IN-LINE CHARACTERIZATION AND RHEOMETRY OF CONCENTRATED SUSPENSIONS USING ULTRASOUND

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ABSTRACT

In this work characterization of model and industrial suspensions is done using ultrasound technique. Experimental investigation of the acoustic properties of the suspensions helps to explain suspension microstructure and its flow behavior. The model fluids consist of corn starch particles suspended in water or oil based systems. Cocoa butter crystalline fat suspensions are selected as industrial suspensions. They are produced in a shear crystallization process with defined solid content. The solid phase of the fat suspensions is determined using Nuclear Magnetic Resonance technique. The sound velocity is found to be a linear function of the solid cocoa butter content and temperature. A software was developed to integrate on-line measurement of flow profiles, pressure difference, temperature and acoustic properties such as velocity of sound.

Beside in-line sound velocity measurements, the shear rate dependent viscosity of suspensions flowing through a pipe is determined by means of pulsed ultrasound based anemometry (UVP: Ultrasound Velocity Profiler) and pressure drop (PD) methods. The measured time delay and frequency of the ultrasound reflected by particles flowing along the axis of the ultrasound beam are used to obtain the velocity profile in the flowing suspension. The rheological models are applied for characterization of the flow velocity profiles determined using the in-line UVP-PD technique. The model parameters can be compared with those obtained from off-line rotational rheometers.

Keywords: Flow profiling, in-line rheometry, suspensions, ultrasound, velocity of sound

INTRODUCTION

A novel method for in-line rheometry involving the measurement of an Ultrasound based Velocity Profile (UVP) using the Met-Flow UVP Monitor [1] and Pressure Difference (PD) in a pipe section was developed and tested at our laboratory by Ouriev and Windhab [2,6] for the flow of a wide variety of model and industrial suspensions. Industrial suspension systems such as chocolate, fat, shampoo and cellulose fibers in water suspensions were characterized by the in-line UVP-PD method. The in-line rheological results were compared with those measured by conventional off-line rheometers [2-9].

The feasibility study of the shear crystallization process and its influence on cocoa butter crystallization is currently under investigation. The result of this research will be used straightforward in the industrial chocolate production. The shear crystallization process developed at our laboratory [10] strongly influences the solid fat content of the cocoa butter suspension. It can be increased from 0% (liquid cocoa butter at 45°C) to almost 30%. Therefore the acoustic properties (attenuation, sound velocity, signal distortion due to multiple scattering) vary drastically. Beside the acoustic properties, the process parameters of the crystallization influence the rheology of the fat suspension and consequently change shape of the flow velocity profiles.

MATERIALS AND METHODS

Ultrasound transducer adapter cells

For acoustic investigations an in-line double wall steel adapter was constructed (figure 1). The suspension flows inside a 25 mm diameter tube. The adapter construction allows the installation of two transducers opposite to each other and can be also used for static acoustic measurements. In this case the adapter tube is closed from one side and a small sample (50ml) is sufficient for the acoustic measurement. Based on this design, a PVC cell for two transducers with a Doppler angle of 20° was developed. This cell allows a simultaneous measurement of the flow velocity profile and monitoring of the transmitted pulse (thus velocity of sound calculated from the time of flight of the pulse and the transducer distance, and signal quality). Pressure and temperature dependencies of the sound velocity in water were used for calibration purpose. Measuring the sound velocity at different temperatures gives precise information about the actual distance between the two opposite transducers. Such distance calibration was applied to both steel and PVC flow adapters. The transmitting and receiving ultrasound transducers have a 4 MHz base frequency and are equipped with a 5mm active diameter acoustic element. One of them is connected to the Ultrasound Velocity Profile (UVP) Monitor and the other to the oscilloscope.

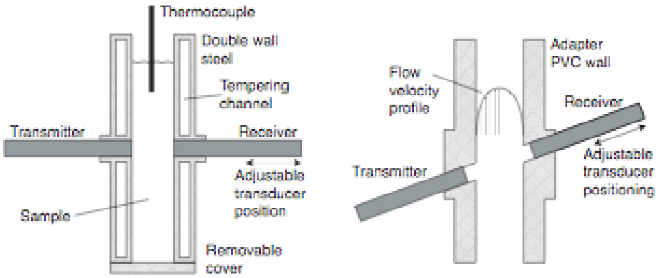

Figure 1: Measurement cells: double wall stainless steel adapter (left) in the configuration for a static measurement and PVC adapter with 20° Doppler angle (right). The transmitting transducer is used for velocity profile and echo amplitude measurement while the second transducer is used as receiver to measure the velocity of sound and other acoustic properties.

Data acquisition and control software

The current measurement setup (figure 2) consists of the UVP instrument (Met-Flow UVP-DUO), oscilloscope (Yokogawa DL 1520, 200 MS/s, 20 GS/s in repetitive mode, 150 MHz), thermocouples and pressure sensors connected to a National Instruments "FieldPoint" data acquisition system and a thermostat for the temperature control of the static measurements in the flow adapter. All the devices are controlled from a National Instruments LabView application. The communication with the UVP-DUO is implemented with an ActiveX library provided by MetFlow SA. As discussed earlier, the velocity of sound in cocoa butter depends on the temperature and the crystal content. Therefore the UVP parameters are set-up in time instead of distance units. For the display of the current state of the flow profile (average and standard deviation), a variable size first-in-first-out buffer was implemented. The waveform data of the transmitted pulse from the oscilloscope are used to calculate the instantaneous velocity of sound. This parameter is automatically applied to calculate the distances along the velocity profile and the volume flow rate. The amplitude and power spectra are used for characterization of the received signal quality.

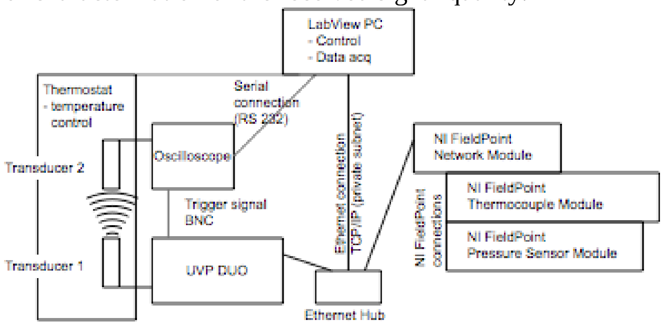

Figure 2: Scheme of the data acquisition and control hardware.

The temperature of a thermostat is controllable from LabView. This allows the automated measurement of the dependency of acoustic properties of liquids by using the double wall cell and driving a temperature ramp over several hours. Figure 3 shows an example of such a measurement.

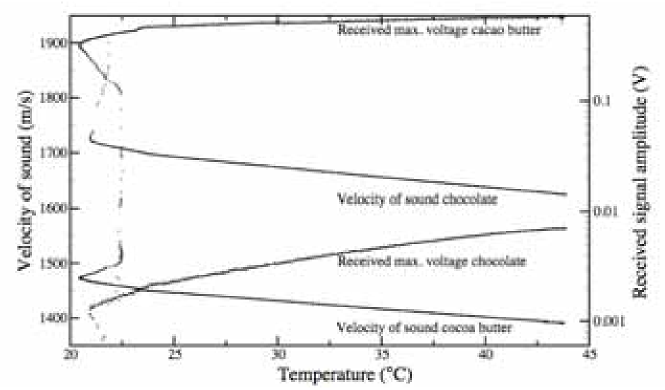

Figure 3: Velocity of sound and amplitude of the received signal plotted as a function of the fluid temperature.

The whole LabView application is implemented as a flexible modular system. The single modules abstracting the peripheral hardware, run in separate threads and can be activated according the requirements of the measurement. A central part of the application provides the graphical user interface for the adjustment of the parameters and the real time display of the measurement data, data logging and the infrastructure for the communication amongst the modules (e.g. the velocity of sound from the oscilloscope that is needed by the UVP module).

Cocoa butter fat suspension

Cocoa butter used for chocolate production is a mixture of triglycerides with a polymorphic crystallization behavior that has a melting range between 33 and 37°C. During the shear crystallization developed at our laboratory the crystal content is increased to up to 30%. The processing of cocoa butter results in the growth of crystals creating a needle like microstructure, which builds up to agglomerates. As the cocoa butter solidifies at room temperature, it is necessary to implement a temperature control of all the measurement equipment in direct contact with the cocoa butter.

Flow loop and crystallization procedure

The temperature controlled flow loop shown in figure 4, discussed in detail elsewhere [10], consists of a stirred cocoa butter feed tank, shear crystallizer and double wall steel transducer adapter cell.

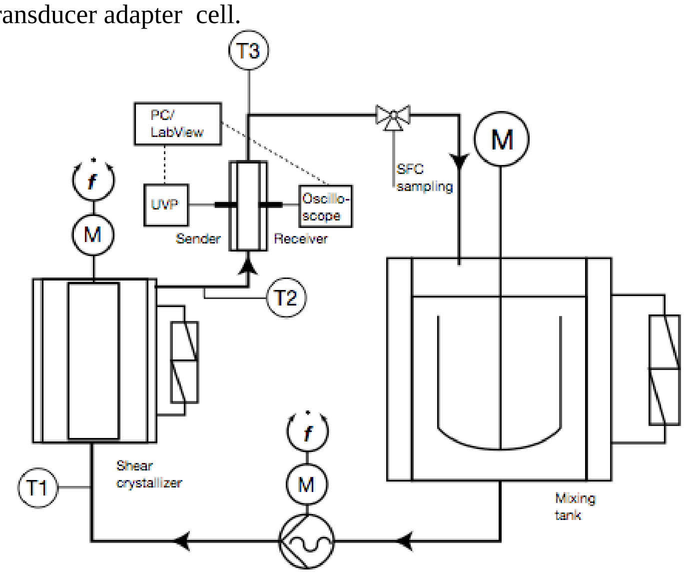

Figure 4: Crystallizer flow loop and instrumentation.

Prior to the crystallization process, cocoa butter was melted in the feed tank at 45°C. The molten suspension was pumped through the shear crystallizer flow loop for at least one hour. This was necessary to ensure that all cocoa butter in the flow loop was melted and a steady state was reached. In order to start the crystallization, the temperature of the shear crystallizer cooling water was set to 15°C and the shear crystallizer was switched on. The feed tank, pipes, and the ultrasound cell were all kept at 32.5°C in order to avoid solidification of cocoa butter.

The temperature, velocity of sound and signal amplitude of the ultrasound pulse were recorded as a function of time. An off-line Nuclear Magnetic Resonance (NMR) instrument was used to determine the corresponding solid fat content (SFC) of the cocoa butter suspension, which is the concentration of the cocoa butter crystals formed by drawing a sample at the outlet of the shear crystallizer.

Model suspensions

For a better understanding of the influence of suspension properties (e.g. kind of the continuous phase, concentration and microstructure of the disperse phase) on acoustic properties and finally the quality of the profile measurements, the model suspensions were used. In contrast, such acoustic investigations would be difficult due to the complexity of the cocoa butter. The temperature dependency of the kinetics of the rheological and micro-structural parameters makes it difficult to compare in-line and off-line measurements.

In a first series of experiments three model suspensions of corn starch particles (size range between 5 to 35 µm) dispersed in (i) sucrose solution with different sugar concentrations, (ii) silicon oil with different shear viscosities and (iii) rapeseed oil were considered. Concentration of disperse phase was varied between 0 and 40%w.

RESULTS AND DISCUSSION

Effect of solid fat content on sound velocity in cocoa butter

During crystallization, the cocoa butter temperature was kept constant over a wide range of crystal content. Therefore it was possible to measure the sound velocity c as a function of the solid fat content (SFC). In figure 5 this measured dependency (symbols) is represented by the straight line: $c = 1349.7+4.862$ SFC with a correlation coefficient of 0.993. It can be seen that 1% increase in SFC results in an increase of sound velocity by 5 m/s. The temperature dependency of the velocity of sound in the liquid cocoa butter is 3.5 m/s per °C.

The velocity of sound in a suspension of particles or emulsion of droplets depends on the mean density and mean compressibility and is given by the Urick equation [11]:

$$c = \frac{1}{\sqrt{\kappa\rho}}, \quad \kappa = \sum_j \phi_j \kappa_j, \quad \rho = \sum_j \phi_j \kappa_j \qquad (1)$$

where c: velocity of sound, κ: adiabatic compressibility, ρ: density, ϕ: volume fraction of the dispersed phase.

For a two phase system (e.g. solid cocoa butter crystals dispersed in liquid cocoa butter) one gets

$$\kappa = \phi\kappa_s + (1-\phi)\kappa_l, \quad \rho = \phi\rho_s + (1-\phi)\rho_l \qquad (2)$$

where subscript l: liquid/continuous phase; subscript s: solid/dispersed phase.

According to equations (1) and (2) it is possible to determine the composition of a suspension when the velocity of sound c, the adiabatic compressibility κ and the density ρ of the solid and liquid phases are known. The densities of liquid and solid cocoa butter are known. In the present case, the velocity of sound in solid coca butter had to be extrapolated. Compressibility of the liquid and solid cocoa butter can be also calculated. The velocity of sound from equation (1) is nearly linear for the range between 0% and 30% SFC. This corresponds qualitatively to the measured data shown in figure 5.

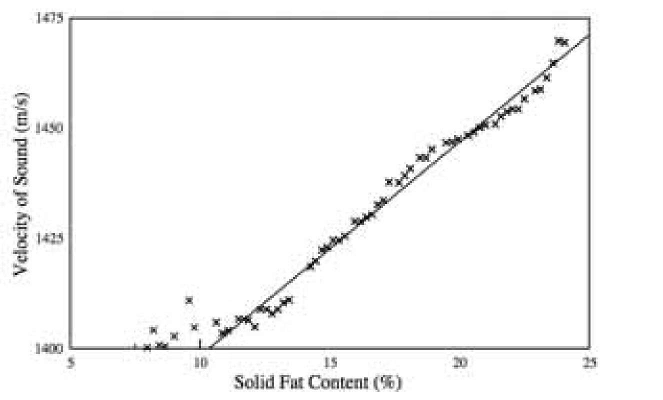

Figure 5: Velocity of sound as a function of the solid fat content (both quantities originally measured as functions of the time).

Acoustic properties of oil and water based model suspensions

Table 1 gives an example of the basic acoustic properties of suspensions degassed under vacuum with the same concentration of corn starch as dispersed phase. The signal transmitted through silicon oil based suspension is visibly distorted. Even for a 4 cycle pulse the amplitudes of the trough and crest are smaller in the center probably due to signal deletion caused by multiple scattering.

Table 1: Acoustic properties of model suspensions (S) of 10%w corn starch and their continuous phases (C) at 27 °C.

	Sound velocity, c		Ampl., a		Freq. Peak, f	
	C	S	C	S	C	S
	m/s		mV		MHz	
Sucrose solution (60%w)	1798.5	1876.1	4653	546	4.0	4.0
Rapeseed oil	1450.0	1479.0	3980	214	4.0	4.0
Silicon oil AK5000	1009.3	1055.5	2950	166	4.0	3.5

CONCLUSIONS

The sound velocity in cocoa butter is found to be a linear function of solid fat content and temperature. The attenuation of sound in model suspensions is found to be affected by the continuous phase - particle interface and whether the continuous phase is hydrophilic or hydrophobic. For instance, rapeseed and silicon oils when used as continuous phases contributed to the significant attenuation compared with that for sucrose solutions. The UVPPD technique developed at our laboratory [2,6] can be used for in-line characterization and rheometry of concentrated suspensions. The present work involved the further development of a new software which integrated the simultaneous in-line measurement of flow velocity profiles, pressure difference, temperature and acoustic properties such as velocity of sound.

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