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Numerical and experimental study of the drying of bi-component droplets under various drying conditions

Holger Grosshans [a], Matthias Griesing [b], Mathias Mönckedieck [c], Thomas Hellwig [b], Benjamin Walther [b], Srikanth R. Gopireddy [d], Robert Sedelmayer [b], Werner Pauer [b], Hans-Ulrich Moritz [b], Nora A. Urbanetz [d], Eva Gutheil [a,*]

[a] Interdisciplinary Center for Scientific Computing (IWR), University of Heidelberg, Germany
[b] Institute for Technical and Macromolecular Chemistry, University of Hamburg, Germany
[c] Department of Pharmaceutics and Biopharmaceutics, University of Kiel, Germany
[d] Daiichi-Sankyo Europe GmbH, Munich, Germany

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ABSTRACT

This paper presents a combined experimental and numerical study of the evaporation and solid layer formation of single bi-component mannitol–water droplets in hot air. Experimentally, the process of droplet evaporation and drying is studied in a custom-built acoustic levitator. The experimental results are compared with numerical simulations of spherically symmetric bi-component droplets in an unsteady, one-dimensional configuration. The model includes evaporation and solid layer formation. This approach enables the comparison of the temporal variation of the droplet size and the porosity, which are related to the final particle sizes. The study is performed for various drying conditions and initial droplet sizes as well as compositions of the droplets. The objective of this paper is the derivation and validation of a suitable model to predict the properties of spray-dried mannitol particles, depending on their drying conditions. A design of experiments (DoE) is used to define suitable drying conditions and to analyze the results. The study includes initial droplet diameters varying from 350 μm to 550 μm and initial mannitol mass fractions in water droplets ranging from 5% to 15%. The surrounding air temperature is varied from 80 °C to 120 °C. Additionally, different relative humidity of the surrounding air between 1% and 7.5% is studied. Based on the DoE, correlations for results from both experiments and simulations including the temporal evolution of the droplet surface area and the final particle size are derived and discussed. Major influences are identified that dominate particle drying characteristics.

1. Introduction

Spray drying is a widely used technique in industry to convert a multi-component spray into a powder. For the industrial use of the powder, the properties of the individual particles, e.g. size, porosity, shape and surface, are of utmost importance. Therefore, there is a high interest to predict and to control the final particle properties through particular drying conditions. Mannitol powders are produced industrially in spray dryers on a large scale. This sugar is extensively used in food product preparation, and it is under investigation for use in inhalation drug formulations in pharmaceutics [8,18,19]. A system of coarse carrier particles combined with small cohesive drug particles (e.g. salbutamol sulfate) is established for dry powder inhalers, which are widely used to treat lung diseases such as asthma and chronic obstructive pulmonal disease [27]. Exploiting its cohesiveness, this carrier is utilized to overcome dosing problems. The drug particles are detached from the carrier by shear forces during inhalation, and entrained by the air flow to the deeper parts of the lungs, where the pharmacological effect takes place. In this system, commercial products usually consist of lactose monohydrate as carrier particles. Drawbacks such as lower storage stability due to the amorph structure after the spray drying of lactose monohydrate and lactose intolerance led to the investigation of spray dried mannitol drug carrier particles within the last decade [43]. Mannitol is completely crystalline after spray drying, which simplifies the storage. The potential use of mannitol as carrier particles for dry powder inhalers encouraged many research groups to focus on the properties of the mannitol particles. Maas et al. [29] and Littringer et al. [25,28,26] dealt with the spray drying of mannitol at different drying conditions to gain particles of different size, porosity, and surface morphology.

* Corresponding author. Tel.: +49 6221 546114.
E-mail address: gutheil@iwr.uni-heidelberg.de (E. Gutheil).

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Significant dependencies on the drying temperature, mass fraction, and droplet size were detected, but the shell formation of the single droplets is still under investigation. In a preliminary study [41], first insight in the solid layer formation and surface texture of levitated mannitol droplets at different drying conditions was presented, but the influence of the initial droplet diameter is an open question.

The study of single droplet drying provides valuable insight into important elementary processes, which helps to understand the complex spray drying process especially regarding the shell formation.

A previous numerical study [12] showed that the evaporation and solid layer formation of a single polyvinylpyrrolidone–water and mannitol–water droplet can be modeled in a suitable way, and this model now is used as base to investigate the drying behavior of single mannitol–water droplets in the context of new experiments using an acoustic levitator to study single droplet drying.

The acoustic levitator is a technique for containerless positioning of small samples, and it is widely used in various applications such as analytical [51] and bioanalytical [38,50] chemistry, material sciences [24,47,46,49], environmental sciences [52,17], and pharmaceutical applications [11,39,36]. A detailed overview is given by Priego-Capote and de Castro [35]. Acoustic levitation for the study of drying kinetics of bi-component droplets includes some unwanted side-effects. The exposure of a droplet to an acoustic field causes an inhomogeneous distribution of forces, leading to an oblate droplet shape. Moreover, the acoustic field causes an acoustic streaming around the droplet [48], which has an effect on the mass transport of the boundary layer in the vicinity of the droplet surface [20,54]. Convective vortex generation inside the droplet is induced by acoustic streaming [53], which does not occur during typical spray drying. However, acoustic levitation is considered to be the most useful method to investigate the single droplet drying behavior among container-less techniques.

Sano and Keey [37] studied the drying behavior of colloidal material into a hollow sphere by considering the convection of solid matter towards the center of the droplet [34]. In other studies [34,3,33,10], the solid formation and the temperature gradient inside the droplet were neglected.

The drying characteristics of a colloidal silica droplet, including the formation of a solid layer on the surface, was considered in the study of Nešić and Vodnik [34]. They classified various stages of droplet evaporation and drying, which serve as a base for numerical models [10,5,37,3,12]. A review is provided by Gopireddy and Gutheil [12]. A detailed survey of existing models of evaporation and drying of single droplet containing dissolved and insoluble solids is given by Mezhericher et al. [31].

The aim of the present study is to determine significant drying parameters and their influence on the drying kinetics of a mannitol–water droplet and the final particle. The parameters under consideration correspond to the variable conditions of a spray drying process, i.e. the diameter and solute content of the initial droplet and the temperature and humidity of the surrounding air. For this purpose, a combined experimental and numerical approach is chosen.

The influence of the initial mannitol mass fraction in water, the drying temperature and droplet size on the drying process is investigated experimentally. From these results, the relevant physical phenomena are identified for each drying condition, and the numerical simulations are performed for validation purposes.

Both experimental and numerical results are analyzed using DoE to provide a detailed statistical analysis of the influence of the considered parameters on the surface reduction during evaporation and the final particle diameter. The significance of the individual parameters and their interaction is analyzed, and correlations are provided to predict the final particle properties based on the drying conditions and the initial droplet properties.

2. Experiment

This section provides the experimental setup, the choice of experimental conditions in the framework of DoE and the methods to extract the relevant information for comparison with the numerical simulations.

2.1. Experimental setup

The experiments concern Mannitol Pearlitol 200 SD, which was donated by the company Roquette Pharma and used without any modifications. The single droplet drying experiments were carried out using a custom-built acoustic levitator at a frequency of 42 kHz, since all commercial available levitation systems have some disadvantages such as a low temperature range or limited range of application. The present study makes use of different peripheral equipment including dispensers, process chambers for controlling ambient conditions, or different illumination techniques, and it was possible to design the levitator system to suit all present requirements. The setup is described in more detail by Laackmann [22]. Fig. 1(a) shows a schematic of the experimental configuration.

The acoustic levitator essentially consists of the transducer, the sonotrode and the reflector, which are surrounded by a process chamber of dimensions 60 mm × 60 mm × 200 mm. The process chamber ensures the monitoring and adjustment of the ambient conditions given in the next section. The gas temperature is controlled by a heating band on the outside of the process chamber as well as a heated gas flow regulated by a mass flow controller (MFC 8711, Bürkert GmbH & Co. KG). The equipment is suitable for flow rates up to 5 L/min. The gas (compressed air or inert gas) is enriched with vapor by a custom-built vaporizing unit to guarantee fixed conditions. The amount of vapor can be adjusted by using a HPLC pump (type 301, Flom). The humidity was monitored by a humidity probe (HMT337, Vaisalla). Before an experiment was initiated, the temperature in the pressure node is controlled by a thermocouple (type K, diameter 150 μm, ES Electronic Sensors GmbH).

The mannitol was dissolved to the required mass fractions in distilled water. This solution was inserted into the acoustic levitator by using a piezo-driven dispenser (MD-K-130, microdrop Technologies GmbH), which generates droplets in the picoliter-range, and these were accumulated during a droplet build-up period lasting up to 1 s until the desired initial droplet size was achieved. An exemplary image of the droplets in the levitator is displayed in Fig. 1(b).

2.2. Experimental methods

Shadowgraphy images of the droplet were taken to investigate its drying behavior using a CMOS camera (PL-A741, PixelLINK). Every 80 ms, an image was evaluated using an online data processing program, which is written in LabVIEW. Due to the acoustic force distribution in an acoustic field, the shape of the droplet was somewhat ellipsoidal. Hence, online analysis was used to fit an ellipse over the droplet shadow and to determine the major and minor axes of the ellipse. These diameters were used to derive an equivalent spherical diameter in the presentation of the results and for comparison with the simulations.

When the initial droplets are generated, there is a certain tolerance in their initial diameters, i.e. the specified values of 350 μm, 450 μm and 550 μm are usually not met exactly (±5%). In