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Mechanisms of thermal process of zinc ultrafine powder preparation by inert gas condensation

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ABSTRACT

Zinc ultrafine powder is widely used as key raw material for anti-corrosion paint in recent years. Inert gas condensation (IGC) is one of the primary means for preparing zinc ultrafine powder with a clean interface. As condensation is the most important process that determines the powder's size, this paper presents new models for radiative and convective cooling in condensation process by studying the microphysical mechanisms. In order to study the microphysical mechanisms of zinc nucleation process in the inert gas condensation, this paper established mathematical expressions to deduce the bonding probability of collisions between condensation nuclei particles and vapor molecules during the nucleation process. The two models have been used to predict the diameter of zinc condensation nucleus. Compared with experiments that have been done, the prediction errors are only less than 7%. This may provide a theoretical method for prediction of ultrafine powder diameter in IGC.

1. Introduction

Metal ultrafine powders have generated considerable recent research interest because of their unique properties, such as superior catalytic, magnetic and electronic properties [1]. As zinc ultrafine powder plays an important role in the production of anti-corrosion paint, the production of zinc ultrafine powder attracts much attention. IGC is a popular method to fabricate zinc ultrafine powder. Zinc is vaporized into low density gas by thermal plasma and other ways of evaporation. Then the generated vapor is condensed into ultrafine particles by cold inert gas [2,3].

The IGC process includes metal evaporation, nucleation mechanism, nuclei growth, crystalline structure and other principles. This study is focused on the nucleation mechanism and nuclei growth. Three theoretical analyses were adopted to explore ultrafine powder particle nuclei growth in IGC. Some researchers applied classical nucleation theory (CNT) to explain condensation process [4,5]. Some researchers have chosen density functional theory (DFT) to study gas–liquid nucleation [6]. Others simulate the nucleation process by molecular dynamics simulation (MD) [7,8].

These studies focused on either the macroscopic thermodynamics and kinetics analysis, or the qualitative analysis of ultrafine powder particle's size distribution by statistical method or molecular dynamics simulations by the classical equations of motion. They are worthwhile. Nevertheless, few studies have been done on the microphysical mechanisms of nucleation process and nuclei growth process in IGC.

In order to provide a theoretical method for the calculation of ultrafine powder diameter in IGC, the microscopic mechanisms of nucleation are needed to be studied. The purpose of this paper is to explore the microscopic mechanisms of homogeneous nucleation process in zinc vapor condensation process.

2. Collision bonding probability

The physical process of ultrafine powder preparation by IGC can be described as [9,10] the vapor of material condenses into nuclei through phase transition in a specific system space. The condensation nucleus is formed from the collision and combination among the vapor molecules. First, this study should investigate the frequency of collision between condensation nucleus and vapor molecules. Second, the probability model of collision and bonding should be established in order to calculate the heat transfer in the process of collision and bonding. Collision bonding is related to collision angle, collision velocity and collision section. The collision bonding probability is inversely proportional to the collision velocity, and is proportional to collision angle and collision section. Collision without binding is defined as invalid collision. We assume that invalid collision decreases exponentially with the increase of the molecule number inside a condensation nucleus. The max value of collision bonding probability is 1, so the collision bonding probability between condensation nuclei and vapor molecules can be defined as:

$$P_{mp} = 1 - e^{-mxi} \tag{1}$$

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where m is constant and i is the molecule number in a single condensation nucleus. The first term on the right represents the max value of collision bonding probability, and the second one represents invalid collision probability. The physical significance of the above equation is that the collision bonding probability between condensation nuclei and vapor molecules will increase exponentially with the increase of the molecule number inside the condensation nucleus.

3. Energy analysis of nucleation and growth processes

The zinc ultrafine powder preparation process by IGC is shown in Fig. 1. Based on an energy analysis it's possible to establish the process of condensation nuclei growth.

3.1. Collision frequency between condensation nuclei and vapor molecules

In real industry production, there are two ways to produce zinc powder. In periodic production line, a certain amount of zinc is provided at a time, so the number of zinc molecules is fixed in the space. In continuous production line, zinc is provided continuously; in this case, the density of zinc vapor is fixed in the space. The two conditions are formulated in order to describe the two production patterns and compare their production results.

The relationship between molecular volume and condensation nuclei volume is:

$$i \times \frac{\pi}{6} d^3 = \frac{\pi}{6} D^3 \times K_b \tag{2}$$

where d is the molecule's diameter, D is the nuclei's diameter, and K_b is molecule density inside the nuclei. A condensation nucleus is small at nucleation stage, so it can be regarded as a multi-molecular particle. Each condensation nucleus grows up uniformly with the same growth rate. The density number of vapor molecules is n_v and that of condensation nuclei is n_c before the condensation. The average temperature of both vapor molecules and condensation nuclei is represented as T_v. When there is a certain number of vapor molecules, combined with Eq. (2) and based on two different sizes of particles, the total collision frequency between condensation nuclei and vapor molecules can be obtained as follow [11–13]:

$$Z_i = n_c(n_v - i \times n_c) \times \frac{\pi}{4}\left[\left(\frac{i}{K_b}\right)^{\frac{1}{3}} + 1\right]^2 d^2 \cdot \sqrt{\left(1 + \frac{1}{i}\right)\frac{8kT_v}{\pi m_v}} \tag{3}$$

where n_c—the density number of condensation nuclei, m^{-3}; n_v—the density number of vapor molecules, m^{-3}; k—Boltzmann's constant, $k = 1.38 \times 10^{-23}$ J/K; T_v—the average temperature between vapor molecules and condensation nuclei, K; and m_v—molecular mass of vapor, k_g.

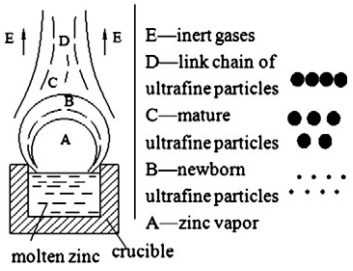

Fig. 1. The process of IGC.

Similarly, when there are a certain density number of vapor molecules, the total collision frequency between condensation nuclei and vapor molecules can be obtained as follow:

$$Z_i' = n_v n_c \times \frac{\pi}{4}\left[\left(\frac{i}{K_b}\right)^{\frac{1}{3}} + 1\right]^2 d^2 \times \sqrt{\left(1 + \frac{1}{i}\right)\frac{8kT_v}{\pi m_v}}. \tag{4}$$

If we assume that the degree of temperature for under cooling in the system is 150 K, then the temperature is $T_v = T_b + 150$ K, where T_b is the material boiling temperature, and condensation nuclei number is one thousandth of total vapor molecular number, that is $n_c = n_v / 1000$. Then substituting zinc's parameters into Eqs. (3) and (4), the variation of collision frequency between condensation nuclei and vapor molecules can be obtained, as shown in Fig. 2.

3.2. Energy analysis of vapor molecules condensation

At the beginning of the homogeneous nucleation, the condensation nuclei are tiny with high temperature and, although there is no regular crystal structure formed between them, the relative position is more stable.

According to the solid state physics [14], most of the coordination numbers in crystal structures are 8 or 12 and can decrease 2–3 in molten state. Analyzing the small scale of condensation nuclei by theory of valency [15], the number of unsaturated bonds of its molecules surface is superior, generally 4–6. At this point, if only taking the nearest layer molecular attraction into consideration, 5–7 atom dissociations will be needed before a vapor molecule enters the condensation nucleus surface, that is breaking 3 pairs of molecule pair potential energy $w(r_0)$, while absorbing $3\varepsilon_b$ energy, where ε_b is the absolute value of the minimum potential energy between molecules.

After the molecule enters into the condensation nucleus, it will form 5–7 pairs of molecule pair potential energy, while releasing (5–7) ε_b energy. So the whole process of molecular condensation is an exothermic process, and each molecule will release about $3\varepsilon_b$ bonding energy. Accordingly, if the vapor molecule enters into the condensation nucleus of zinc, $5\varepsilon_b$ energy is absorbed at the beginning and $10\varepsilon_b$ energy is released after condensation, which makes the overall effect releasing about $5\varepsilon_b$ bonding energy.

From the micro-scale, when a condensation nucleus collides with the vapor molecule, the bonding energy releases only by radiation. If the vapor molecule enters and stays on the surface of condensation nucleus, the bonding energy will dissipate in two ways, as shown in Fig. 3. If the bonding energy radiates to the adjacent molecule, it will characterize heat conduction of the condensation nucleus q_{cond}, of which part of the bonding energy maintains the condensation nucleus at high temperature or raises it, and then dissipates by radiation and

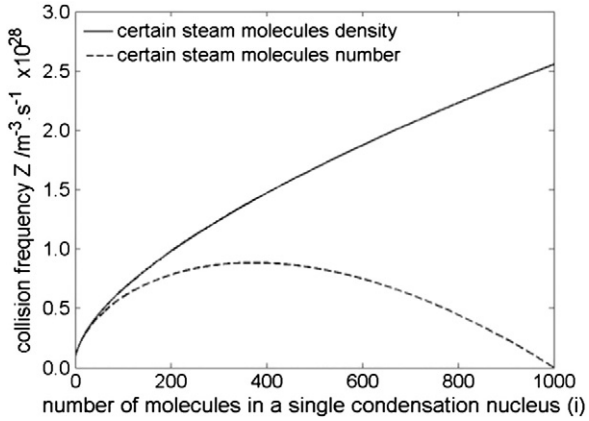

Fig. 2. Variation of collision frequency between condensation nuclei and vapor molecules.