Drying Behavior of High Load Multiphase Droplets at High Temperature Conditions in an Acoustic Levitator

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Abstract

Spray drying is the process by which a fluid feed material is transformed into a dry powder by spraying the feed into a hot drying medium. The drying behavior of suspension droplets is important in many industrial applications (material processing, ceramic, chemical or food industry). The main aim of drying by this method is to obtain dry particles with desired properties. In this work, the drying behavior of acoustically levitated multiphase droplets has been experimentally investigated. The experiments have been performed using ceramic suspensions like those used in the manufacture of porcelain tiles. High load and temperature conditions close to those found in industrial applications have been used. The experiments have been carried out in an acoustic tube levitator modified in order to allow experiments at high temperature conditions. The flow rate, temperature and relative humidity of this air stream can be controlled by an air conditioning system. A CMOS camera and a back-light illumination system are used to measure the droplet cross-sectional area and vertical position of the droplet during the drying process. An experimental matrix based in a factorial design 2k has been chosen to study the effect of the flocculation state (flocculated-deflocculated), solid mass load ($0.65 < Y_S < 0.70$), particle size distribution of the porcelain composition ($1.95 < d_{P50} < 3.25$ μm), ambient air temperature (70 ºC < T < 100 ºC) and initial droplet volume (0.4 μl < V_0 < 0.7 μl) on the mean porosity of the grain and its mechanical strength.

Introduction

In the chemical, food and material processing industries, powder products are produced by spray drying of liquid-solid suspensions. Spray drying is the process by which a fluid feed material is transformed into a dry powder by spraying the feed into a hot drying medium. The spray drying process comprises three major phases. The first one is the atomization of the liquid stream by an appropriate device. Next, fine droplets of the feed are subjected to the interaction with a drying gas at adequate temperature. During this drying phase, the solvent contained within the dispersion droplets is vaporized, which results in the formation of solid product particles. During the last phase, the dried particles must become separated from the drying gas by an appropriate device and must be collected by a receptacle tank. Each of the above-mentioned phases, as well as the conditions under which they are conducted, has a huge effect on the drying process efficacy and the final product properties. It is an efficient means of drying due to the large surface area available for heat and mass transfer as a result of atomizing the liquid into very small droplets. Properly done, spray drying is an economical and continuous operation which produces a powder of uniform and repeatable characteristics.

The spray drying is a complex industrial process that includes physical processes such as spray atomization, heat and mass transport from the droplets to the surrounding gas, drop-wall interactions, etc. Drying behavior of droplets of liquid-solid suspensions in a gas is of significant importance. This fact is particularly significant for high load and temperature conditions close to those found in industrial applications. Droplet drying models can be used to relate the final powder properties (such as the grain diameter distribution, mean porosity, morphology, etc) with the spray dryer design and process parameters.

The drying process of a liquid-solid suspension droplet is characterized by two drying periods. In the first period, the evaporation is produced in the droplet surface and the mass transfer rate equals the mass transfer rate of an equally sized droplet containing the liquid component of the suspension droplet. In this constant rate period, the droplet diameter, d, decreases following the d^2-law. The surface of the droplet is completely wetted and this leads to the d^2-behavior virtually uninfluenced by the solid phase. In the second drying period a crust is formed in the droplet surface and the volume of the droplet remains constant. Although the volume is constant, the mass still changes due to the evaporation of liquid from inside the droplet through the pores of the crust. In this falling rate period a hollow droplet, collapse or explosion can occur, modifying the final grain morphology.

It is not possible to obtain detailed information about the droplet drying process in industrial or laboratory-scaled dryers. For the study of the drying behavior of the suspension droplets, single droplets experiments are

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needed. In this regard levitator tubes present some advantages with respect to conventional methods since there is no contact between the droplet and the device. In the last decades acoustic levitator tubes have been extensively used to study the drying behavior of pure liquid, multi-component liquid and liquid-solid suspension droplets. In this regard, Yarin et al. [1] predicted the droplet shape and evaporation rate of acoustic levitated liquid droplets. Kastner et al. [2] developed the experimental procedure to measure evaporation rates in both drying periods. Yarin et al. [3] modeled the drying behavior, obtaining the duration of the first drying period and an experimental correlation for the final grain porosity. Finally, Brenn [4] modeled the solid concentration fields inside the droplet; predicting the appearance of hollow grains. Most of the available data on the drying behavior of suspension droplets in acoustic levitators were obtained for moderate temperatures (T<80 ºC) using water suspensions containing glass particles. However, higher temperature conditions can be found in important industrial applications. Recently, Mondragon et al. [5] have developed a levitator tube modified to allow experiments at high temperature condition. This system allows temperatures up to 150 ºC.

In case of using spray dryers in the ceramic industry, it is very important that the drying process produces certain final powder characteristics which favor later stages of the manufacturing process, increase the production and also improve the quality of the resulting ceramic tiles.

Porcelain tile is a product characterized by low water absorption (usually less than 0.1%) and excellent mechanical properties. To enhance tile aesthetic qualities, much of the porcelain tile production is polished to provide a high-gloss surface finish, in which certain closed pores in the tile body become visible. This apparent porosity of the polished tile, which had been closed porosity before polishing, sometimes lowers the product's stain resistance.

In this work experiments have been performed using ceramic suspensions like those used in the manufacture of porcelain tiles. The effect of the flocculation state, solid mass load (Y_S), particle size distribution of the porcelain composition (d_{P50}), ambient air temperature (T) and initial droplet volume (V_0) on the mean porosity of the grain (ε) and its mechanical strength (σ_R) has been studied. Anova method has been used to analyze the results and to obtain the optimal conditions of the process. The values of the input variables needed to achieve optimal output variables have been discussed. For this material, the optimal conditions established according to its technical properties and manufacturing process are low porosity and high mechanical strength. The former is the most important and the lowest porosity is desirable, while the latter has to be high enough to resist handling and processing but not so high to deform easily during pressing process.

Experimental set-up

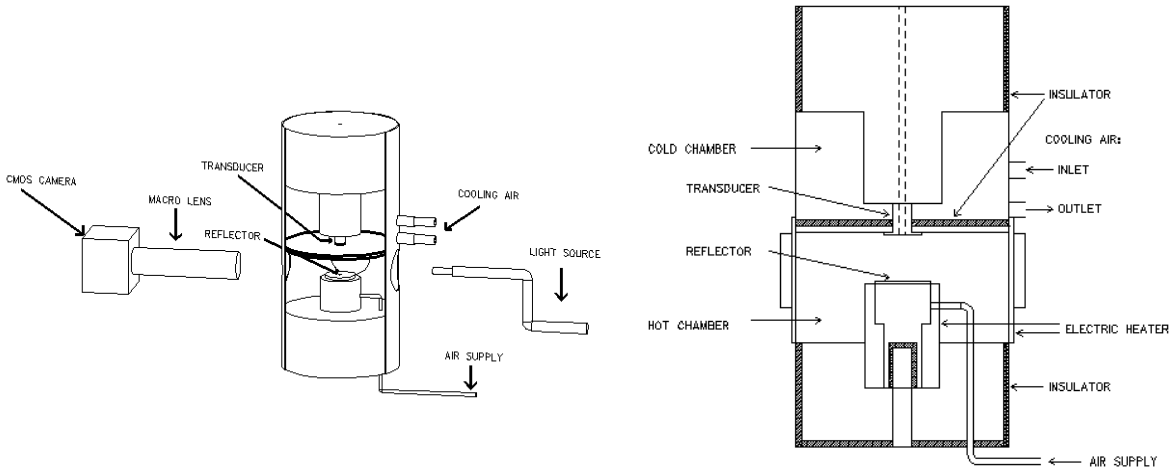
The experimental facility has also been detailed previously in [5]. The experimental setup is composed of three systems (see Figure 1 a):

- An acoustic levitator consisting of an ultrasonic 58 kHz horn and a concave reflector. The levitator tube (tec5 AG Sensorik und Systemtechnik) produces a standing wave and pressure nodes where the droplet can be located. The original levitator tube has been modified in order to allow experiments at high temperature conditions (see Figure 1 b). In this way, two separated metallic chambers can be found in the levitator tube. The section identified as "cold chamber" contains the ultrasonic transducer of the levitator. The temperature of this chamber can not be higher than 60ºC in order to prevent damages in the piezoelectric transducer and it is controlled by forced convection using cold air. The section identified as "hot chamber" contains the levitator reflector and the multiphase droplet. The temperature of this chamber is controlled by an electric heater at the wall and by an air stream that enters the levitator tube through an array of holes located in the reflector. The flow rate of the air stream is set to 0.5 l/min and it is used to ensure constant drying conditions around the droplet and deplete the acoustic vortices system around the droplet from liquid vapor. The droplet is inserted into the acoustic field with a syringe. The needle of the syringe can be introduced into the acoustic field using a hole centered in the horn (dotted line in Figure 1 b). Using this levitator tube configuration it is possible to work with temperatures up to 150 ºC. However, the droplet insertion method limits the maximum temperature working condition to 120 ºC (for higher temperatures the droplet is dried before it is expelled from the syringe tip).

- An optical system consisting of a white light source with a diffuser and a CMOS camera with a macro lens. The CMOS camera (UI-1220M, IDS-Imaging Development Systems GMBH) (752×480 pixels, 87 frames per second) and the back-light illumination system are used to measure the droplet cross-sectional area and vertical position of the droplet during the drying process. The spatial resolution of the images can be varied with the macro lens. A resolution range between 250 and 500 pixels/mm has been used in the experiments depending on the initial droplet volume value.

- A gas conditioning system (not shown in the figure) controlling the temperature, flow rate and relative humidity of the air inside the levitator tube. The air conditioning system (CEM System W-202A, Bronkhorst High-Tech B.V) is composed of an air-drying cartridge, a two-mass flow controllers and a mixer/evaporation unit. This system allows temperatures up to 200ºC and a humidity up to a dew point of T=80 ºC. In order to prevent

the air stream cooling from the conditioning system to the levitator tube the tube that connect both devices as well as the reflector need to be heated using electric heaters.



a) b)

Figure 1. a) General sketch of the experimental set-up, b) modified acoustic levitator tube

Measurement techniques

The two output variables measured and analyzed in this work have been the mean grain porosity, ε, and the mechanical strength, σ_R.

The porosity has been calculated from the processing of the images that has been implemented in Matlab. Thus, the equivalent diameter and position of the droplet during the drying process are measured. With this information and knowing the initial properties of the droplet it is possible to obtain the mean porosity of the grain which can be calculated using the following equation [6]:

$$\varepsilon = 1 - \frac{V_S}{V_G} \tag{1}$$

where V_S and V_G are the volumes occupied by the solid phase and the dried grain, respectively and,

$$V_S = V_0 Y_S \frac{\rho_D}{\rho_S} \tag{2}$$

where ρ_D and ρ_S are the densities of the liquid-solid suspension and solid phase respectively.

In order to obtain the mechanical strength of the granules, diametric compression tests of single granules were performed. Typical load-displacement curves were obtained for each experimental condition by which the fracture load of the grains was obtained. With this information, the mechanical strength of spherical bodies loaded diametrically can be calculated by means of the following equation [7]:

$$\sigma_R = 0.7 \frac{F_{max}}{\pi R^2} \tag{3}$$

where F_{max} is the maximum force resisted by the grain before breaking and R is its radius.

Materials

All the experiments were carried out with water-based ceramic suspensions. The composition is the same used in the manufacture of porcelain tiles (clays, sand and feldspar in the proper proportion). A standard suspension with mean particle size, initial solid mass load and flocculation state similar to those found in industrial processes is established with values 3.25 μm, 0.65 and deflocculated suspension respectively. These variables have been modified to analyze their effect on the output variables (ε and σ_R).

The powder compositions are prepared milling all the raw materials in a ball mill. The milling time is adjusted so the final particle size distribution has the desirable mean diameter. For the standard composition only

10 minutes have been needed. However, to achieve a mean particle size of the composition less than the standard value, the milling time has been increased, being necessary 4 hours to obtain an appreciable reduction in the particle size. Figure 2 shows the particle size distribution of both compositions used to prepare the ceramic suspensions: the standard composition and the reduced size composition. The mean particle size, d_{P50}, is the diameter value corresponding to a 50 % of cumulative mass.

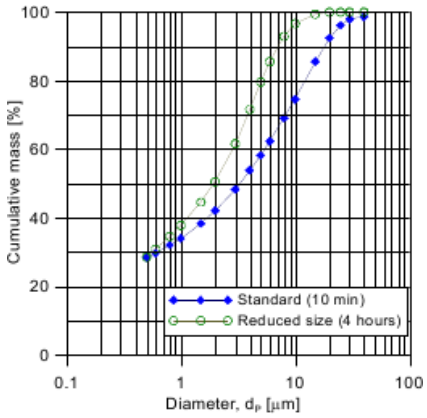


Figure 2. Particle size distribution of the standard and the reduced size composition

The flocculation state of the suspensions has been modified by addition of different amounts of deflocculant additive. The deflocculant used has been a $Na_5P_3O_{10}$: $Na_2O{\cdot}SiO_2$ (1:3) mixture. Both additives produce an electrostatic and steric mechanism of repulsion between particles.

For each composition and solid mass load (four combinations in total) the deflocculation curve has been obtained as shown for d_{P50} = 3.25 μm, Y_S = 0.70 in Figure 3 a). The viscosity has been measured by mean of a torsion viscosimeter (Gallenkamp). The amount of deflocculant needed to achieve the deflocculated state for each case was that one which provided the minimum viscosity and thixotropy (difference in viscosity measured after one and six minutes), while to achieve the flocculated state an amount far from the minimum was chosen. For each condition (eight suspensions in total) the rheogram was measured by means of a rotary viscosimeter (Figure 3 b). In these curves the different rheological states can be seen at low shear rates. At high shear rate, suspensions with the same mass load have the same viscosity independently of its state.

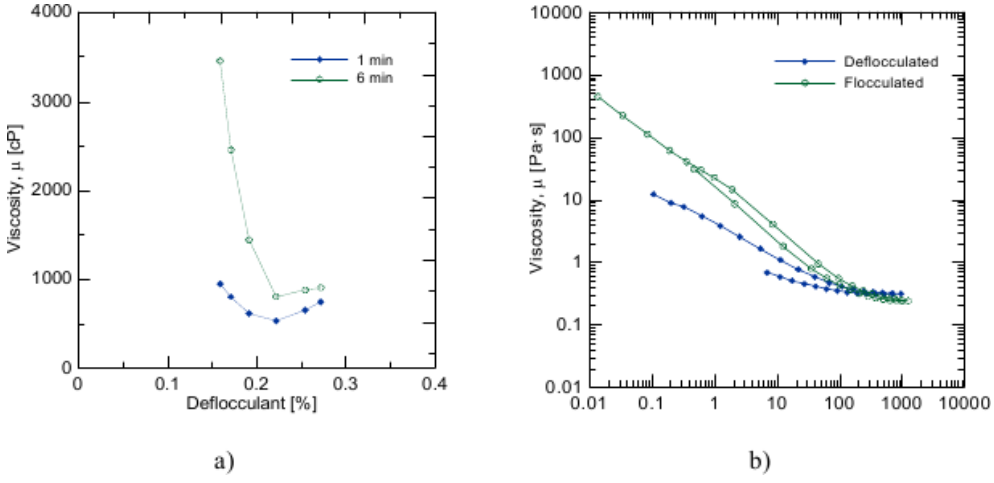


Figure 3. a) Deflocculation curve and b) rheogram for the suspension with d_{P50} = 3.25 μm and Y_S = 0.70

Results and Discussion

The effect of mean particle size of the composition (d_{P50}), initial solid mass load (Y_S), flocculation state, ambient air temperature (T) and initial droplet volume (V_0) on the mean grain porosity and its mechanical strength have been studied. Table 1 summarizes the input variables and their corresponding covered ranges.

Table 1. Input variables and their corresponding experimental ranges

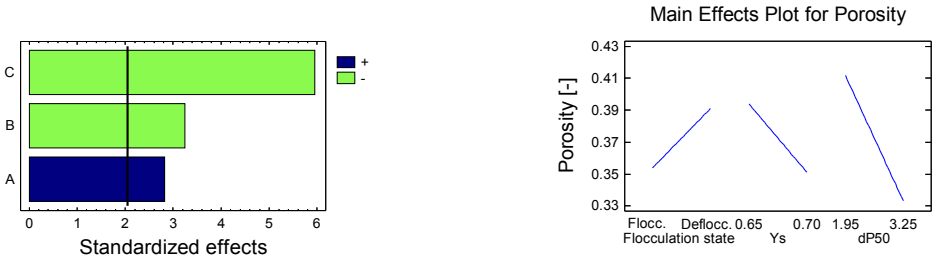
Input variables	Experimental Range
d_{P50}	1.95 – 3.25 µm
Y_S	0.65 – 0.70
Flocculation state	Flocculated – Deflocculated
T	70 – 100 ºC
V_0	0.4 – 0.7 µl

An experimental matrix based in a factorial design 2^k has been chosen. As a result, a total number of 32 test cases were carried out. All the results were analyzed using Anova method by means of Statgraphics. The effects of inputs on outputs are shown in the Pareto charts, where the most sensitive input variables can be identified. Each of these graphs shows an ordered bar chart of the absolute effects scaled by P-values, essentially the number of standardized effects beyond the mean response. The line at 2.31 P-value represents a significant level for achieving 95 % confidence that a given effect did not just occur by chance.

Analysis and optimization for porosity

Figure 4 a) shows the Pareto chart for porosity, where the following codes have been used for the input variables: A for flocculation state, B for initial solid mass load, Y_S and C for particle size of the composition, d_{P50}. It can be seen that these three variables have significant effects on the porosity. On the contrary, the influence of ambient air temperature and initial droplet volume on the porosity is not relevant.

The significant variables for porosity are analyzed in Figure 4 b), which shows the variation of the porosity when main effects change from lower to upper values.



a) b)

Figure 4. a) Standardized effects and b) main effects for porosity

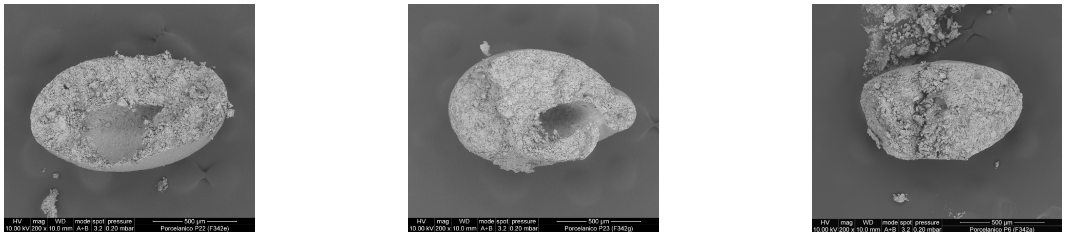
The degree to which particles are able to rearrange during the drying process influences the final granules porosity. The most important effect on the porosity is the particle size of the composition. It is observed from the particle size distribution (Figure 2) that the composition with higher mean particle size (standard composition) has also a wider size distribution, defined by the ratio between d_{P90} and d_{P10}. It means that there are more particles with different sizes and thus, the small ones can fill the voids created between the big ones when these latter approach and contact them during the drying process. Therefore, an increase in the mean particle size leads to a better particle-packing and a decrease in final grain porosity.

The increase in the initial solid mass load leads to a decrease in the porosity. Duffie and Marshall [9] demonstrated that the variation of the grain density between two different conditions depends on the relationship between mass load ratio and grain diameter ratio, the granules being denser when the former is bigger than the latter. It is obvious that in droplets with equal initial volume, the higher the mass load, the higher the number of particles inside the droplet when the shell is formed. As a result, the arrangement of the particles when the droplet dries is more uniform and the packing leads to a less porosity.

Finally, deflocculation of the suspension involves an increase in the mean grain porosity. The granule formation is determined by the strength of the floc structure. When the slurry is deflocculated, a crater may form from the inward collapse of the surface of a forming granule when the particle-packing density in a droplet continues to increase after the droplet size become fixed by the formation of a rigid shell, leaving an internal void and a hollow grain [8]. Moreover, the deflocculant chosen acts by a steric mechanism in which sodium silicate forms a protector colloid and sodium tripolyphosphafe adsorbs over the particles leading to an increase in the effective particle volume. In this case the particle-packing is worst than in the flocculated one.

Cross section of some grains has been observed by Scanning Electron Microscopy (SEM). The biggest grains with $V_0 = 0.7$ µl (Figure 5 a, b) present hollow morphology while the smallest one with $V_0 = 0.4$ µl (Figure 5 c) present a bigger shell thickness which makes the grain almost solid with a more homogeneously distrib-

uted porosity. Figure 5 a) and b) also show the difference porosity obtained when the flocculation state is changed, as mentioned before.



a) b) c)

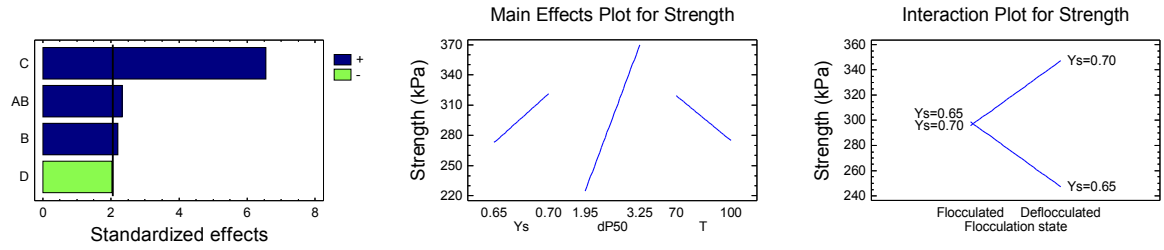
Figure 5. Cross section of grains under different conditions: a) V_0 = 0.7 µl, Deflocculated, b) V_0 = 0.7 µl, Flocculated and c) V_0 = 0.4 µl, Deflocculated

With all this information, if a low porosity condition is established as the optimum for the manufacturing process, the input variables must be the following: standard particle size distribution (wider distribution with higher mean particle size), high initial solid mass load and flocculated suspension. Moreover, although the initial droplet volume is not of significance, it is desirable a small droplet volume in order to achieve a more uniformly distributed porosity. Under these conditions, the expected value for porosity obtained by means of the Anova method is 0.294 ± 0.014.

Analysis and optimization for mechanical strength

Figure 6 a) shows the Pareto chart for mechanical strength, where the following codes have been used for the input variables: A for flocculation state, B for initial solid mass load, Y_S, C for particle size of the composition, d_{P50} and D for ambient air temperature, T. It can be seen that the most significant effect that influence the mechanical strength is the particle size of the composition. Effects also important are the initial solid mass load, the ambient air temperature and the cross effect of flocculation state and initial solid mass load.

The main effects for mechanical strength are shown in Figure 6 b). Figure 6 b) and c), where the variation of the mechanical strength when main and cross effects change from lower to upper values, can be respectively seen.



a) b) c)

Figure 6. a) Standardized effects, b) main effects and c) cross effect for mechanical strength

The obtained results are related to the effect that the same variables have on the porosity. As mentioned before, an increase in the particle size of the composition and in the initial solid mass load leads to a decrease in porosity. This leads to denser grains with higher mechanical strength. Also, an increase in the particle size involves more attractive Van der Waals forces between particles which make grains less breakable.

The increase in the ambient air temperature leads to a decrease in the mechanical strength of the grains. This is consequence of the increase in the drying rate that generates an internal stress in the grains which break easier under an external force.

Finally, from the cross effect (Figure 6 c) it can be seen that when the suspension is flocculated, the initial solid mass load has no effect on the mechanical strength of the grains. However, when the suspension is deflocculated two opposite effects appear. These effects are also related with the porosity. As mentioned before, a deflocculated suspension has a higher porosity and, thus, a lower strength. That happens when the initial solid mass load is low, but increasing the initial solid mass load has the opposite effect and this is predominant when the solid content is high.

With all this information, if a high mechanical strength condition is searched as the optimum for the manufacturing process, the input variables must be the following: standard particle size distribution (wider distribution

with higher mean particle size), high initial solid mass load, low ambient air temperature and deflocculated suspension. Under these conditions the expected value for mechanical strength obtained by means of the Anova method is 442 ± 24 kPa.

The two first conditions are the same that those required for a low porosity, but the deflocculation state is different. However, in this case the highest strength is not desirable. Actually, it has to be high enough to resist handling and processing but small enough to allow an easy deformation during pressing process. Therefore, is more important to know the conditions that provide the lowest porosity and check if the mechanical strength is appropriate for the process. Considering that the maximum load that supports a grain inside a conventional silo is 100 kPa, the optimum value obtained is a very good one.

Conclusions

The final properties of grains obtained from liquid-solid suspension droplets dried in experimental conditions close to those found in ceramic industry have been studied. A standard levitator tube modified to work at high temperature conditions (drying temperature up to 120 ºC) has been used. A ceramic suspension with the same characteristics that those used in the manufacture of industrial porcelain tiles has been used as a reference. The flocculation state, the initial solid mass load, the particle size distribution of the porcelain composition, the ambient air temperature and the initial droplet volume have been modified to study their effect on the mean porosity of the grain and its mechanical strength and to establish the optimal conditions of the process.

In this way, the significant effects that influence porosity are the particle size of the composition, the initial solid mass load and the flocculation state. If low porosity grains are desirable, standard particle size distribution (wider distribution with higher mean particle size), high initial solid mass load and flocculated suspension is needed. Moreover, small initial droplet volume gives a more uniformly distributed porosity.

For the mechanical strength, the significant effects are the particle size of the composition, the initial solid mass load, the ambient air temperature and the cross effect of flocculation state and initial solid mass load. If grains with high mechanical strength are desirable, standard particle size distribution (wider distribution with higher mean particle size), high initial solid mass load, low ambient air temperature and deflocculated suspension is required.

Nomenclature

d	diameter
F	force
R	radius
T	temperature
V	volume
Y	mass fraction
ε	porosity
μ	viscosity
ρ	density
σ_R	mechanical strength

Subscripts

D	droplet
G	grain
max	maximum
P	particle
S	solid
0	initial
10	10% (mass) of particles below d_{P10}
50	50% (mass) of particles below d_{P50}
90	90% (mass) of particles below d_{P90}

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