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RAPID DENDRITIC GROWTH INVESTIGATED WITH ARTIFICIAL NEURAL NETWORK METHOD*

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Rapid dendritic growth of γ-(Ni, Fe) phase, β-CoSb intermetallic compound and α-Fe phase was realized by undercooling Ni-10%Fe single phase alloy, Co-60.5%Sb intermetallic alloy and Fe-40%Sn hypomonotectic alloy to a substantial extent. Their experimentally measured dendrite growth velocities were 79.5m/s, 12m/s and 0.705m/s, corresponding to undercooling levels of 303K($0.18T_L$), 168K($0.11\ T_L$) and 219K($0.15\ T_L$) respectively. Since the usual dendrite growth theory deviates significantly from reality at great undercoolings, an artificial neural network incorporated with stochastic fuzzy control was developed to explore rapid dendrite growth kinetics. It leads to the reasonable prediction that dendritic growth always exhibits a maximum velocity at a certain undercooling, beyond which dendrite growth slows down as undercooling increases still further. In the case of Fe-Sn monotectic alloys, α-Fe dendrite growth velocity was found to depend mainly on undercooling rather than alloy composition.

Keywords: dendritic growth, neural network, undercooling, solidification
PACC: 6125M, 6150C, 8640Z

I. INTRODUCTION

Dendritic growth is the most common structural formation mechanism during crystal growth and alloy solidification. Because of its great significance in both fundamental research and practical applications, the growth kinetics of dendrites has been extensively investigated for almost five decades.[1−6] And it is still a challenging subject in the frontiers of condensed matter physics and materials science.[7−15] The "constitutional supercooling"(CS) theory[1−3] set up the first milestone in analytical study on the evolution of dendritic structures, which has been widely applied to directional solidification process. The most up-to-date breakthrough was brought about by Lipton, Kurz and Trivedi[4] (LKT), who established the theoretical model for rapid dendritic growth in undercooled alloy melts. This LKT model consists of two coupled equations, which determine the interrelationships among dendrite growth velocity (V), tip radius (R) and alloy undercooling(ΔT). In the last decade, a significant progress has also been achieved in experimental measurements of dendrite growth velocity versus undercooling.[5−9] In principle, the experimental results confirm that LKT model is quite successful in describing dendritic growth during rapid solidification. But they also reveal that this model will significantly deviate from reality when undercooling becomes sufficiently large[16]. Therefore, it is highly desirable to obtain a more universal theoretical model, which can depict rapid dendritic growth within any attainable undercooling regime. Because quantitative experiments at extremely great undercoolings are very difficult to perform, the new model is also expected to be capable of predicting the dendritic growth velocity for larger undercooling condition on the basis of experimental data at smaller undercoolings.

Since rapid dendritic growth is a typical complex nonlinear dynamic process characterized by transient heat and mass transfer, it is incredibly difficult to develop a normal model by traditional mathematical physics method. The artificial neural network (ANN) technique may provide an unusual approach to solve this problem. ANN itself is an important research field in automatic control engineering. And it has also found many applications in other scientific areas. For instance, Sun and coworkers[17] successfully predicted the thermophysical properties of high temperature metallurgical melts by ANN method. Li and Xu[18] acquired satisfactory results when they used ANN technique to study the CVD/SiC coating formation process of C/C composites. So far, there seems to be no attempt to investigate rapid dendritic growth kinetics with ANN method.

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The utmost important task of rapid dendritic growth research is to reveal the interrelation between dendrite growth velocity(V) and alloy melt undercooling(ΔT). This paper makes an effort toward such an end in three respects. At first, a systematic experimental study is accomplished to explore the actual variation of V versus (ΔT). Theoretical calculations are then performed in the light of LKT dendrite growth model. Meanwhile, a stochastic fuzzy neural network (SFNN) is developed and applied to predict the V-ΔT relation at large undercoolings. A combined analysis of these three aspects provides a more comprehensive understanding about the rapid growth kinetics of dendrites in substantially undercooled alloys.

Ⅱ. EXPERIMENTAL PROCEDURE

The experiments were performed with a glass flux undercooling facility under 80kPa He-20%H_2 atomosphere.[19,20] In order to get a better comparison, three different kinds of alloys were studied in the present work. Ni-10%Fe alloy is a simple single-phase alloy with an equilibrium freezing temperature range of only 8K, and it merely involves the dendritic growth of γ-(Ni, Fe) solid solution phase. Co-60.5%Sb alloy was also chosen for this research because it displays the dendritic growth of an intermetallic compound β-CoSb phase. Fe-40%Sn hypomonotectic alloy was finally investigated to reveal the dendrite growth characteristics in monotectic systems. An *in situ* alloying procedure was utilized to prepare alloy samples from 99.998% pure Ni, 99.998% pure Fe, 99.998% pure Co, 99.9999% pure Sb and 99.999% pure Sn by RF induction melting. Each sample had a mass of 1g and was immersed within a pool of molten boronsilicate glass in an 8mm ID×10mm OD×15mm alumina crucible. Both alloy melt undercooling and dendrite growth velocity were measured by infrared detecting techniques.[9,16] After experiment the solidification microstructures were analyzed following conventional metallographic procedure.

Ⅲ. ARTIFICIAL NEURAL NETWORK MODEL

The SFNN model is schematically presented in Fig.1. This is a forward neural network with multi-inputs and single output. It is composed of one input layer, one output layer and two hidden layers. Input parameters mainly involve such independent variables as alloy composition and melt undercooling, whereas its output result is dendrite growth velocity. The output versus input relationship is determined by the following reflection function[21,22]

$$f(x)=\frac{\sum_{l=1}^{M}\frac{\overline{y}_{g^l}}{\delta_{g^l}}\exp\left[-\left(\frac{\overline{y}_{g^l}-m_{g^l}}{\delta_{g^l}}\right)^2\right]\prod_{i=1}^{n}\exp\left[-\frac{(x_i-m_{F_i^l})^2}{\sigma_{F_i^l}^2+\sigma_{x_i^l}^2}\right]}{\sum_{l=1}^{M}\frac{1}{\delta_{g^l}}\exp\left[-\left(\frac{\overline{y}_{g^l}-m_{g^l}}{\delta_{g^l}}\right)^2\right]\prod_{i=1}^{n}\exp\left[-\frac{(x_i-m_{F_i^l})^2}{\sigma_{F_i^l}^2+\sigma_{x_i^l}^2}\right]}. \tag{1}$$

The back propagation (BP) learning method is used to train the above SFNN model. The corresponding target function is:

$$E=\frac{1}{2}[f(x)-y_d]^2 \tag{2}$$

In order to minimize the error E, the weights during parameter learning are modified according to the following rule:

$$W(k+1)=W(k)-\alpha\frac{\partial E}{\partial W_k}-\eta\frac{\partial E}{\partial W_{k-1}}. \tag{3}$$

In Eqs. (1)–(3), $\overline{y}_{g^l}$, m_{g^l}, σ_{g^l}, $m_{F_i^l}$, $\sigma_{F_i^l}$ and $\sigma_{x_i^l}$ are adjusting parameters, $\alpha\epsilon[0,1]$ is learning coefficient, and $\eta\epsilon[0,1]$ is momentum factor. In terms of Eqs. (2) and (3), the parameters can be adjusted as follows:

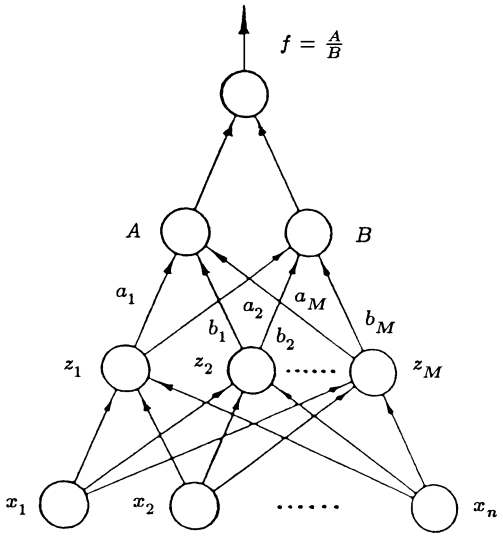


Fig.1. Schematic of artificial neural network.

$$\overline{y}_{g^l}(k+1) = \overline{y}_{g^l}(k) - \alpha \left[\frac{\partial E}{\partial \overline{y}_{g^l}}\right]_k + \eta \Delta \overline{y}_{g^l}(k-1), \tag{4}$$

$$m_{g^l}(k+1) = m_{g^l}(k) - \alpha \left[\frac{\partial E}{\partial m_{g^l}}\right]_k + \eta \Delta m_{g^l}(k-1), \tag{5}$$

$$\delta_{g^l}(k+1) = \delta_{g^l}(k) - \alpha \left[\frac{\partial E}{\partial \delta_{g^l}}\right]_k + \eta \Delta \delta_{g^l}(k-1), \tag{6}$$

$$\delta_{x^l}(k+1) = \delta_{x^l}(k) - \alpha \left[\frac{\partial E}{\partial \delta_{x^l}}\right]_k + \eta \Delta \delta_{x^l}(k-1), \tag{7}$$

$$m_{F^l}(k+1) = m_{F^l}(k) - \alpha \left[\frac{\partial E}{\partial m_{F^l}}\right]_k + \eta \Delta m_{F^l}(k-1), \tag{8}$$

$$\delta_{F^l}(k+1) = \delta_{F^l}(k) - \alpha \left[\frac{\partial E}{\partial \delta_{F^l}}\right]_k + \eta \Delta \delta_{F^l}(k-1). \tag{9}$$

IV.RESULTS AND DISCUSSIONS

As shown in Fig.2, Ni-10%Fe alloy has been undercooled by up to 303K($0.18T_L$). The maximum growth velocity of γ-(Ni, Fe) dendrites detected through experiment attains 79.5m/s. When undercooling is below about 170K, the calculated results of LKT dendritic growth model (dashed line) agree well with experimental data. As undercooling increases still further, LKT theory deviates significantly from experiments. Although the temperature dependence of solute diffusion coefficient has been taken into account, the calculated dendrite growth velocity of LKT model still rise up monotonously. Because dendritic growth is a solidification process controlled by atomic diffusion, the growth velocity of dendrites cannot increase infinitely. The actual possibility is that dendrite growth velocity falls down after reaching a certain maximum, since atomic diffusivity is reduced with the increase of undercooling. It is evident in Fig.2 that the calculated results of SFNN method (solid line) are consistent with the measured dendritic growth velocities in the whole undercooling regime achieved here. Moreover, SFNN method predicts that the growth velocity of γ-(Ni, Fe) dendrites displays a maximum of about 72m/s at an undercooling of 350K. If undercooling level is further enhanced, the dendritic growth velocity calculated by SFNN method will decrease gradually.

The experimental and theoretical results for rapid dendritic growth of β-CoSb intermetallic compound are presented in Fig.3. Co-60.5%Sb alloy has achieved an undercooling level of 168K($0.11T_L$). And the corresponding dendrite growth velocity was measured as 12m/s. LKT dendrite growth model is in agreement with experimental results only when undercooling is smaller than 120K. As undercooling increases, the calculated dendrite growth velocity of LKT theory rises up indefinitely. In contrast, SFNN method produces results which are in much better consistence with experimental data. Furthermore, SFNN calculations predict that β-CoSb intermetallic compound has a maximum growth velocity of 53.3m/s at 250K undercooling. If undercooling goes beyond 250K, the dendritic growth of β-CoSb intermetallic compound will be slowed down.

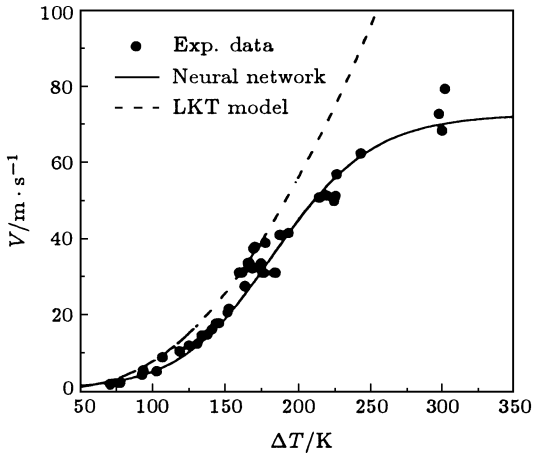


Fig.2. Growth velocity of γ-(Ni,Fe) dendrites versus undercooling.

The maximum undercooling of Fe-40%Sn hypomonotectic alloy attained was 219K($0.15T_L$), which indicates that the metastable undercooled liquid state can be achieved at temperatures far below its monotectic point. As seen in Fig.4, the growth velocity of α-Fe dendrites reaches 0.705m/s corresponding to the actual largest undercooling. Apparently, α-Fe dendrites grow far more slowly than γ-(Ni, Fe) dendrite and β-CoSb intermetallic compound. This is because the growth of α-Fe dendrites within undercooled Fe-

40%Sn hypomonotectic alloy is always controlled by solute diffusion, whereas the dendritic growth of γ-(Ni, Fe) and β-CoSb phases is mainly controlled by thermal diffusion at higher undercoolings.

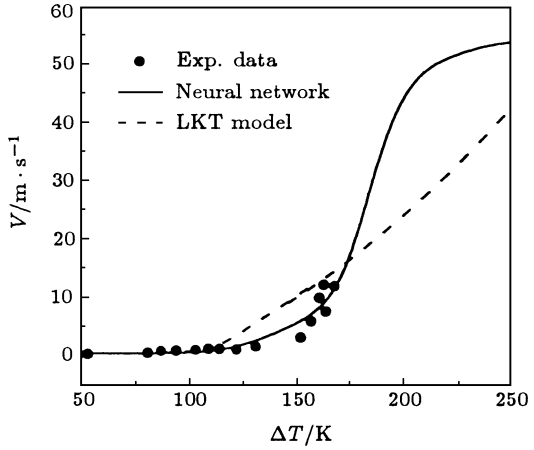


Fig.3. Growth velocity of β-CoSb intermetallic compound versus undercooling.

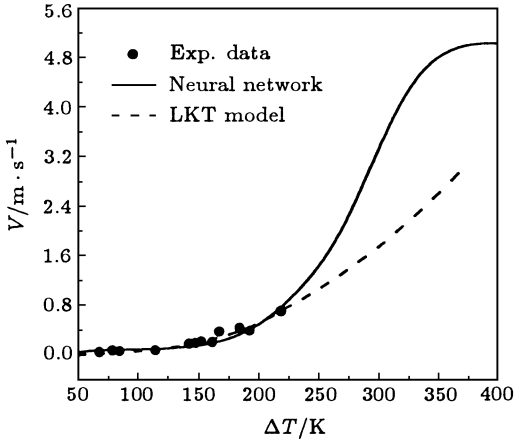


Fig.4. Growth velocity of α-Fe dendrites versus undercooling.

Similar to the previous two cases illustrated in Figs. 2 and 3, the calculated α-Fe dendrite growth velocity of LKT theory increases with undercooling monotonously, although it agrees with experiment in the undercooling range obtained in the present work. Again, SFNN method predicts a maximum growth velocity of 5m/s for α-Fe dendrites at 400K undercooling. Obviously, even this estimated maximum dendrite growth velocity is also far smaller than the corresponding values for γ-(Ni, Fe) dendrite and β-CoSb intermetallic compound.

It should be mentioned that monotectic type alloys are usually more difficult to be undercooled than other kinds of metallic melts. The reason lies in that the nucleation of a new liquid phase (L_2) within a parent melt (L_1) is much easier than the formation of a solid phase (S) nucleus in the same parent liquid phase. The monotectic transformation $L_1 \rightarrow S + L_2$ involves the simultaneous formation of one solid phase and one liquid phase inside a previous liquid phase. Since the thermodynamic barrier preventing L_2 phase nucleation is relatively small, it will nucleate in preference to S phase. But the preferential nucleation of L_2 liquid will soon induce the formation of S nucleus, because both local composition and actual undercooling conditions become favourable for S phase nucleation. Consequently, monotectic type alloys are prone to solidify at smaller undercoolings. Therefore, it is extremely desirable to predict the variation of α-Fe dendrite growth velocity versus the undercooling and composition of Fe-Sn alloys on the basis of actually obtainable experimental results. Figure 5 illustrates the prediction of SFNN method in this respect. Obviously, if undercooling is kept constant, the variation of alloy composition only shows a slight influence on α-Fe dendrite growth velocity at least in the content range of 36%–54%Sn. For a given alloy composition, α-Fe dendrite always exhibits a maximum growth velocity around 400K undercooling.

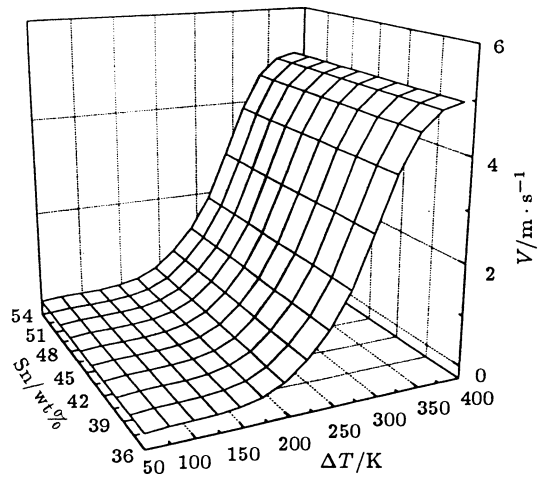


Fig.5. Predicted α-Fe dendrite growth velocity versus undercooling and alloy composition.

V. CONCLUSIONS

In summary, quantitative experimental investigations have been accomplished on the rapid dendritic growth characteristics under high undercooling conditions. Ni-10%Fe single phase alloy, Co-60.5%Sb intermetallic compound alloy and Fe-40%Sn hypomonotectic alloy achieve large undercoolings up to 303K($0.18T_L$), 168K ($0.11T_L$) and 219K ($0.15T_L$) respectively. The actual dendritic growth velocities attained of γ-(Ni, Fe) phase, β-CoSb intermetallic

compound and α-Fe phase are 79.5m/s, 12m/s and 0.705m/s respectively. LKT dendritic growth model agrees with experiment within a moderate undercooling range, but significantly deviates from reality if an alloy melt is substantially undercooled. In contrast, the stochastic fuzzy neural network (SFNN) method results in more reasonable predictions. According to SFNN calculations, the dendritic growth in the three different types of alloys always exhibits a maximum velocity at a certain undercooling, beyond which dendritic growth slows down as undercooling is continually increased. In the case of Fe-Sn monotectic alloys, it is revealed that dendritic growth velocity mainly depends on undercooling rather than alloy composition. Meanwhile, the actual calculations indicate that the incorporation of stochastic fuzzy control into artificial neural network can effectively reduce the bad influences of experimental errors during parameter learning and results in a much higher convergence rate for computations.

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