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Short Communication

Sonocrystallisation of lactose in concentrated whey

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ABSTRACT

Whey concentrated to 32% lactose was sonicated at 30 °C in a non-contact approach at flow rates of up to 12 L/min. Applied energy density varied from 3 to 16 J/mL at a frequency of 20 kHz. Sonication of whey initiated the rapid formation of a large number of lactose crystals in response to acoustic cavitation which increased the rate of crystallisation. The rate of sonocrystallisation was greater than stirring for approximately 180 min but slowed down between 120 and 180 min as the metastable limit was reached. A second treatment with ultrasound at 120 min delivering an applied energy density of 4 J/mL stimulated further nuclei formation and the rate of crystallisation was maintained for >300 min. Yield on the other hand was limited by the solubility of lactose and could not be improved. The crystal size distribution was narrower than that with stirring and the overall crystal size was smaller.

1. Introduction

Lactose is the most abundant carbohydrate found in milk (4.4–5.2%) and a major constituent of many concentrated and dried milk and whey products. Lactose must first be crystallized before many of these products can be spray dried. Commercial manufacture of whey powder involves concentration of whey, often by evaporation, followed by batch crystallisation which is initiated by rapid cooling or by seeding directly with lactose over many hours (up to 20 h) to ultimately yield up to 80% crystallized lactose [1]. These processes offer limited control and improving the efficiency of crystallisation will benefit the dairy industry [2]. Crystallisation of lactose consists of three phases; the first is supersaturation followed by nucleation (appearance of crystals) and crystal growth. During the crystallisation process it is critical to control crystal purity, shape and size but traditional paddle mixers are known to create non-uniform mixing. Irregularities cause random fluctuations in supersaturation, resulting in uneven and irregular crystal size and growth occasionally forming agglomerates [3,4].

The overall crystallisation process is slow and lactose recovery can be improved. Sonication is known to reduce crystallisation induction times and increase the rate of nucleation in a number of processes including the crystallisation of fats [5] and pharmaceutical lactose [6] in a process known as sonocrystallisation. Sonocrystallisation is most effective when ultrasound is delivered at the nucleation phase [7].

Ultrasonic cavitation can enhance the rate of reaction and facilitate mass transfer in liquid. Studies have shown that sonocrystallisation generally exhibits four characteristics which are not typical of crystallisation without sonication. These are faster primary nucleation, ease of nucleation, initiation of secondary nucleation and production of smaller and purer crystals [8]. Ultrasound in the presence of an anti-solvent such as ethanol was used to increase the yield of lactose crystallisation [7,9–11], in acetone [12] and in glycerine solution [4]. More recently, these characteristics were reported in a simple aqueous system without anti-solvent [13].

Much of the laboratory data reported in literature is based on direct contact sonication. In this approach, a titanium ultrasonic probe was immersed directly into the product. Because the energy density is greatest at the surface of the sonotrode it will cause gradual pitting and degradation. Although the risk associated with such practice is minimal, there is concern that erosion of the sonotrodes may result in product contamination [14]. A non-contact alternative to direct contact sonication exists and this design permits modular implementation and in-line operation. These sonication cells are designed with multiple low power transducers attached to the outer surface of the metal cell, eliminating the need for sonotrodes. Sound waves propagate through the metal surface overcoming sonotrode erosion and improving energy distribution. These generate lower power densities than sonotrodes but efficiently initiate lactose nucleation and have been implemented industrially outside the food industry [6,15].

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Sonocrystallisation of lactose is known to occur in the presence of anti-solvents and in aqueous solutions but the effect of ultrasound on lactose crystallisation in concentrated whey remains unknown. Since the use of anti-solvent in the manufacture of food grade lactose is unlikely to be feasible at a commercial scale, the aqueous lactose study [13] is probably the most relevant reference publication for industry. In the current study, commercially manufactured whey concentrate was sonicated at pilot scale using non-contact equipment to study the effects on lactose crystallisation.

2. Materials and methods

Concentrated whey was sourced directly from a commercial dairy factory (Northern Victoria, Australia). Whey was concentrated to 32 ± 2% lactose by evaporation at 55 °C. Concentrated whey was then flash cooled to 30 ± 1 °C to initiate lactose crystallisation. Two sonication studies were conducted (Fig. 1).

In the first, sonication was performed with a 20 kHz Sonolab SL250 non-contact sonicator (Prosonix Ltd., Oxford, UK). The processing chamber was 15.4 cm in diameter with a capacity of 2.1 L. Concentrated whey was treated with high intensity low frequency ultrasound at 30 ± 1 °C delivering an applied energy density of 3–16 J/mL in a single pass. The whey concentrate was sampled immediately following the industrial flash cooler and pumped through the SL250 at various flow rates using a peristaltic pump (Masterflex L/S model 7554-95, Illinois, USA). The control solution (T0) was pumped through the ultrasonic rig at the appropriate flow rate without sonication. Three flow rates were explored (0.75, 1.2 and 2 L/min; achieving residence times of 168, 105 and 63 s) at two power settings (100 and 200 W). The applied energy density (J/mL) was calculated as described by Zisu et al. [16] and the corresponding energy densities are shown in Table 1 ($n \geqslant 2$). Sonicated whey (400 g) was transferred to 400 mL glass beakers (6.5 cm diameter) and allowed to crystallize at room temperature (~22 °C) for 60 min.

The magnitude of the experiment was scaled up in the second study based on energy density. A Prosonitron P500 (Prosonix Ltd.) non-contact sonicator was installed in-line with the commercial manufacturing process immediately following the flash cooler. The processing chamber was 15.4 cm in diameter with a capacity of 6.4 L. Concentrated whey (30 ± 1 °C) was diverted to the sonicator at the desired flow rate. Sonication was performed in a single pass at 250–600 W and flow rates of 4–12 L/min (residence time of 96–32 s, respectively) to achieve applied energy densities of 3–15 J/mL. Control samples were passed through the ultrasonic rig at the appropriate flow rate without sonication. Sonicated and control whey (400 g) were then placed in 400 mL glass beakers (6.5 cm diameter) and transferred to a 30 °C water bath. Samples were cooled to 15 °C by lowering the temperature by 2 °C every 30 min (4 °C/h) then holding at temperature for up to 24 h. Whey was stirred continuously during the cooling period and for the entire holding time using an overhead stirrer (RZR 2020, Heidolph Instruments GmbH & Co., Schwabach, Germany) fitted with a 30 mm three paddle operating at 650 rpm.

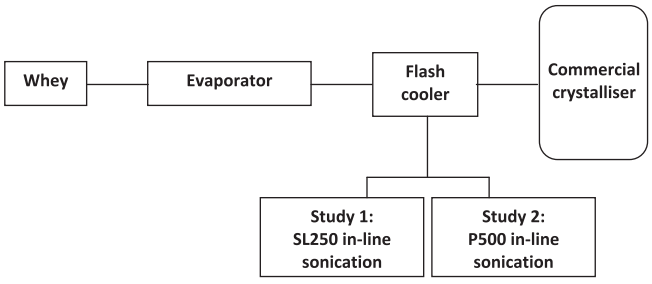

Fig. 1. Process flow diagram for two approaches to sonocrystallisation.

Table 1
Applied energy density (J/mL) delivered by the SL250 at various flow rates and power settings.

Flow rate (mL/min)	Electrical power (W)	
	100 (J/mL)	200 (J/mL)
750	8	16
1200	5	10
2000	3	6

When a second off-line sonication treatment was required, a 1 kW (UIP100hd) 200 mm radial sonotrode (BS2d34SPEC) was used to deliver an applied energy density of 4 J/mL (Hielscher Ultrasonics GmbH, Teltow Germany).

Total dissolved solid (Brix) were measured at 23 °C as an indicator of crystallisation (Refracto 30GS, Mettler Toledo, Schwerzenbach, Switzerland). Samples were frozen immediately and sent to the Dairy Technical Services laboratories (Kensington, Victoria, Australia) for total solids (Test number: MOIS 21 10.00) and lactose by enzyme analysis (Test number: LACT 02 04.93) measurements.

Crystallisation was calculated according to Westergaard [2]:

$$\% \text{ Crystallisation} = \frac{(S_1 - S_2) \times 9500 \times 100}{L \times \text{TS} \times (95 - S_2)} \quad (1)$$

where S_1 = % sugar (Ref. index) of the concentrate direct from the evaporator, S_2 = % sugar (Ref. index) of the crystallized concentrate, L = % lactose and TS = total solids content in %.

Complementary to absorbance, whey solutions were viewed under a light microscope (Olympus BH-2, Tokyo, Japan) fitted with and without a blue light filter at 10× magnification immediately after sonication (T0) and 60 min (T60) of treatment. Whey was also examined after 30 min (T30) in some experiments. Images were captured with a 3.2 mega pixel digital camera (Pro-MicroScan Model DCM310, Oplenic Co., Hangzhou, China) and were used to measure crystal size by Scope Photo image analysis software (Version 3.0, Oplenic Co., Hangzhou, China). The size of crystal was reported as the length of a crystal in the *b* direction (defined by Fries et al. [17]) and the average crystal size was measured as the average size of all the particles viewed under the microscope. The average growth rate of the (0 1 0) face (defined by Michaels and Van Kreveld [18]) was calculated from the slope of the average crystal size as a function of time. Saturated lactose solution was used to dilute the whey when necessary to allow accurate measurement of crystal size.

3. Results and discussion

Crystallisation of lactose in commercially concentrated whey was significantly increased by the application of ultrasound at a low energy density of 3 J/mL and a flow rate of 2 L/min (Fig. 2). A similar observation was made for the various flow rates (0.75, 1.2 and 2 L/min) and power inputs explored (3–16 J/mL) (data not shown). Regardless of the sonication intensity and flow rate, the least number of lactose crystals was observed in the control solutions at T0. A greater number of lactose crystals were present in whey immediately after sonication (T0) at all energy densities (3–16 J/mL). Although some nucleation occurred in the control solution after 30 and 60 min of crystallisation, the number of crystals observed in sonicated solutions was far greater at an equivalent time. Ultrasound generated a large number of nuclei resulting in the growth of many small crystals, differing to the growth of fewer but larger crystals without treatment. Unlike aqueous solutions of reconstituted lactose, a lower energy density