CHEMICAL ENGINEERING TRANSACTIONS

VOL. 39, 2014

A publication of AIDIC
The Italian Association of Chemical Engineering
www.aidic.it/cet

Guest Editors: Petar Sabev Varbanov, Jiří Jaromír Klemeš, Peng Yen Liew, Jun Yow Yong

ISBN 978-88-95608-30-3; **ISSN** 2283-9216

DOI:10.3303/CET1439167

Study on Spray Drying for the Production of High Value Particles

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The drying mechanism of high-value particles of hydroxyapatite in an industrial spray dryer was investigated numerically using the developed models and compared with the drying mechanism of silica droplets of the same final size. Similar mechanisms were observed for the drying of slurry droplets of both materials, but the rate of heat and mass transfer between air and droplets was lower, and the heat transfer resistance and heat accumulation in the dry crust was larger in the case of drying of hydroxyapatite droplets. The hydroxyapatite droplets were dried in a longer axial distance in the dryer, and the air temperature decreased and humidity increased more slowly during drying of hydroxyapatite droplets than in the case of silica. The temperature difference between the outer droplet surface and evaporation interface was significantly larger in the hydroxyapatite droplets than that in the silica due to the difference in thermal properties of solid materials. Less water vapour was accumulated in the dry crust of hydroxyapatite than in the crust layer of silica of the same thickness.

1. Introduction

Spray drying has recently been applied for production of various high value particles in the form of agglomerates of nanoparticles (Sollohub and Cal, 2010). Such agglomerates can be utilized in dry phase for further processing on industrial scale due to their enhanced flowability (Oi et al, 2013). At the same time, they possess the superior physical, chemical and mechanical properties owing to their nanosized constituents (Nandiyanto and Okuyama, 2011).

The agglomerate morphology, size and porous structure are determined by the rates of heat and mass transfer between the droplet and the drying medium, and inside the partially dried agglomerate. The drying mechanism is defined by the process conditions, slurry composition and droplet trajectories in the drying chamber (Wang et al., 2009).

Hydroxyapatite was selected as a promising material for a wide range of biomedical and other advanced applications. The composition of hydroxyapatite, as one of the thermodynamically stable phosphate salts in a body fluid, is similar to the mineral component of natural bones and teeth (Sadat-Shojai et al., 2013). It is used in the form of coating in manufacturing of composite bioceramic materials (Yang et al., 2014), microspheres for sustained release of drugs (Sun et al., 2009), nanoparticles for preparation of effective pH-responsive particulate emulsifier (Fujii et al., 2007), etc.

The knowledge of a detailed drying mechanism of a slurry droplet is essential for selection of operational conditions for production of advanced material. The drying kinetics of slurry droplets is conventionally described separately for two drying periods (Elperin and Krasovitov, 1995). Water evaporates from the outer droplet surface of the slurry at constant rate during the first period of drying resulting in droplet diameter shrinking, simultaneous increase of solid concentration inside the droplet and, eventually, the formation of a layer of solid component called a crust (Charlesworth and Marshall, 1960). This is a beginning of the second, falling rate, period of drying. In this period, the evaporation interface is located inside the wet particle and the drying rate is controlled by heat and mass transfer resistances in the growing crust layer.

The comprehensive mathematical model of spray drying of a slurry droplet was developed and validated in our previous studies (Julklang and Golman, 2012). The temperature and moisture concentration

Please cite this article as: Golman B., Julklang W., 2014, Study on spray drying for the production of high value particles, Chemical Engineering Transactions, 39, 997-1002 DOI:10.3303/CET1439167

distributions inside the crust and wet core regions were simulated taking into account both internal and external mass and heat transfer resistances during the constant and falling rate periods (Golman and Julklang, 2013). Furthermore, Julklang and Golman (2013) established a mathematical model for spray drying of slurry droplets of nanoparticles in the industrial-scale spray dryer. The model is based on the combination of a model for the heat and mass transfer for a single droplet with a model for the flow of droplets and drying gas, and the heat and mass transfer in the spray dryer.

The objective of the present study is to investigate the drying mechanism of slurry droplets of hydroxyapatite nanoparticles in an industrial-scale spray dryer for production of high-value agglomerates using the developed comprehensive mathematical model and compare with the drying mechanism of droplets of nanosized silica particles.

2. Methodology

2.1 Simulation setup

The computer simulation studies are carried out on the drying of droplets including solid particles in an industrial-scale spray dryer with a chamber of total height of 3.730 m consisting of cylindrical, 2.005 m high and 2.215 m in diameter, and conical sections. The slurry feed is supplied to the chamber concurrently with a drying air and a centrifugal-pressure nozzle is used for droplet generation.

The production rate of dried agglomerates is set at 100 kg/h and the final agglomerate size at 50 μm. The operational parameters are summarized in Table 1.

Table 1: A summary of operational parameters

Parameter	Value
Drying air mass flow rate	3.7 kg/s
Drying air temperature	110 ^oC
Drying air humidity	kg/kg
Slurry concentration	30 wt.%
Slurry temperature	30 ^oC

2.2 Drying process simulation

A comprehensive FORTRAN computer code is developed for the purpose of simulating the drying process in spray dryer. The spray dryer chamber is divided on small control volumes along the chamber height. The temperature, humidity and velocity of drying air and droplet velocities are assumed to be constant in the control volume. The drying model of a slurry droplet is then used in each volume for calculation of heat and mass transfer between the drying medium and the droplets. The system of partial differential equations with a moving boundary describing the conservation of energy and mass in the droplet is solved numerically by a finite difference method using an implicit scheme (Crank, 1984). The droplet moisture content and the average temperature are calculated using the moisture and temperature distributions in the radial direction of the droplet (Dalmaz et al., 2007). The droplet trajectories, and the temperature and humidity profiles of drying air in the chamber are calculated by numerical solution of the system of ordinary differential equations.

3. Results and discussion

The simulation results confirm that drying of hydroxyapatite slurry can successfully be accomplished in the present industrial-scale spray dryer to produce the final agglomerates of 50 micron under the drying conditions summarized in Table 1. The drying of droplets will be finished in short radial and axial distances in the chamber thus avoiding contact of moist droplets with a side wall of cylindrical and a bottom wall of conical sections and subsequent build-up of deposit, as shown in Figure 1.

The silica product particles of the same size and porosity as hydroxyapatite ones are selected for comparison purposes. However, the droplets of hydroxyapatite and silica are of different initial sizes because of the dissimilar material densities. The density of silica is 2,220 kg/m^3 and that of hydroxyapatite 2,462 kg/m^3. The initial size of droplets $d_{p,init}$ is evaluated by fixing the slurry concentration C_{sl}, porosity ε and radius of final product r_{fp} as

$$d_{p,init} = 2r_{fp}\left[(1-\varepsilon)+\frac{\rho_s(1-\varepsilon)}{\rho_w}\left(\frac{100}{C_{sl}}-1\right)\right]^{\frac{1}{3}} \quad (1)$$

where ρ_s and ρ_w are the densities of solid and water.

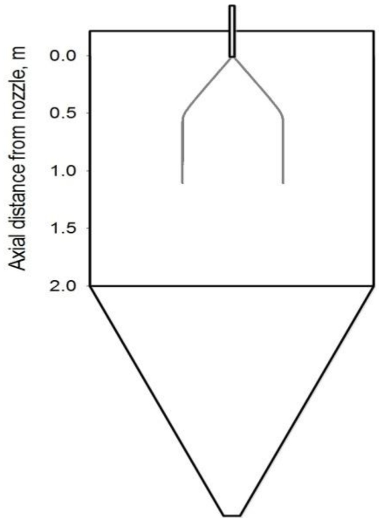


Figure 1: Trajectories of hydroxyapatite droplets in the dryer

A hydroxyapatite droplet is of larger initial size and it contains a larger amount of water than that of silica. As a result, the hydroxyapatite droplets are dried in a longer axial distance in the dryer in comparison with silica droplets, as illustrated in Figure 2(a).

The relative velocities of droplets of both material types bit differ in the initial heating-up period, but the difference vanishes in the constant and falling rate periods. Therefore, the effect of relative velocity can be neglected on the convective heat and mass transfer from the drying air to the droplet outer surface.

The droplets of both materials are of the same initial moisture contents specified by inlet slurry concentration. However, the moisture content of silica droplet decreases faster than that of hydroxyapatite droplet in the constant and falling rate periods, as illustrated in Figure 2(b), due to the high rate of convective heat and mass transfer between droplets and drying air.

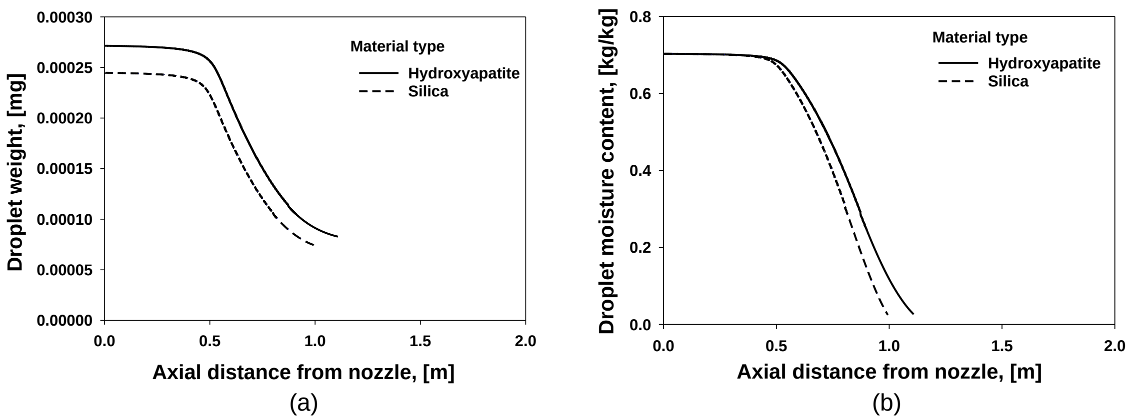


Figure 2: Profiles of (a) weight loss and (b) moisture content of hydroxyapatite and silica droplets in the axial direction of spray dryer

In the constant rate period, the rate of heat and mass transfer is high owing to the small size of silica droplets. Although the droplet sizes of both materials are identical in the falling rate period, the enhanced rate of heat and mass transfer in case of silica particles can be attributed to the high air temperature and low air humidity, as illustrated in Figure 3.

The air temperature decreases and humidity increases more slowly during drying of hydroxyapatite droplets than in case of silica, as indicated in Figure 4 (a) and (b). These results also confirm the lower rate of heat and mass transfer between the drying air and hydroxyapatite droplets. The larger humidity and lower temperature of outlet air in the drying of hydroxyapatite droplets results from the higher initial amount of water in the droplet in comparison with drying of silica droplets.

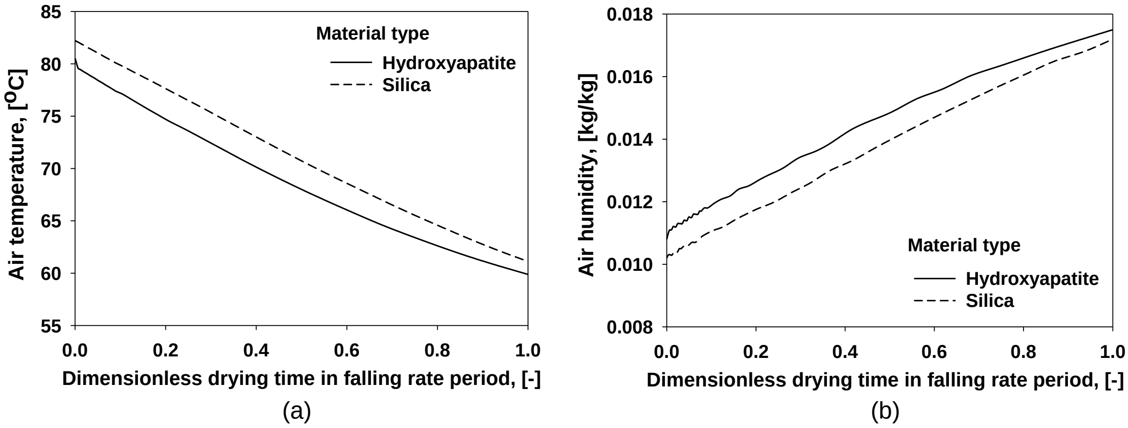


Figure 3: Variation of air (a) temperature and (b) humidity with drying time in the falling rate period

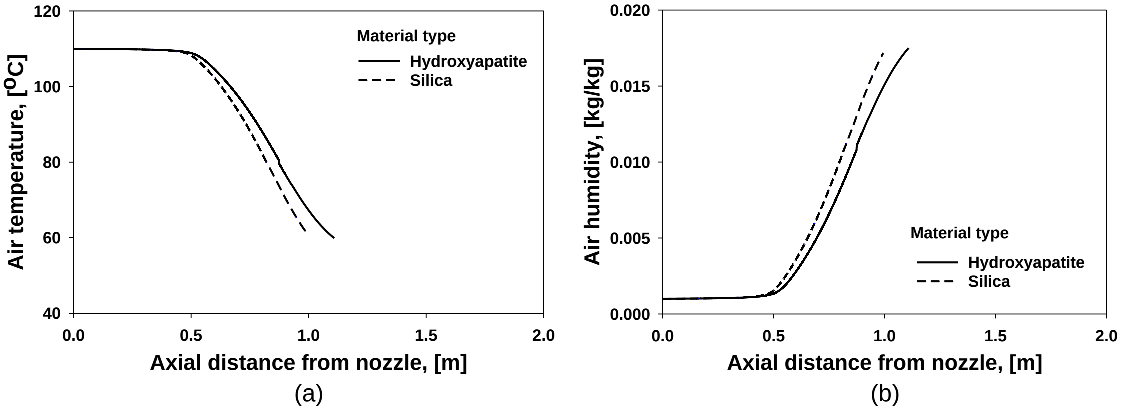


Figure 4: Profiles of air (a) temperature and (b) humidity in spray drying of hydroxyapatite and silica droplets

The hydroxyapatite droplets travel a longer axial distance in the dryer than silica droplets in the constant rate period due to the lower rate of heat and mass transfer of large droplets and larger amount of water to be evaporated during drying, as shown in Figure 5. In the falling rate period, the rate of average temperature increase of hydroxyapatite droplet is slower than that of silica owing to the declining rate of heat and mass transfer by low air temperature. The difference of average and surface temperatures in case of hydroxyapatite droplets is significantly larger than that of silica due to the difference in thermal properties of solid materials.

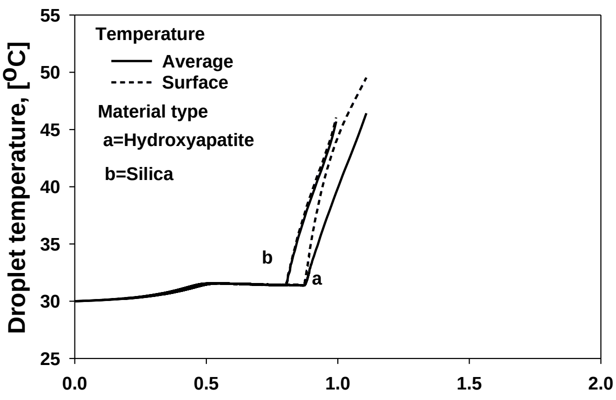
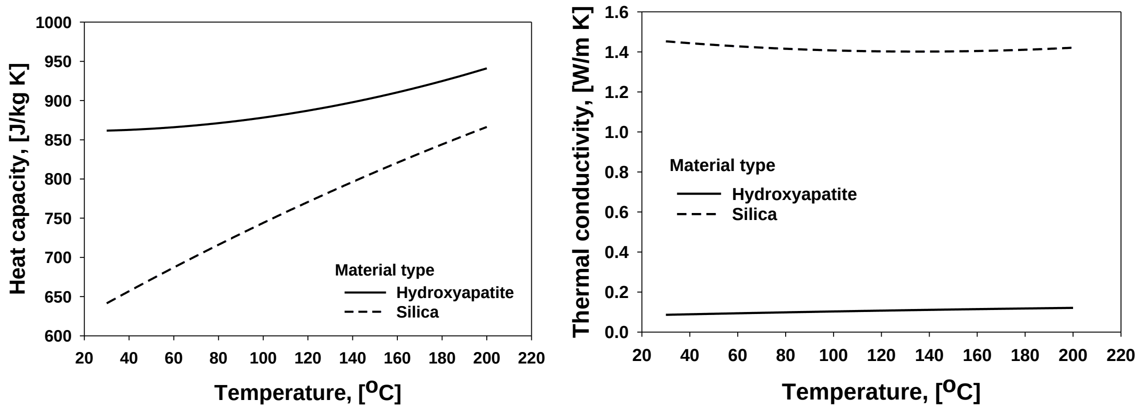


Figure 5: Profiles of average and surface temperatures of hydroxyapatite and silica droplets in spray dryer

A high heat capacity and low thermal conductivity of hydroxyapatite in comparison with silica, as illustrated in Figure 6, lead to the low rate of heat transfer from the outer droplet surface to evaporation interface by conduction through the crust layer and considerable heat accumulation in dry crust which results in a high surface temperature.

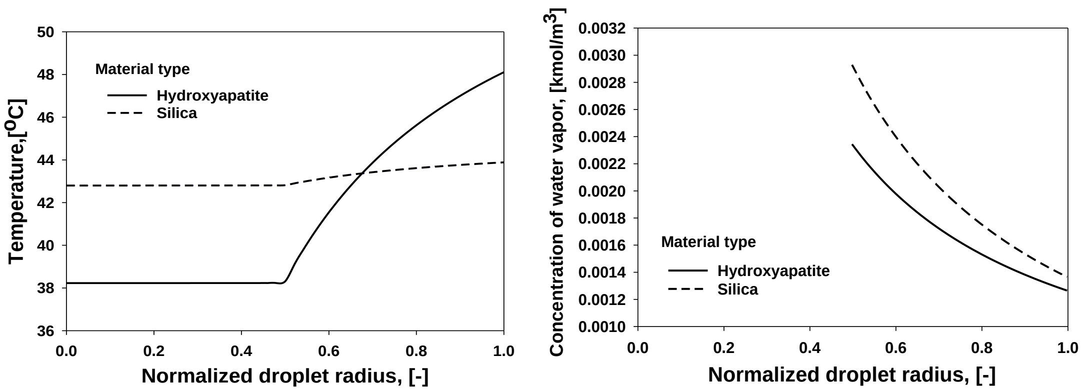


(a) (b)

Figure 6: Temperature dependencies of (a) heat capacities and (b) thermal conductivities of hydroxyapatite and silica

The temperature difference between the outer surface of hydroxyapatite droplet and evaporation interface is significantly larger than the one in the silica droplet, as shown in Figure 7 (a) for the same crust layer thicknesses. An appreciable heat transfer resistance in the crust layer of hydroxyapatite droplet may account for large temperature difference and high droplet surface temperature due to the low thermal conductivity.

The concentration profiles of water vapour in the dry crust layer of droplets are shown in Figure 7 (b). The concentration of water vapour at the evaporation interface of hydroxyapatite droplet is lower than that of silica droplet due to the low rate of water evaporation associated with the low heat supply to evaporation interface. A high crust temperature of hydroxyapatite leads to a high rate of mass transfer through the dry crust resulting in the lower water vapour concentration at the same thickness of dry crust.



(a) (b)

Figure 7: Distributions of (a) temperature and (b) water vapour concentration in hydroxyapatite and silica droplets

4. Conclusions

Using the developed model, the drying behavior of slurry droplets of high-value particles of hydroxyapatite and silica in the spray dryer were comparatively investigated during the initial heating-up, constant rate and falling rate periods by analyzing the profiles of air temperature and humidity, and the droplet velocity, average temperature and moisture content in the dryer axial direction as well as the distributions of temperature and water vapor concentration in the droplet.

The simulation results confirmed that drying of hydroxyapatite slurry can successfully be accomplished in the present industrial-scale spray dryer to produce final agglomerates of 50 μm without contact of partially dried particles with dryer walls.

Similar mechanisms were observed for the drying of hydroxyapatite and silica slurry droplets, but the rate of heat and mass transfer between air and droplets was lower, and the heat transfer resistance and heat accumulation in the dry crust was larger in the case of drying of hydroxyapatite droplets.

The hydroxyapatite droplets are dried in a longer axial distance in the dryer in comparison with silica droplets. The air temperature decreased and humidity increased more slowly during drying of hydroxyapatite droplets than in the case of silica. The moisture content of silica droplet decreased faster than that of hydroxyapatite droplet in the constant and falling rate periods. The temperature difference between the outer droplet surface and evaporation interface was significantly larger in the hydroxyapatite droplets than that in the silica due to the difference in thermal properties of solid materials. The lower amount of water vapor was accumulated in the dry crust of hydroxyapatite than in the silica crust layer of the same thickness.

Acknowledgements

The authors would like to thank the National Research Council of Thailand for their funding of this research.

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