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Influence of Drying Kinetics on Particle Formation: A Personal Perspective

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Taking the special opportunity of an anniversary issue of *Drying Technology*, a topic that may be expected to gain much importance in the future, namely, the influence of drying on particle formation processes and, thus, on the morphology and properties of the resulting product, is outlined. This article is not an in-depth review; it is intended as a capsule discussion based on recent experimental and computational results from the author's group on agglomeration, layering granulation, and coating in spray fluidized beds and spray drying. Reference is made to recent book chapters for more details. Thematically relevant articles from the 2011 volume of *Drying Technology* are quoted as a frame of reference for the placement of the discussion in the broader landscape of current drying research activities.

Keywords Coating; Drying rate; Particle morphology; Particle porosity; Spray drying; Spray fluidized bed; Wet granulation

INTRODUCTION AND SCOPE

An anniversary issue celebrating 30 years of *Drying Technology* offers a truly unique opportunity of providing perspectives on a topic that is expected to gain further importance in the next several decades, namely, product formulation. *Formulation* means to consistently aim at favorable end-use properties of solid products; for example, mechanical strength, kinetics of rehydration, or active ingredient release. Such properties typically depend on the internal structure of the solids. In some cases this structure is given and only needs to be preserved during drying. In other cases, however, the solid phase emerges during the drying process, so that drying kinetics on the one hand, and the structure and properties of the resulting solid particles on the other hand, are intimately interrelated. Then, drying is not just a separation process that takes place under restrictions defined by product quality considerations but the main step in product formulation.

A thorough treatment of formulation by drying could fill several review articles, but this is not the aim of this article. Instead, interesting effects shall be illustrated by referring to just a few examples of processes investigated recently by the

author's group. These are spray fluidized bed processes (spray fluidized bed agglomeration and layering granulation or coating) and spray drying processes (for solutions or suspensions, concentrating on single droplet behavior, not on the tower). The goal is to point out and explain as briefly and simply as possible the influence that the drying rate has on particle formation in these specific cases. Citation of recent publications will be given for readers interested in detailed documentation. Such readers may, additionally, take a look at two thematic threads of the *Modern Drying Technology* series on spray processes, one treating conventional spray drying^[1–3] and the other spraying on fluidized particles.^[4–6] Two of these chapters are especially interesting in the context of product quality and formulation, namely, Peglow et al.^[6] and Walzel and Furuta,^[3] for spray fluidized beds and spray towers, respectively. Some of the data and the ideas in this article also appear there, along with all of the essential primary literature. Though the present contribution has the character of a personal perspective, it is still interesting to see how intensively and in which way the discussed topics appear in the current research literature. Such a placement is attempted by a brief evaluation of relevant content from the last issue of *Drying Technology*, which should provide both representative and high-quality results, before summarizing the main conclusions of the article.

SPRAY FLUIDIZED BED AGGLOMERATION

In the process termed *spray fluidized bed agglomeration*, droplets, which usually contain water and a dissolved binder, are sprayed on fluidized particles. After a relatively short flight, the droplets are deposited on the particles and become sessile, creating wet spots (caps). Because the particles are mobile they collide with each other. When such collisions take place at an accessible wet spot, the particles may rebound, or they may stick together by a liquid bridge. Bridges of this kind solidify, creating agglomerates that may have superior properties with regard to, for example, their “instant” behavior (as in the case of instant coffee).

An agglomerate granule produced by spraying binder solution on fluidized primary particles (γ -Al₂O₃ of about 600 μ m in diameter) is depicted in Fig. 1a. The picture

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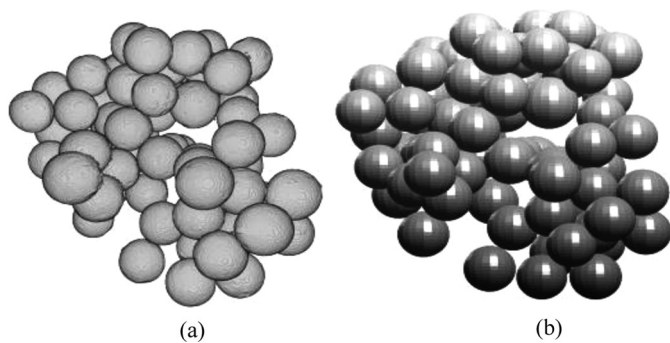


FIG. 1. Visualization of the three-dimensional structure of a spray fluidized bed agglomerate granule by means of X-ray μ -CT: (a) μ -CT volume image and (b) image after MATLAB processing (unpublished data of Dadkhah et al., from the author's group in Magdeburg).

was obtained by X-ray micro-computed tomography (μ -CT) and is thus three-dimensional, so that arbitrary cross sections can be visualized and studied. Furthermore, it can be processed in MATLAB to images (Fig. 1b), matrices, and files that contain all of the essential information about agglomerate structure. This can be used to derive a variety of morphological descriptors, such as radius of gyration, fractal dimension and pre-factor, porosity, coordination number distribution, and coordination angle distribution.

It is well known that the rate of particle formation in spray fluidized bed agglomeration—that is, the rate with which the diameter of the agglomerates grows in time—is influenced by, among other parameters, the temperature of the fluidizing gas. One can relatively easily anticipate that this dependence should have something to do with drying, but detailed explanations or even model description were missing until the very recent work by Terrazas-Velarde et al.^[7,8]; see also Peglow et al.^[6]

Some experimental results taken from this work are shown in Fig. 2. The experiments were carried out in a lab-scale fluidized bed, with primary particles made of glass and an aqueous solution of hydroxypropyl-methylcellulose (HPMC; binder) in top-spray. As the symbols show, the agglomeration rate decreases significantly with increasing gas (air) inlet temperature. Why should this happen? The qualitative explanation is that as the inlet air becomes hotter, the drying of sessile droplets becomes faster. Consequently, the droplets become smaller, so that they cover a smaller portion of particle surface and have a lower ability to dissipate the kinetic energy of colliding particles when such a collision occurs. Therefore, more particles will rebound without agglomerate formation at high air inlet temperatures, slowing down the process.

This explanation has been quantified in Terrazas-Velarde et al.^[7,8] in the frame of a comprehensive discrete model (constant volume Monte Carlo method, CVMC) for the process. Among other events, the model considers sessile droplet drying and the so-called Stokes criterion

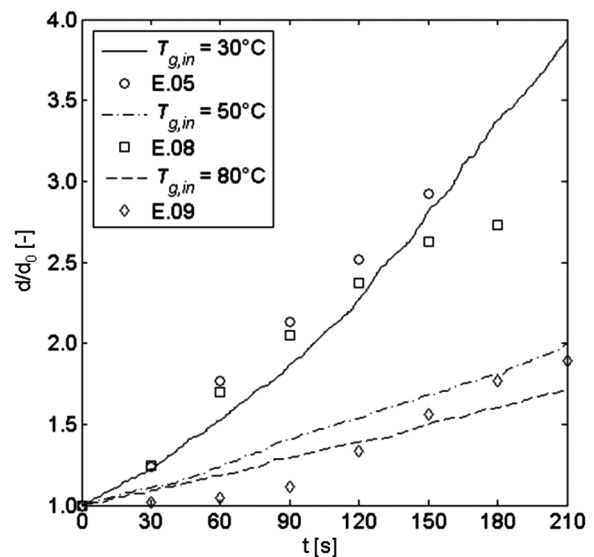


FIG. 2. Influence of drying kinetics (via the gas inlet temperature) on the agglomeration rate in spray fluidized beds according to measurements and computations by Terrazas-Velarde et al.^[7] (d = agglomerate diameter, d_0 = diameter of primary particles, t = time, $T_{g,in}$ = inlet gas temperature).

for rebound or agglomeration after viscous dissipation of the collision energy according to Ennis et al.^[9] This criterion reads

$$St = \frac{2Mu_c}{3\pi\mu_1 d^2} < St_{cr} = \left(1 + \frac{1}{e}\right) \ln\left(\frac{h}{h_a}\right) \quad (1)$$

Here, M and d are the average mass and diameter of the colliding particles, respectively; u_c is the collision velocity; μ_1 and h are the viscosity and height of the sessile droplet, respectively; e is the restitution coefficient; and h_a is the height of surface asperities on the particles. When the actual Stokes number, St , on the left-hand side of Eq. (1) is smaller than the critical value of St_{cr} on the right-hand side of the relationship, then viscous dissipation can absorb the collision energy and the collision partners will form an aggregate; otherwise, they will rebound. Because properties in a particle system, represented by something like 2,000 aggregates in the simulation box of the CVMC, are distributed, Eq. (1) does not describe a sharp change but a gradual transition, where some particles may fulfill the criterion and some others may not. Increasing the drying rate by higher gas temperatures means a smaller sessile droplet height, h , at the time point of collision, which shifts the threshold of St_{cr} to lower values, so that less collision events will fulfill it, and the agglomeration rate will decrease.

The consequence is that the model (solid, dashed, and broken curves in Fig. 2) describes the trend of the influence of gas inlet temperature correctly. With regard to Terrazas-Velarde et al.'s^[7,8] data, an even closer agreement between

computations and measurements than in Fig. 2 is attained, implying predictive ability of the model. This is achieved by using the results of just one measurement for a certain combination of materials in order to fit just one model parameter, which has to do with the collision frequency of particles, to a value that then remains constant for any variation of operating conditions.^[7,8]

Having recognized the large influence of drying kinetics on the rate of spray fluidized bed agglomeration, one may ask: Is this influence always going in the discussed direction of decreasing agglomeration rate with increasing speed of drying? With proven accuracy of the CVMC model, the answer to this question can be illustrated on computational results and is no. To recognize this, simulation results that consider or neglect drying are compared with each other in Fig. 3. The former correspond to usual drying kinetics under common experimental conditions, and the latter (denoted by ND: no drying) represent extremely slow drying kinetics. We can see the already discussed influence of drying at high mass fractions of binder in the sprayed solution ($x_b = 8\%$ or 10%): Faster drying (here: the mere presence of drying) decelerates the agglomeration significantly. However, at a low mass fraction of binder ($x_b = 2\%$), this trend is inversed completely: Particles do not agglomerate at all when drying is slow (so that the ratio d/d_0 remains nearly equal to unity and the respective curve is almost flat in Fig. 3), but they do agglomerate when drying is reasonably fast. The reason is that faster drying does not only reduce the surface coverage and the height of deposited

droplets (as already discussed), but it also increases their viscosity. The latter effect opposes the previous ones, because more viscous droplets are stickier. In other words, the left-hand side of Eq. (1) becomes smaller because μ_1 increases. This does not matter too much at high binder mass fractions, because the droplets are sticky even without drying (i.e., the Stokes number, St , is small enough). However, it prevails when the sprayed liquid is thin. Then, drying pushes St below St_{cr} by thickening the solution and triggers the agglomeration, enabling successful aggregation events (instead of rebounds) after interparticle collisions.

As pointed out, the non-drying (or slow-drying) thin-solution condition of Fig. 3 does not lead to agglomeration. What is going to happen with the (few but existing) solute after solidification in this case? The answer is that this material will form layers on the surface of the primary particles, still leading to particle growth, albeit much slower (consequently, the respective curve of Fig. 3 is not strictly horizontal, having a small but finite slope). The resulting product has then an onion-like structure, completely different from the blackberry-like morphology of agglomerates. It may therefore be concluded that drying kinetics does not only influence the rate of particle formation but may also define the kind and properties of the particles formed. In case of layering, the process is called *spray fluidized bed granulation* (or coating), which is discussed in the next section.

SPRAY FLUIDIZED BED LAYERING GRANULATION OR COATING

The term *layering granulation* is used when initially small particles (nuclei) develop in much larger granules. The application of just one or only a few thin layers on relatively large particles (cores) is called *coating*. The layer and core are different in coating, whereas nucleus and the added layers are made of the same material in granulation. Anyway, the onion-like particles resulting from both processes are conventionally considered to consist of compact layers. The growth rate of such particles would very straightforwardly result from the rate of solute addition into the fluidized particle system, the solvent (water) being dried out in some way that does not really matter. In fact, particles with a rather compact structure and smooth surface can be obtained by spray fluidized bed granulation, as the pictures on the left-hand side of Fig. 4 show. However, it is equally possible to produce very porous and rough granules (right-hand side of Fig. 4). The porous and rough particles have different properties than the compact and smooth ones, and they grow faster, because a significant part of the added volume is void. Notably, the products shown on both sides of Fig. 4 were produced in the same equipment (lab-scale with top spray) with the same materials (aqueous solutions of sodium benzoate sprayed on cores consisting of $\gamma\text{-Al}_2\text{O}_3$; see Hoffmann et al.^[10]).

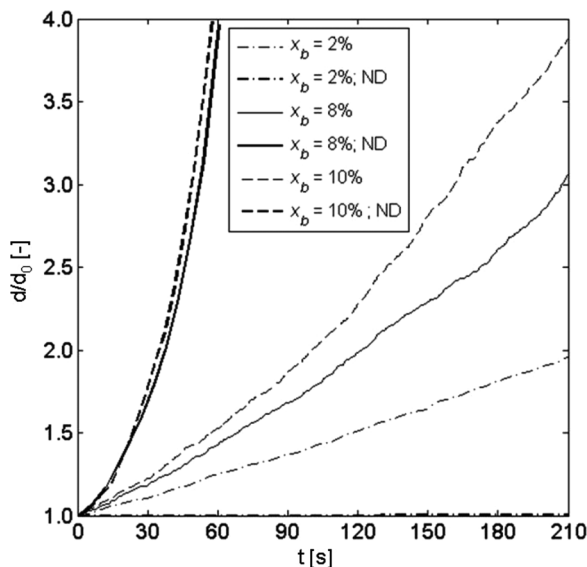


FIG. 3. Comparison of agglomeration rates computed for common drying conditions and for very slow drying (ND: no drying) according to Terrazas-Velarde et al.^[7] Faster drying decreases the agglomeration rate at high binder mass fractions (x_b) but increases the agglomeration rate when the binder mass fraction is low.

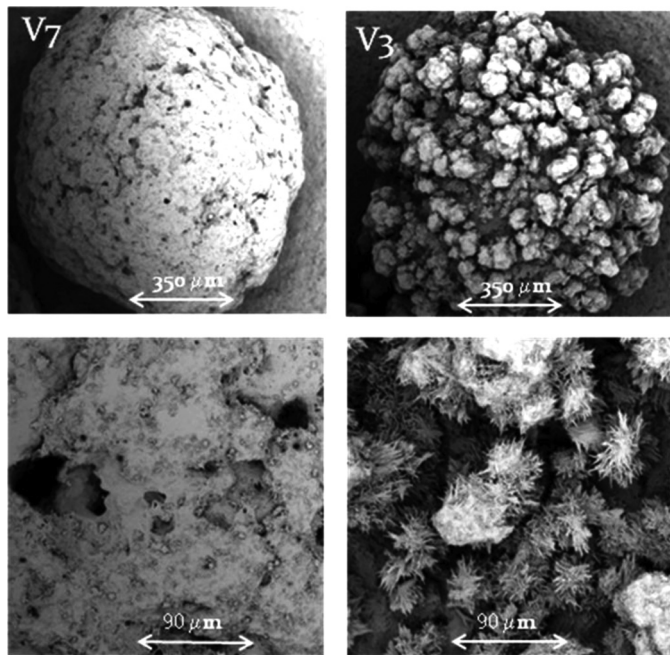


FIG. 4. In spray fluidized bed granulation, fast drying leads to compact and smooth particles (left-hand side), whereas porous and rough particles result from slow drying (right-hand side). Pictures obtained using SEM.

The difference behind the completely different looking particles lies only in the operating conditions, namely, in the liquid flow rate and the gas temperature. If the liquid flow rate is small and the gas temperature is high (95°C in case of Fig. 4, left-hand side), then compact and smooth particles are obtained, whereas porous and rough granules are formed at a high spraying rate and low gas temperature (50°C in Fig. 4, right-hand side). The physical background is obviously fast drying in one and slow drying in the other case, which shows that drying kinetics governs particle formation also under conditions of layering granulation. Similar observations were reported in Walzel and Furuta^[3] and Peglow et al.,^[6] including the drying-dependent kinetics of precipitation or crystallization as a possible mechanistic explanation. However, a model providing comparable accuracy and insight to the previously mentioned CVMC model for agglomeration does not exist yet.

SPRAY DRYING OF SOLUTIONS

In spray drying, the droplets evaporate during flight and not on fluidized particles. The great influence of drying kinetics on particle formation is well known and documented both empirically and with the help of some models.^[3] Here, the discussion will focus on a new model, based on previous approaches by Sloth et al.^[11] The model refers to the single droplet, which can be experimentally investigated in an acoustic levitator (cf. with Peglow et al.^[2]), assuming that its results can be integrated into computational fluid dynamics codes for tower computation.^[11]

The main features of the model, illustrated in Fig. 5, are as follows:

- In a first drying period the droplet is considered to shrink by gas-side controlled evaporation of water. Emerging profiles of solute concentration are computed by numerical solution of the diffusion equation under moving boundary conditions.
- As soon as a critical concentration of solute has been reached at the surface of the droplet, the formation of a solid crust begins, and the shrinking droplet is transformed to a drying particle of constant outer size (“locking point”).
- In a second period of drying, the boundary that separates the dry crust from the still liquid core of the particle propagates inwardly. Transport phenomena are treated in a quasistationary way in the crust, whereas the complete diffusion equation continues to be solved in the core.

It should be noted that the model is, although here not considered an effect of aggregation, identical to the model for the drying of nanosuspension droplets according to the next section, so that a detailed mathematical description can be found in the literature cited there.

Selected model results are depicted in Fig. 6 for fast drying and in Fig. 7 for slow drying, whereby “fast” and “slow” are always meant in relation to solute/solvent diffusion. The upper plots in these figures show that outer diameter and liquid core diameter are identical and decrease during the first period of drying. In the second period, after the locking point, the outer diameter remains constant, whereas the diameter of the liquid core continues to shrink, which corresponds to an increase in the thickness of the crust. The plots differ from each other in that the locking point is much earlier for fast than for slow drying. This is due to the fact that diffusion cannot follow fast drying, so that steep water content profiles (and complementary profiles of solute concentration) build up. This is shown by the large difference between average water content in the liquid core and local water content at the surface during the first drying period) in the lower plot of Fig. 6. Notably, this difference decreases after the locking point and finally

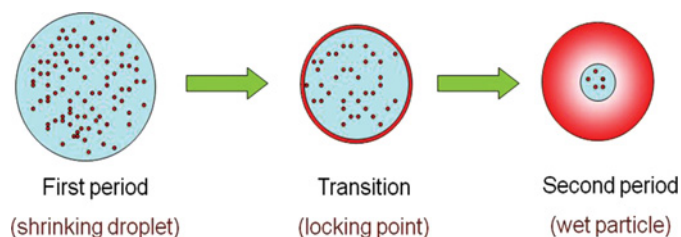


FIG. 5. Illustration of the model for the drying of a single solution droplet (color figure available online).

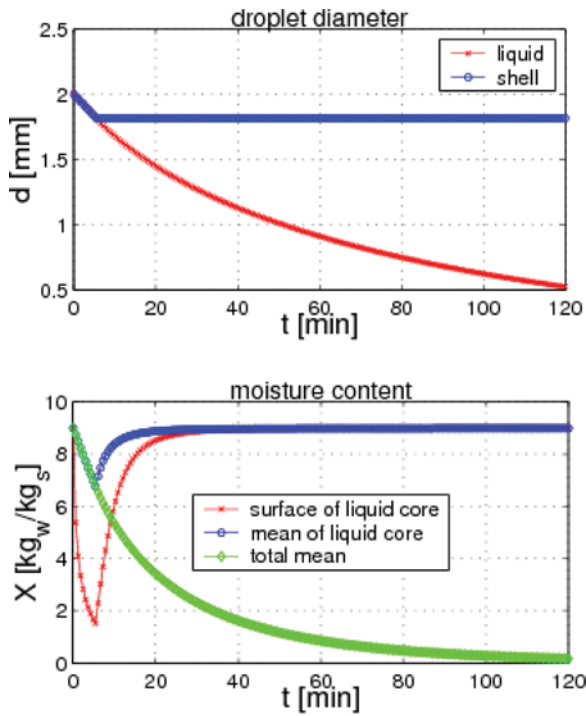


FIG. 6. Model predictions for fast drying of a solution droplet (X =dry basis water content) (color figure available online).

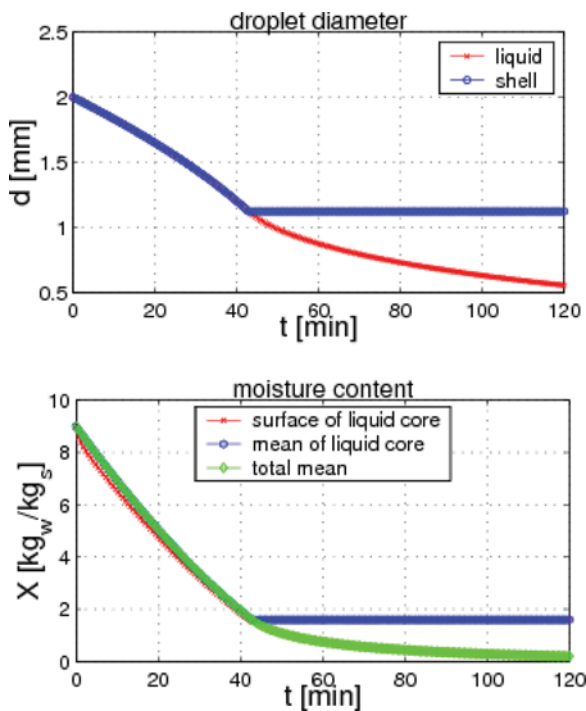


FIG. 7. Model predictions for slow drying of a solution droplet (color figure available online).

disappears, because of the increasing mass transfer resistance of the dry crust, which results after some time in slow drying, even if it had originally been fast. This condition prevails from the very beginning in the case of slow drying, because the diffusion is then always relatively fast and can equilibrate the concentrations of water and solute in space (lower diagram in Fig. 7).

The aforementioned differences in the temporal evolution of concentration profiles have dramatic consequences for the porosity and structure of the resulting dry particles. As the upper diagram in Fig. 8 shows, the porosity increases strongly from the surface to the interior of rapidly dried particles, so that such particles can be expected to be very porous in the average and even hollow. On the other hand, slow drying results in homogeneous and quite compact products (lower plot in Fig. 8). This is in good qualitative agreement with both the discussion by Walzel and Furuta,^[3] and the primary literature cited therein (for example, Brenn et al.^[12]). A hollow particle produced by drying a lactose-in-water

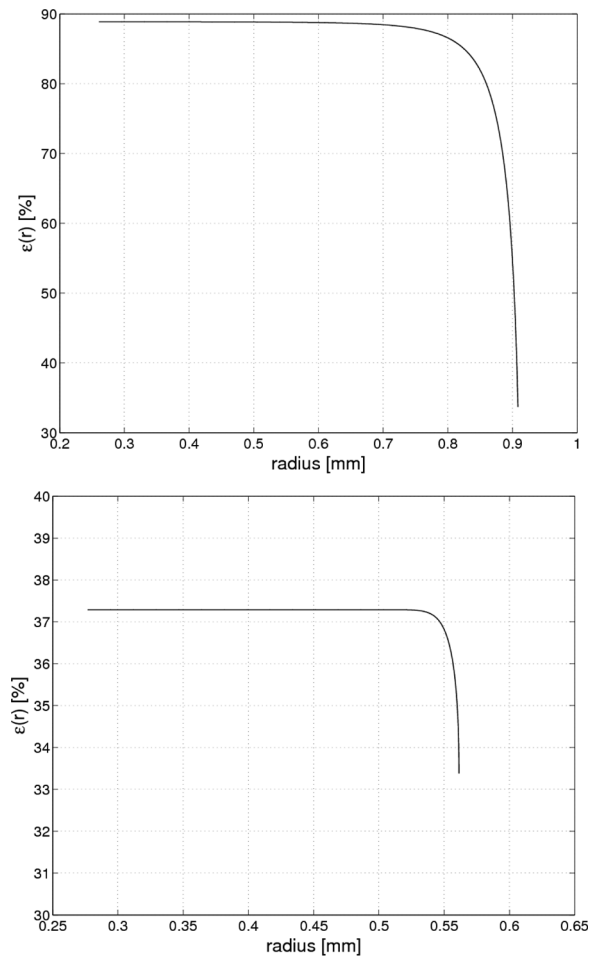


FIG. 8. Porosity profiles in particles produced by fast drying (upper plot) or by slow drying (lower plot), corresponding to the computations of Fig. 6 and Fig. 7, respectively.

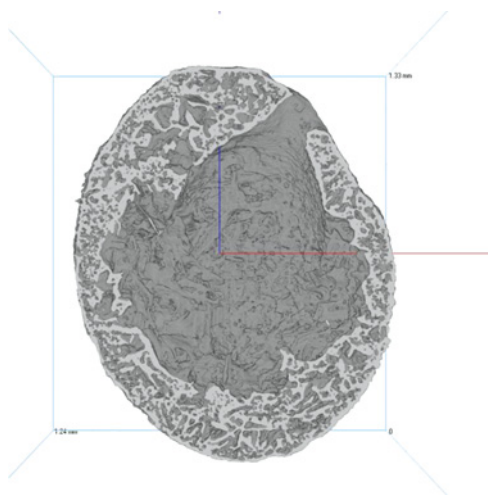


FIG. 9. X-ray tomogram of hollow particle produced by drying a 10 mass-% lactose-in-water droplet in an acoustic levitator (unpublished data of Ihlow et al., from the author's group in Magdeburg) (color figure available online).

droplet in an acoustic levitator is shown in Fig. 9. The image is cross-sectioned, by use of the already mentioned X-ray μ -CT.

This discussion demonstrates once more the influence of drying kinetics on the formation of particles from sprays, and it also indicates that the resulting structure can—to a certain extent—be quantified by models.

SPRAY DRYING OF NANOSUSPENSIONS

The previous computation can be upgraded by additional consideration of primary particle aggregation in the droplet. This is typical for the drying of more or less efficiently stabilized nanosuspensions. Because the mobility of nanoparticles depends on their diameter (according to Stokes-Einstein), and the diameter depends on the degree of aggregation, an interrelation exists between aggregation and nanoparticle diffusion in the droplet.

Modeling, as recently presented by Naumann et al.^[13] and Bück et al.,^[14] requires in this case the solution of population balance equations. Resulting porosity profiles at the locking point are shown exemplarily in Fig. 10. In the computations, the initial droplet diameter and the diameter of primary nanoparticles were set to 1 mm and 1 nm, respectively. Different values of a constant aggregation kernel β_0 were used. A high value of $\beta_0 = 0.1$ 1/s means higher diameters of aggregates, so that diffusion in the droplet becomes slower in relation to drying. Large locking point diameters with steep porosity profiles are predicted in this case. At a low $\beta_0 = 0.001$ 1/s, the aggregates are smaller and more mobile. Then, smaller droplet diameters with a flatter porosity profile are obtained at the locking point.

It should be noticed that the high-aggregation ($\beta_0 = 0.1$ 1/s) result of Fig. 10 is very similar to the upper plot in

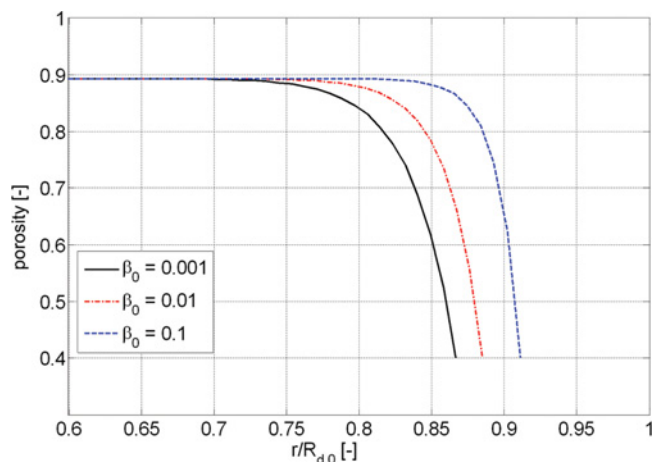


FIG. 10. Porosity profiles in drying nanosuspension droplets at the locking point according to Bück et al.^[14] (β_0 = aggregation rate, r = radius, $R_{d,0}$ = initial radius of the droplet) (color figure available online).

Fig. 8. Lower aggregation relaxes the porosity profile. This relaxation could be further increased by taking smaller primary particles, accounting for the intensification of mass transport by internal flow in the droplet (which can be done in terms of higher effective diffusion coefficients) or using milder drying conditions. All of these measures are in favor of porosity profiles similar to the one in the lower plot in Fig. 8, leading to small, compact, and homogeneous product particles.

In total, particle formation from sprayed nanosuspension droplets is similar to particle formation from solutions and, again, is distinctly dependent on the drying kinetics. The briefly discussed models are certainly coarse, but they can be extended in several directions in order to consider, for example, a crust that is still flexible after the start of its formation and, thus, continues shrinking for some time,^[15] crust buckling, blow holes, or even crust creeping and compaction by transition from the glassy to the rubbery state.^[3] What is really important is the ability of the models to describe the emerging structural morphology on physical grounds, because this is a presupposition for scale-up from, for example, the artificial conditions of levitator drying to real spray tower conditions, and it is the essential joint between processes and equipment on the one hand and end-user properties on the other.

A BRIEF LITERATURE SURVEY

For a concise review, a glance at *Drying Technology* should be sufficient to recognize how intensively and in which way the discussed topics appear in the current research literature. The contents of the most recent volume of the journal from the year 2011 have been evaluated, with the following results.

A first striking result is that spray drying is the most frequently treated topic in the journal, with a plethora of

papers in 2011.^[16–29] Most of these papers are very similar to each other: The solution or suspension of an industrially interesting material is spray dried, relevant powder properties are measured, and the number of trials is kept bearable by application of some method of design of experiments, which also provides an empirical correlation between variable process conditions and the variable properties of the particulate product. The range of investigated properties is large: Particle size distribution in almost all investigations, density (bulk, tapped, and—less frequently—particle density),^[17,23,25] porosity (as a conjugated property to particle density),^[23] pore size, volume and surface area by adsorption,^[25] various flowability and compressibility indices,^[17,18] color,^[17,27] antioxidant activity,^[16,17] other kinds of ingredient activity or active ingredient content,^[21,23] viability in case of microorganisms,^[23] solubility,^[23] wettability,^[25] crystallinity,^[25] thermal properties by differential scanning calorimetry,^[23] or even electromagnetic properties, such as dielectric permittivity and electrical conductivity.^[25] Particle shape and surface structure were in some cases visualized by scanning electron microscopy (SEM; cf. Figure 4), which can also be applied to broken or cut particles in order to obtain an impression of their inner structure.^[20,23] Nondestructive tomographic measurements were not reported in 2011. Some research works aim at specific formulations, such as encapsulation^[19,24,27] or the production of inhalable micropowders.^[20] Therefore, they evaluate specific product properties, such as encapsulation efficiency, active ingredient concentration, retention or release after production (loss during storage), or the aerodynamic diameter.

A trend from research in process design to research in product design had been identified in 2003, with a still more comprehensive list of investigated product properties.^[30] The present evaluation shows that this trend has been significantly increased in the meantime. Certainly, aspects such as drying time, dryer capacity, and component yield were also considered in the evaluated papers, along with aspects of material behavior that are important for the process, such as stickiness^[18] and its avoidance by, for example, the use of novel additives,^[28] but the main focus is nevertheless on product properties. A certain exception is Schmitz et al.,^[29] who dealt with the deactivation of lysine, an essential amino acid that cannot be metabolically produced by the human organism and thus must be contained in infant formulae. In Schmitz et al.,^[29] deactivation kinetics was determined on small samples, not in the spray dryer, so that application of this kinetics requires a model for single droplet drying—perhaps similar to the previously discussed model—and—in a two-scale approach (cf. Blei and Sommerfeld^[1])—a second one for the tower.

Modeling is otherwise appearing too rarely in the evaluated literature. This leaves open questions about the

transferability of results on product properties from the lab-scale dryers used in research work to the large—and continuously operated—spray dryers in practice. Concerning our main topic of the influence of drying rate on particle formation, it should be noted that the drying rate is varied in experiments by changing the inlet air temperature. However, the temperature itself can have a direct influence on, for example, ingredient deactivation or glass transition of the matrix, so that it is not possible to say—not even empirically and at the lab scale—which part of an effect is caused by the drying rate and which one by the temperature level. The so-called reaction engineering approach^[18] can remedy some of these deficiencies, but only partially, due to its empirical nature. Consequently—and as previously pointed out—there is a clear and continuing need for the further development of models (cf. Mezhericher et al.^[31]), which—if physically justified—will be able to describe all aspects of the process: Drying kinetics, the resulting solid phase formation, and the end-user properties of the particulate product.

Only five papers on spray fluidized beds were published in volume 29 of *Drying Technology*,^[32–36] fewer than on spray drying. This is somehow surprising, because fluidized bed processing is much more intense in terms of, for example, evaporation capacity per unit volume of equipment. An attitude of “simplest things first” during the development of new formulations may explain this finding, because fluidized bed equipment may require a higher degree of engineering than a lab-scale spray drying facility. All of the mentioned references deal with coating, aiming at the production of either coated particles^[32,33] or entrained fine powder after the in situ destruction of emerging coating layers by attrition, due to interparticle collisions.^[34–36] Articles on spray fluidized bed agglomeration were not included in the evaluated journal volume. Product properties were discussed, even very intricate ones, such as the sensory perception of tropical fruit pulp powders in yogurts,^[35] but they are in total less dominant in the overall picture than in case of spray drying, because processing aspects, such as residence time distribution^[36] or the influence of operating pressure^[34] were also treated. Even modeling was provided,^[32] albeit based on the conventional assumption of compact and smooth layers (cf. Hoffmann et al.^[37]). As we have seen before, this is not necessarily true (Fig. 4). An indication for the succession of porous layer formation and compaction steps in time was reported in Komoda et al.,^[33] where coating thickness variation was proposed as an instrument for monitoring of different formation stages. Despite such advances, we are still far from a reliable modeling of drying rate-dependent layer structures. On the other hand, the need for such models is huge, because they would be applicable not only to the coating of particles in spray fluidized beds but also to every other wet coating process.

CONCLUSIONS

The intensity of drying has a strong impact on particle formation in spray fluidized beds. It influences the agglomeration rate, determines under certain conditions the kind of particles to be formed (agglomerates or layered granules), and—in case of granulation or coating—influences the morphology of the resulting layers. In spray dryers, the intensity of drying correlates with the average value and the spatial distribution of porosity in product particles formed from solution or nanosuspension droplets.

Models that can capture some aspects of the described behavior exist already, but they need to be further improved. Additionally, some completely new models need to be developed in order to describe, for example, the porosity and morphology of coating layers formed by drying. Empirical approaches to formulating drying processes, which dominate the contemporary literature, will certainly maintain their importance, but they can be beneficially supported and guided by an appropriate frame of physically based modeling. In such models, the solid phase structure is the joint between process and equipment conditions on the one hand and end-user product properties on the other. Therefore, nondestructive methods for the determination of internal particle structure, such as X-ray μ -CT, will play a significant role in the future.

The opportunities offered by processes that combine drying with particle formation are huge, including reverse engineering, which means the design of equipment and processes upon given specifications for the end-user properties of powdery and particulate materials. This is one of the topics that may be expected to define the face of *Drying Technology* in the next 30 years, contributing to the continuing and increasing success of the journal.

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