Simulation of the evolution of agglomerated SiO$_2$ particles in thermal plasma flow

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Abstract. The paper studies the behavior of agglomerated SiO$_2$ particles in thermal plasma processing. The main stages are determined for their transformation to the hollow melt droplets. The proposed model, which includes partial encapsulation of gas, allows describing and analyzing the evolution stages for the particle size and the shell thickness in the ranges of 50-150 µm agglomerated particles and their 0.2-0.6 porosity.

1. Introduction

The development of thermal process technologies for various powder materials is becoming more relevant in synthesizing hollow spherical particles of tens of microns due to their wide application in various industries [1-3]. Among the most important application fields is the production of composite constructional and heat-protection materials, catalysts, adsorbents, sound insulators, radar-transparent ceramics, etc [4-7]. Traditional heat sources are not always acceptable, because their temperature modes are insufficient for processing refractory materials with the melting point over 1600°C. Thermal plasma with high energy concentration and temperature range of 3000–10000 K is a promising direction of the powder thermal treatment. Today, plasma technologies are being intensively developed in the production of ceramic hollow microspheres from zirconium dioxide powders [8], silicon dioxide [9] and other materials. Also, a method of aerosol spray pyrolysis is of great interest for the production of metal oxide powders from the solution precursor [10].

Optimum selection of operating parameters for plasma generators producing the particles with controlled parameters requires experimental research and numerical simulation of the evolution of the initial powder particles in a plasma flow. Gulyaev [11] investigated the heating and acceleration dynamics of solid and hollow zirconium dioxide (ZrO$_2$) particles using plasma processing. In [12, 13], hollow microspheres were obtained from agglomerated particles consisting of refractory oxides and silicates processed by a thermal plasma flow. Control for thermophysical parameters of the plasma flow and agglomerated particles allowed synthesizing hollow microspheres with the specified geometric characteristics.

The aim of this work is mathematical simulation of the synthesis of hollow SiO$_2$ particles in a plasma flow. The dynamics of motion, heating, melting and evaporation of the initial powder particles depending on their size and porosity as well as the processes associated with the particle evolution after heating (expansion of the hollow melt droplet, shell thickness formation) are studied in this work.
2. Agglomerate evolution in high-temperature environment

In contrast to known models of generating the hollow particles from agglomerates, we propose the model that includes the encapsulation of gas in a porous particle (αΠ), where α varies in the range of 0÷1. When the agglomerate enters the plasma jet, the particle passes through four stages presented in Figure 1.

![Figure 1. Dynamics of hollow particle produced from the agglomerate processed by thermal plasma.](image)

- **Stage 1.** The particle heating from the initial \( T_0 \) to melting \( T_{melt} \) temperature and the formation of the microporous structure. The gaseous phase (air) in the pores expands and gas releases through the open pores in the particle.

- **Stage 2.** The formation of the primary outer shell \( \delta_1 \). When the agglomerated particles are heated up to \( T_{melt} \), the formation of the molten shell made of coalesced grains occurs. This shell partially encapsulates gas. The material inside the formed particle melts and deposits onto the inner side of the liquid shell under the gas pressure and surface tension.

- **Stage 3.** The formation of the final shell \( \delta_{fin} \). When the particle heating reaches the final \( T_{fin} \) temperature, the pressure in the gaseous cavity increases resulting in the diameter increase of the hollow particle and the thickness reduction of its shell. The final temperature should not exceed the evaporation temperature, viz. \( T_{fin} < T_{vap} \). The formation of the outer \( D_{fin} \) and the inner \( d_{fin} \) diameters of the hollow particle and the shell thickness \( \delta_{fin} \) is driven by three forces: gas pressure \( p \) inside the shell, surface tension \( \sigma_p \), and external pressure \( p_0 \). The hollow particle size and the shell thickness are determined from the equilibrium criterion of these forces.

- **Stage 4.** The shell cooling after the particle moving out from the plasma jet.

3. Hollow particle formation model

The parametric characteristics \( (D_{p,m}, d_{p,m}, \delta) \) of the agglomerate-based hollow particle are evaluated by the system of equations [13]:

\[
\begin{align*}
\left( \frac{d_{p,m}}{D_{p,m}} \right)^3 + K \left( \frac{D_{p,m} - D_{p,0}}{d_{p,m}} \right) &= \alpha \Pi \frac{T_p}{T_0} + (1 - \alpha) \Pi, \\
\left( \frac{D_{p,m}}{D_{p,0}} \right) - \left( \frac{d_{p,m}}{D_{p,0}} \right)^3 &= 1 - \Pi,
\end{align*}
\]

where \( D_{p,0} \) is the initial size of the agglomerated particle; \( D_{p,m} \) and \( d_{p,m} \) are the outer and inner diameters of the hollow microsphere; \( \Pi \) is volumetric porosity of particle; \( \alpha \) is the gas volume fraction in closed pores of the agglomerated particle; \( T_0 \) and \( T_p \) are the particle initial and operating...
temperatures, respectively; $K = (4\sigma_p)/(p_0 D_{p0})$ is the nondimensional parameter of the surface tension. The film thickness of the molten material can be written as $\delta = 0.5(D_{m,p} - D_{p,m})$.

It is important to note that $K$ parameter is very small. Actually, if $D_{p0} = 5 \times 10^{-5}$ m, $\sigma_p = 0.37$ N/m and $p_0 = 10^5$ Pa, this parameter is 0.3 and it lowers down to 0.1 with the initial diameter increased up to $1.5 \times 10^{-4}$ m.

While neglecting the terms containing $K$ parameter in (1), we obtain the following approximated equations:

$$d_{p,m} = \frac{\alpha \Pi T}{T_0} + \left( 1 - \alpha \right) \Pi \right)^{1/3}, \quad D_{p,m} = \frac{1 + \alpha \Pi \left( \frac{T}{T_0} - 1 \right)}{2 \left( \frac{D_{p,m}}{D_{p0}} - \frac{d_{p,m}}{D_{p0}} \right)} \quad \text{(2)}$$

At $K = 0.3$, the error in (2) does not exceed 10% for $D_{p,m}/D_{p0}$ and 20% for $\delta/D_{p0}$. With decreasing $K$ parameter (increasing initial diameter), the error also decreases.

The system of equations (1) is solved numerically for SiO$_2$ in the parameter range of $50 \leq D_{p0} \leq 150$ µm; $\rho_p = 2.65$ g/cm$^3$; $0.2 \leq \Pi \leq 0.6$; $T_{\text{melt}} = 1993$ K; $T_{\text{vap}} = 3048$ K; $\sigma_p = 0.37$ N/m; $p_0 = 10^5$ Pa; $T_{\text{melt}} \leq T \leq T_{\text{vap}}$, $0 \leq \alpha \leq 1$. The parameter $K$ for the problem of interest varies between $0.1 \leq K \leq 0.3$. Figures 2 and 3 present the results of numerical simulation of the agglomerate-based particle evolution depending on the initial porosity of the particles processed by thermal plasma.

![Figure 2](image2.png)

**Figure 2.** Calculated characteristics of hollow particle: a) dependences between the final particle size and the precursor particle porosity; b) dependences between the final relative shell thickness and the precursor particle porosity. Solid and dashed lines indicate $K$ parameter of 0.3 and 0.1, respectively. Gas volume fraction: 1 – 0, 2 – 0.1, 3 – 0.5, 4 – 1.

Figure 3 shows that in changing the initial porosity ($\Pi = 0\text{–}0.6$), the particle properties ($D_{p,m}$, $\delta$) significantly differ due to the different content of closed and open pores.

When the particle has no closed pores ($\alpha = 0$), the final particle size does not differ from the initial. At $a \leq 50$ µm particle size and a significant surface tension ($K \geq 0.3$), the final particle size can be smaller than the initial. When the concentration of closed pores is insignificant, the difference between $D_{p0}$ and $D_{\text{fin}}$ values does not exceed 10%. In the case of the substantial amount of closed pores ($\alpha = 1$), this difference can achieve 70%. Further heating of the hollow microsphere up to $T_{\text{vap}}$ causes a slight expansion ($\leq 5\%$) of the gaseous cavity. The outer and inner diameters of the molten film as well as its thickness are then mainly determined during the melting process ($T_P \geq T_{\text{melt}}$), and change slightly after further heating up to the evaporation temperature of the condensed material. The proposed model allows controlling the parametric characteristics of the synthesized microspheres due to the thermophysical parameters of the plasma jet provided by the operating modes of the plasma generator.
**Figure 3.** Temperature dependence of the hollow particle size (a) and relative shell thickness (b) after melting.

**4. Conclusions**

Based on the studied behavior of SiO$_2$ particles in the thermal plasma flow, it can be concluded that the evolution of the agglomerated particle into the hollow microsphere occurred in several stages. Using the proposed process model which involved the partial encapsulation of gas, the evolution of the hollow particle size and shell thickness was analyzed in the range of 50-150 µm agglomerated particles and their 0.2-0.6 porosity. This paper has clearly shown that:

- The outer and inner diameters and the thickness of the molten film were mostly determined during the melting process and changed slightly with further heating.
- The final particle size strongly differed in the presence of open and closed pores. That difference could reach 70%.
- When the pore concentration was low ($\alpha < 0.1$), the final particle size insignificantly differed from the initial size of the agglomerated particle.

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