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Original Research Paper

Monitoring concept of single-frequency ultrasound and its application in dynamic crystallization processes

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ABSTRACT

The supersaturation in liquid as well as the mean particle size and solid content (also called as slurry, magma or suspension density) of solids can be monitored simultaneously by single frequency ultrasound. A concept of mathematical modeling based on the ultrasound velocity and attenuation measured with two sensors (one for liquid and one for solid state) has been developed and was proofed by experimental data at different temperature levels. The model structure is characterized by a minimized effort for the parameter identification and can be easily implemented. A turn-key probe with heated cage avoiding incrustations is presented to enable the exclusive investigation of the solution in dynamic suspensions. Examples for application of single frequency ultrasound probes as PAT tools are demonstrated by case studies for industrial urea suspensions and for sugar crystallization.

1. Introduction

Optimal crystallization processes require the control of the supersaturation as driving force. Dynamic process effects such as a reduced crystal growth rate in solution due to accumulated impurities have to be considered in industry and demand for process regulation. The overall crystal surface influences the crystallization kinetics and has to be controlled, too. Consequently, the solid state characteristics, the particle size and the solid content, have to be quantified in order to maintain continuous processes within the operational window or to evaluate the progress of batch processes. The demand for process control implies the need for adequate measuring techniques. Since the PAT (process analytical technology) initiative by the FDA in 2002 real-time process monitoring has been explored and developed even in the pharmaceutical sector [1–4]. Several physical methods (e.g. conductivity, density) or spectroscopic PAT tools (UV–Vis, Raman, IR, ultrasound) are available to measure the concentration and, consequently, to determine the supersaturation [5]. In case of the solid state characterization, in-line methods (e.g. ultrasound spectroscopy [6], in-situ imaging [7], FBRM [7–9]) are generally preferred because errors during sampling are avoided and process information are promptly available.

However, most of the aforementioned PATs show obstacles with electrically non-conducting, dense, concentrated, or optically opaque solutions [10]. Here, the ultrasound-based technologies have clear advantages, because most materials are ultrasonically transparent and hence allow the analysis of a broad variety of sample types [1,11–14]. The overlapping effects of liquid and solid state, however, make the analysis of ultrasound challenging. Titiz-Sargut and Ulrich [15] and Stelzer et al. [10] demonstrated the use of a self-made protected sensor (covered by a cage that hinders the particles to pass the measurement section of the sensor) in order to investigate exclusively the liquid state in suspensions. Nevertheless, the protected sensor tends to become incrusted. In this study a turn-key protected probe featured with a heated cage avoiding incrustations will be applied for the first time.

Pertig et al. [14] have shown that the solid state can be measured by SFUS as comparatively robust and inexpensive PAT in dense and opaque systems without diluting. The SFUS monitoring requires the identification of adequate mathematical models [14]. Based on an optimal model structure, the effort for the model identification can be reduced [10]. Stelzer et al. [10] already applied a model with 5 parameters for the solid state characterization of urea. In this study the development of a basic model concept for two ultrasound sensors is published in detail opening further modification and applications. The model structure and its applicability at different temperature levels will be proven by experimental data and will be demonstrated at dynamic conditions. Stelzer et al. [10] demonstrated that the liquid and solid state

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List of Symbols and Abbreviations

a	attenuation (dB)	v	ultrasound velocity (m/s)
a_1	coefficient (wt.%)	z	identified index (–)
a_2	coefficient (wt.%/°C)	β_1	coefficient (1/wt.%)
a_3	coefficient (wt.%/(m/s))	β_2	coefficient $(1/(\text{wt.\%})^2)$
a_4	coefficient (wt.%/°C/(m/s))	β_3	coefficient $(\text{m/s}/(\text{dB})^{\beta 4})$
a_5	coefficient $(\text{wt.\%}/(°\text{C})^2)$	β_4	coefficient (–)
a_6	coefficient $(\text{wt.\%}/(\text{m/s})^2)$	c	solute concentration (wt.%)
$\overline{d_{43}}$	mean particle diameter (m)	c_1	coefficient (–)
$f()$	function of	c_2	coefficient (μm)
FBRM	focused beam reflectance measurement	c_3	coefficient (μm)
k	adiabatic compressibility (ms^2/kg)	c_4	coefficient (–)
ORM	optical reflectance method	φ	solid content (wt.%)
PAT	process analytical technology	ρ	density (g/L)
SFUS	single frequency ultrasound		
T	temperature (°C)		

of urea can be measured simultaneously under dynamic process conditions only by one ultrasound device equipped with two sensors. In case of sugar crystallization the integral signal effects of liquid and solid state are very specific. It will be shown how the presented measuring concept based on two sensors deals with this issue.

2. Experimental section

2.1. Materials

The compounds chosen for this study are industrial grade Urea (chemical purity > 99.6%) provided by SKW Piesteritz, Germany and food grade sugar (chemical purity > 99.96%). Distilled water is used for all experiments.

2.2. Experimental setup

A jacketed reactor (diameter of 115 mm and 200 mm height) with external temperature control equipped with a marine impeller (diameter of 50 mm, placed 25 mm above the bottom) was used for the experiments. The ultrasound probe (LiquiSonic 30 immersion sensor) was inducted slightly skewed from the top and connected to a controller, developed by SensoTech GmbH, Magdeburg, Germany. Fig. 1 illustrates the experimental setup.

Further details about the experimental setup are published by Pertig et al. [14]. In case of the sugar crystallization an additional encased probe, so called protected ultrasound probe was additionally inserted into the reactor. The unprotected probe measured the ultrasound velocity and attenuation in suspension. The second protected probe (LiquiSonic OCM) was encased in a basket with a mesh size of 35 μm (see Fig. 2).

Fig. 2. Ultrasound sensor with crystallization adapter and exchangeable membrane (with kind permission of Senso Tech GmbH), see Nitschke et al. [16].

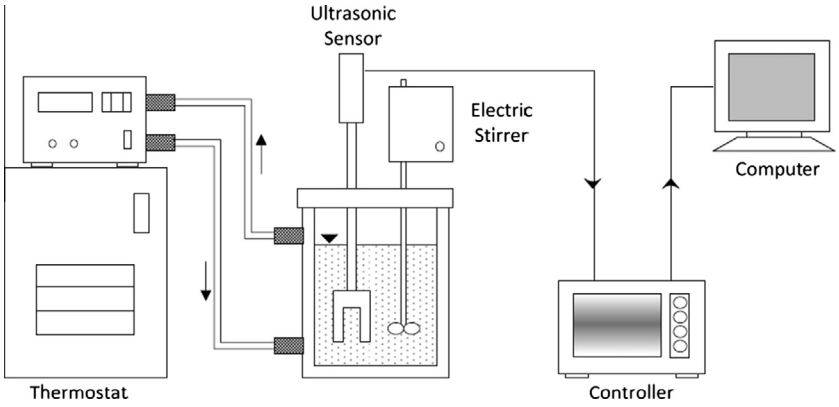

Fig. 1. Experimental setup with ultrasound sensor and controller measuring ultrasound velocity and attenuation in stirred suspensions.