J. Dairy Sci. 98:1–10
http://dx.doi.org/10.3168/jds.2015-9959

Water-lactose behavior as a function of concentration and presence of lactic acid in lactose model systems

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ABSTRACT

The presence of high amounts of lactic acid in acid whey restricts its ability to be further processed because lactose appears to remain in its amorphous form. A systematic study is lacking in this regard especially during the concentration step. Hence, the main aim of the study was to establish the structure and behavior of water molecules surrounding lactose in the presence of 1% (wt/wt) lactic acid at a concentration up to 50% (wt/wt). Furthermore, the crystallization nature of freeze-dried lactose with or without lactic acid was established using differential scanning calorimetry and Fourier transform infrared spectroscopy. Two mechanisms were proposed to describe the behavior of water molecules around lactose molecules during the concentration of pure lactose and lactose solutions with lactic acid. Pure lactose solution exhibited a water evaporation enthalpy of ~679 $J \cdot g^{-1}$, whereas lactose + lactic acid solution resulted in ~965 $J \cdot g^{-1}$ at a 50% (wt/wt) concentration. This indicates a greater energy requirement for water removal around lactose in the presence of lactic acid. Higher crystallization temperatures were observed with the presence of lactic acid, indicating a delay in crystallization. Furthermore, less crystalline lactose (~12%) was obtained in the presence of lactic acid, indicating high amorphous nature compared with pure lactose where ~50% crystallinity was obtained. The Fourier transform infrared spectra revealed that the strong hydration layer consisting lactic acid and H_3O^+ ions surrounded lactose molecules via strong H bonds, which restricted water mobility, induced a change in structure of lactose, or both, creating unfavorable conditions for lactose crystallization. Thus, partial or complete removal of lactic acid from acid whey may be the first step toward improving the ability of acid whey to be processed.

Key words: lactose, acid whey, lactic acid

INTRODUCTION

Lactose is the major carbohydrate in milk and consequently one of the main components of whey streams. Whey is the serum or liquid portion of milk remaining after coagulation and casein removal. Moreover, this whey contains more than half of the nutrients in original whole milk (Vasiljevic and Jelen, 1999). Depending on the manufacturing process, whey has been classified into 2 major groups, namely sweet whey, which originates from hard cheese and rennet casein production, and acid whey with a pH of ~4.5 expelled during production of acid coagulated dairy products such as cream, ricotta, and cottage cheeses or strained (Greek style) yogurt (Schmidt et al., 1984). In the past, whey streams were disposed of by spreading onto the fields or simply dumping into the ocean. However, the presence of milk solids, mainly lactose in whey, created a significant environmental issue due to its high organic load and a high biological oxygen demand required for its decomposition. The only sustainable way to reduce this environmental effect has been to extract and concentrate solids from whey. Hence, sweet whey has been widely used for the production of different powders (lactose, whey proteins), which are currently used as ingredients in many food and pharmaceutical applications (Chandrapala et al., 2015).

In contrast to sweet whey, an industrial solution for acid whey processing has not been found likely due to its compositional difference (Chandrapala et al., 2015). Acid whey contains higher levels of minerals, but less proteins and lactose depending on different processing conditions, and more importantly, greater concentration of lactic acid than sweet whey (De Wit, 2001). Acid whey is considered a hard to process stream mainly due to failure of lactose to crystallize, which apparently remains in its amorphous form during concentration and thus hinders further processing. Amorphous lactose exists in a glassy, solid state or in syrup-like, super-cooled liquid state. The state transition of an amorphous solid is described by a second order known as glass transition (T_g); T_g is defined as the temperature at which an amorphous system changes from glassy to the rub-

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Received June 14, 2015.
Accepted September 2, 2015.
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bery state. This can either take place by increase in temperature or increase in the plasticizer amount (e.g., water content) or both. Molecular mobility is limited below T_g, whereas molecular mobility increases above T_g, resulting in a greater potential for chemical interactions between components. This in turn can substantially change structural properties (Bhandari, 2008).

Relative low sweetness, crystallization properties, flavor-emphasizing abilities, and protein stabilization properties make lactose an important ingredient for various food applications. The industrial extraction of lactose is traditionally achieved by concentration and crystallization of whey, whey permeates, or milk permeates. Whey is pasteurized and concentrated to about 60 to 65% (wt/wt) of TS by vacuum evaporation (De Wit, 2001). Consequently, the concentrated whey is subjected to rapid cooling followed by slow cooling to induce lactose nucleation and crystal growth, which are the main steps of the crystallization process. Subsequently, lactose crystals are separated by centrifugation or sent to spray drying. Although this process appears simple, the lactose recovery from whey depends on numerous factors such as the degree of supersaturation, solution temperature, viscosity, concentration of lactose, presence of minerals, presence of organic acids, and pH (Jelen and Coulter, 1973b; Bhargava and Jelen, 1996; Ganzle et al., 2008).

Whereas the presence of inorganic acids improves the rate of lactose crystallization, the presence of lactic acid appears to act as an inhibitor hindering the crystallization process (Jelen and Coulter, 1973a; Ganzle et al., 2008). Increasing the lactic acid concentration from 1 to 20% (wt/wt) decreased the yield of spray-dried lactose powders (Saffari and Langrish, 2014). Furthermore, presence of minerals in lactose solutions may have a contradicting outcome, resulting in an enhanced or diminished crystallization rate depending on the concentration and the type of the salt present (Jelen and Coulter, 1973a). A change in solubility of lactose was assumed to be the cause for the crystallization behavior changes in the presence of impurities (Mullin, 1979). These solubility changes can be due to structural changes of water molecules in bulk water. The presence of ions with high charge density (Ca^{2+}, Ba^{2+}) are believed to orient the water structure due to their ability to electrostatically organize the structure of water. However, some ions with low charge density (Cl^-, Br^-, I^-) led to perturbation and structure breaking of water molecules (Von Hippel and Schleich, 1969). Hence, changes in water structure directly affect the solubility and thereby the supersaturation of the lactose solution, which is one of the leading factors governing the lactose crystallization (Mullin, 1979).

Current understanding highlights the importance of the presence of minerals, whey proteins, and lactic acid for elucidating kinetics of lactose crystallization. Because one of the major compositional differences between acid and sweet whey is the presence of lactic acid, it would be beneficial to establish the influence of the presence of lactic acid toward the change in the behavior of water and thereby predict the crystallization behavior of lactose model solutions. A systematic study is lacking in this regard especially during the concentration step, because this is also considered a crucial step during processing of acid whey and thus recovery of lactose crystals. Hence, the present study aimed at establishing the structure and behavior of water molecules surrounding lactose as a function of concentration (15 to 50% wt/wt) in the presence or absence of lactic acid. Furthermore, the crystallization nature of lactose obtained from 50% (wt/wt) lactose solutions with or without lactic acid was established.

MATERIALS AND METHODS

Materials

A commercial food-grade lactose powder and an analytical-grade lactic acid solution were obtained from Murray Goulburn Co-operative Co. Ltd. (Brunswick, Australia) and Sigma-Aldrich Pvt. Ltd. (Castle Hill, NSW, Australia), respectively. Milli-Q water was used at all times for the preparation of solutions.

Preparation of Solutions

Lactose powder (5 g) was dissolved in 95 g of Milli-Q water at room temperature to obtain a 5% (wt/wt) lactose solution. Lactose plus lactic acid (lactose + lactic acid) solution was prepared by the addition of 1 g of lactic acid to the 5 g of lactose powder and made up to a total weight of 100 g. Concentrations of 5% and 1% (wt/wt) were selected to imitate the lactose and lactic acid concentrations present in acid whey, respectively. The solutions were stirred using a magnetic stirrer for about 30 min until clear solutions were obtained with no visually observable powder particles. All the solutions were filtered using 0.45-μm Whatman filter papers to remove any undissolved particles. The pH of the lactose and lactose + lactic acid solutions were 5.8 and 4.9, respectively.

Concentration of Lactose and Lactose + Lactic Acid Solutions

Concentration of lactose and lactose + lactic acid solutions was performed using a rotary evaporator