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Unusual drying behaviour of droplets containing organic and inorganic solutes in superheated steam

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ABSTRACT

There are currently limited studies on the effect of superheated steam (SHS) on the solidification behaviour of droplets with dissolved solids which is a core phenomenon in spray drying. In this work, the single droplet drying technique was used to investigate how SHS affects the particle formation process of organic sugar droplets (lactose and mannitol), protein droplets (whey protein isolate) and droplets containing inorganic solute (sodium chloride). Unexpected droplet drying phenomenon was observed differing from drying behaviour typically assumed in the literature. While SHS is well known to result in slower drying rates below the inversion temperature, it also exhibits a lower potential to remove moisture at the end point 'pseudo-equilibrium' stage of drying. This led to lactose and mannitol droplets encountering incomplete moisture removal challenges in its drying process. The higher constant rate drying temperature experienced by droplet in SHS led to the precipitation of ultrafine protein particles. This higher constant rate drying temperature also led to the production of inverted pyramidal shaped salt crystals highlighting the potential of SHS as a useful medium for spray drying crystallization control.

1. Introduction

Hot air is the typical standard medium used in most drying related processes. For specific applications and product requirements, the usage of different drying mediums were also reported (Mujumdar et al., 2010). Some of the reported studies include the use of humid air to control the evaporation rate in the dehydration process (Ebrahimi and Langrish, 2015; Islam et al., 2010). There were also reports focusing on the use of carbon dioxide and ethanol vapour to enhance product properties and induce crystallization and precipitation of particles (Brown et al., 2008; Mansouri et al., 2012; Mansouri et al., 2013; Tan et al., 2014; Tan et al., 2015). The use of nitrogen as a dehydration medium is also widely adopted to prevent oxidation and as a safety blanket during the dehydration of volatiles (Mujumdar, 2006). The premise of this report is on the application of superheated steam on the dehydration process, specifically on liquid droplets.

The use of superheated steam (SHS) in drying processes has been gaining much research attention in the past decades. To date, the application of SHS in various drying processes has covered a diverse range of products, predominantly in food processing, timber processing as well as in the waste treatment industry (Pang and Pearson, 2004; Shiravi et al., 2007; Speckhahn et al., 2010; Van Deventer and Heijmans, 2001; Yamsaengsung and Sattho, 2008; Zielinska et al., 2015; Devahastin et al., 2004). Studies have shown that the substitution of hot air with SHS have resulted in the enhancement of either product quality or process efficiency and thus is able to counteract some of the shortcomings in conventional hot air-drying process. A study on SHS drying of parawood produced a significant decrease in drying time, from 8 to 16 days to less than 35 h and less energy usage while maintaining the same dried wood quality (Bovornsethanan and Wongwises, 2007). Another study on beef drying under both hot air and SHS condition showed that SHS drying resulted in a shorten drying time as well as the reduction of lipid oxidation reactions which has minimized undesirable quality changes (Speckhahn et al., 2010). Investigations on the use of SHS in the drying of waste sludge revealed several advantages such as simultaneous disinfection during drying which extends the storage life of biomass, eliminate odour as well as extensive energy recovery as high as 80% (Hoadley et al., 2015; Jarrett et al, 1996). A drying study on reconstituted whole milk also showed minimisation of lipid oxidations and greater nutritional retention in milk

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powders as a result of SHS drying (Ghazwan Mahdy, 2014). Apart from the potential to improve product quality, investigations on the energy efficiency of a SHS pilot study also revealed the possible recovering nearly 85% of the input energy (Robert, 1985). SHS drying models and trials have also shown positive energy savings with one case study involving SHS impinging drying of paddy rice study, reporting approximately 25–30% energy reduction as compared to air drying (Li et al., 2016; Romdhana et al., 2015).

While most of these reports mainly focused on the drying of solids or suspensions, there are currently minimal reports on investigating the fundamental use of SHS for liquid drying such as those in spray drying. The potential application of SHS in spray drying was first discovered by Gauvin. W.H (Gauvin, 1981, 1983). Subsequent reports in the literature on this area, which were rare, focused mainly on measuring or understanding quantitatively the heat and mass transfer of droplets dried under superheated steam conditions. These studies ranged from SHS single droplet studies (Trommelen and Crosby, 1970) to large scale SHS computational fluid dynamics studies (Frydman et al., 1998, 1999); Ducept et al., 2002. Although these reports examined the mass and temperature changes droplets with dissolved solute, the reports neglected detailed investigation into the entire solidification process of droplets. The direct effect of SHS on the solid formation from a solute containing droplet and the final particle morphology have yet to be fundamentally examined. Therefore, this presents a gap in the literature in which this report aimed to fill.

In a previous publication, using the single droplet drying technique, Lum et al. (2018) investigated in detail how single milk droplet is solidified under SHS conditions. It was found that SHS produced single particles which were more hydrophilic and displayed improved wettability characteristics when compared to hot air drying. Milk is a complex system constituting carbohydrates, fats, proteins and minerals. In this work, we extended the same experimental technique to investigate in a more fundamental approach on individual classes of materials, namely droplets containing: slow to crystallize organic solutes (lactose), fast to crystallize organics solutes (mannitol), inorganic solutes (salt) and dissolved protein (whey isolate). The aim here was so that a more general conclusion can be made on how SHS affects the different types of materials typically found in food and pharmaceutical spray dried products.

2. Materials and method

2.1. Sample preparation drying

Lactose, mannitol, protein and salt solutions with 10 wt% were prepared with a minimal stirring of 4 h for lactose (α lactose monohydrate, Sigma Aldrich) and 15 min for mannitol (D-Mannitol, Sigma Aldrich). Whey protein isolate (WPI) was used as the model protein material (100% Whey Protein Isolate, Bulk Nutrients Pure Supplements) and table salt was used (Natural Rock Salt 99.8% NaCl, SAXA).

2.2. Single droplet drying experiments

A detailed schematic of the experimental setup is given in the previous report (Lum et al., 2018). In the experiment, a single droplet of the sample solution was suspended via a glass filament designed for single droplet studies. The single droplet with a volume of 2 ± 0.05 μL was generated with a 5-μL plunger syringe (Fixed Needle - Plunger Protection Syringe, 5F, SGE Analytical science). The suspended droplet was exposed to a stream of hot air or SHS at the desired temperature and allowed to dehydrate until a specific time duration. This time duration was determined from preliminary tests in which there were no further changes in the morphology of the droplet during the drying process. Experimental conditions used are tabulated in Table 1. Each sets of drying conditions were repeated 5 times to ensure the observations presented were reproducible.

Table 1
Summary of drying conditions used in the study.

Drying Conditions	
Medium Flow Rate	0.75 m/s
Solution Concentration	10 wt%
Droplet size	2 μL
Drying Temperatures in both mediums	
Lactose and Mannitol	110 °C, 130 °C and 145 °C
Protein	60 °C (air only), 110 °C
Salt	110 °C, 130 °C and 145 °C

2.3. Drying history analysis

The entire drying process of each droplet was monitored and recorded with a digital HD video camera recorder (Sony HDR- PJ430) with 3 full sets of close up-lens (Hoya 46 mm Multicoated Close-up Lens Set including +1, +2, +4 Diopter Strengths). On top of providing visual monitoring of the solidification process, the effect of air and SHS at different temperatures on the drying rate of the droplet was also analysed, delineated by the rate of shrinkage of the droplet. The rate of shrinkage of the droplet was quantified from the initial constant rate period of drying in which the droplet was still liquid like prior to the onset of droplet surface skin or crust formation; beyond which the drying behaviour does not correlate with shrinkage. Changes in droplet volume were analysed using ImageJ (National Institutes of Health, Bethesda, MD). For air-dried droplets, the rate of volume shrinkage of the droplet was determined from volume changes in the first 10 s of dehydration. In SHS runs, quantification of the rate of droplet volume shrinkage was determined after the initial condensation period, with the analysis obtained from changes in droplet size between the time period of 15–45 s of the drying process. The rates of volume shrinkage calculated are reported as the percentage of volume shrinkage per second, averaged over triplicate runs. The uncertainty reported describes the span of the data from the triplicate runs.

2.4. Scanning electron microscopy (SEM) analysis

Structural features and morphologies of the dehydrated and solidified droplet were analysed with Scanning Electron Microscopy (Phenom, Australia). Samples were fixed onto a SEM holder with a double-sided adhesive carbon tape mounted on the SEM stubs. The samples were sputter coated with platinum for electron conduction prior to the analysis and viewed under 5 kV of accelerating voltage.

3. Results and discussions

3.1. Organic material: lactose and mannitol

Comparison of lactose droplets experiments with hot air and SHS at the same temperature of 110 °C revealed a delay in shrinkage or size reduction when the lactose droplets were dried under SHS with an average percentage volume shrinkage per second of 1.45 ± 0.64% and 3.78 ± 0.71% for SHS and air, respectively. This can be further observed in Fig. 1 by comparing the size of the droplet throughout the shrinking phased of the droplet drying. Similar reduced SHS evaporation rates, particularly for low superheating temperature below the inversion temperature was also previously reported in the literature (Haji and Chow, 1988; Bond, 1991; Devahastin and Mujumdar, 2014). Drying of droplet containing various types of dissolved solids at 150 °C conducted by (Trommelen and Crosby, 1970) had also revealed lower drying rates in SHS when compared to air. In their work, the inversion temperature was found to be at approximately 250 °C.

The possible effect of initial condensation of SHS in delaying droplet