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Characterization of cooling rate and microstructure of CuSn melt droplet in drop on demand process

Yi XU[1], N. ELLENDT[2], Xing-gang LI[3], V. UHLENWINKEL[2], U. FRITSCHING[2]

1. School of Materials Science and Engineering, Southwest Jiaotong University, Chengdu 610031, China;
2. Foundation Institute of Materials Science, Bremen University, Bremen 28359, Germany;
3. National Engineering and Technology Research Center for Nonferrous Metal Matrix Composites, General Research Institute for Nonferrous Metals, Beijing 100088, China

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Abstract: Different sized single droplets of Cu−6%Sn alloy were prepared by drop on demand (DOD) technique. The secondary dendrite arm spacing was measured and correlated with the droplet cooling rate by a semi-empirical formula. The microstructure of droplets was observed by optical microscopy (OM) and electro backscatter diffraction (EBSD). The dendrite feature of single droplets depends on solidification rate, cooling medium and flight distance. When droplets collide with each other at temperatures between solidus and liquidus, the dendrites and grains are refined obviously possibly because the collision enhances the heat transfer. The cooling rate of colliding droplets is estimated to be more than 4×10^4 K/s based on a Newton's cooling model. The dendrites grow along the colliding direction because of the temperature gradient induced by the internal flow inside the droplets.
Key words: impinging droplet; secondary dendrite arm spacing; cooling rate; heat transfer; drop on demand

1 Introduction

Rapid solidification techniques have been extensively developed in the last decades as they enable to obtain an original microstructural and constitutional features in the final products which cannot be formed under conventional solidification processes [1]. Among the other methods, atomization is an innovative approach that is capable of producing droplets of controlled sizes with a relatively narrow distribution and a predictable cooling rate [2]. This technique has successfully atomized metallic materials such as aluminum, zinc, copper, cobalt, nickel and their alloys as well as steel [3].

The large deviations from thermodynamic equilibrium induced by rapid solidification significantly alter the solidification conditions compared with those obtained at or close to equilibrium. The final properties can be modified through microstructure morphology change [1], extended solute solubility, nonequilibrium phase formation [4] or structure refinement.

Together with undercooling and cooling rate, the resulting microstructure is process dependent. CHEN et al [5] investigated the microstructure evolution of atomized powders of Fe-dosed Al alloy, and studied the effect of Fe content on the dendrite growth direction. CIFTCI et al [6] quantified the amorphous fraction of atomized powders and computed the critical cooling rate of approximately 5000 K/s. ILBAGI and HANI [7] effectively utilized various 2D and 3D characterization techniques to investigate the effect of cooling rate on the phase fractions. KHATIBI DELSHAD and HENI [8] produced D2 tool steel powders using a drop tube-impulse atomization, measured the in-situ droplet velocity and droplet size, and deduced the droplet cooling rate using a thermal model of droplet cooling. BEDEL et al [1] investigated the development of the dendrite arms occurring in most droplets along (111) crystallographic axes, and discussed the impact of different processing parameters on the final distribution of dendrite morphologies. ELLENDT et al [9] used the impulse atomization for spray deposition, and compared

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Corresponding author: Yi XU; Tel: +86-28-87600782; E-mail: xybwbj@swjtu.cn
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different spray characteristics and impinging conditions of the droplets with gas atomization technique which is the common technique used for spray deposition. ZHAI and WEI [10] investigated the effect of substantial undercooling condition on the direct nucleation and growth of peritectic phase of Cu−70%Sn alloy.

Nowadays, the drop on demand (DOD) technique has attracted more and more attention from researchers as a kind of three-dimensional(3D) printing technology. In the present study, we aim to prepare single impinging Cu−6%Sn droplets, to investigate the dendrite feature, to calculate the effects of droplet diameter and flight distance on the cooling rate of single droplets based on the Newton's cooling law. A semi-empirical formula will be derived to describe the relationship between dendrite arm spacing and droplet cooling rate.

2 Experimental

Drop on demand (DOD) experimental facility was set up in Bremen University where a liquid alloy jet emanating from a capillary can be destabilized when disturbed with a specific wave. The unit included a gas pulser which pushed the molten metal through a nozzle at the bottom of the graphite crucible. Nitrogen was used as impulse gas and cooling gas in this experiment. The Cu−6%Sn alloy was heated upto and held at 1150 °C in the crucible, and the melt temperature near the atomizing nozzle was recorded by a thermocouple immersed in the crucible. The impulses generated discontinuous streams of molten metal, which broke up into fine droplets subsequently. A video camera pointing at the crucible bottom recorded each single droplet.

Single droplets with a uniform diameter (d) of 320 μm can be obtained at a specific pulse frequency. The initial velocity of single droplets was 0.5 m/s. These droplets flew through a stagnant nitrogen gas atmosphere, fell into different beakers filled with two kinds of cooling medium, i.e., oil or water, and then solidified as spherical particles. These beakers were located at 0.22, 0.48 and 0.82 m away under the crucible bottom, respectively. The collected droplets samples in different beakers were marked by h_1-o (0.22 m, oil), h_1-w (0.22 m, water), h_2-o (0.42 m, oil), h_2-w (0.42, water), h_3-o (0.82, oil) and h_3-w (0.82 m, water), respectively. The heat transfer occurred between the static nitrogen gas environment and the droplets in the whole flight stage. The heat transfer intensified when these droplets fell into the cooling liquid.

Different sized droplets can be fabricated by the drop generator in a range between 220 and 741 μm. The secondary dendrite arm spacing of different sized droplets was measured, respectively, and was related to the droplet cooling rate based on the mathematical methods and experimental results. In addition, according to the calculated solidification rate of melt droplets, the distance between the deposit substrate and the crucible bottom was selected as 0.15 m where droplets consisted of solid and liquid phases when these droplets collided with each other. One purpose of the present contribution is to study the effect of impinging on the microstructure characteristics and the solidification rate of Cu−6%Sn alloys.

An optical microscope Olympus BX51 was used to take photomicrographs of samples of each particle size range. The characteristic lengths of secondary dendrite arm spacing and cell spacing were measured. More detailed observations of the microstructure were carried out using a JXA−8200 backscatter scanning electron microscope (BSEM). For each micrograph, four lines perpendicular to the growth direction of secondary dendrite were drawn across the particle microstructure. The number of cell/dendrite intercepts (n) was counted for a line of known length (l). Most of the secondary dendrite arm spacings (λ) were calculated using 40 lines to get a statistically meaningful value. The cell/dendrite spacing (λ) is then given by Eq. (1):

$$\lambda = \frac{l}{n-1} \tag{1}$$

3 Mathematical model

A mathematical droplet-cooling model based on the Newton's cooling law was used to calculate thermal history of an alloy droplet during its free fall in a static gas. According to the formula derivation, the thermal history can be determined using the following Eq. (2):

$$R_\text{C} = \frac{\mathrm{d}T}{\mathrm{d}t} = \frac{3h(T-T_0)}{Rc_p\rho} \tag{2}$$

where h is the effective heat transfer coefficient and consists of the additive contribution of convection and conduction heat transfer mechanisms in the environment, the cooling rate (R_C) greatly depends on the heat transfer coefficient between droplet surface and gas; T is the droplet surface temperature; T_0 is the free stream gas temperature; ρ is the density of alloy material; c_p is the specific heat capacity; and R is the powder particle radius. Because undercooling and microsegregation had a negligible effect on the calculated cooling rate [11], the effect of undercooling on the cooling rate and the microstructure is thus ignored in order to simplify the