

Cooling Rate Calculation of Non-Equilibrium Aluminum Alloy Powders Prepared by Gas Atomization

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Abstract: The cooling rate of aluminum alloy powders prepared by ultrasonic gas atomization process was calculated through the convection heat transfer principle. A simple and theoretical model is established, which can be expressed as

$$\left| \frac{dT_d}{dt} \right| = \frac{12}{\rho \cdot C_p} \cdot (T_d - T_f) \cdot \frac{k_g}{d^2}$$

The average cooling rates of Al-Ni-Ce-Fe-Cu alloy powders prepared by argon gas atomization and by helium gas atomization are about $10^4 \sim 10^7$ K/s and $10^5 \sim 10^8$ K/s, respectively. The critical cooling rate is calculated to be 3.74×10^5 K/s for Al-Ni-Ce-Fe-Cu alloy amorphous powders prepared by argon gas atomization. The cooling rates of gas-atomized powder particles estimated from secondary dendrite arm spacing are in consistence to those predicted from the theoretical model.

Key words: gas atomization; cooling rate; aluminum alloy; amorphous; secondary dendrite arm spacing

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Ultrasonic gas atomization has become a popular and preferred method for the production of rapidly solidified powders of metals and alloys^[1]. Atomized powders have such outstanding properties, as decreased segregation, increased solid solubility, elimination of segregation phases and fine microstructure. A key factor in gas atomization which influences powder microstructure is the cooling rate. The average cooling rate of rapid solidification processes has been indicated to be in the range of 10^7 to 10^4 K/s. Most reported cooling rates are based either on dendrite arm spacing measurements for microcrystalline materials^[2,3] or on indirect estimation for amorphous alloys^[4,5]. Direct measurement is often subject to inherent restrictions of the measuring method. On the other hand, traditional theoretical calculation of cooling rates necessitates construction of time-temperature-transformation (TTT) curves, which is time-consuming and requires many parameters. Thus, it is of great significance to calculate the cooling rates of rapidly solidified powders using an easy-to-manuever model.

In this study, an analytical simulation Al-Ni-Ce-Fe-Cu alloy powders of convection heat transfer in the rapid solidification process by ultrasonic gas atomization is proposed, and a simply theoretical model leads to excellent agreement between the simulation and experimental results.

1 Theoretical Model

In the ultrasonic gas atomization process the highly energized atomizing gas at ultrasonic frequencies is achieved by an atomizer which is based on the Hartmann shock-wave generator. The molten metal stream is broken up into fine spherical droplets immediately after the liquid metal stream comes in contact with the ultrasonic, pulsed impinging gas. Quantitative modeling of heat transfer phenomena in ultrasonic gas atomization is rather complex and difficult because of the uncertainties about the nature and roles of various fluid dynamic phenomena that may take place. In ultrasonic gas atomization, the cooling method of droplets follows Newton heat transfer mode^[6]. Further simplifications of

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analysis can be achieved by the following assumptions:

- (a) droplets have a spherical shape which is rigid and constant;
- (b) droplets form immediately at the nozzle outlet;
- (c) the velocity field of gas flow is constant;
- (d) droplets are supposed to move a straight path on the flow line.

The heat balance condition of molten droplets during solidification can be regarded as: the heat that molten droplets release equals to the thermal flux from the molten droplets surfaces to the ambient environment. The balance of heat equation is given as [7]:

$$-V \cdot \rho \cdot C_p \cdot \frac{dT_d}{dt} = h \cdot A \cdot (T_d - T_f) \quad (1)$$

where V , ρ , C_p , h , A , t , T_d and T_f are the droplet volume (m^3), density of molten droplet (kg/m^3), specific heat ($J/(kg \cdot K)$), heat transfer coefficient ($W/(m \cdot K)$), surface area of droplet (m^2), time (s), droplet temperature (K) and atomizing gas temperature (K), respectively.

Suppose that the droplets are spherical, whose diameter is d , Eqs. (1) may be changed into

$$\frac{dT_d}{dt} = -\frac{h \cdot A}{V \cdot \rho \cdot C_p} \cdot (T_d - T_f) = -\frac{6h}{\rho \cdot C_p \cdot d} \cdot (T_d - T_f) \quad (2)$$

From Eqs. (2), the cooling rates of droplets are affected by the density of molten droplet, specific heat, the size of droplet, heat transfer coefficient and the temperature difference between the droplets and the gas.

According to Szekely^[8], the heat transfer coefficient (h) between inert gas and molten droplets could be calculated by the formula

$$h = \frac{k_g}{d} (2.0 + 0.6 \sqrt{R_e} \cdot \sqrt[3]{P_r}) \quad (3)$$

where k_g is the gas thermal conductivity, P_r the gas Prandtl number given by $(C_g \mu_g / k_g)$ and R_e the Reynolds number, defined by $(\rho_g d / \mu_g) |v_d - v_g|$.

Reynolds number is dimensionless, and according to Estrada^[9], R_e can be expressed as

$$R_e = \frac{U \cdot \rho_g \cdot d}{\mu_g} \quad (4)$$

where ρ_g , μ_g , d and U are the density of gas, kinetic viscosity, droplet diameter and the relative velocity between the inert gas and molten droplets, respectively.

From Eqs. (3) and (4), we can get

$$h = \frac{2.0 k_g}{d} + 0.6 k_g \sqrt{\frac{U \cdot \rho_g}{\mu_g \cdot d}} \cdot \sqrt[3]{P_r} \quad (5)$$

And from Eqs.(2) and (5), we have

$$\left| \frac{dT_d}{dt} \right| = \frac{6}{\rho \cdot C_p} \cdot (T_d - T_f) \cdot \left(\frac{2.0 k_g}{d^2} + 0.6 \frac{k_g}{d} \sqrt{\frac{\rho_g U}{\mu_g d}} \cdot \sqrt[3]{P_r} \right) \quad (6)$$

The melt disperses into different size droplets after atomizing. And the droplets are subsequently accelerated in the gas stream. The smaller droplets can gain larger acceleration, thus which can reach quickly the atomizing gas velocities and solidify swiftly. While the larger droplets gain smaller acceleration, so they can not keep up with the high-speed gas, but they disengage the fast gas beam area. Based on the two cases, we suppose that the relative velocity (U) between droplets and atomizing gas could be approximately equal to zero during gas-atomized process^[10]. Eqs.(6) is then simplified as follows:

$$\left| \frac{dT_d}{dt} \right| = \frac{12}{\rho \cdot C_p} \cdot (T_d - T_f) \cdot \frac{k_g}{d^2} \quad (7)$$

We can gain the following results according to the model just developed. The calculation of cooling rates is simple, which does not depends on parameters such as solidification time, nucleation rate and the quantity of heat (ΔH), etc. The calculation of cooling rates is dependent only on the powder particle size, the temperature difference between the molten droplet and the ambient, and the intrinsic thermophysical properties of the melt and the atomizing gas.

2 Cooling Rate of Aluminum Alloy Powder

To test the above model, the Al-Ni-Ce-Fe-Cu alloy was used, because the authors have already studied the alloy^[11].

The following assumptions are also considered, which are introduced for the sake of simplicity.

- (1) We substitute the specific heat of common aluminum alloy for the value of Al-Ni-Ce-Fe-Cu alloy.
- (2) The temperature of droplets is considered as 1473 K, which approaches the as-atomized temperature.
- (3) The ambient temperature is considered as 300 K.

Considering aluminum alloy droplets atomized by argon gas, coefficient of heat conductivity of argon gas is $1.79 \times 10^{-2} W / (m \cdot K)$ at 0.1 MPa and 300 K^[12]. The thermophysical properties of aluminum are shown in Tab.1^[12].

If we substitute the properties in Tab.1 into Eqs.(7), we can obtain the cooling rate as follows:

$$\left| \frac{dT_d}{dt} \right| = \frac{12}{3400 \times 880} \cdot (1473 - 300) \cdot \frac{1.79 \times 10^{-2}}{d^2} = \frac{8.42 \times 10^{-5}}{d^2} \quad (8)$$

Fig.1 is the surface morphology of Al-Ni-Ce-Fe-Cu alloy powders prepared by argon gas atomization. The microstructures of alloy powders in Fig.1(a), which is smaller than 15 μm , are amorphous. The size of powder particle in Fig.1(b) is about 65 μm , being crystalline, and the cooling rate is $2.0 \times 10^4 K/s$ according to Eqs.(8).

Table 1 Thermophysical properties of aluminum alloy

Density/ kg·m ⁻³	Specific heat /J·kg ⁻¹ ·K ⁻¹	Droplet temperature/K	Ambient temperature/K
3400	880	1473	300

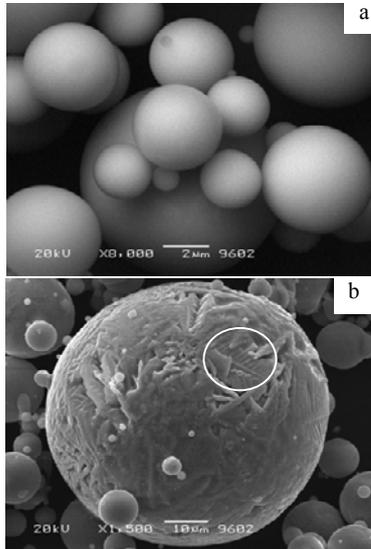


Fig.1 Surface morphology of Al-Ni-Ce-Fe-Cu powder: (a) amorphous powders and (b) about 65 µm

When the size of as-atomized powder particle is smaller than 15 µm, the microstructure of Al-Ni-Ce-Fe-Cu alloy powders is amorphous^[11]. Thus we can obtain the critical cooling rate of Al-Ni-Ce-Fe-Cu amorphous alloy powder which is 3.74×10^5 K/s according to Eqs.(8). The cooling rates of gas atomization affect obviously the microstructures of alloy powders. When the cooling rates are larger than 3.74×10^5 K/s, non-equilibrium aluminum alloy powders, such as amorphous and nanocrystal, may be obtained. While the cooling rates are smaller than 3.74×10^5 K/s, the microstructures may be crystalline.

At the same time, we can verify the cooling rates by the relationship between the cooling rate and the secondary dendrite arm spacing. SDAS could be determined by the following equation^[13]:

$$\lambda_2 = B(CR)^{-n} \tag{9}$$

where λ_2 is secondary dendrite arm spacing, µm; CR the cooling rate, K/s. Here, the constants B and n are 47 and 0.33^[7], respectively.

It can be measured that the secondary dendrite arm spacing is about 1.75 µm from the circle in Fig.1b. Thereby the cooling rate is 2.1×10^4 K/s according to Eqs.(9). The experimental result is consistent with the calculated result (2.0×10^4 K/s).

Suppose that the atomizing gas is helium gas, since

its coefficient of heat conductivity is $0.15 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ at 0.1 MPa and 300 K^[12], and then the cooling rate will be

$$\left| \frac{dT_d}{dt} \right| = \frac{12}{3400 \times 880} \cdot (1473 - 300) \cdot \frac{0.15}{d^2} = \frac{7.06 \times 10^{-5}}{d^2} \tag{10}$$

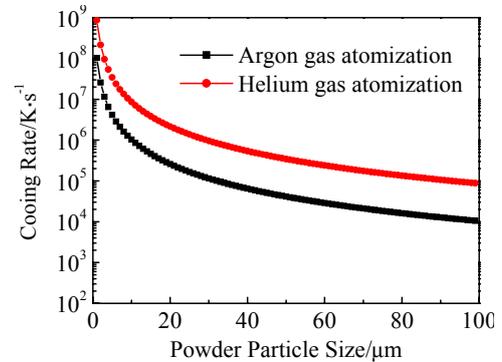


Fig.2 The relationship between cooling rates and powder particle sizes of as-atomized aluminum alloy powder by different ambient gas

Fig.2 shows the relationship between the sizes of as-atomized powder particles and the cooling rates in different atomizing gas according to the simple model. It indicates that the cooling rate has a significant effect on the powder particle sizes of as-atomized aluminum alloy powders. The average cooling rate of aluminum alloy powders prepared by argon gas atomization is between about 10^4 to 10^7 K/s. While the average cooling rate of aluminum alloy powders prepared by helium gas atomization is between about 10^5 to 10^8 K/s. Aluminum alloy powders prepared by helium gas atomization are indicated to have about one order of magnitude higher cooling rates as compared with aluminum alloy powders prepared by argon gas atomization. The calculated result is consistent with that of Wang^[14]. The model also correctly predicts that the cooling rate is proportional to the thermal conductivity of the atomizing gas.

3 Conclusions

1)The cooling rate of aluminum alloy powders prepared by gas atomization process through the convection heat transfer principle can be expressed simply as follows:

$$\left| \frac{dT_d}{dt} \right| = \frac{12}{\rho \cdot C_p} \cdot (T_d - T_r) \cdot \frac{k_g}{d^2}$$

2) Aluminum alloy powders prepared by helium gas atomization are indicated to have about one order of magnitude higher cooling rates as compared with aluminum alloy powders prepared by argon gas atomization.

3) The critical cooling rate is 3.74×10^5 K/s to obtain aluminum amorphous alloy powders prepared by argon gas atomization.

4) The theoretical model leads excellent agreement between the simulation and experimental results.

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气雾化制备非平衡态铝合金粉末冷却速度的计算

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摘要: 依据对流换热原理, 对超音速气体雾化非平衡态铝合金粉末的冷却速度进行了理论计算。获得了一个较简单的理论计算公式, 其表达式为 $\left| \frac{dT_d}{dt} \right| = \frac{12}{\rho \cdot C_p} \cdot (T_d - T_f) \cdot \frac{k_g}{d^2}$ 。根据理论公式, 氩气和氦气雾化制备铝合金粉末的冷却速度分别为 $10^4 \sim 10^7$

和 $10^5 \sim 10^8$ K/s, 其结果与前期科研者的计算结果相符, 且计算公式更简化。对于氩气雾化制备 Al-Ni-Ce-Fe-Cu 合金而言, 获得非晶态粉末其临界冷却速度为 3.74×10^5 K/s。通过测定合金晶态粉末的二次枝晶臂间距, 并利用冷却速度和枝晶臂间距之间的经验关系, 验算了合金粉末的冷却速度, 其结果与理论计算相吻合。

关键词: 气雾化; 冷却速度; 铝合金; 非晶; 二次枝晶

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