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Prediction of fat globule particle size in homogenized milk using Fourier transform mid-infrared spectra[1]

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ABSTRACT

Our objective was to develop partial least square models using data from Fourier Transform mid-infrared (MIR) spectra to predict the particle size distributions d(0.5) and d(0.9), surface volume mean diameter D[3,2], and volume moment mean diameter D[4,3] of milk fat globules and validate the models. The goal of the study was to produce a method built into the MIR milk analyzer that could be used to warn the instrument operator that the homogenizer is near failure and needs to be replaced to ensure quality of results. Five homogenizers with different homogenization efficiency were used to homogenize pasteurized modified unhomogenized milks and farm raw bulk milks. Homogenized milks were collected from the homogenizer outlet and then run through an MIR milk analyzer without an in-line homogenizer to collect a MIR spectrum. A separate portion of each homogenized milk was analyzed with a laser light-scattering particle size analyzer to obtain reference values. The study was replicated 3 times with 3 independent sets of modified milks and bulk tank farm milks. Validation of the models was done with a set of 34 milks that were not used in the model development. Partial least square regression models were developed and validated for predicting the following milk fat globule particle size distribution parameters from MIR spectra: d(0.5) and d(0.9), surface volume mean diameter D[3,2], and volume moment mean diameter D[4,3]. The basis for the ability to model particle size distribution of milk fat emulsions was hypothesized to be the result of the partial least square modeling detecting absorbance shifts in MIR spectra of milk fat due to the Christiansen effect. The independent sample validation of particle size prediction methods found more variation in d(0.9) and D[4,3] predictions than the d(0.5) and D[3,2] predictions relative to laser light-scattering reference values, and this may be due to variation in particle size among different pump strokes. The accuracy of the d(0.9) prediction for routine quality assurance, to determine if a homogenizer within an MIR milk analyzer was near the failure level [i.e., d(0.9) >1.7 μm] and needed to be replaced, is fit-for-purpose. The daily average particle size performance [i.e., d(0.9)] of a homogenizer based on the mean for the day could be used for monitoring homogenizer performance.
Key words: particle size, homogenization, mid-infrared, light scattering

INTRODUCTION

Annually, in the United States, millions of kilograms of milk are produced and tested daily for the determination of the concentration of the main components (i.e., protein, fat, and other solids). The results are used by processors to determine the payment of dairy farmers, and by the farmers for dairy herd management (Lynch et al., 2004; Barbano and Lynch, 2006). Mid-infrared (**MIR**) milk analyzers have been used and have positively affected the dairy industry and farmers by providing rapid, cost effective, and direct determination of milk components (Barbano and Clark, 1989; Lynch et al., 2004, 2006; Adams and Barbano, 2015).

A laboratory homogenizer made it possible for homogenized emulsions to be analyzed by an MIR milk analyzer (Phipps 1960, 1975). Today, all MIR milk analyzers have an internal homogenizer that breaks the native fat globules to smaller sizes (Goulden, 1961; Biggs, 1967; Biggs et al., 1987). The main reason why fat globules need to be reduced to smaller sizes is that large fat globules increase light scattering, leading to an inaccurate estimate of fat, protein, and lactose content of milk (Barbano and Clark, 1989; Smith et al. 1993). Furthermore, large fat globules can also lead to the Christiansen light-scattering effect (Goulden, 1964; Smith et al., 1993), which causes a change of in the refraction of light at wavelengths near maximum ab-

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sorption by the carbonyl and carbon-hydrogen groups. The Christiansen effect causes a shift in the apparent wavelength of maximum light absorption to a longer wavelength. This effect can be reduced by decreasing the fat globule diameter. Ideally, after homogenization, the fat globule diameter of milk should be less than one-third of the wavelength of fat B (3.48 μm), which is the shortest wavelength used for fat analysis (Goulden, 1964; Smith et al., 1993). Different types of homogenizers have different efficiency; at the same pressure, a single-stage homogenizer will be less efficient than a double stage homogenizer. Other factors that may affect homogenizer efficiency are milk temperature, pump speed, pump stroke length, fat content, and time of usage (Goulden and Phipps, 1964; Walstra and Jenness, 1984). The deterioration of a homogenizer's mechanical components over time may be difficult for the operator to detect and will have a negative effect on analytical repeatability and accuracy (Lynch et al., 2006).

Various tests to evaluate homogenizer efficiency have been used over the years. One test is called the recycle test, where an unhomogenized milk is run through an MIR milk analyzer, the readings are recorded, and the homogenized milk is collected from the instrument outlet tube. Next, the collected homogenized milk is rerun through the instrument and the readings are recorded. If the difference in readings for the fat test on the unhomogenized milk and the instrument homogenized milk is <0.05%, then the homogenizer meets minimum performance standards (AOAC International, 2000; International Dairy Federation, 2000). As the homogenizer performance decreases, the difference between the 2 results gets larger, but as the homogenizer performance continues to get worse across time, the difference in results becomes <0.05% again. Thus, a homogenizer that does not homogenize at all or very poorly will pass the evaluation (Barbano and Clark, 1989; Smith et al., 1993; Lynch et al., 2006). Therefore, the recycle test is easy to preform but, because of this weakness, is not very good.

Another method to evaluate homogenizer efficiency is the determination of milk fat globule size distribution using a laser light scattering particle size analyzer after unhomogenized milk is homogenized through the MIR milk analyzer (Lynch et al., 2006). Laser light scattering uses Mie theory to calculate the particle size distribution, assuming a volume equivalent sphere model. Mie theory predicts scattering intensity as a function of the angle at which light is scattered at the point of interaction with a spherical particle (Horvath, 2009).

The milk fat globule diameter distribution reported by laser light-scattering particle size analyzers is based on the volume of the sphere (i.e., according to the volume of each fat globule present in the sample the instrument calculates a diameter). The parameter reported as particle size distribution d(0.5) is the median of volume distribution [i.e., half of the total fat globules volume in the sample comes from particles with diameter smaller than the d(0.5) value and half of the total fat globules volume in the sample comes from particles with diameter larger than the d(0.5) value]. The parameter reported as particle size distribution d(0.9) indicates that 90% of the total fat globules volume in the sample comes from particles with diameter that lies below the d(0.9). The surface volume mean diameter D[3,2] and volume moment mean diameter D[4,3] are calculated from the particle size distribution (Allen, 1990). The surface volume mean diameter D[3,2], also known as Sauter mean diameter, is calculated using the equation $x_{SV} = \Sigma x^3 dN/\Sigma x^2 dN$, where x = average particle diameter lying in the size range x_i to x_n (μm) and dN = the percentage of the total number of particles lying in the size range x_i to x_n. The volume moment mean diameter D[4,3], also known as De Broucker mean diameter, is calculated using the equation $x_{VM} = \Sigma x^4 dN/\Sigma x^3 dN$, where x = average particle diameter lying in the size range x_i to x_n (μm) and dN = the percentage of the total number of particles lying in the size range x_i to x_n.

Particle size of the milk produced by a homogenizer within an MIR should result in a mean globule diameter d(0.9) <1.7 μm; if d(0.9) ≥1.7 μm, then the homogenizer performance has deteriorated and should be replaced (Smith et al., 1995; Lynch et al., 2006). Our objective was to develop models using data from Fourier transform MIR spectra to predict the particle size distributions d(0.5) and d(0.9), surface volume mean diameter D[3,2], and volume moment mean diameter D[4,3] of milk fat globules and validate the partial least square (**PLS**) model performance. The goal of the study was to produce a method built into the MIR milk analyzer that could be used to warn the instrument operator that the homogenizer is near failure and needs to be replaced to ensure quality of results.

MATERIALS AND METHODS

Experimental Design

Five different homogenizers with different homogenization efficiency (i.e., produced different milk globule size distributions) were used to homogenize 2 types of milk samples sets. The first type of milk sample set contained 12 pasteurized, preserved, modified, and unhomogenized milks ranging from 1.0 to 5.7% fat, as described by Kaylegian et al. (2006), and the second set contained 12 different preserved raw bulk milks obtained from the USDA Federal Milk Market Laboratory (Cleveland, OH) that ranged in fat from about 2.6