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Density, ultrasound velocity, acoustic impedance, reflection and absorption coefficient determination of liquids via multiple reflection method

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ABSTRACT

The accuracy of density, reflection coefficient, and acoustic impedance determination via multiple reflection method was validated experimentally. The ternary system water–maltose–ethanol was used to execute a systematic, temperature dependent study over a wide range of densities and viscosities aiming an application as inline sensor in beverage industries.

The validation results of the presented method and setup show root mean square errors of: 1.201E−3 g cm^{-3} (±0.12%) density, 0.515E−3 (0.15%) reflection coefficient and 1.851E + 3 kg s^{-1} m^{-2} (0.12%) specific acoustic impedance. The results of the diffraction corrected absorption showed an average standard deviation of only 0.12%. It was found that the absorption change shows a good correlation to concentration variations and may be useful for laboratory analysis of sufficiently pure liquids.

The main part of the observed errors can be explained by the observed noise, temperature variation and the low signal resolution of 50 MHz. In particular, the poor signal-to-noise ratio of the second reflector echo was found to be a main accuracy limitation. Concerning the investigation of liquids the unstable properties of the reference material PMMA, due to hygroscopicity, were identified to be an additional, unpredictable source of uncertainty. While dimensional changes can be considered by adequate methodology, the impact of the time and temperature dependent water absorption on relevant reference properties like the buffer's sound velocity and density could not be considered and may explain part of the observed deviations.

1. Introduction

In the past, several methods to investigate the density via ultrasonic were investigated [1–8]. In particular the non-invasive characteristic suits the buffer-rod techniques (BRTs) to be applied as process analytical technology (PAT) in food and beverage industries, to determine the density and the ultrasonic velocity of multicomponent mixtures [9]. The basis of the BRTs is the plane wave propagation across one or more interface and the knowledge of the reference's (buffer) material properties. Four BRT sub-groups could be identified: the multiple reflection method (MRM), the transmission methods (TM), the reference reflection methods (RRM) and the angular reflection methods (ARM). In case of a process application in beverage industries, moderate attenuation, inconstant process conditions, and temperature gradients have to be considered. It was found that the MRM is the best choice, particularly considering the minor calibration effort, the sensor design and the analytical output.

To calculate the reflection coefficient (RC), the density, the absorption and the specific acoustic impedance (SAI) via MRM, the time-of-flight (TOF) and the amplitudes of three echo pulses have to be evaluated. We may specify them as: A_{r1} – the 1st of the multiple echo signal which are reflected at the buffer- liquid interphase, A_{e11} – the 1st echo signal which was transmitted into the sample liquid and reflected by the reflector, and A_{e21} – the 1st echo signal which was transmitted into the sample liquid and passed the liquid volume twice before being received (compare Fig. 3). Further details concerning the method including the series expansion of the echo description will be found in [8,10–15]. The details concerning the amplitude and TOF evaluation will follow in next paragraph. Knowing the relevant amplitudes, the reflection coefficient of a plane wave passing the interface from medium 1 (buffer) to medium 2 (fluid), r_{12} can be calculated via:

$$r_{12} = \sqrt{\frac{x}{x-1}} \quad \text{and} \quad x = \frac{A_{r1} \cdot A_{e21}}{A_{e11}^2}, \tag{1}$$

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whereby the indices of the amplitude values define only the position within the complete signal (see Fig. 3). The indices of the other parameters define the corresponding medium: 1 – buffer material, 2 – sample liquid, 3 – reflector material; or medium combination at the interphase. From the TOF in the sample liquid and the known distance, l_2 between buffer and reflector, the sample liquid's ultrasonic velocity (c_2) can be calculated:

$$c_2 = \frac{2l_2}{TOF_2}. \tag{2}$$

Knowing both variables, the buffer's sound velocity and density at the actual temperature, the liquid's density can be calculated:

$$\rho_2 = \frac{\rho_1 c_1}{c_2}\frac{(1+r_{12})}{(1-r_{12})}. \tag{3}$$

The acoustic impedance, Z is the product of density and sound velocity:

$$Z_2 = c_2\rho_2 = \rho_1 c_1 \frac{(1+r_{12})}{(1-r_{12})}. \tag{4}$$

And, in case that reflector and buffer are made of similar material and assuming that both the sample liquid's composition and temperature is similar at both interfaces, the sample liquid's attenuation, α can be calculated by:

$$\alpha_2 = ln\left(\frac{B}{C\cdot r_{12}^2}\right)\cdot\frac{1}{2l_2}. \tag{5}$$

The investigated liquids are solutions and can be considered as a homogeneous medium. As well reflection and transmission losses are considered and diffraction effects will be corrected. Accordingly, the calculated loss coefficient corresponds to the absorption coefficient which is mainly caused by viscous energy absorption and thermal conduction.

2. Materials, methods and experimental setup

According to the methods requirements an experimental setup was designed that provides all parameters to characterize ternary component mixtures and to validate the methods accuracy (see Fig. 1). A vibrating U-tube density meter (L-Dens 313, Anton Paar, accuracy: 1E−3 g/cm^3, 0.1 °C) was used to determine the density. The temperature is provided by a measurement chain of TTI-22 (Isothermal Technology Ltd.) and a standard platinum resistance thermometer (SPRT 909Q, 25 Ω, Isothermal Technology Ltd.) resulting in an certified accuracy of ⩽5 mK (resolution: 0.1 mK). The time-of-flight (TOF) in the liquid is determined between the echoes A_{r1} and A_{e11} via pulse-echo method, cross correlation and zero crossing approximation [16]. The ultrasonic velocity is calculated from periodical reflector distance (RD) calibrations with demineralized water [17]. And the temperature controlled environment is provided by a cooling thermostat (Lauda RP3530 C). The applied trial procedure provided following reproducible conditions at each concentration combination: average temperature variation: ±5 mK, temperature gradients across the sound propagation path: ⩽0.05 K, and sound velocity errors ⩽0.05 m/s over the investigated temperature range of 10–30 °C. To monitor the temperature uniformity across the propagation path and to ensure a sufficient stability for the measurements six waterproofed Pt100 were immersed in different depth. The complete methodical details are presented in [9].

2.1. The ultrasonic measurement cell (USVMC)

The USVMC was especially designed to allow extensive temperature supervision, the investigation of varying liquids, a simultaneous reference density measurement of acceptable accuracy and to offer the investigation of varying reflector distances, buffer materials, and buffer dimension. For the sake of completeness and reproducibility, all MRM relevant details of the USVMC are provided in the following section. The USVMC consists of two Poly(methyl methacrylate) (PMMA)-cylinders flanged at each side of a PMMA tube. As visualized in Fig. 2, the transducer was pressed waterproofed to the PMMA cylinder by an additional flange. Materials, dimensions, and specifications were chosen according to the design considerations as stated in [8]. Dimension changes of the propagation path due to thermal expansion and hygroscopicity of PMMA [18,19] were considered by cyclic calibrations with demineralized water at each temperature [9]. The mean values of the USVMC at 20 °C are given Table 1.

The temperature dependent sound velocity of PMMA was evaluated preliminary to the main trial (validity: 10–30 °C):

$$USV_{PMMA}(T) = 2811.107\frac{m}{s} - 2.074\cdot 10^{-3}\frac{m}{^\circ C\cdot s}\cdot T - 2.544\cdot 10^{-5}\frac{m}{^\circ C^2\cdot s}\cdot T^2, \tag{6}$$

whereby T represents the temperature in °C. The sound velocity of PMMA at 20 °C results in 2759.43 m/s which is in good agreement with values found in literature. The temperature dependent density

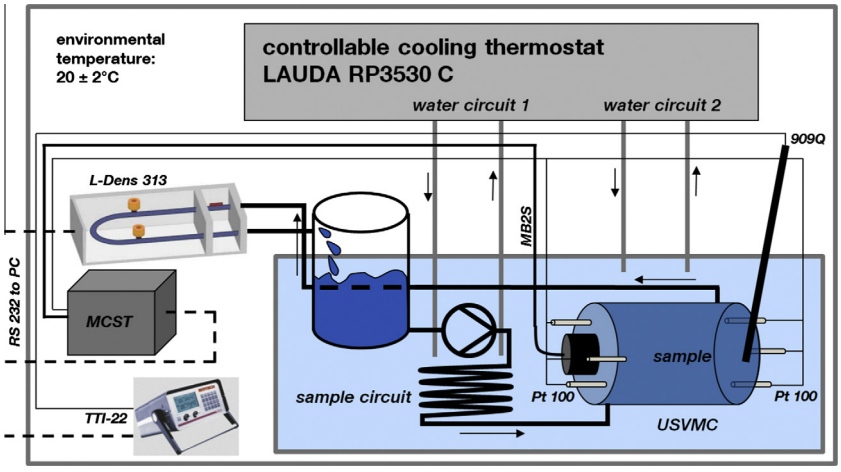

Fig. 1. Scheme of the experimental setup to measure on-line measurement the ultrasonic signals, the temperature and the density (MCST: Multi-Channel-Signal-Transformer, USVMC: Ultrasound Velocity Measurement–Measurement-Cell).