Erciyes Üniversitesi Fen Bilimleri Enstitüsü Dergisi 26(1): 71-76 (2010)

Erciyes University Journal of the Institute of Science and Technology
Erciyes Üniversitesi Fen Bilimleri Enstitüsü Dergisi
http://fbe.erciyes.edu.tr
ISSN 1012-2354
Cilt (Volume) 26, Sayı (Issue) 1, Mart/March-2010

Comparison of experimental results with theoretical eutectic model in In-Bi-Sn ternary alloy

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Anahtar Kelimeler
Ternary alloys
Directional solidification
Eutectics
Microstructure
Theoretical models

ABSTRACT
In present work, values of eutectic spacing (λ) for In-Bi-Sn eutectic alloy were calculated with Wilde-Froyen-Witusiewicz-Hecht's (WFWH) theoretical model by using the physical parameters of In-Bi-Sn eutectic alloy. The theoretical values of eutectic spacing have been compared with the experimental results obtained from In-Bi-Sn and In-Bi-Cd ternary alloys. It was found that, the values of theoretical eutectic spacings are very close the values of the experimental eutectic spacing for In-Bi-Sn eutectic alloy. Moreover, it was also found that, the theoretical eutectic spacing values at high-speed ranges are highly compatible with the experimental results obtained by different researchers at low-speed ranges for In-Bi-Sn eutectic alloy, in the other hand, the experimental values of λ for In-Bi-Cd eutectic alloy are fairly higher than experimental and theoretical values of λ for In-Bi-Sn eutectic alloy.

In-Bi-Sn üçlü alaşımında deneysel sonuçların teorik ötektik model ile karşılaştırılması

Keywords
Üçlü alaşımlar
Doğrusal katılaştırma
Ötektikler
Mikroyapı
Teorik modeller

ÖZET
Bu çalışmada, In-Bi-Sn ötektik alaşımı için ötektik mesafe değerleri, (λ) bu alaşımın fiziksel parametreleri kullanılarak Wilde-Froyen-Witusiewicz-Hecht (WFWH) teorik modeli ile hesaplandı. Ötektik mesafelerin teorik değerleri, In-Bi-Sn ve In-Bi-Cd üçlü alaşımları için elde edilen deneysel sonuçlarla karşılaştırıldı. Hesaplanan ötektik mesafe değerlerinin deneysel değerlere oldukça yakın olduğu tespit edildi. Ayrıca, In-Bi-Sn alaşım sistemi için yüksek hızlarda teorik olarak hesaplanan ötektik mesafe değerlerinin farklı araştırmacılar tarafından düşük hızlarda deneysel olarak ölçülen değerlerle oldukça uyumlu olduğu, diğer taraftan, In-Bi-Cd ötektik sistemi için elde edilen deneysel λ değerlerinin In-Bi-Sn öteklik sistemi için teorik ve deneysel olarak elde edilen λ değerlerinden daha büyük olduğu tespit edildi

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1. INTRODUCTION

Multiphase solidification of multicomponent materials attracts pronounced academic interest as well. The study of the solidification behaviour of multicomponent and multiphase systems is an important question in understanding the different properties of these materials. Whereas fundamental knowledge on solidification has been developed mainly for pure materials and for binary alloys exhibiting single-phase growth (solid solution) and/or two-phase growth in eutectic and peritectic class reactions, the process of microstructure formation during solidification is less understood for cases where multiphase reactions occur along the solidification path of these alloys. A comprehensive overview with respect to the last topic is given by Hecht et al. [1].

The industrial and fundamental aspects of multiphase microstructure evolution in multicomponent alloys can only be addressed with the hope for success, if a close interaction is maintained among scientists investigating the thermodynamic, thermophysical, and microstructural properties of these alloys. The In–Bi–Sn ternary system is technologically important for soldering applications. Alloys of eutectic composition may also serve as model alloys for the investigation of pattern formation during directional solidification of eutectics [2–6]. Prior studies indicate that several invariant reactions exist in this alloy system at temperatures below 100 Celsius degree [7-9].

Purpose of this paper is to determine the eutectic spacing from theoretical model by using the physical parameters of In-Bi-Sn eutectic alloys and compare the theoretical results with the previous similar experimental results.

2. THEORETICAL BACKGROUND

Directional solidification of binary or pseudo-binary eutectics may result in regular structures of fibrous, rod or lamellar type. In fibrous and rod growth, one of the phases grows in the form of fibres embedded into a continuous matrix of the other phase, while in the case of lamellar growth; two phases grow cooperatively side by side, in the form of lamellae.

At the present, there are available two theoretical models which were developed by Jackson-Hunt, JH (at low velocity) [10] and Trivedi-Magnin-Kurz, TMK (at high velocity) [11] to determine the microstructure parameters for binary eutectic alloys.

The Jackson and Hunt's (JH) eutectic theory [10] gives the following relationship between the average undercooling ΔT, the growth rate V and the lamellar spacing (λ) for an isothermal solidification front as;

$$\Delta T = K_1 V \lambda + \frac{K_2}{\lambda} \qquad (1)$$

where K_1 and K_2 are the physical parameters of the alloy. In addition, applying the condition of growth at minimum undercooling, $(\partial \Delta T / \partial \lambda)_V = 0$, to equation (1) gives

$$\lambda_e^2 V = K_2 / K_1 = \text{constant}_1 \qquad (2)$$

$$\frac{\Delta T^2}{V} = 4K_1 K_2 = \text{constant}_2 \qquad (3)$$

$$\lambda_e \Delta T = 2K_2 = \text{constant}_3 \qquad (4)$$

where λ_e is the extremum eutectic spacing and ΔT is the undercooling. The experimentally confirmed inter-relationship between the eutectic spacing, growth rate and the undercooling in eutectic system implies that a mechanism is available for changing the rod spacing when the growth rate and/or ΔT varied.

The Trivedi–Magnin–Kurz (TMK) model [11] for eutectic growth at high velocity are based on the equations;

$$\lambda^2 \upsilon = \frac{\alpha^L}{Q^L} \qquad (5)$$

$$\lambda \Delta T = m\alpha^L \left[1 + \frac{p}{p + \lambda(\partial p / \partial \lambda)} \right] \qquad (6)$$

λ is the lamellar spacing, and the definitions of the other terms are as follows, ΔT is the non-equilibrium eutectic undercooling;

$$\alpha^L = 2\left\{\frac{\alpha^L_\alpha}{fm_\alpha} + \frac{\alpha^L_\alpha}{f(1-f)m_\beta}\right\} \qquad (7)$$

$$Q^L = \frac{1-k}{f(1-f)D}\left(p + \lambda\frac{\partial p}{\partial\lambda}\right) \qquad (8)$$

$$p = \sum_{n=1}^{\infty}\left(\frac{1}{n\pi}\right)^3 \left[\sin(n\pi f)\right]^2 \frac{p_n}{\sqrt{1+p_n} - 1 + 2k} \qquad (9)$$

$$p + \lambda\left(\frac{\partial p}{\partial\lambda}\right) = \sum_{n=1}^{\infty}\left(\frac{1}{n\pi}\right)^3 \left[\sin(n\pi f)\right]^2 \frac{p_n}{\sqrt{1+p_n} - 1 + 2k}\,\frac{p_n}{(1+p_n)^{1/2}}$$

(10)

where P_n = 2nπ/P, P=νλ/2D, f is the volume fraction of the α-phase in the α+β eutectic, α^L_α and α^L_β are the capillarity constants, P is the solutal Peclet number. D is the liquid inter-diffusion coefficient, m is the liquidus slope, k is the solute distribution coefficient, ν is the interface velocity.

More recently, a theoretical model was developed to determine the microstructure parameters for two eutectic phase's growth from ternary eutectic liquid by Wilde et al. [12]. This analytical model describes the steady-state regular planar two-phase coupled growth with a lamellar morphology in the case of a univariant eutectic reaction in ternary alloys. The Wilde-Froyen-Witusiewicz-Hecht's (WFWH) model [12] gives the following relationship between the growth rate (V) and the eutectic spacing (λ) for an isothermal solidification front as;

$$\lambda^2 V = \frac{1}{\sum_{i=1}^{\infty}\frac{\sin^2(i\pi f_\alpha)}{i^3\pi^3}}\left[\frac{\Gamma_\alpha \sin\theta_\alpha}{\frac{|m_B^\alpha|}{D_B}\left|C_B^\alpha + C_B^\beta\right| + \frac{|m_C^\alpha|}{D_C}\left|C_C^\alpha + C_C^\beta\right|} + \frac{\Gamma_\beta \sin\theta_\beta}{|m_A^\beta|\left|\frac{C_B^\alpha + C_B^\beta}{D_B} + \frac{C_C^\alpha + C_C^\beta}{D_C}\right| + \frac{|m_C^\beta|}{D_C}\left|C_C^\alpha + C_C^\beta\right|}\right] \qquad (11)$$

Where Γ is the Gibbs–Thomson coefficient, m is the liquidus line slope, C is the alloy concentration, D is the diffusion coefficient in the liquid, A, B and C subscripts are different components, α and β are any phase growth from ternary alloy. Equation (11) dependents only the constant values of the selected alloy [12]. In other hand, anyone theoretical model could not be developed for calculated the values of eutectic spacing to growth three separate phase from ternary eutectic liquid, yet.

3. RESULT and DISCUSSION

The eutectic spacing for directionally solidified In-Bi-Sn eutectic alloy were measured by different researchers [2, 6 and 13]. Typical optical images for In-Bi-Sn ternary eutectic alloy are shown in Figure 1. As can be seen from Figure 1, the microstructure consists of regular lamellar γ-Sn in In_2Bi intermetallic solution matrix.

In present work, firstly, the eutectic spacing in the In-Bi-Sn eutectic system was calculated with WFWH theoretical model by using the physical parameters. Later, calculated λ values in this work have been compared with the similar experimental results obtained for ternary alloys. Some physical parameters used in the prediction of eutectic spacing for In-Bi-Sn eutectic system are given in Table 1.

Eutectic spacing were determined for In–33.1%Bi–15.6%Sn (wt.) and In–32.2%Bi–15.6%Sn (wt.) in high temperature gradient and low growth rates (V=0014–1 μm/s and G=8 K/mm) by Witusiewicz et al. [2]. Samples were prepared into a mold and the solidification experiments were carried out into two-dimensions (2D) by Witusiewicz et al. [2] and into the three-dimensions (3D) by Ruggerio and Rutter [6]. Directional solidification experiments for In-Bi-Cd eutectic alloys for different eutectic compositions, 77.5 °C and 61.5 °C melting temperatures were carried out by Snugovsky et al. [17] and they have observed regular eutectic spacing between the BiIn-$BiIn_2$ and $BiIn_2$-ε phases. Comparisons of the calculated eutectic spacing with experimental results for In-Bi-Sn and In-Bi-Cd eutectic alloys are shown in Figure 2. The λ values, shown as a black line in Figure 2 are theoretical calculated eutectic spacing values from WFWH model. Other symbolic representations are the experimental λ values and grey lines are the average slope of this experimental λ values. As can be seen from Figure 2, values of λ calculated by WFWH model for In-Bi-Sn alloys are very close the values of λ obtained by Çadırlı et al. [13]. The experimental results obtained by Çadırlı et al. [13] and theoretical results obtained in this work for In-Bi-Sn eutectic alloy at high-speed ranges (V≅ 3-160 µm/s) are highly compatible with the experimental results obtained by Witusiewicz et al. [2] and Ruggerio and Rutter [6] at low-speed ranges (V≅001-1 µm/s) for In-Bi-Sn eutectic alloy, respectively.

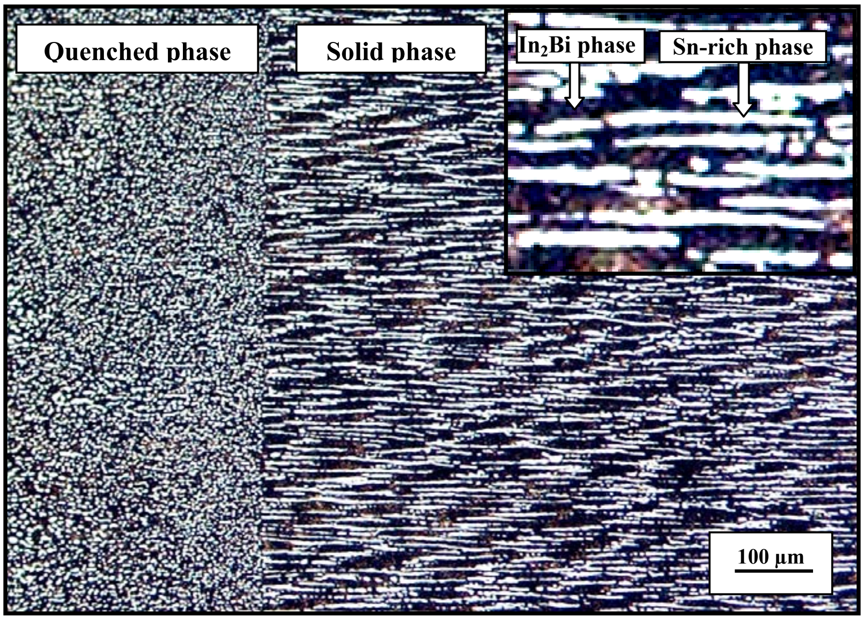

Figure 1. Typical optic images of the growth morphologies of directionally solidified In-32.2%Bi-15.6%Sn (at.) ternary eutectic alloy (V=3.2 µm/s, G=0.91 K/mm).

Table 1. Some physical parameters of In-Bi-Sn eutectic alloy

In-33.1%Bi- 15.6%Sn (wt.) $(i = A, B, C)$, $\alpha = Sn(\alpha)$, $\beta = In_2Bi$				
Symbol	**Unit**	**A: In**	**B: Bi**	**C: Sn**
C_i^o	(% mol)	60.7	21.5	17.78
C_i^α	(% mol)	55.6	9.32	35.62
C_i^β	(% mol)	66.63	29.87	3.50
m_i^α	(K/ mol %)	.....	2.363 [14]	- 2.218 [14]
m_i^β	(K/ mol %)	- 0.345 [14]	…..	2.550 [14]
D_i	(m^2/s)	…..	10.96×10^{-12} *	4.73×10^{-12} *
		$\alpha = Sn(\alpha)$	$\beta = In_2Bi$	
f_i	(…..)	0.28	0.72	
Γ_i	(K. m)	7.68×10^{-8} [15]	1.42×10^{-7} [16]	
θ_i	(o)	14.26 [15]	14.02 [16]	

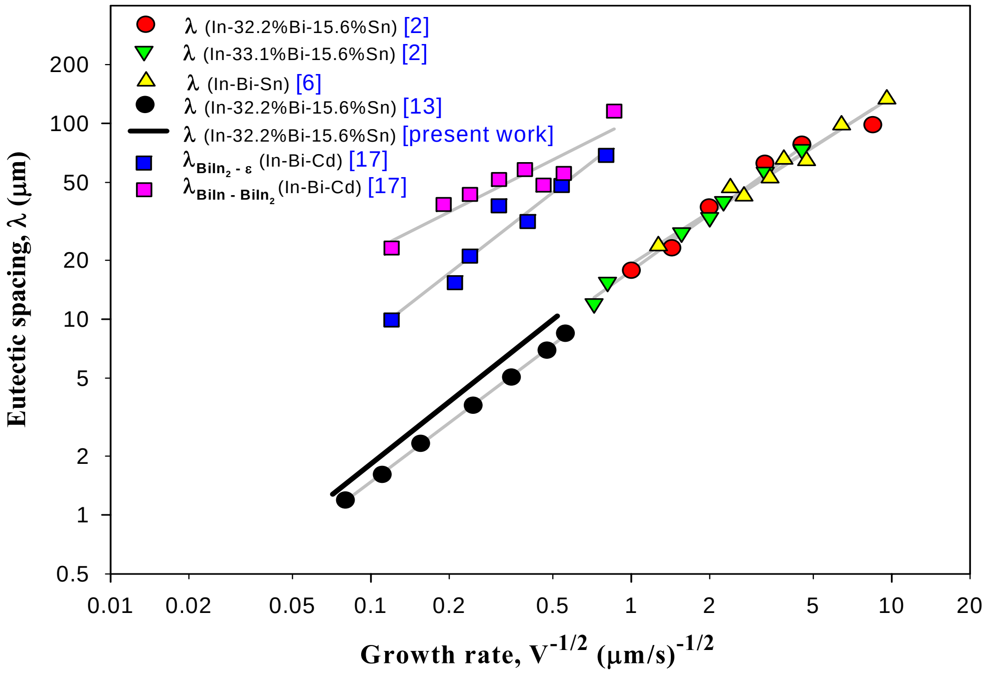

Figure 2. Comparison of calculated eutectic spacing with previous.

The maximum values of growth rate used by Witusiewicz et al. [2] and Ruggerio and Rutter [6] are about three times smaller than the value of growth rate used by Çadırlı et al. [13]. Values of λ for In-Bi-Cd eutectic system obtained by Snugovsky et al. [17] is very higher than the values of λ for In-Bi-Sn eutectic system obtained by Çadırlı et al. [13] and calculated by WFWH theoretical model.

4. CONCLUSIONS

The eutectic spacing in the In-Bi-Sn eutectic system was calculated with WFWH theoretical model for In-Bi-Sn eutectic system. The values of theoretical eutectic spacing for In-Bi-Sn eutectic alloy have been compared with the similar experimental results obtained for ternary alloys. The values of λ calculated by WFWH theoretical model for In-Bi-Sn alloys are very close the experimental values of λ obtained by Çadırlı et al. [13]. The experimental result obtained by Çadırlı et al. [13] and theoretical results obtained in present work for In-Bi-Sn eutectic alloy at high-speed ranges (V≅ 3-160 μm/s) are highly compatible with the experimental results obtained by Witusiewicz et al. [2] and Ruggerio and Rutter [6] at low-speed ranges (V≅001-1 μm/s) for In-Bi-Sn eutectic alloy, but experimental values of λ obtained for In-Bi-Cd eutectic system by Snugovsky et al. [17] is higher than the experimental and theoretical values obtained in previous works for In-Bi-Sn eutectic system.

ACKNOWLEDGEMENT

This research was supported by Erciyes University Scientific Research Project Unit under Contract No: FBT 07–65. The authors are grateful to Erciyes University Scientific Research Project Unit for their financial supports.

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