Journal of Alloys and Compounds 744 (2018) 606–614

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Response and prediction of microstructures indicating deceleration growth condition in a Sn-Ni peritectic alloy

Peng Peng [a, b, *]

[a] Institute of Materials Science and Engineering, Lanzhou University, Lanzhou 730000, China
[b] School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, China

ARTICLE INFO

Article history:
Received 3 December 2016
Received in revised form
10 February 2018
Accepted 12 February 2018
Available online 14 February 2018

Keywords:
A. Metals and alloys
Sn-Ni alloy
Intermetallics
C. Microstructure

ABSTRACT

The Sn-36 at.%Ni peritectic alloy in which both the primary and peritectic phases are intermetallic compounds was directionally solidified attending different deceleration growth conditions. The cellular-dendrite growth of the primary Ni_3Sn_2 phase was studied in this analysis, and the influence of the deceleration rate on the microstructure length scales including primary/higher order dendrite arm spacing (λ_1, λ_2, λ_3) and dendrite tip radius (R) were analyzed. Experimental results showed that although these length scales all decreased with the increasing deceleration rate, the responses of the secondary dendrite arm spacing λ_2 and the tip radius of primary dendrite R are faster than that of λ_1. This difference in response leads to a larger variation in the λ_1/λ_2 ratio with the increasing deceleration rates, as compared with the λ_2/R ratio. Furthermore, the ratio of the thickness of primary dendrite stem d to the primary dendrite arm spacing λ_1, d/λ_1 was used to predict the melt concentration at the solid/liquid interface C_l. These length scales were related to the melt concentration C_l which is obtained through the d/λ_1 ratio.

1. Introduction

Dendritic feature is a frequently observed morphology of the growing interface in most solidification conditions. This kind of microstructure is usually characterized by specific length scales such as the primary/higher order dendrite arm spacing (λ_1, λ_2, λ_3) and the dendrite tip radius (R). Analyses of steady-state directional solidification have developed a number of models to relate them to solidification processing parameters such as the growth velocity v and temperature gradient G [1,2]. In addition, a lot of work has been done in the last two decades using phase field modeling to describe directional solidification [3–5]. Using a phase-field variable and a corresponding governing equation to describe the state (solid or liquid) in a material as a function of position and time, the diffusion equations for heat and solute can be solved without tracking the solid-liquid interface [3]. However, the models mentioned above do not precisely represent the realistic conditions prevalent during the industrial casting, which normally occurs under rapidly changing growth conditions. The existence of an allowable range of stable primary dendrite arm spacing (λ_1) has been demonstrated in numerous studies [6,7] during the past decades. These experimental and numerical results show that a range of spacings can be found for a given processing condition, which is distinct from the unique prediction on primary dendrite arm spacing by these geometric models [1,2]. Besides, it has been shown that when solidifying complex shapes like turbine blades [8] or aerospace engine blades [9], the cross sectional area changes resulted in the fluctuations of both directional growth velocity (v) and temperature gradient, leading to undesirable structures that adversely affect the performance of the directionally solidified component. Furthermore, it is also demonstrated that the growth of dendrites, thus the variation in these length scales is history-dependent. This means that the scheduled growth conditions can not be achieved instantaneously [7]. Therefore, it is of critical importance to investigate the responses of these length scales to the variations in solidification conditions.

A number of studies has been carried out on the response, or modification of these length scales reflecting the variations in the solidification conditions. Although response of the length scales to the change in growth conditions can be monitored well, the difference in their response speed is not negligible, reflecting the fact that the growth parameters have different influences at different

* School of Physical Science and Technology, Lanzhou University, Lanzhou 730000, PR China.
E-mail address: pengp@lzu.edu.cn.

https://doi.org/10.1016/j.jallcom.2018.02.160
0925-8388/

length scales. Liu et al. [10] carried out a series of experiments with a continuous decrease in growth velocity to assess the influence of velocity changes on these structural length scales. According to their observations, both the tip radius and the initial secondary dendrite arm spacing respond more rapidly on the changing growth velocity, while the primary dendrite arm spacing responds rather slowly. Therefore, there is a considerable hysteresis between the transient adjustments. It is now well established that the steady state primary spacings of the cellular and dendrite structures at any given velocity are dependent on their solidification history [6]. In contrast, the cell or dendrite tip radii are found to be independent of the path by which the final velocity was reached. The growth of dendrites is usually composed of the growth of primary dendrite stems and the secondary branches grow out from them, the former part is also influenced by the growth of the dendrite tips. Thus, the differences in the response of these length scales to growth parameters lead to the morphology change of dendritic structures during solidification.

The dendrite morphology has been characterized in many peritectic systems [11,12]. Since the liquidus slope is the dominating factor determining whether faceted solid/liquid morphology exists [13], the non-faceted dendrite growth can be found even in peritectic systems where both the primary and the peritectic phases are intermetallic compounds with nil or narrow solubility ranges, such as Sn-Ni [12] and Sn-Mn [14] alloys. The restriction of the coarsening process of the secondary dendrite arm of primary phase enclosed by peritectic phase [15] has also been confirmed in Sn-Ni peritectic alloys [12]. In addition, for the peritectic systems containing intermetallic compounds with a narrow solubility range, the modified solute distribution coefficient k^* has been demonstrated to be in good agreement with experimental results [16]. Since both the primary Ni_3Sn_2 phase and the peritectic Ni_3Sn_4 phase are most commonly formed in solder joint reactions in Sn based solder materials [17], investigation of Ni_3Sn_2 and Ni_3Sn_4 phases is helpful to control solder joint quantity in Sn based solder materials [18]. For solder alloys, the dendrite structures with different arm spacing are essentially important to enhance the mechanical properties of a solder alloy. Extensive theoretical and experimental researches have been conducted on the relationships between microstructure, physical properties, and solidification processing parameters of solder materials [17–20]. In recent years, increasing environmental and health concerns combined with legislation forbidding the use of lead-based solders have motivated the development of lead-free solder alloys. These analyses have clearly demonstrated the close connection between the dendrite structures with different arm spacings and the mechanical properties of the solder alloy.

In our previous work, the length scales were investigated in Sn-Ni peritectic alloys under steady-state growth conditions [21], while the response of a series of length scales and relationships has never been discussed under non steady-state growth conditions in this peritectic system. Neither did the solidification characteristics of intermetallic compounds such as Ni_3Sn_2 and Ni_3Sn_4 phases have been concerned, and the available information is insufficient for describing the microstructural length scales in peritectic systems. Compared with the temperature gradient, the growth velocity can be more easily changed, thus the growth velocity (v) is the most frequently used alternating parameter in solidification under non-steady state growth conditions. The present study is motivated by the scientific conjectures mentioned above, and aims to investigate the response of these length scales, thus their relationships to the variation in growth velocity. The responses of the length scales to different deceleration growth conditions are compared and studied. Furthermore, the reasons why the length scales show different responses to the variation in the growth velocity have also been explained. In addition to that, the melt concentration C_l which is different at different local positions is always assumed to be equal to the initial concentration of alloy, C_0. However, the actual melt concentration C_l is not equal to C_0 for the solute redistribution of alloys. Due to the influence of the quenching process on the solidified structures, C_l obtained through a post-mortem examination may not be correct either. Thus, to precisely predict the melt concentration C_l, the ratio of the thickness of the primary dendrite stem d to the primary dendrite arm spacing λ_1, d/λ_1 is introduced as a parameter describing growth of the dendrites. In this study, the melt concentration C_l is obtained using d/λ_1, and the reliability of the proposed ratio is also evaluated.

2. Experimental procedures

2.1. Sample preparation process

The Sn-36 at.%Ni alloy was induction melted from pure Ni and Sn (99.9%), and the as-cast rods with the diameter and length of 3 mm and 110 mm respectively were machined from the ingot by a spark machining. The actual compositions of the as-cast alloy and phases are analyzed using an electron micro probe analyzer (EPMA) EPMA-1610. Experiments can be divided into two steps: melting of the samples and subsequent directional solidification. The experiments are carried out in a Bridgman-type furnace consisting of a resistance furnace as a previously described one [12,21]. The samples were placed into the alumina crucibles with a purity of 99.99 pct. The inside/outside diameter of the alumina crucible is 4/5.5 mm diameter and the length of it is 150 mm. A PtRh30-PtRh6 thermocouple which is inserted down the center of the samples is used to measure the temperature gradient close to the solid/liquid interface, which was approximately 40K/mm. For each experiment, the furnace is heated to 1250 °C to melt the alloy, and then is held for 30 min to homogenize the melt. Subsequently, the samples are solidified at a constant growth velocity for a growth distance of 10 mm and then subjected to controlled deceleration. Deceleration growth continues until the final growth velocity is reached. As illustrated in Table 1, solidification of Sn-36 at.%Ni peritectic alloys is carried out at the same initial (5 μm/s) and final growth velocities (1 μm/s) in the Bridgman-type apparatus. When the predetermined solidification time is achieved, the samples are quenched into a liquid Ga-In-Sn alloy quickly to obtain a good solid/liquid interface. For a more clear comparison, the analysis on the sample directionally solidified at a constant growth velocity of 1 μm/s (deceleration rate a is 0) is also presented. Since the growth distance in the present work is more than three times longer compared to the distance required for the initiation of steady state growth [22], the influence of the solidification fraction (pulling distance) on λ_1, λ_2 and R can be neglected.

2.2. Measurements of length scales

Scanning electron microscopy (SEM (Quanta-200F)) is used to study the solidification microstructure of both the longitudinal and transverse sections of the solidified samples. The primary dendrite arm spacing λ_1 is measured using the area method: $\lambda_1 = \sqrt{A/N}$ [23], where N is the number of the primary dendrites in the area A of the transverse section. Secondary dendrite arm spacing (λ_2) is measured in the vicinity of the primary dendrite tip by averaging the distance between the adjacent side branches on the longitudinal section of a primary arm. Therefore, the influence of the coarsening process on secondary dendrite arm spacing can be minimized. Both the thickness of the primary dendrite stems d and secondary dendrite arm spacing (λ_2) are measured in longitudinal