urick equation prediction of velocity and measured sample density (Fig. 4C). Again, three lines are shown representing the mean and upper and lower confidence limits, but here the lines are more closely grouped.

The experimental data fit the Urick prediction well for both velocity and reflectance data, indicating that the surface properties measured by reflectance are representative of the bulk properties measured by velocity. When a wave is reflected at a surface, the depth of the surface layer affected (ξ) is given by

$$\xi = \frac{\lambda_2}{2\pi\sqrt{\left(\frac{\sin^2 \vartheta_1 c_1^2}{c_2^2} - 1\right)}} \qquad [4]$$

where λ_2 is the wavelength in the second medium, ϑ_1 is the angle of incidence, and c_1 and c_2 are the sound velocities of the first and second medium, respectively (17). In our case, the reflectance measurement should be sensitive to the surface up to 80 µm inside the sample, which is of the same order of magnitude as the fat structures observed in microscopy and light scattering; therefore, the reflectance measurements of SFC could reasonably be expected to be sensitive to the bulk.

The rheological properties of all dispersions were investigated by oscillatory viscometry. All dispersions of types A and B were Newtonian over the frequency range investigated (1–12 rad·s^{-1}) with the exception of the most concentrated systems (samples A and B at a concentration of 12.5 wt% and sample A at 10 wt%), which showed a weak frequency dependence and an elastic component at very low frequency that became negligible with increasing frequency. The most dilute samples were indistinguishable from the continuous phase (i.e., corn oil), but for the measurable samples, viscosity increased with the solids content (Fig. 5). Sample B was always more viscous than sample A at a similar solids content (Fig. 5). The viscosity of a dispersion is proportional to the square of the specific surface area of the crystals present (18,19). Small crystals are characterized by a higher specific surface area and are therefore more viscous.

The dynamic viscosity was also measured by shear ultrasonic reflectance. The complex shear acoustic impedance (Z_2) can be calculated as follows:

$$Z_s = R_s + iX_s = \frac{1 - r^2 + 2ir \sin \Delta\theta}{1 + r^2 + 2r \cos \Delta\theta} \qquad [5]$$

where r is the measured normalized reflectance and $\Delta\theta$ is the phase difference between the sample and the calibrating material (20). The shear modulus (G'') and the viscosity (η') can then be calculated as follows (20):

$$G''(\omega) = \frac{2R_sX_s}{\rho} \qquad [6]$$

$$\eta' = \frac{G''}{\omega} \qquad [7]$$

Density, in this case, was measured independently (Fig. 1), but because it is very well correlated with SFC, which in turn is a simple function of ultrasonic velocity or reflectance, it could easily be determined simultaneously with a longitudinal ultrasonic transducer. In fact, G'' was only weakly affected by small changes in density, and good measurements could be made using an average value.

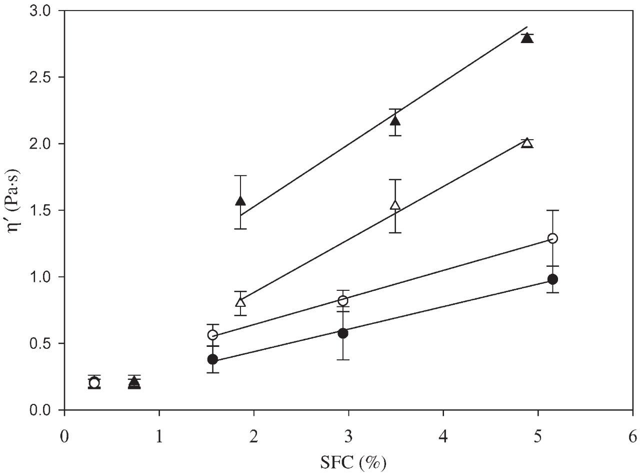

FIG. 5. Dynamic viscosity by ultrasonic shear reflectance (filled symbols) and oscillatory viscometry (open symbols) of sample A (●,○) and sample B (▲,△) dispersions at 10°C vs. SFC by NMR. Regression lines are shown alongside measurable data. Error bars represent SD. For abbreviation see Figure 1.

Similar to the conventional dynamic viscosity measurements, the ultrasonically measured viscosity of the most dilute samples was indistinguishable from the continuous phase (presumably due to a lack of sensitivity of both methods), and the viscosity of the more concentrated samples increased apparently linearly with the solids content (Fig. 5). The lack of measurable viscosity at low levels of solids may be because the techniques were insensitive to the low levels.

The viscosity of sample A was again lower than the viscosity of sample B at a similar solids contents. The dynamic viscosity by ultrasound correlated well with the dynamic viscosity from oscillatory viscometry ($r^2 > 0.99$) for both systems; however, the numerical correlation was different for each. The dynamic viscosity of type A samples measured by ultrasound was slightly lower than when measured by oscillatory rheometry, but the difference was not significant ($P > 0.05$). The dynamic viscosity of type B samples was significantly higher ($P < 0.05$) than with oscillatory rheology but was still of the same order of magnitude. It seems that although the longitudinal ultrasonic reflectance properties of a fat are not dependent on fat microstructure, the shear ultrasonic properties are.

Previously, we used a similar method to measure the dynamic viscosity of sucrose syrups (21). In that work, we also saw a good correlation between ultrasonic and conventional measurements, but the numeric agreement became increasingly poor as the viscosity increased. We hypothesized that at ultrasonic frequencies, the more viscous samples were behaving in a glassy manner and hence had a lower dynamic viscosity. The Maxwell relaxation frequency (f_m) can be calculated as follows:

$$f_m = \frac{G_\infty}{\eta} \qquad [8]$$

where G_∞ is the instantaneous shear elastic modulus (for most liquids ~10^9 Pa) and η the low shear viscosity (21). The fat samples used here were of relatively low viscosity, and we would expect a glass transition at approximately 1000 MHz. At our measurement frequencies (10 MHz), they should behave as Newtonian fluids with a similar dynamic viscosity, as measured using conventional techniques at low frequencies. The ultrasonic data for sample A agreed with this prediction, whereas the viscosity of sample B (small crystals) was significantly higher than that measured by conventional viscometry. This result could be because the surface of the fat suspension interrogated by the ultrasonic waves was not representative of the bulk. Longitudinal wave studies have shown that the surface 80 μm is representative of the bulk, but for shear waves the depth of penetration is only approximately 10 μm and is therefore perhaps not representative of the structures present in sample B.

In summary, ultrasonic velocity and reflectance were strongly dependent on SFC but were not significantly affected by differences in crystal microstructure. The Urick equation provided a good model for the changing velocity/reflectance data. Attenuation showed some sensitivity to sample microstructure and may therefore be a more useful approach to structural characterization. However, at this stage it is premature to speculate as to which structural properties attenuation is sensitive. Although in our work there was limited to spectral information available in the measurable range (0.5–3.5 MHz), it would be interesting to measure more complete attenuation spectra of well structurally characterized, semicrystalline fats.

Shear ultrasonic reflectance can be used to give a useful measurement of the rheological properties of fat crystal dispersions when the viscosity is relatively low (<~20 Pa·s) and therefore when the sample remains fluid at ultrasonic frequencies. It is somewhat surprising that a complex suspension such as a fat dispersion responds to the ultrasonic wave, since we have previously seen that polymers are entirely glassy under these conditions and only indirectly affect the viscosity of the solvent. Perhaps the viscosity perceived by the ultrasonic wave is due to the movement of the liquid oil through a matrix of solid fat crystals, which themselves do not respond to the ultrasonic measurement. Unlike longitudinal measurements, shear ultrasonic reflectance is sensitive to changes in microstructure, possibly because of their limited penetration depth.

A combined shear and longitudinal ultrasonic reflectance sensor could be readily applied on-line to monitor the processing of semicrystalline lipids (e.g., chocolate, shortenings). The longitudinal reflectance could be used to measure the SFC and density of the suspension, whereas the shear wave could measure its rheological properties. The scaling behavior of rheological properties with SFC could be used to measure the fractal properties of fat suspensions (22).

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