Materials Characterization 109 (2015) 9–18

Contents lists available at ScienceDirect

Materials Characterization

journal homepage: www.elsevier.com/locate/matchar

In situ synchrotron X-ray studies of the coupled effects of thermal and solutal supercoolings on the instability of dendrite growth

Faguo Li [a,b], Jiao Zhang [a,*], Qing Dong [a], Yongbing Dai [a], Yanan Fu [c], Honglan Xie [c], J. Mi [d], Fucheng Yin [e], Baode Sun [a,b]

[a] State Key Laboratory of Metal Matrix Composites, Shanghai Jiao Tong University, Shanghai 200240, China
[b] Shanghai Key Laboratory of Advanced High-temperature Materials and Precision Forming, Shanghai Jiao Tong University, Shanghai 200240, China
[c] Shanghai Synchrotron Radiation Facility, Shanghai Institute of Applied Physics, CAS, Shanghai 201204, China
[d] School of Engineering, University of Hull, Cottingham Road, East Yorkshire, HU6 7RX, UK
[e] School of Materials Science and Engineering, Xiangtan University, Xiangtan 411105, China

ARTICLE INFO

Article history:
Received 5 March 2015
Received in revised form 23 August 2015
Accepted 4 September 2015
Available online 12 September 2015

Keywords:
Morphology
Dendrite formation
X-ray radiography
Aluminum alloys
Doublon

ABSTRACT

Special growth pattern representing unique growth conditions is a vital clue to investigate the morphology evolution mechanism in metallic alloy solidification. The dendrite pattern and growth orientation of dendritic doublons in the hypoeutectic Al-Cu alloy have been studied by synchrotron radiation imaging technology and electron back scatter diffraction. The results show that this kind of doublon is two-dimensional and the secondary arms grow perpendicularly to the primary stem. This doublon morphology can appear as an equiaxed grain, columnar dendrite, or coexist with a regular dendrite. The growth directions of the dendritic doublon tips and the secondary arms are <110> and <001>, respectively. Rising cooling rate or Cu concentration in the alloys facilitates the formation of the doublonic structure. According to the kinetic morphology diagram of Al-Cu alloys (pattern-dimensionless supercooling-anisotropy) obtained from the experimental data, the dendritic doublon forming region was above the boundary of fractal dendrite and compact dendrite.

1. Introduction

Dendritic growth is an important process in structure formation during solidification [1]. Due to the anisotropy of the solid–liquid interfacial energy, a growing nucleus generally becomes unstable when its radius is several times larger than the critical radius, and then evolves into a dendrite [1,2]. The crystalline anisotropy determines the crystalline orientation and the supercooling drives the phase interface advance. With regard to the selection of the crystal pattern, Brener et al. proposed a kinetic morphology diagram for 2D single component systems in diffusional growth (see Fig. 7a) [3] to illustrate the relationship between the dimensionless supercoiling and the strength of anisotropy to the pattern.

The dendritic structure and seaweed structure are the basic patterns. The building block of the dendritic structure is a dendrite with a parabolic tip and has a pronounced orientational order; however, the basic element of the seaweed is a doublon without any apparent orientational order. The internal structures of the patterns are further classified into fractal and compact. A fractal pattern consists of a self-similar or self-affine internal structure [4]. Thus, four patterns are included in the proposed diagram, i.e., compact dendrite (CD), compact seaweed (CS), fractal dendrite (FD), and fractal seaweed (FS). There are some basic relationships in the transition between crystal structures.

(i). There exists two critical values, ε^* and Δ^*.

When the $\varepsilon > \varepsilon^*$, the compact dendrite has no side-branches without noise and shows a needle crystal structure [3].

$$\varepsilon^* \approx |\ln \Gamma|^{-8/7} \tag{1}$$

Γ is the Gibbs–Thomson coefficient [5].

The black dot line separating the CS and FS structures in Fig. 7a corresponds to the Δ^* [3].

$$\Delta^* \approx \varepsilon^{*1/2} + 1/2 \tag{2}$$

* Corresponding author.
E-mail address: zj119@sjtu.edu.cn (J. Zhang).

http://dx.doi.org/10.1016/j.matchar.2015.09.008
1044-5803/

Nomenclature

Δ	dimensionless supercooling.
Δ^*	critical value of dimensionless supercooling.
ε	intensity of anisotropy.
ε^*	critical value of strength of anisotropy.
Γ	Gibbs–Thomson coefficient [5].
B	scaling exponent.
D_f	fractal dimension.
G	temperature gradient.
Rc	cooling rate.
N	total number of dendrites shown in view field of 7.58×4.75 mm^2 for each experiment.
N_d	number of dendritic doublons.
Pi	$P_i = \frac{N_d}{N} \times 100\%$, percentage of the occurrence of a dendritic doublon for each experiment.
$\overline{P}$	$\overline{P} = \frac{\sum_{1}^{10} P_i}{10}$, average percentage of the dendritic doublon occurrence for each kind of alloy.
C_0	original concentration.
$C_{Cu}(t)$	Cu concentration of a selected pixel in liquid at time t.
$C_{Al}(t)$	Al concentration of a selected pixel in liquid at time t.
$I(t)$	transmitted beam intensity at time t.
$I(t_0)$	transmitted beam intensity at time t_0.
$I(te)$	transmitted beam intensity at time t_e.
I_0	incident beam intensity.
μ_{Cu}	absorption coefficient of Cu.
μ_{Al}	absorption coefficient of Al.
d	thickness of the sample.
σ	dimensionless dendritic tip selection parameter.
σ^*	dendritic tip marginal stability parameter.
D_l	solute diffusion coefficient in liquid.
d_0	capillary length.
R	dendritic tip radius.
R^*	critical value of the dendritic tip radius.
V	tip growth velocity.
k_0	partition coefficient.
T_M	Melting temperature.
T_L	Liquidus temperature.
C_p	Specific heat of the liquid phase.
L	Latent heat of melting.
m_L	liquidus slope.
ΔT	total supercooling.
ΔT_t	thermal supercooling.
ΔT_c	constitutional supercooling.
ΔT_r	curvature supercooling.
ΔT_k	kinetic supercooling.
Δ_t	dimensionless thermal supercooling.
Δ_c	dimensionless constitutional supercooling.
C_L^*	solute concentration of the liquid in front of dendrite tip.
P_c	solute Péclet number at dendrite tip.
$\Delta\varepsilon\%$	percentage of anisotropy increment.

(ii). The black solid line separating the CD and CS structures in Fig. 7a is given by the following condition [3].

$$\Delta \approx 1/2 + \varepsilon^{1/2} \tag{3}$$

(iii). The black dashed line separating the FD and FS structures in Fig. 7a is given by the following condition [3].

$$\Delta \approx |\ln\Gamma|^{\psi_\Gamma} \varepsilon^{\psi_\varepsilon} \tag{4}$$

$\psi_\Gamma = (2-D_f)/[1-\beta(2-D_f)]$,
$\psi_\varepsilon = [1+(5/2-\beta)(2-D_f)]/4[1-\beta(2-D_f)]$,
The scaling exponent $0 \le \beta < 1/2$ and D_f are fractal dimensions ($1.5 \le D_f \le 1.71$ for anisotropic dendritic fractals [6,7]).

(iv). The transition line between CD and FD structures is as follows [3].

$$\Delta \sim \left(\frac{\varepsilon}{\varepsilon^*}\right)^{\Phi} \tag{5}$$

$\Phi = (7/8)(2-D_f)$.
The CD structure has no fractal feature and its mean fractal dimensions is equivalent to Δ,and $0 \le \Delta \le \Delta^*$. Note that, the $0 \le \varepsilon/\varepsilon^* \le 1$; hence, the transition boundary (the blue dot line in Fig. 7a) from CD to FD can be defined as

$$\Delta = \Delta^* \left(\frac{\varepsilon}{\varepsilon^*}\right)^{\Phi} \tag{6}$$

$\Phi = (7/8)(2-\Delta)$.
In fact, the shape of the morphology boundary in 3D is qualitatively similar to the one in 2D [8]. The above control equations of morphology boundary have a certain universality.

Literature shows that some special dendritic structures appear on the morphology transition boundary.

(i) Above the boundary of FS and CS, the symmetry-broken (SB) double finger (or called doublon) is found [9]. The selection of doublons to be strongly dependent on solute concentration and sample orientation [10].
(ii) Above the boundary of CD and CS, the dendritic doublon (or double doublon) is found in CBr_4-8 mol% C_2Cl_6 alloy experiments, which retain the triangular shape of the symmetric dendrites and remain as the stable growth direction [11].
(iii) Numerical studies indicate that the doublons, as independent patterns, exist on the boundary of FD and FS and the structure transition occurs with marked change in the growth velocity [4].
(iv) Neither Wheeler Model [12] nor Karma Model [13] had described the transition morphology between CD and FD dendritic structures.
(v) Under high temperature gradient (about 10 K mm^{-1}) and solidification speed (about 1 mm s^{-1}), a doublon-type morphology i.e., twinned feathery grains, is found in aluminum alloys (Al-Zn, Al-Cu, Al-Mg, Al-Si) [14–18].

However, to the best of our knowledge, reports mentioning transitional forms between the CD and FD are absent. The crystalline structure of the α-phase in the hypoeutectic Al-Cu alloys is face centered cubic (FCC), and the possible growth directions of the primary arm are <100>, <110>, or <111> [15,19]. There are six directions equivalent to [100] by symmetry if it grows along the <100> direction. Similarly, there are twelve equivalent directions for <110> [20]. The regular dendrite has a needle tip and growth direction is <100>. We observed a doublon-type dendrite ("anaxial dendrite") clearly in an Al-15 wt.% Cu alloy in our previous work [21,22] with *in situ* synchrotron X-ray radiography. This was a new type of doublon, different from any reported previously [4,9,10,11,15], i.e., each finger consists of a parabolic dendritic tip, only the primary arm splits at its center with a liquid channel, and the angle between the secondary arms and the primary stem is 90°. According to the methods of defining a dendritic doublon provided in the reference [11], it is appropriate to name the anaxial dendrite as dendritic doublon because of its splitting tip and dendritic profile. To identify a