5-2005

Effect of Fat and Casein Particles in Milk on the Scattering of Elliptically Polarized Light

Czarena L. Crofcheck
University of Kentucky, crofcheck@uky.edu

Jeremie Wade
University of Kentucky

Janakiraman N. Swamy
University of Kentucky, jswamy@cmrhoutex.com

Mustafa M. Aslan
University of Kentucky

M. Pinar Mengüç
University of Kentucky, menguc@engr.uky.edu

Repository Citation
Crofcheck, Czarena L.; Wade, Jeremie; Swamy, Janakiraman N.; Aslan, Mustafa M.; and Mengüç, M. Pinar, "Effect of Fat and Casein Particles in Milk on the Scattering of Elliptically Polarized Light" (2005). *Biosystems and Agricultural Engineering Faculty Publications*. 33.
https://uknowledge.uky.edu/bae_facpub/33

Effect of Fat and Casein Particles in Milk on the Scattering of Elliptically Polarized Light

Notes/Citation Information
Published in *Transactions of the ASAE*, v. 48, issue 3, p. 1147-1155.

Digital Object Identifier (DOI)
https://doi.org/10.13031/2013.18488

EFFECT OF FAT AND CASEIN PARTICLES IN MILK ON THE SCATTERING OF ELLIPTICALLY POLARIZED LIGHT

C. Crofcheck, J. Wade, J. N. Swamy, M. M. Aslan, M. P. Mengüç

ABSTRACT. *In this article, we present an experimental approach to determine the milk fat content using scattered light intensity profiles. The elements of the scattering (Mueller) matrix have been shown to provide valuable information about variation of the optical properties of scattering particles. The scattering behavior of fat and casein in terms of the scattering matrix elements was experimentally determined for milk with varying fat levels ranging from 0.05 wt% (skim) to 3.20 wt% (whole). Three of the scattering Mueller matrix elements, specifically* S_{11}, S_{12}/S_{11}, *and* S_{33}/S_{11}, *were found to be sensitive to the number of fat particles in milk. These results indicate that it should be possible to develop a reliable sensor based on the measurement of these scattering elements, which will allow for the development of a robust, in-line sensor to be used in food processing. In addition, an attempt was made to model the phenomena using a relatively simple approach based on single scattering with a size distribution. The disagreement between the model and experiments suggests that a more comprehensive model is needed which can account for multiple scattering.*

Keywords. *Depolarization, Fat content, Light scattering, Mueller matrix.*

An essential requirement for the automation of processes in the food industry is the continuous evaluation of the composition and physical properties of liquid mixtures during crucial processing steps. However, the physical characteristics of many food products are inherently difficult to measure using traditional process monitoring schemes, due to the food material being sticky, highly viscous, or containing particles of varying sizes and shapes. Therefore, the need exists for the development of robust, versatile, in-line sensors capable of rapid and continuous monitoring of these food materials. Optical measurement techniques are attractive for this purpose, as they are clean, effective, and can be designed to function non-intrusively.

There is significant motivation to investigate new methods for measuring fat content, in an attempt to decrease milk processing cost and to improve control. Milk fat concentration can currently be measured by existing commercial systems at a cost of as much as $250,000 per measurement system, for an infrared transmission/reflectance (IFIR) based system. Devices that utilize near-infrared transmission technology to measure and control milk fat content can range in cost from $20,000 to $60,000, while devices that utilize full transmission NIR wavelengths for milk standardization can range from $60,000 to $100,000 per measurement system. In addition, milk fat measurement in the dairy industry requires considerable precision because of the economic loss associated with producing milk products with too high fat content. Generally speaking, the milk fat content measurements must have an accuracy of approximately 0.02%. While existing technologies exist for measuring fat content, an easy to calibrate, compact, and less expensive system would have great advantages, especially if the level of accuracy could be increased as well (personal communication with Tony Suda, ESE, Inc., Marshfield, Wisc.).

Fiber optic sensors have been shown to provide reliable and inexpensive methods for measuring the diffuse reflectance of multiple-scattering fluids (Meeten and Wood, 1993). Such sensors have previously been developed for monitoring changes in backscatter (diffuse reflectance) during enzymatic coagulation of milk (Payne, 1995; Payne et al., 1993) and the culture of cottage cheese (Crofcheck et al., 1999; Payne et al., 1997). In addition, similar sensors may be used for transition sensing of dairy products, when product lines are switched from water to milk (Payne et al., 1999) and for determining the milkfat content in skim milk (Crofcheck et al., 2000).

These previous studies have focused on light scattering without taking the changes in the polarization of scattered light into account. A technique that also encompasses changes in polarization may provide additional information about the scattering particles and/or increase the accuracy of measurement. By defining the relation between the properties of the incident and scattered intensities (i.e., measurable quantities) in terms of physical characteristics of the scatterer, one can arrive at these characteristics using a rigorous inverse analysis (Aslan et al., 2003a; Aslan et al., 2003b; Manickavasagam and Mengüç, 1997; Manickavasagam et al., 2002; Mengüç and Manickavasagam, 1998). The work described in this article is a preliminary step prior to developing monitoring systems based on elliptically polarized light scattering to distinguish fat and casein concentrations in dilute milk samples.

Milk is a complex biological fluid composed of water, fat, protein, lactose, citric acid, and inorganic compounds (Walstra

---

Article was submitted for review in November 2004; approved for publication by the Food & Process Engineering Institute Division of ASAE in May 2005. Presented at the 2003 ASAE Annual Meeting as Paper No. 036016.

The authors are **Czarena Crofcheck, ASAE Member Engineer,** Assistant Professor, **Jeremie Wade,** Senior Engineering Technician, and **Janakiraman N. Swamy, ASAE Member Engineer,** Graduate Student, Department of Biosystems and Agricultural Engineering, University of Kentucky, Lexington, Kentucky; **Mustafa M. Aslan,** Assistant Research Professor, and **M. Pinar Mengüç,** Professor, Department of Mechanical Engineering, University of Kentucky, Lexington, Kentucky. **Corresponding author:** Czarena Crofcheck, 212 CE Barnhart Building, Lexington, KY 40546-0276; phone: 859-257-3000, ext. 212; fax: 859-257-5671; e-mail: crofcheck@uky.edu.

Transactions of the ASAE

Vol. 48(3): 1147−1155

and Jenness, 1984). Light scattering by fat globules and casein micelles causes milk to appear turbid and opaque. These two components scatter light differently based on differences in size, number, and optical properties (e.g., index of refraction) of the particles. The average particle diameter of casein micelles falls in the range of 0.13 to 0.16 μm (Ruettiman and Ladisch, 1987), and milkfat in the form of globules falls in the range of 0.1 to 10 μm for unhomogenized milk, with a mean diameter of 3.4 μm ($\bar{d}_{32}$) (Mulder and Walstra, 1974). The mean diameter reported for particles in skim milk is around 0.210 to 0.225 μm (Attaie and Ritcher, 2000). Skim milk appears slightly blue because the small casein micelles predominately scatter the shorter (blue) wavelengths of visible light. On the other hand, whole milk appears white because the larger fat globules multiple-scatter all wavelengths of incident light.

The polarization of a beam of light can be described in terms of the Stokes parameters. The differences in the intensity and polarization of light between the incident (before encountering the particle, $[\boldsymbol{I}]_{inc}$) and scattered (after encountering the particle, $[\boldsymbol{I}]_{sca}$) light can be characterized by a 4 × 4 property matrix, known as the scattering or Mueller matrix, $[\boldsymbol{S}(\theta)]$, (Bohren and Huffman, 1983) using the following equation:

$$[\boldsymbol{I}]_{sca} = \frac{1}{k^2 r^2}[\boldsymbol{S}(\theta)][\boldsymbol{I}]_{inc} \qquad (1)$$

where $k = 2\pi/\lambda$, and r is the distance between the light source and the detector. This property matrix can be reduced from a 16-element matrix (denoted as S_{ij} values) down to a 6-element matrix (S_{11}, S_{12}, S_{22}, S_{33}, S_{34}, and S_{44}) after considering all averaging effects (Govindan et al., 1996; Mengüç and Manickavasagam, 1998):

$$[\boldsymbol{S}(\theta)] = \begin{bmatrix} S_{11}(\theta) & S_{12}(\theta) & 0 & 0 \\ S_{12}(\theta) & S_{22}(\theta) & 0 & 0 \\ 0 & 0 & S_{33}(\theta) & S_{34}(\theta) \\ 0 & 0 & -S_{34}(\theta) & S_{44}(\theta) \end{bmatrix} \qquad (2)$$

The scattering matrix elements (S_{11}, S_{12}, S_{22}, S_{33}, S_{34}, and S_{44}) can be calculated from the scattering amplitudes that relate two perpendicular components of incident electromagnetic (EM) wave with two perpendicular components of scattered EM wave using the equations in Bohren and Huffman (1983). Similar methodologies have been used to characterize colloidal metallic particles (Aslan et al., 2003b) and coal particles (Mengüç and Manickavasagam, 1998).

In this article, the normalized scattering matrix elements (M_{ij}) were calculated, which are defined as:

$$[\boldsymbol{M}] = \begin{bmatrix} M_{11} & M_{12} & 0 & 0 \\ M_{12} & M_{22} & 0 & 0 \\ 0 & 0 & M_{33} & M_{34} \\ 0 & 0 & -M_{34} & M_{44} \end{bmatrix}$$

$$= \begin{bmatrix} \frac{S_{11}}{S_{11}^{o}} & \frac{S_{12}}{S_{11}} & 0 & 0 \\ \frac{S_{12}}{S_{11}} & \frac{S_{22}}{S_{11}} & 0 & 0 \\ 0 & 0 & \frac{S_{33}}{S_{11}} & \frac{S_{34}}{S_{11}} \\ 0 & 0 & \frac{-S_{34}}{S_{11}} & \frac{S_{44}}{S_{11}} \end{bmatrix} \qquad (3)$$

where S_{11}^{o} is the value of S_{11} at the first scattering angle measured, and the remaining S_{ij} values are all measured as a function of scattering angle, e.g., $M_{22}(85^\circ) = S_{22}(85^\circ)/S_{11}(85^\circ)$.

Taken in totality, the scattering elements reflect the effect of the medium on the scattering of light. Taken individually, the scattering elements can indicate specifics about the properties of the medium. The differential scattering cross-section (S_{11}) is a measure of how an incident beam of light is scattered into different directions. S_{12} represents the depolarization due to the scattering by the medium and the scattering particles, which depends on the size, geometry, and optical properties. S_{34} is related to the fraction of the elliptically polarized light transformed to circularly polarized light. When the particles are perfectly spherical, S_{22} is equal to S_{11}, and any deviation from a spherical shape is reflected by the ratio S_{22}/S_{11}: the greater the deviation of the ratio from 1, the greater the deviation of the particles from a spherical shape. S_{33} is a measure of how much of the light polarized at +45° is retained through the medium. Each one of these S_{ij} parameters can be related to milk properties, such as particle size, shape, and number of the fat globules and casein micelles. Typically, the S_{22} and S_{33} elements are sensitive to the particle aspect ratio, while the other elements are sensitive to particle size and distribution (Yang et al., 2003). A sensitivity analysis of each of these S_{ij} parameters may find correlations with the properties of interest.

The overall objective of our current research is to develop a light-based in-line sensor that can monitor the particulate concentrations, for example fat and protein, in milk. The specific objectives were to:

- Determine and evaluate several possible correlations between the experimentally determined scattering matrix elements and milk fat level.
- Determine the sensitivity of the scattering matrix elements to other parameters, such as particle size distribution, particle index of refraction, and the presence of casein particles based on single-scattering model predictions.
- Determine which matrix elements and scattering angles should be investigated further with a prototype in-line sensor.

RESEARCH METHODS

EXPERIMENTAL SETUP

Experiments were carried out with the setup shown in figure 1. The details of the system can be found in the literature (Aslan et al., 2003b). Optical components included two polarizers (P1 and P2) and two retarders (R1 and R2) that were used to modulate incident and scattered light so that the polarized light scattered by the diluted milk sample could be measured. The system was calibrated and validated using latex particles of well-known size and concentration (Aslan et al., 2003b). Polarization angle and type of incident light was modulated by retarder R1, and the first polarizer (P1) was fixed at 45° in the incident beam path. Scattered light from the sample was filtered by retarder R2 and polarizer P2. A 20 mW helium neon laser (λ = 632 nm) was employed as the light source. This wavelength is compatible with the particle sizes expected in milk and would also be cost-effective for future sensor development.

Scattered light that passed through R2 and P2 was detected by a photomultiplier tube (PMT; Hamamatsu R446) as a function of the scattering angle (θ). Since the incident light was plane polarized at +45°, the Stokes vector for scattered light that carried both the intensity and the polarization information in normalized form can be written as (Aslan et al., 2003b):

$$\begin{aligned}&\left[\boldsymbol{I}^{out}(\theta,\alpha,\beta_1,\beta_2)\right]\\&\quad=\left[\boldsymbol{M}^{sys}(\theta,\alpha,\beta_1,\beta_2)\right]\left[\boldsymbol{I}^{in}\right]\\&\quad=\left[\boldsymbol{M}_{\mathrm{P2}}(\alpha)\right]\left[\boldsymbol{M}_{\mathrm{R2}}(\beta_1)\right]\left[\boldsymbol{S}(\theta)\right]\left[\boldsymbol{M}_{\mathrm{R1}}(\beta_2)\right]\left[\boldsymbol{I}_{\mathbf{0}}\right]\end{aligned}\qquad(4)$$

where $[\boldsymbol{M}_{\mathrm{R1}}]$, $[\boldsymbol{M}_{\mathrm{R2}}]$, and $[\boldsymbol{M}_{\mathrm{P2}}]$ make up the Mueller matrix of retarder R1, retarder R2, and polarizer P2, oriented at angles β_2, β_1, and α, respectively. The scattering (Mueller) matrix, $[\boldsymbol{S}(\theta)]$, of the medium (diluted milk) is given by equation 3 for an isotropic and symmetric medium. The intensities are measured at a given scattering angle (θ) for six different combinations of α, β_1, and β_2. The scattering matrix elements (S_{ij}) are then calculated by solving six equations for six unknowns. The components of the Stokes vector of light at the sensor, derived for our optical system, are (Aslan et al., 2003b):

$$\begin{aligned}\left[\boldsymbol{I}_i^{out}(\theta,\alpha,\beta_1,\beta_2)\right] &= \left[\boldsymbol{M}_{i2}^{sys}(\theta,\alpha,\beta_1,\beta_2)\right]\\&\quad+\left[\boldsymbol{M}_{i4}^{sys}(\theta,\alpha,\beta_1,\beta_2)\right],\quad i=1,2,3,\text{ and }4\end{aligned}\qquad(5)$$

Experimental Procedure

Three separate batches of milk were tested (sorted by expiration date) for all factors, in triplicate for each batch. Skim and whole milk were purchased from a local grocery store (Kroger Co.) and stored at 4°C until tested (within four days of the purchase date). The two milks were mixed in appropriate portions to obtain different fat levels (1.15 and 2.2 wt%). The fat and protein contents of the milk samples were determined by the University of Kentucky Regulatory Services using a Milkoscan FT 120 (Foss Electric, Denmark) in order to have consistent calibration.

Labview 6i (National Instruments, Austin, Texas) was used to control light input and record scattering data from the experimental system. All experiments were performed in a no-light environment. The data were recorded and plotted versus the scattering angle.

Preliminary tests were completed with skim and whole milk samples at multiple volume fractions. The purpose of these tests was to find the optimum volume fraction to use for the future experiments and sensor development. Data were recorded and plotted versus the scattering angle, 25° < θ < 145°. For these preliminary tests, the samples were diluted in 100 mL of skim milk ultrafiltrate (SMUF; Jenness and Koop, 1962).

Based on the preliminary results, a modified scattering angle range 75° < θ < 125° and a milk volume fraction of 3.0×10^{-3} was used for the correlation experiments. To prepare each sample, 300 µL of milk of various fat levels was diluted with deionized water to a final volume of 100 mL. The diluted milk solution was then poured into the sample

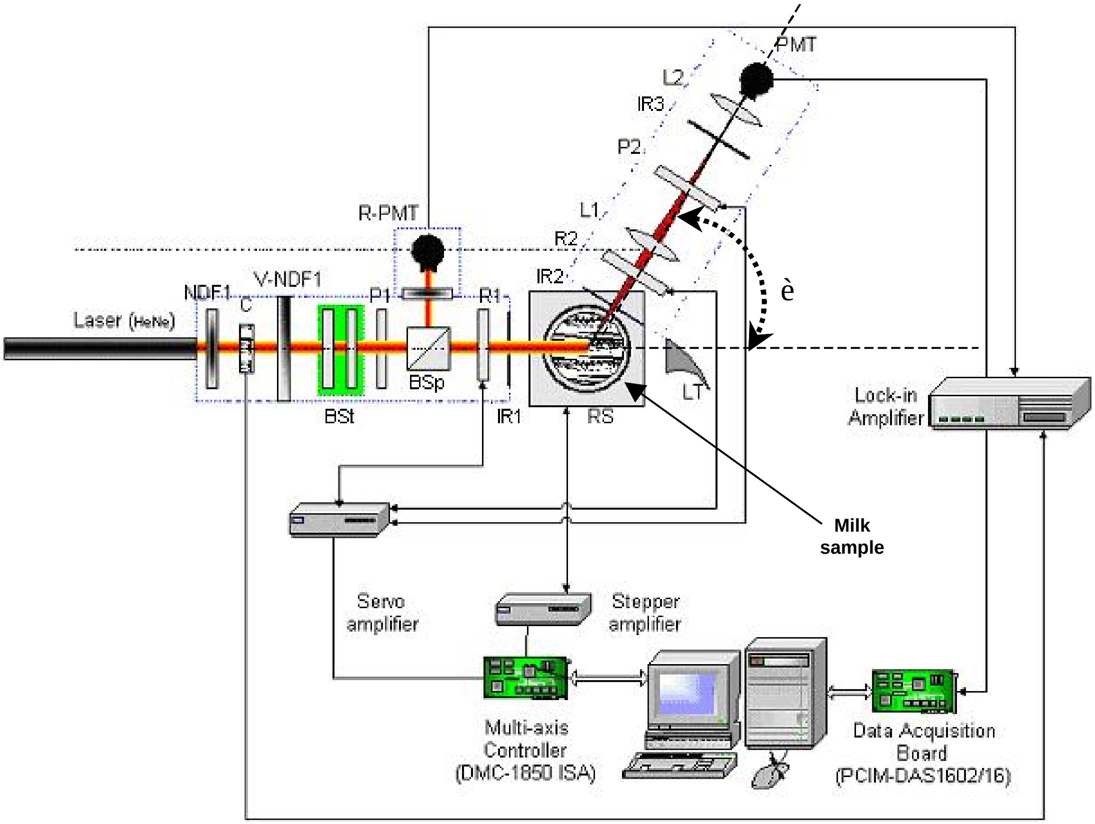

Figure 1. Experimental setup to measure elliptically polarized light scattered by fat and casein particles in diluted milk (Aslan et al., 2003b); C = collimator; NDF1 and V-NDF1 = filters; P1 and P2 = polarizers; BSp = beam splitter; R1 and R2 = retarders; IR1, IR2, and IR3 = iris; L1 and L2 = lenses; RS = rotation stage; LT = beam dump; PMT = photomultiplier tube; R-PMT = reference PMT; and θ = scattering angle.

cup, followed by lowering the light trap into the solution to trap the laser after passing through the cup. The laser power, input and scattering, were then fine-tuned by adjusting NDF1, V-NDF1, R-PMT, and the lock-in amplifier in order to ensure signal stability. A table of six sets of angles was created for the two retarders (R1 and R2) and the first polarizer (P1). The program was used to acquire the light input and scattering data over the range of scattering angles for all six sets of angular inputs. The resulting M_{ij} values were then calculated.

MODEL PREDICTIONS

Changes in the concentration of the casein and fat globules can be detected in milk only if we have a comprehensive predictive capability. Thus, a model could be helpful to predict how fat and casein particles scatter the incident light and how the polarization changes as a function of particle properties. Such a computational model requires accurate prediction of elliptically polarized light scattering by particles in single or multiple scattering media. Our milk samples were diluted in order to ensure that the final solution was optically thin with an optical thickness of less than 0.5 (Agarwal and Mengüç, 1991); hence, contributions due to multiple scattering effects were not considered here.

Modeling of light scattering by the individual fat and casein particles was performed using the Lorenz-Mie theory (Bohren and Huffman, 1983). The sensitivity of the M_{ij} values to the size distribution, index of refraction, and the presence of casein micelles was investigated.

The sensitivity of the scattering elements to changes in size distribution was tested by varying the effective diameter model input. Distributions encountered in particle characterization are notoriously non-Gaussian; however, a log-normal distribution often provides an improved fit to the observations. Log-normal distributions have proven useful for the size distributions of all kinds of small particles, aerosols, and many other cases. The general form for a log-normal distribution is given by (Hansen and Travis, 1974):

$$F(d) = \sqrt{\frac{2}{\pi}}\,\frac{fv}{\sigma_g}\,\frac{1}{d}\exp\left(-\frac{\left[\ln\left(\frac{d}{2}\right)-\ln R_g\right]^2}{2\,\sigma_g^2}\right) \tag{6}$$

where fv is particle volume fraction, σ_g is variance distribution, and R_g is the characteristic dimension. They can be related with effective diameter (d_{eff}) and effective variance (υ_{eff}) as:

$$d_{eff} = 2R_g\exp\left(\frac{5\sigma_g^2}{2}\right) \tag{7}$$

$$\upsilon_{eff} = \exp\left(\sigma_g^2\right) - 1 \tag{8}$$

A base-case size distribution was developed by fitting a log-normal distribution to a size distribution found in the literature and input into the model as an effective diameter. The size distribution for the fat globules was taken from Walstra (1975), while the size distribution for casein micelles was taken from Schmidt et al. (1977). The amount of casein in the samples was assumed to be 80% of the total protein content (Walstra and Jenness, 1984). The measured wt% of fat and protein in the milk samples was converted to a volume fraction, assuming densities of 1.11, 0.92, and 1.03 g mL^{-1} for protein, fat, and milk, respectively (Walstra and Jenness, 1984). The resulting effective diameter was 0.125 μm for the base case. Four additional effective diameters tested in the model were 0.075, 0.1125, 0.1375, and 0.175 μm. For the 0.075 and 0.175 mm distributions, it was assumed that the distribution varied such that the distribution curve narrowed or widened by 10%. For the 0.1125 and 0.1375 μm distributions, a shift in the average of the distribution was assumed; specifically, the effective diameter was shifted by 0.0125 μm. The minimum diameter was zero for all cases, while the maximum diameter was 1.625 μm for the base case and 1.575, 1.463, 1.788, and 1.675 μm, respectively, for the other distributions.

Using the base-case size distribution, two different casein indices of refraction were considered. The first, 1.5713, was measured by Griffin and Griffin (1985), while the second, 1.4682, was assumed in previous work (Crofcheck, 2001), where 1.4682 is the measured index of refraction of fat. Measurements of the refractive index of milk indicate that the imaginary part of the index of refraction, which is related to the amount of absorption that can be expected from the material, is negligible (Jaaskelainen et al., 2001). Hence, the effect of absorption on the Mueller matrix elements has been assumed negligible in our study. Finally, the casein micelles were left out of the model simulation in order to determine the effect of scattering due to the fat alone.

RESULTS AND DISCUSSION

EXPERIMENTAL RESULTS

The relationships between varying fat level (0.05, 1.15, 2.20, and 3.20 wt% before dilution) for the six independent normalized matrix elements (M_{11}, M_{12}, M_{22}, M_{33}, M_{34}, and M_{44}) as a function of scattering angle (75°, 85°, 95°, 105°, 115°, and 125°) are shown in figure 2. In our experiments, forward scattering angles are 0° to 70°, side-scattering angles are 70° to 110°, and backscattering angles are 110° to 180°. Each data point represents the average over the three batches, where each batch was tested in triplicate, for a total of nine observations. The error bars, based on standard error, associated with the M_{22} and M_{34} plots are the most prominent, while the greatest statistical difference between the batches was found with the M_{12} and M_{44} data. In general, the repeatability for all six M_{ij} elements was deemed acceptable.

These results show clearly that the M_{ij} elements are quite sensitive to fat concentration. The results also reflect that these elements show a definite trend with respect to the scattering angle. Such variation can be used to select specific angles for optimized measurement sensitivity. For example, M_{11} values, indicative of the scattering properties, show better sensitivity to fat concentration at backscattering angles (115° and 125°), while M_{12} values show sensitivity over a fairly large range except the backscattering angles. The sensitivity of other M_{ij} elements to fat concentration is fairly independent of the scattering angle. Thus, by measuring the M_{ij} values at predetermined angles, definite correlations can

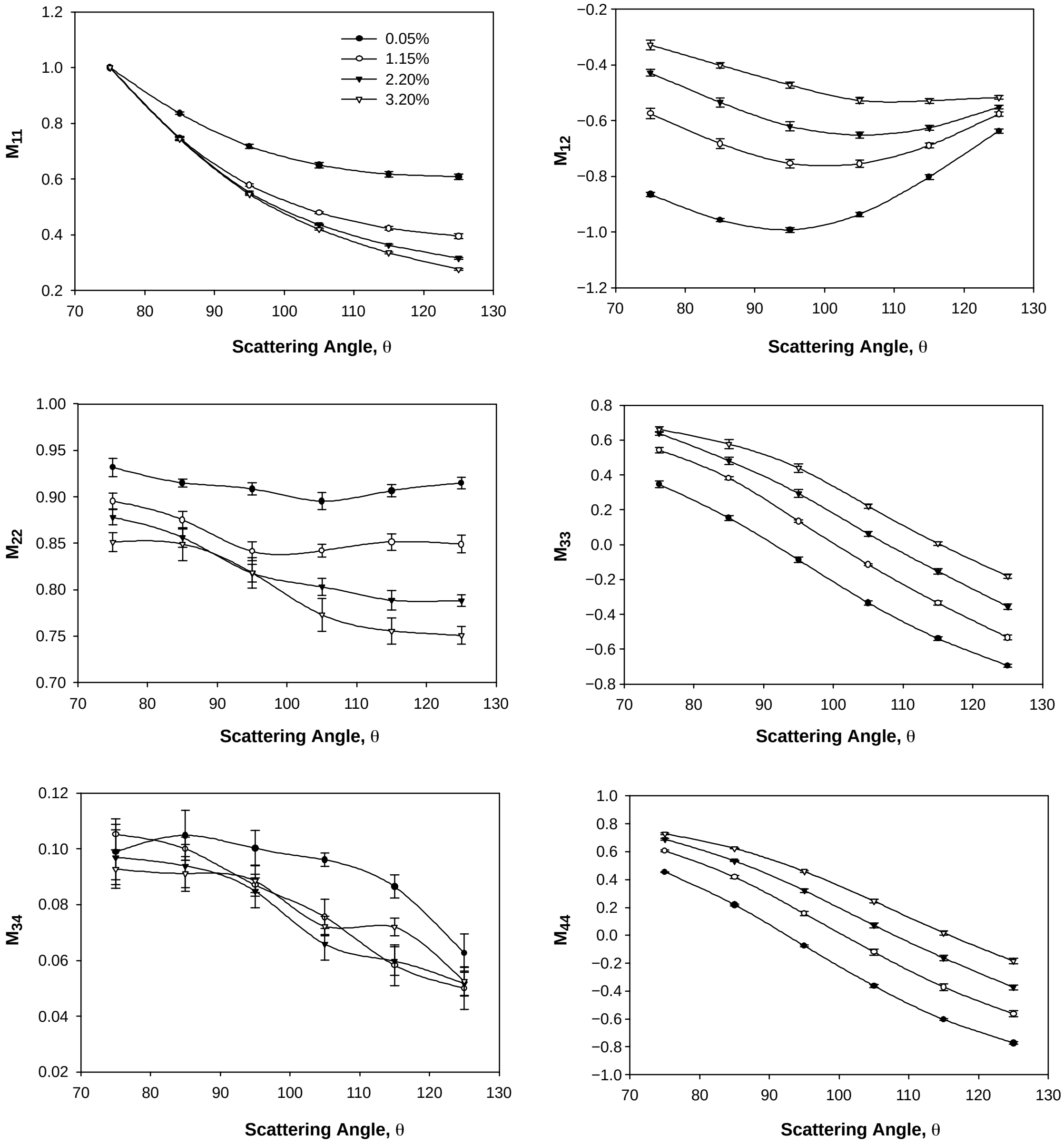

Figure 2. Scattering elements as a function of scattering angle for four different fat levels (averaged over three batches and tested in triplicate for each batch). Error bars represent standard error (nine observations).

be obtained between these M_{ij} profiles and fat concentrations. The deviation of M_{22} from 1 indicates that the fat globules are not spherical in shape. The variation in data for a given fat concentration is reflective of the arbitrariness and non-uniformity of shape. Further, a drop in M_{22} values at higher fat concentrations is thought to be a result of multiple scattering effects. The agglomeration of fat globules into more random shapes at higher fat levels may also be a factor.

The variations of M_{ij} elements with respect to fat level were tested at specific scattering angles. The resulting curves for fat level based on the M_{11}, M_{12}, and M_{33} values at four different scattering angles are depicted in figures 3 through 5. The forward scattering (<90°) results showed little dependence on fat level and have been omitted from the figures. Each data point represents the average for a single batch (n = 3). The other M_{ij} profiles also show some sensitivity to the change in fat level; however, they are not as encouraging as the M_{11}, M_{12}, and M_{33} profiles. A logarithmic curve was used to fit the data for the M_{11} correlation (R^2 values from 0.977 to 0.990), and linear fits were used for M_{12} (R^2 values of 0.937 to 0.967) and M_{33} (R^2 values of 0.970 to 0.990). The dynamic range and linearity of the M_{12} and M_{33} plots indicate that these elements might prove useful in terms of sensor development. The results clearly indicate better sensitivity of M_{11} at backscattering angles. The M_{33} plot shows no variation in slope for different scattering angles. For the M_{12} plot, slope increases with decreasing scattering angle.

In figure 2, the M_{12} curve for each fat level has an inflection point at a different scattering angle. Based on this observation, the correlation between the scattering angle at which M_{12} has an inflection point and the fat level, as shown in figure 6, was investigated. The dynamic range and linearity of the curve (R^2 = 0.9447) are very promising for accurate and repeatable measurements. With this curve, the sensor would need to take measurements for a range of

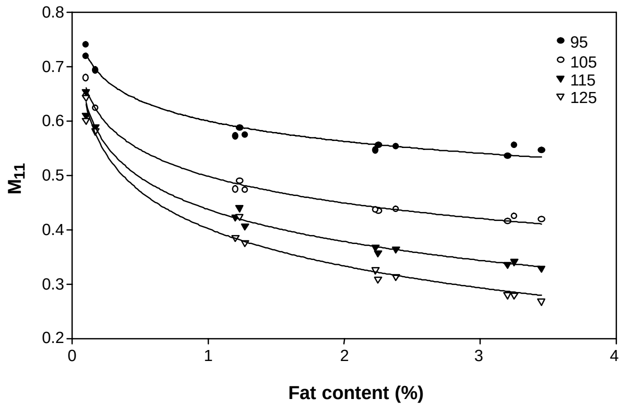

Figure 3. Logarithmic calibration curves relating the scattering element M_{11} and fat level (wt%) at four scattering angles (95°, 105°, 115°, and 125°). Each point represents three tests with the same milk batch ($n = 3$).

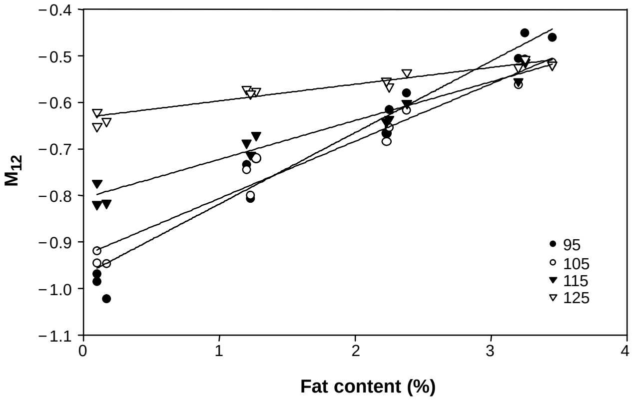

Figure 4. Linear calibration curves relating the scattering element M_{12} and fat level (wt%) at four scattering angles (95°, 105°, 115°, and 125°). Each point represents three tests with the same milk batch ($n = 3$).

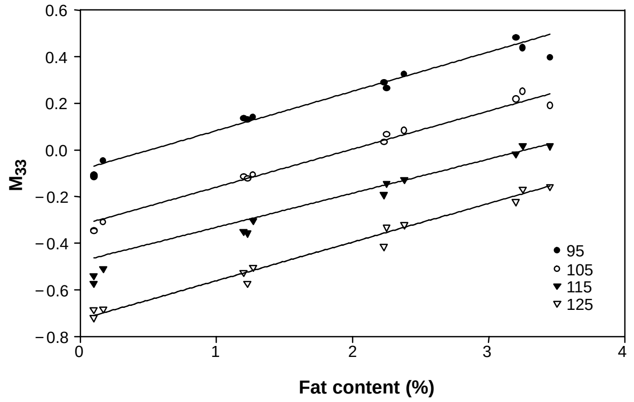

Figure 5. Linear calibration curves relating the scattering element M_{33} and fat level (wt%) at four scattering angles (95°, 105°, 115°, and 125°). Each point represents three tests with the same milk batch ($n = 3$).

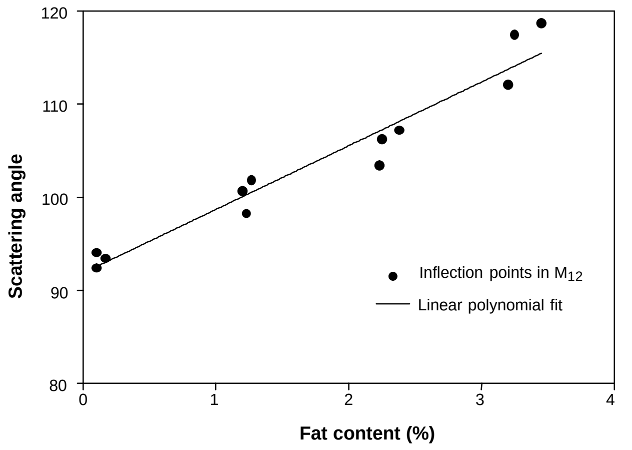

Figure 6. Linear calibration curve relating the scattering angle at which the M_{12} curve has an inflection point for the various fat levels (wt%). Each point represents three tests with the same milk batch ($n = 3$).

scattering angles, but it could prove to be more robust and sensitive.

MODEL PREDICTIONS

The experimental results suggest that it is possible to use the M_{ij} parameters obtained at discrete scattering angles to detect the variations in the fat level in diluted milk samples. It is, however, important to test the reliability of these predictions, especially if there are variations in the structural and optical properties of samples considered. To have a more comprehensive and reliable predictive power, we examined the sensitivity of the M_{ij} parameters to size distribution of fat particles, casein index of refraction, and the presence of casein particles with a series of model simulations. The model simulations for M_{11}, M_{12}, and M_{33} for whole milk, assuming five different size distributions, are shown in figure 7. The trends were identical for the skim milk simulations; hence, skim milk results are not shown. The trends are quite similar to the experimental results, although the values are dramatically different. The differences can be attributed to the structural and optical properties of the sample, which could not be controlled or physically measured during experimentation. Further, the model is based on the Lorenz-Mie theory, which assumes the scattering particles to be spherical, and that may be an over-simplification. Finally, and possibly most importantly, the model should be expanded to include multiple scattering before direct comparisons between the experiments and model predictions can be made, a formidable task. However, the simple model utilized here shows the same trends as the experiments and can be used for a careful analysis of the sensitivity of single-scattering model M_{ij} elements to sample properties, This sensitivity analysis will prove useful as a tool to further identify which elements are worthy of further investigation for sensor development.

The dramatic differences in the model predictions illustrate that these scattering matrix elements are sensitive to size distribution profiles. Changes in the average of the log-normal distribution (0.1125 and 0.1375 μm) appear to be less dramatic than changes due to the variance of the log-normal distribution (0.075 and 0.175 μm). Interestingly, for the positive offset distribution results (larger particles), the M_{ij} predictions are quite different from the remaining predictions. This illustrates that the presence of larger particles will have a substantial influence on the M_{ij} values.

M_{ij} Sensitivity to Casein Particles

The model simulations for M_{11}, M_{12}, and M_{33} for both skim and whole milk, considering the effect of both fat and casein particles and compared to fat particles alone, are shown in figure 8. The effect of casein micelles on M_{11} is negligible, while the effects are dramatic for M_{12} and M_{33}. Hence, M_{11} may prove useful for determining the milk fat content of milk independent of casein content. For the case

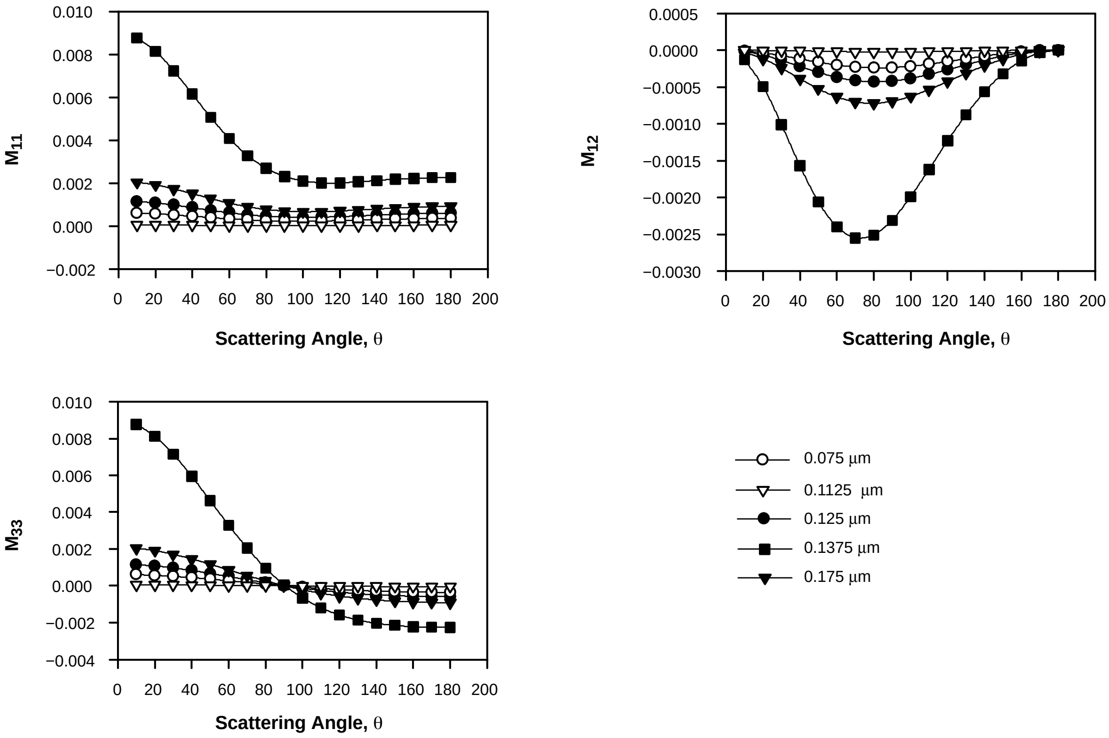

Figure 7. Model-predicted scattering elements (M_{11}, M_{12}, and M_{33}) as a function of scattering angle for whole milk, assuming five different size distributions.

of M_{12}, the differences are prominent at side-scattering angles (90°) and may prove useful for determining the fat and protein content simultaneously. On the other hand, the M_{33} values at 90° are fairly consistent for both fat levels with and without casein. A measurement at this point could prove useful for comparison to other measurements.

M_{ij} Sensitivity to Index of Refraction

The model simulations for M_{11}, M_{12}, and M_{33} for both skim and whole milk considering two different casein indices of refraction (1.462 and 1.5713) are shown in figure 9. The M_{11} plot shows a shift in values, although the trend is consistent for the two values of refractive index considered.

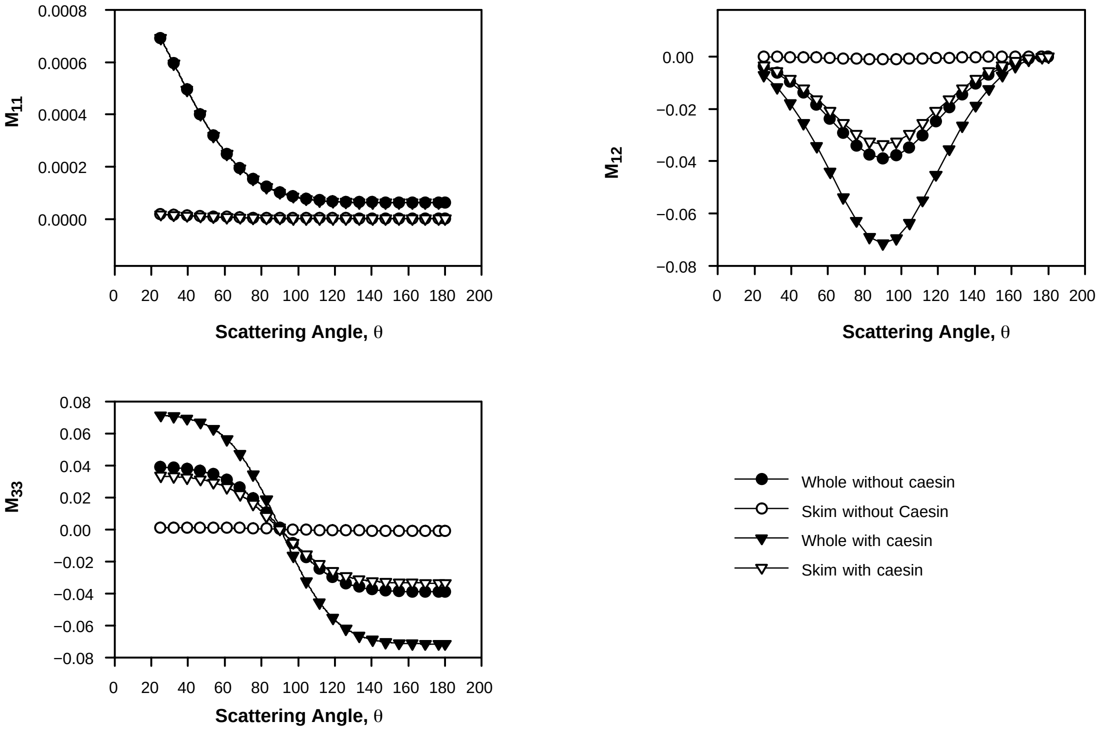

Figure 8. Model-predicted scattering elements (M_{11}, M_{12}, and M_{33}) as a function of scattering angle for whole and skim milk with and without casein particles.

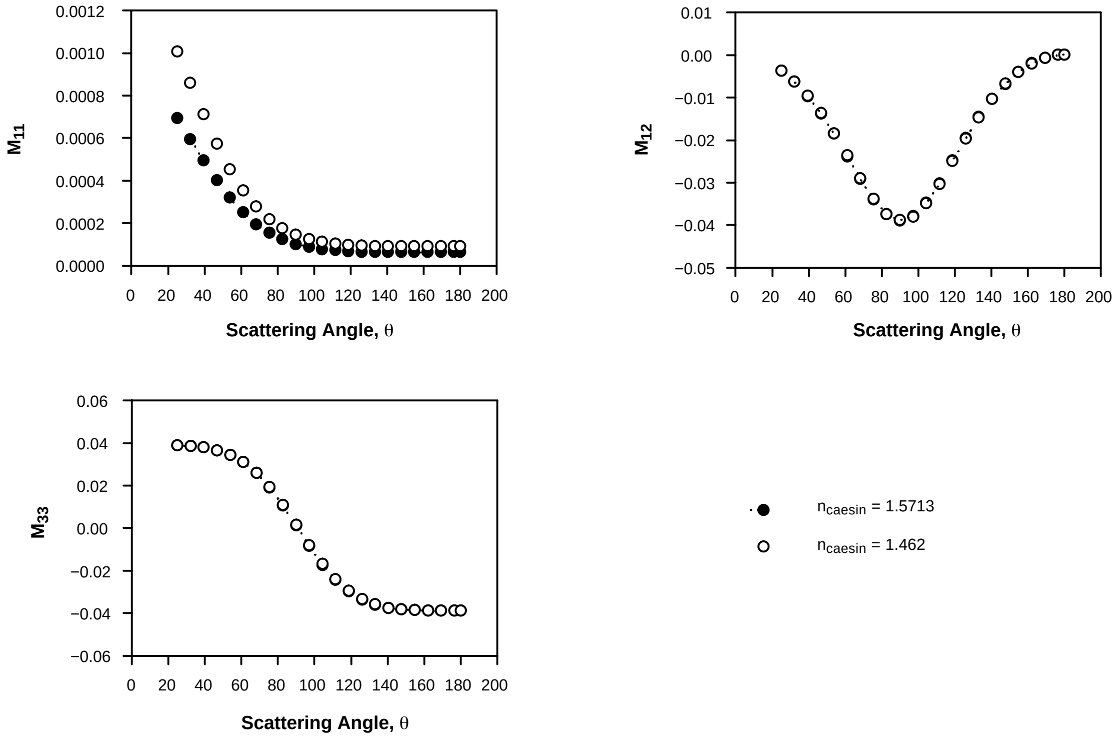

Figure 9. Model-predicted scattering elements (M_{11}, M_{12}, and M_{33}) as a function of scattering angle for whole milk with two indices of refraction for casein.

Computed M_{12} and M_{33} values are insensitive to the change in refractive index. This may prove desirable, because fluctuations in process conditions can lead to small changes in refractive index of the biomaterials. Further, the refractive indices of biomaterials like casein can only be measured with reasonable accuracy, as they are difficult to isolate, and their properties change with conditions. Thus, these model predictions support further investigation of M_{12} and M_{33} for sensor development.

CONCLUSION

A series of experiments were carried out to characterize milk samples using the profiles of the scattered elliptically polarized light. Experimental results indicate that the fat level in milk has an influence on the scattering matrix values. There is a strong correlation between fat level and the values of M_{11}, M_{12}, and M_{33}. The linear correlation and the dynamic range associated with the M_{12} and M_{33} values make these elements candidates for further sensor development. However, the calibration curve based on M_{12} at multiple scattering angles results in a substantial dynamic range and linearity. The sensitivities of the various scattering elements to the particle size distribution, particle index of refraction, and the presence of casein micelles were investigated with single-scattering model simulations. Based on these simulations, M_{12} and M_{33} show additional beneficial qualities for sensor development, since these elements are sensitive to casein particles and insensitive to particle index of refraction, especially if the final sensor is to provide information about the protein and fat levels.

The agreement between experimental values and model predictions will be improved once multiple scattering due to fat globules and casein particles is also included in the model (currently under investigation). Additionally, we are expanding a Monte Carlo vector radiative transfer model (Vaillon et al., 2004) to be applicable to turbid media, like milk.

REFERENCES

Agarwal, B. M., and M. P. Mengüç. 1991. Forward and inverse analysis of single and multiple scattering of collimated radiation in an axisymmetric system. *Int. J. Heat Mass Transfer* 34(3): 633-647.

Aslan, M. M., J. Yamada, M. P. Mengüç, and A. Thomasson. 2003a. Characterization of individual cotton fibers via light scattering: Experiments. *J. Thermophys. Heat Transfer* 17(4): 442-449.

Aslan, M. M., M. Pinar Mengüç, S. Manickavasagam, and C. Saltiel. 2003b. Predictions of size and structure of colloidal metallic particles from elliptically polarized light scattering experiments. In *Proc. 7th Conference on Electromagnetic and Light Scattering by Nonspherical Particles: Theory, Measurements, and Applications.* T. Wriedt, ed. Bremen, Germany: University of Bremen + Institut für Werkstofftechnik.

Attaie, R., and R. L. Ritcher. 2000. Size distribution of fat globules in goat milk. *J. Dairy Sci.* 83(5): 940-944.

Bohren, C. F., and D. R. Huffman. 1983. *Absorption and Scattering of Light by Small Particles.* New York, N.Y.: Wiley.

Crofcheck, C. L. 2001. Characterization of milk by light scattering for fiber optic sensor applications. Unpublished PhD diss. Lexington, Ky.: University of Kentucky, Department of Biosystems and Agricultural Engineering.

Crofcheck, C. L., F. A. Payne, C. L. Hicks, M. P. Mengüç, and S. E. Nokes. 2000. Fiber optic sensor response to low levels of fat in skim milk. *J. Food Process Eng.* 23(2): 163-175.

Crofcheck, C. L., F. A. Payne, and S. E. Nokes. 1999. Predicting the cutting time of cottage cheese using backscatter measurements. *Trans. ASAE* 42(4): 1039-1045.

Govindan, R., S. Manickavasagam, and M. P. Mengüç. 1996. On measuring the Mueller matrix elements of soot agglomerates. In *Proc. First International Symposium on Radiative Heat Transfer.* Presented at Kusadasi, Turkey, August 1995. New York, N.Y.: Begell House.

Griffin, M. C. A., and W. G. Griffin. 1985. A simple turbidimetric method for the determination of the refractive index of large colloidal particles applied to casein micelles. *J. Colloid Interf. Sci.* 104(2): 409-415.

Hansen, J. E., and L. D. Travis. 1974. Light scattering in planetary atmospheres. *Space Sci. Rev.* 16(4): 527-610.

Jaaskelainen, A. J., K. E. Peiponen, and J. A. Raty. 2001. On reflectometric measurement of a refractive index of milk. *J. Dairy Sci.* 84(1): 38-43.

Jenness, R., and J. Koop. 1962. Preparation and properties of a salt solution which simulated milk ultrafiltrate. *Netherlands Milk Dairy J.* 16(3): 153-164.

Manickavasagam, S., and M. P. Mengüç. 1997. Scattering matrix elements of fractal-like agglomerates. *Applied Optics* 36(6): 1337-1351.

Manickavasagam, S., M. P. Menguc, Z. Drozdowicz, and C. Ball. 2002. Size shape and structure - Analysis of fine particles. *American Ceramic Society Bulletin* 81(7): 29-33.

Meeten, G. H., and P. Wood. 1993. Optical fibre methods for measuring the diffuse reflectance of fluids. *Meas. Sci. and Tech.* 4(6): 643-648.

Mengüç, M. P., and S. Manickavasagam. 1998. Characterization of size and structure of agglomerates and inhomogeneous particles via polarized light. *Int. J. Eng. Sci.* 36(12-14): 1569-1593.

Mulder, H., and P. Walstra. 1974. *The Milk Fat Globule Emulsion as Applied to Milk Products and Comparable Foods.* Wageningen, The Netherlands: Pudoc.

Payne, F. A. 1995. Automatic control of coagulum cutting in cheese manufacturing. *Applied Eng. in Agric.* 11(5): 691-697.

Payne, F. A., C. L. Hicks, and P.-S. Shen. 1993. Predicting optimal cutting time of coagulating milk using diffuse reflectance. *J. Dairy Sci.* 76(1): 48-61.

Payne, F. A., Y. Zhou, R. C. Sullivan, and S. Nokes. 1997. Radial backscatter profiles in milk in the wavelength range of 400 to 1000 nm. ASAE Paper No. 976097. St. Joseph, Mich.: ASAE.

Payne, F. A., C. L. Crofcheck, S. E. Nokes, and K. C. Kang. 1999. Light backscatter of milk products for transition sensing using optical fibers. *Trans. ASAE* 42(6): 1771-1776.

Ruettiman, K. W., and M. R. Ladisch. 1987. Casein micelles: Structure, properties, and enzymatic coagulation. *Enzyme Microb. Tech.* 9(10): 578-589.

Schmidt, D. G., J. Koops, and D. Westerbeek. 1977. Properties of artificial casein micelles. *Netherlands Milk Dairy J.* 31(4): 328-341.

Walstra, P. 1975. Effect of homogenization of the fat globule size distribution in milk. *Netherlands Milk Dairy J.* 29(2-3): 279-294.

Walstra, P., and R. Jenness. 1984. *Dairy Chemistry and Physics.* New York, N.Y.: Wiley.

Vaillon, R., B. T. Wong, and M. P. Menguc. 2004. Polarized radiative transfer in a particle-laden semi-transparent medium via a vector Monte Carlo method. *J. Quant. Spectrosc. Radiat. Transfer* 84(4): 383-394.

Yang, P., B. A. Baum, A. J. Heymsfield, Y. X. Hu, H.-L. Huang, S.-C. Tsay, and S. Ackerman. 2003. Single-scattering properties of droxtals. *J. Quant. Spectrosc. Radiat. Transfer* 79-80(1): 1159-1169.