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Original Research Paper

In-situ crystallization of particles in a counter-current spray dryer ☆

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ABSTRACT

This paper evaluated the potential of counter current spray drying to produce engineered crystalline particles in-situ. One main feature of the counter current spray dryer is in inducing droplet drying history with a progressively increasing drying rate; in contrast to a co-current spray dryer in which the particle is immediately confronted with high driving force of evaporation. In addition, counter current dryer provides higher residence time for particles within the drying chamber. This work explored the manipulation of these unique features of counter current spray drying to control the crystallinity of the particles formed. Sucrose, lactose and mannitol were spray dried as model materials exhibit contrasting crystallization. Counter current spray drying was suitable in producing well defined crystalline lactose particles mainly due to the higher residence time. The produced lactose particles were composed of agglomeration of fine thin lactose crystals. Surprisingly, the counter current spray drying with relatively lower initial evaporation rate resulted in amorphous mannitol particles. Fully crystalline mannitol was produced when the feed spray temperature was elevated leading to rapid supersaturation. This may be an important strategy to control the crystallinity of mannitol particles particularly for pharmaceutical application. Similar feed pre-heating strategy in conjunction with counter current spray drying was used for sucrose drying. Crystalline sucrose was achieved with relatively lower drying temperature. This will be a valuable strategy for producing free flowing sucrose particles specifically for the food industry.

1. Introduction

Crystalline particles in general exhibit lower tendency to agglomerate, cake and undergo phase change; hence, these features provide appropriate stability for such particles. Various methods for producing crystalline particles are available and each of them faces some drawbacks and advantages [1]. Batch crystallization with subsequent milling is the most common method for producing crystalline particle [2]. Although advanced milling techniques are emerging, this process increases surface energy (generation of amorphous phase) of particles which decreases the free flowing behaviour and stability of the particles [3]. Utilization of the anti-solvent precipitation method facilitates production of engineered crystalline particles but flammability of anti-solvent, typically alcohol based, significantly limits its application particularly in larger scales [4,5]. Spray drying is a well-known method for producing engineered amorphous particles. There are reports on the production of amorphous particles in spray drying and followed by subsequent treatment to induce phase transition within particles. During this phase transition the moisture content of the particle may contribute to particles stickiness; subsequently, large particle chunks are produced which reduces the free flowing ability of the particles [6,7]. However, the main advantage of the spray drying process is the flexibility of this method to directly produce particles in specific size ranges in a single stage without further treatment like milling [8].

In situ crystallization in spray drying is a new method of producing engineered crystalline particles. Although crystallization occurs randomly for the particles through drying, this method was scientifically investigated by Shastry and Hartel for producing crystal sucrose by a continuous drying method of 50 μm sucrose stream [9]. They proved that with change in the drying process crystals can be produced in a continuous manner. Chio and Langrish utilized the potential of a one-step crystallization approach and employed spray drying for producing partially crystalline lactose particle [10,11]. Complementary research was carried out using single droplet experiments [7,12] and in lab scale spray dryers [11,13,14] which proved that some degree of crystallinity is feasible via this method.

The challenge of crystalline particle formation is the relatively long time scale nature of crystallization [15]. In contrary, spray

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drying is a fast drying method in which most of the moisture is evaporated and the total residence time in the order of seconds, which conventionally hinders crystallization. To overcome these features one possible approach is to reduce the evaporation rate to avoid early particle formation giving more time for crystallization before the particle solidify; however, formation of slightly non-dried particles may be ensue. The threshold between the formation of amorphous and crystalline particle is very narrow depending on the drying history of the particles [16,17]. On this basis, production of engineered crystalline particle necessitates the understanding and control of the mechanism of heat and mass transfer affecting the crystal formation. In other words, it is important to control the heat and mass transfer to ensure the required time for crystallization but yet produce sufficient drying.

In a typical co-current spray dryer, droplets reach the drying air velocity very rapidly after injection. Therefore, the drying air dominates the particle movement and will subsequently affect residence time of particle in the dryer [18,19]. In the absence or with non-significant relative difference in velocity between the particle and the drying air, effectively, diffusion of water vapour from the droplet is the main mode of heat and mass transfer for the most part of the particle trajectory. It is noteworthy that diffusive heat and mass transfer will be less intense when compared to forced convection. Compounded by the progressively decreasing air temperature as droplet travels to the outlet, this necessitate a high air temperature at the inlet to compensate for the lower evaporation rate. Increasing the drying temperature leading to rapid initial evaporation contributes to early crust formation or solidification and is one of the key factors that would hinder crystal formation (Fig. 1). Most spray drying in-situ crystallization works *hitherto* were reported for co-current dryer.

In a counter current dryer, however, the inverse direction of drying air relative to the trajectory of the particle, amplifies the rate of heat transfer as forced convection is the dominant mode of drying; there is always a relative difference in the opposing velocity between the particles and drying air. In addition, the drag force of the air which is against the gravitational force, resists the falling motion of the droplet to provide a longer residence time (Fig. 1). This can potentially allow the decrement of the inlet drying temperature which will allow a longer time for particle solidification. It is the hypothesis of this work that these factors will provide conditions suitable for crystallization. There is limited published research on the effect of different drying history in a counter current spray dryer on the in-situ particle crystallization process. This research aimed fill this gap in knowledge by providing an insight on how this factor coupled with a longer residence time in a counter current spray dryer can be used to manipulate the in-situ crystallization process for production of crystalline particles.

2. Materials and equipment

2.1. Solution preparation

Three types of sugars sucrose, lactose and mannitol, which are mainly used in pharmaceutical and food industry, were selected for this work. Lactose and mannitol are two main materials employed as excipients and their characteristics play a critical role in the efficacy of drug delivery [20]. The other criterion for selection of these materials was based on the difficulty of these particles to dry and crystallize in spray drying. Compare to other two sugars mannitol has a very low glass transition temperature which allows rapid crystallization [21]. In turn, sucrose is well known to be difficult to dry and crystallize particularly because of its inherently high viscosity during drying. Lactose, although does not exhibit such high viscosity during drying, has a relatively higher glass transition in comparison with other sugars giving it a lesser propensity to crystallize. Therefore, for the purpose of this work, these three materials will provide contrasting crystallization behaviour during the spray drying process. For consistency, all the initial solute solutions were prepared in 10 wt% concentration to reduce the number of variables affecting the spray drying process. The solutions were prepared with distilled water in ambient temperature and stirred for 60 min to ensure feed is totally dissolved. Only lactose solute was stirred for at least 4 h to ensure an equilibrium of α and β has been achieved [22].

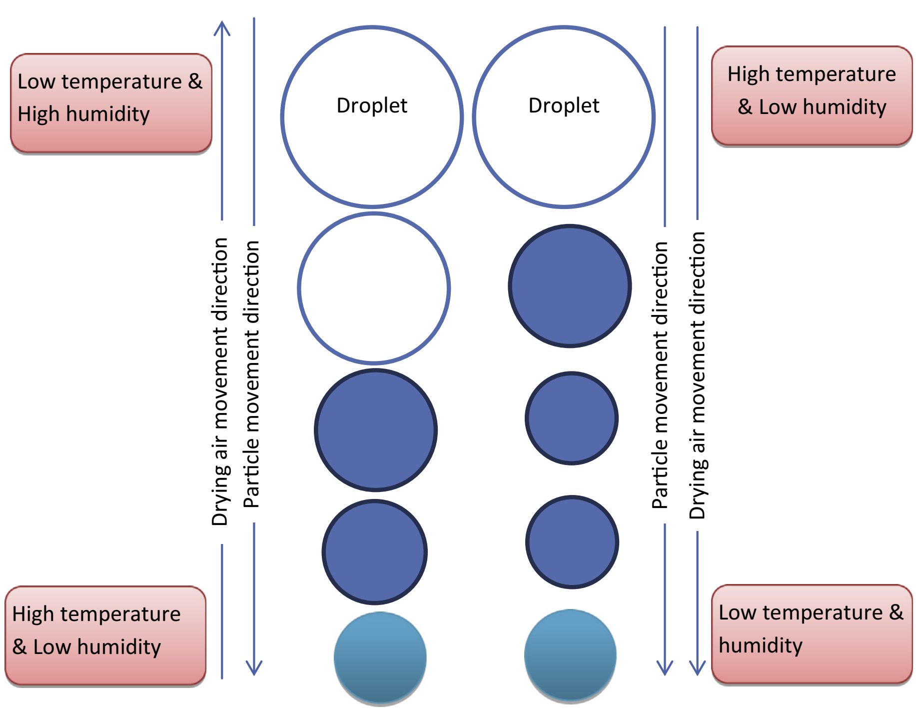

Fig. 1. Comparison of particle drying history: (left) counter current (right) co-current spray dryer.