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Original Research Paper

Spheroidizing mechanisms and simulation of spherical silica in Oxygen-Acetylene flame

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ABSTRACT

In order for heat transferring and spheroidizing mechanisms of silica powders in Oxygen-Acetylene flame to be observed, the experiments were conducted and the Finite Element (FE) simulation method was employed. It has been certified that powder spheroidization occurred only at molten states, primarily depending on two major factors: melting time and particle size. Various particle sizes and various process parameters were studied systematically, whereas following the spheroidization mechanisms were concluded. At a flow rate of 40 L/min, the spheroidization rate percentage of 20 μm silica powders reached the maximum value. At a flow rate of 45 L/min, melting time of all particles were lower than 0.08 s, where it was difficult for 40 μm sized or larger powders to be spheroidized, also sharp corners of bigger powders were still observed. SEM results also revealed that powder spheroidization was affected by flow rate. The silica particle heat transfer time inside the gas flame furnace was reported and compared to SEM results. Through comparative analysis, the simulation results were in agreement with experimental results. The numerical model was capable of particle melting time calculation and can be used in spheroidizing results prediction to a certain extent.

1. Introduction

Spheroidizing process at high temperature is used in a wide range of industries to manufacture particles, such as metal, mineral, ceramic and so on. Over the past years, many methods were developed for spherical powder preparation by high-temperature heat sources, such as radiofrequency thermal plasma and gas flame [1,2]. However, the radiofrequency thermal plasma technique was confined because plasma torches could not easily be enlarged and required higher energy consumptions [3]. As a cost-effective process, the gas flame technique has more promising chance to be a large-scale production technique.

As ceramic particle, spherical silica possess good fluidity, low thermal expansion, high thermal conductivity, low dielectric constant and low stress concentration, it has been widely used and attracted significant research interest in the ultra large scale integration circuit packaging field [4,5]. Elodie et al. [6] indicated that the spherical silica powder was utilized as a unique filler material in the ultra large scale integration circuits packaging field. Sahma et al. [7] utilized Flame Spray Pyrolysis (FSP) to synthesize tin oxide nano-particles from ethylhexanoate precursor in ethanol. Mueller et al. [8] investigated the effects of powder production rates, dispersion gas flow rates and precursor concentration on silica particle size, morphology and carbon content by versatile flame spray pyrolysis. In one of the authors' work [9], a chemical-flame spheroidizing method was proposed for easy low radio activity spherical silicon oxide powder preparation. Mostly, spherical silica powders were obtained by Oxygen-Acetylene flame high-temperature spheroidization routes, as a simple, low-cost, industrialized and suitable route for spherical silica powder production. In this process, the melting silica droplet bonding was the bottleneck and it was affected by characteristics of silica powders, spheroidizing process parameters and spheroidizing furnace structure. Due to complicated heating conditions, an understanding regarding the heat transferring mechanism of silica powders is still not clear enough. Also, it is proven to be difficult for precise temperature and transferring heat measurement at high-temperatures. The numerical simulation method can be applied for heat transferring [10].

With numerical simulation methods for analysis and research reliability can be improved and the cost reduced [11–18]. The Finite Element Method (FEM) constitutes a useful tool in manufacturing process research [19–22]. Kumar et al. [23] conducted a

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theoretical estimation of spheroidization percentage of particles from basic fluid dynamic equations. It was discovered that spheroidization was strongly depended on particle sizes and plasma jet temperatures. Dios et al. [24] utilized Monte Carlo simulations to study the interplay between chemical reaction rates, material inter-droplet exchanges, and the consequences on mechanisms and size distributions of nano-particles synthesized in micro-emulsions. Diarra et al. [25] utilized the finite element method to study certain powders and conduct comparisons between experiments and simulations to improve simulation results and prediction abilities. Thorough numerical simulation methods have a significant advantage in furnace construction and combustion conditions optimization.

Many research contributions were focused mainly on furnaces and interactions between particles. The studies about heat transferring of single silica particle and relative simulations were rarely done. The study regarding heat transferring of single silica particle constitutes a key factor for spheroidizing to be understood and improvement of spheroidizing rate. In this work, the Finite Element Method was used as a predictive tool, as difficulties and modeling choices existed to be addressed for this particular application. Based on previous works [1,9], morphology and microstructure of silica microspheres prepared by Oxygen-Acetylene flame spheroidization were studied. In order for spheroidizing and heat transferring of silica powders to be simulated, FEM was employed. Finally, various heating conditions were introduced in the FEM simulation model for heat transferring mechanics to be clarified and for the corresponding capability evaluation in spheroidizing result prediction. The goal of this paper is for heat transferring and spheroidizing mechanics in Oxygen-Acetylene flame spheroidization to be understood and spheroidizing result prediction.

2. Experimental procedure

The raw material was a natural vein quartz dealt with chemo-mechanical disposal. The raw material purity was 99.9% and the content of the radioactive microelement U was 2.5×10^{-9} g/g. The treated silica powders were inserted into a spheroidization furnace by a powder feeder (Fig. 1). The powders were heated and driven by an oxygen-acetylene flame jet. Following, the powders were melted and spheroidized due to surface tensions and interfacial energy existence. Consequently to removal from the high temperature region, the spheroidized powders were cooled down and were solidified. The silica powders were characterized by scanning electron microscopy (Quanta200, FEI).

The spheroidization furnace consisted mainly of a furnace body and an oxygen-acetylene flame jet. The furnace was constructed of bonded silica carbide bricks that could retain a constant temperature. Silica powders were fed into the flame jet through the powder feeder coaxially fixed to the furnace axis. The initial speed of silica powders varied, according to the adjustments of the carrying gas. The carrying gas was a mixture of both oxygen and nitrogen. The flame temperature could be adjusted by the design of the flame jet and proportion of nitrogen alternation. Infrared temperature sensors and thermocouples were used for flame and furnace body temperature measurement.

The commercial finite element analysis software ABAQUS was used to perform the analysis. The FE simulation was applied for heat transferring and spheroidizing modeling. The model simulates process of silica particle melting and spheroidizing in different work condition temperatures. A 2D axisymmetric model was developed to simplify the 3D model. Therefore, only half of the silica particle was modeled, and symmetry boundary conditions were applied. In the numerical simulations of the silica particle, the geometry was adjustable rectangular region. Five groups of side length were 2 μm × 2 μm, 5 μm × 5 μm, 10 μm × 10 μm, 20 μm × 20 μm, 40 μm × 40 μm, respectively. The mesh was constructed with a four node linear heat transferring quadrilateral elements (DC2D4). The structured technique was used for meshing creation. The optimized model had 10,201 nodes and 10,000 elements. Initially, all particles were at the same room temperature and were heated by fluid convection. The temperature of particles gradually increased until particles are melted or evaporated in the simulations. The physical and chemical properties of utilized silica particles are presented in Tables 1 and 2.

The temperature distribution inside particles was described by conduction as follows [26]:

$$\rho_P C_P \frac{\partial T}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left(k_P r^2 \frac{\partial T}{\partial r} \right) \quad (1)$$

The silica powders were inserted into the spheroidization furnace through the powder feeder and consequently heated and accelerated inside the flame during gas heating. As the flame diffused, the speed of the powder was lowered. The velocity variation with powder airflow temperature was similar to the Ref. [13].

A heat transferring procedure was utilized for the simulation of heat transferring to be performed. The theoretical combustion temperature of the oxygen-acetylene flame was 4451.9 K in the adiabatic system. The temperature could be lowered by proportional adjustment of both oxygen and nitrogen. There were three kinds of heat exchanges between the flame and silica powder, including heat conduction, heat radiation and heat convection. The thermal parameter variety of materials with temperatures was known. In this paper, heat capacity, melting heat and thermal conductivity were considered variables with temperature.

3. Results and discussion

Firstly, temperature distribution was studied inside the furnace. The airflow velocity and temperature distribution inside the furnace could be adjusted by the design of flame jet and flow rate alteration. The furnace temperature could be measured with thermocouples and the temperature of powders could be calculated.

The powder velocity at the middle line is presented in Fig. 2. The temperature of the flame reached up to 2500 °C. The highest temperature appeared at 0.1 m approximately away from the jet, but decreased linearly as the distance between the material location and the jet increased and reached a stable value of approximately 1000 °C at 1 m from the jet.

The surface temperature of powders varied with time following the powder injection into the furnace and it could be calculated according to Fig. 2 and presented in Fig. 3. The temperature of the silica powder surroundings increased sharply following furnace insertion and reached the corresponding maximum value at 0.04–0.05 s, whereas consequently decreased linearly and reached a stable value at 0.5 s approximately.

The difference in temperature between the core and surface of silica powders existed during heating due to finite thermal conductivity. The temperature in sharp corners of silica powders increased faster than the other parts and can be explained by the corresponding large surface area. The larger the surface area, the more heat absorption the parts presented. The silica particles were irregular at the beginning, whereas the temperature differences among various powder parts were apparent. However, during further spheroidization, the temperatures between the core and surface were almost similar.

In this study, the silica particles were considered to be independent during heating to be spheroidized. Therefore, the effects of powder diameter on spheroidization were examined

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