

**Article Arrival Date**

26.09.2021

**Article Type**

Research Article

**Article Published Date**

20.12.2021

**Doi Number:** <http://dx.doi.org/10.38063/ejons.490>**EFFECT OF CU CONTENT AND GROWTH VELOCITY ON THE MICROSTRUCTURE PROPERTIES OF THE DIRECTIONALLY SOLIDIFIED AL-MN-CU TERNARY ALLOYS****Uğur BÜYÜK**Erciyes University, Faculty of Education, Department of Science Education, Kayseri-TURKEY  
ORCID: 0000-0002-6830-8349**Emin ÇADIRLI**Niğde Ömer Halisdemir University, Faculty of Arts and Sciences, Department of Physics, Niğde -TURKEY  
ORCID: ID 0000-0002-8085-9733**Hasan KAYA**Erciyes University, Faculty of Education, Department of Science Education, Kayseri-TURKEY  
ORCID: ID 0000-0003-3529-9762**M. İzzettin YILMAZER**Erciyes University, Faculty of Education, Department of Science Education, Kayseri-TURKEY  
ORCID: ID 0000-0001-8790-902X**ABSTRACT**

In this work, influences of composition (Cu content) and growth velocity (V) on the microstructure (dendritic spacing) of Al–Mn–Cu ternary alloys have been investigated. Al–1.9Mn–xCu (x=0.5, 1.5 and 5 wt. %) alloys were prepared using metals of 99.90% high purity in the vacuum atmosphere. These alloys were directionally solidified upwards under various growth velocities (8.3–978  $\mu\text{m/s}$ ) using a Bridgman-type directional solidification furnace at a constant temperature gradient (7.1 K/mm). Measurements of primary dendrite arm spacing ( $\lambda_1$ ) of the samples were carried out and then expressed as functions of growth velocity and Cu content. Especially, cell-dendritic transition was detected for low growth velocity (41.6  $\mu\text{m/s}$ ) for alloys containing 0.5 and 1.5Cu. It has been found that the values of  $\lambda$  decrease with increasing V and decreasing Cu content.

**Keywords:** Aluminum alloys, Solidification, Cell-dendritic transition, Dendrite arm spacing**1. INTRODUCTION**

Studies on the multiphase solidification of ternary and multicomponent alloys have reached significant levels in the last 20 years. The study of the solidification behavior of multicomponent and multiphase systems is an important point in understanding the different properties of these materials. Physical properties of metallic materials such as mechanical, electrical, thermal, etc. are affected by their morphology. Microstructure evolution during solidification depends on composition, melting conditions, growth velocity and casting process.

When an alloy is solidified, the most common solid morphology is eutectic or dendritic microstructures (Trivedi and Kurz, 1994), which are characterized by microstructure parameters (eutectic or dendritic spacing). Especially in recent years, there has been numerous works on characterizing the microstructure parameters of ternary aluminum alloys depending on the growth velocity (Böyük et al., 2009, Böyük et al., 2011, Böyük, 2012, Böyük et al. 2015).

Manganese is the principal alloying element in the 3xxx aluminium alloys series. A limited percentage of up to 2 wt. % Mn added to Al makes the alloy higher in corrosion resistance and much stronger than the commercial pure aluminium. The improvements in the mechanical properties adapt the alloy for the wide use in moderate strength applications requiring good workability (Davis, 2001, Davis 1998) in various applications.

Aluminum–Manganese–Copper alloy is purported to have an excellent combination of mechanical properties and possesses erosion and corrosion resistance to high velocity seawater. Its foundry and welding characteristics are better than conventional aluminum bronzes (Macken and Smith 1966). Therefore, it is commonly used to casting large section parts, especially propellers for huge ships (Langham, 1962). However, there are very limited studies in the literature on the microstructure parameters of the Al-Mn-Cu alloy depending on the growth velocity and Cu content.

The aim of this work was experimentally investigate the effect of Cu content (Co) and growth velocity (V) on the microstructure (primary dendritic spacing,  $\lambda$ ) of the directionally solidified Al-1.9Mn-xCu (x=0.5, 1.5, 5 wt.%) ternary alloys.

## 2. MATERIALS AND METHODS

### 2.1 Sample Preparation

Al-1.9Mn-xCu (x=0.5, 1.5, 5 wt.%) alloys have been prepared under vacuum atmosphere by using 99.90 % purity metals. After allowing time for melt homogenization, the molten alloy was poured into graphite molds in the hot filling furnace. The molten alloy was directionally solidified from bottom to top to ensure that the crucible was completely full. Then, each sample was positioned in a Bridgman type furnace in a graphite cylinder (Fig. 1). In this technique, the samples were heated to about 100 K above the melting temperature of the studied alloy. After stabilizing the thermal conditions in the furnace, the samples were grown by pulling it downwards at different growth conditions under an argon atmosphere. The samples were solidified under steady-state conditions at a constant temperature gradient and wide range of growth velocities (8.3–978  $\mu\text{m/s}$ ) in the Bridgman-type growth apparatus described elsewhere (Böyük et al., 2009). After 10–12 cm steady-state growth, the samples were quenched by rapidly pulling them down into the water reservoir. The temperature of water in the reservoir was kept at 283 K by using a heating/refrigerating circulating bath. The sample temperature was controlled with an accuracy of  $\pm 0.1$  K using a temperature controller. In order to see the effect of growth velocity and Cu content on microstructure properties, directional solidification experiments were made at four different growth velocities (8.3, 41.6, 166.3 and 978  $\mu\text{m/s}$ ) for each Al-1.9Mn-xCu (x=0.5, 1.5, 5 wt.%) alloy.

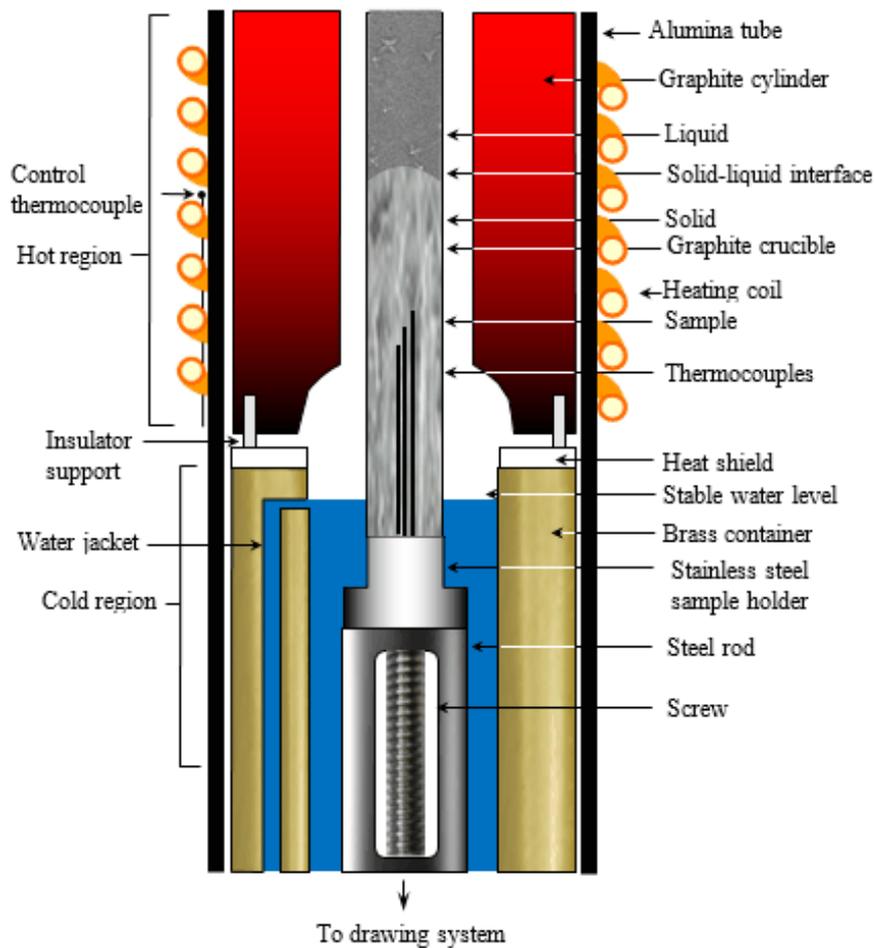
### 2.2 Measurement of Temperature Gradient and Growth Velocity

The temperatures in the samples were measured by three K-type thermocouples. All the thermocouples were connected to the measurement unit consisting of data-logger and computer. The cooling curves were recorded with a data-logger via computer during the solidification process (Fig. 1). When the second thermocouple was at the solid/liquid interface, the temperature difference ( $\Delta T$ ) between the first and second thermocouples was read from data-logger record. The temperature gradient in liquid at the solid-liquid interface ( $G = \Delta T / \Delta X$ ) for each sample was calculated from three cooling curves using the measured value of  $\Delta T$  and the value of  $\Delta X$ .

### 2.3. Microstructure Characterization

The quenched samples were removed from the graphite crucible and cut into lengths of typically 8 mm. The longitudinal and transverse sections of the ground samples were then cold mounted with epoxy-resin. The longitudinal and transverse sections were ground flat with (180, 500, 1000, 2500 and 4000) grit SiC paper, and then polished with (6, 3, 1, 0.25, and 0.05)  $\mu\text{m}$  diamond paste. After polishing, the samples were etched with suitable etching solution (95 ml distilled water + 5 ml HF from 5 to 8 s). The microstructures of the samples were revealed and photographed by the SEM (LEO

model). Primary dendrite arm spacing was measured with the Adobe Photoshop CS3 program. Chemical composition analysis of the samples were carried out with the same SEM equipped with an energy dispersive X-ray (EDX) spectrometer at 20 keV using the X-ray lines as well as a computer controlled image system.



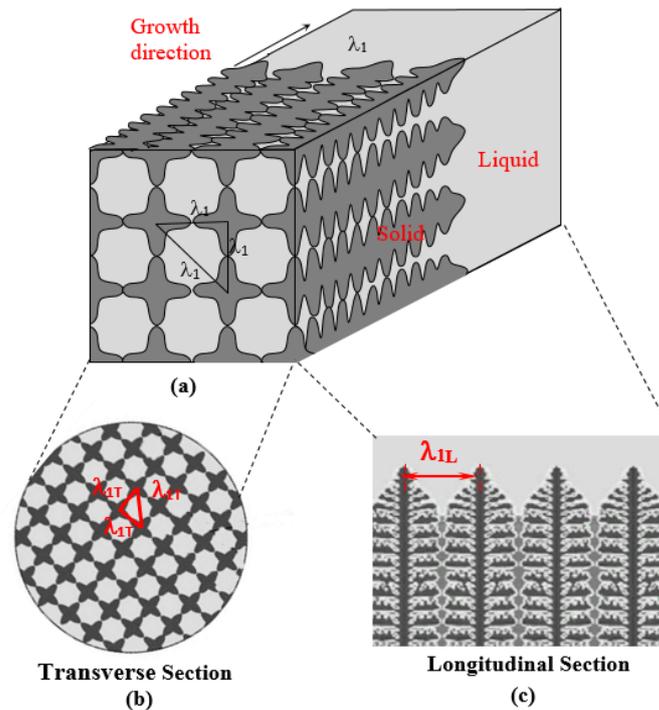
**Figure 1.** The Bridgman–type directional solidification furnace

#### 2.4. Measurements of Dendritic Spacing

Two different methods were used to measure primary dendrite arm spacing,  $\lambda_I$ . In the first method, the primary dendrite arm spacing,  $\lambda_{IL}$  was obtained by measuring the distance between the nearest two dendrites tips (Fig. 2a) on longitudinal section (Ganesan et al., 1992). The second method is the area counting method (Bhat et al, 1995)]. The values of  $\lambda_{IT}$  were measured on the transverse section of the sample (see Fig. 2b). In this method the average primary dendrite arm spacing,  $\lambda_{IT}$  was determined from;

$$\lambda_{IT} = \frac{I}{M} \left( \frac{A}{N} \right)^{0.5} \quad (1)$$

where M is the magnification factor, A is the total specimen cross sectional area and N is the number of primary dendrites on the cross section.  $\lambda_I$  is arithmetic average values of  $\lambda_{IL}$  and  $\lambda_{IT}$ .



**Figure 2** a) Schematic illustration of the dendritic spacing measurements longitudinal and transverse sections (b) transverse section (c) longitudinal section.

### 3. RESULTS AND DISCUSSION

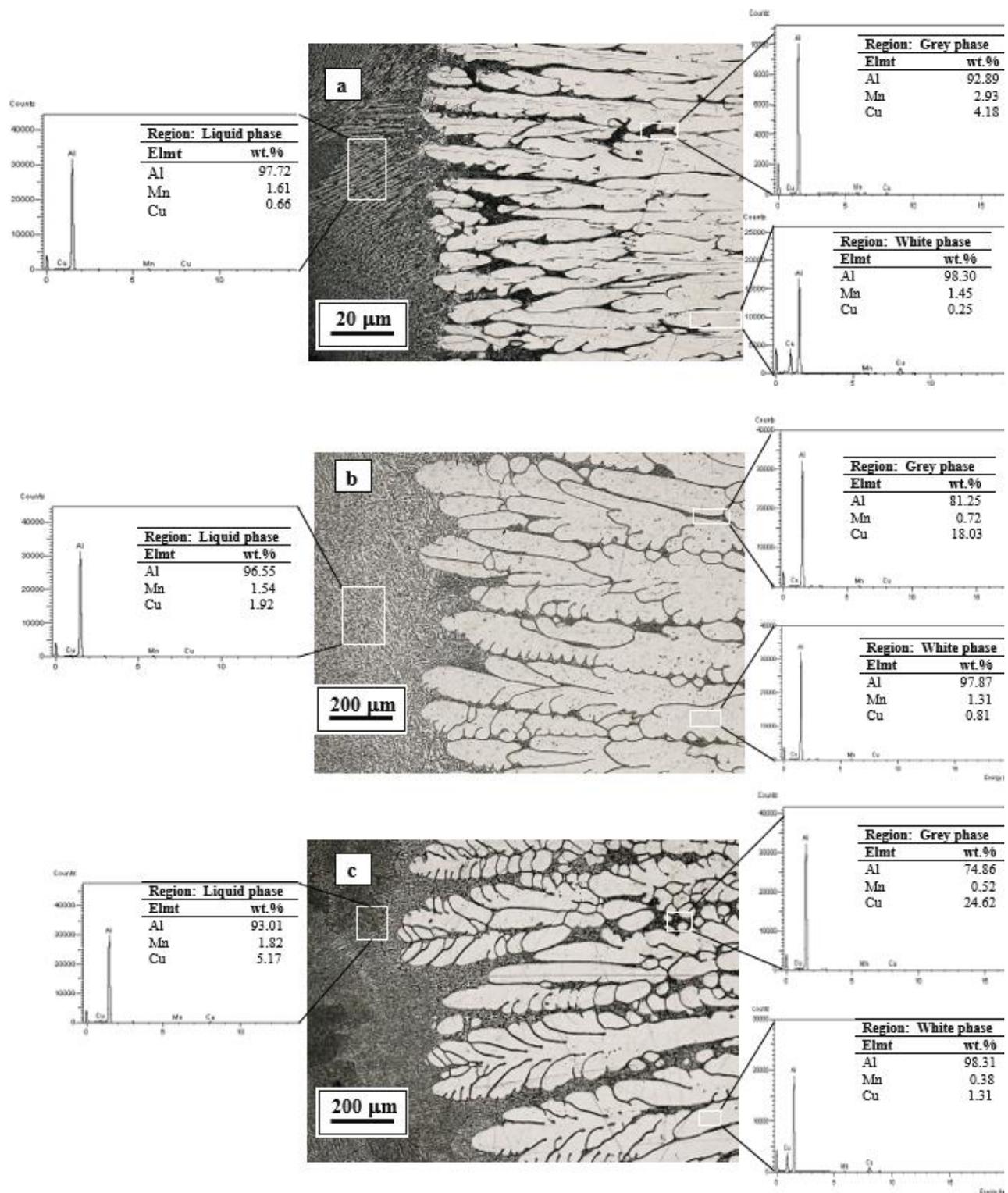
#### 3.1 Microstructural Characterization

The microstructure of Al-Mn-xCu alloys consists of Al-rich cellular and dendrites, as shown in Fig. 3. The EDX analysis in Fig. 3 indicates that the chemical composition of quenched liquid phase is very close to the nominal composition and the inter-cellular and inter-dendritic gray phase belongs to the Al-rich matrix phase. According to the EDX results, the measurements taken from the liquid region of Al-1.61Mn-0.66Cu, Al-1.54Mn-1.92Cu and Al-1.82Mn-5.17Cu (wt. %) are very close to the nominal composition of Al-1.9Mn-xCu ( $x=0.5, 1.5, 5$  wt.%) alloys, respectively. The chemical compositions of dendrite trunks in solid phase are Al-1.45Mn-0.25Cu, Al-1.31Mn-0.81Cu and Al-0.38Mn-1.31Cu (wt.%). As can be seen from Fig. 3(a, b and c), the amounts of Cu in the dendrite trunks increased and contributed to the formation of Cu-rich regions (Region: grey phase).

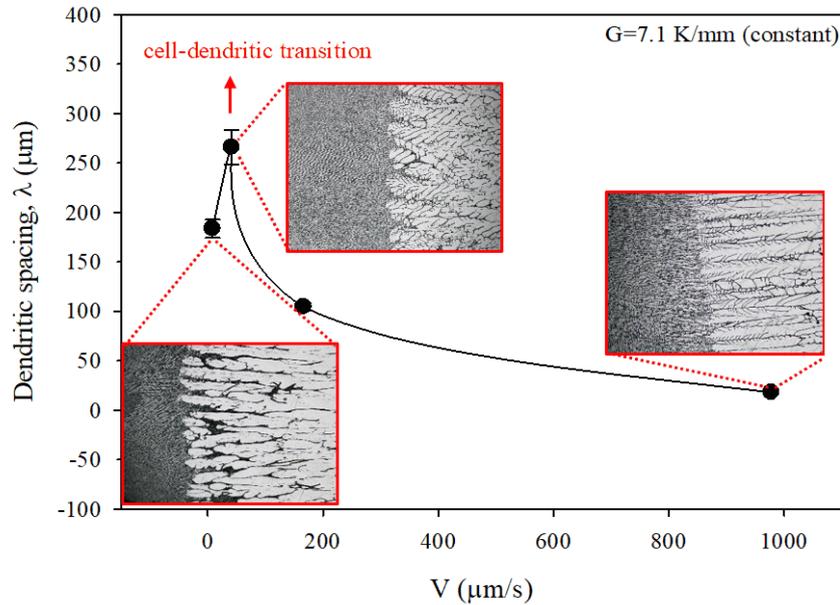
On the other hand, cell-dendritic transition was detected for low growth velocity ( $41.6 \mu\text{m/s}$ ) for 0.5 and 1.5 Cu%. Then with increasing growth velocity, the microstructure is completely converted to a dendritic form. The cell-dendritic transition observed (Büyük et al., 2017) at growth rate of  $41.6 \mu\text{m/s}$  for Al-1.9Mn-0.5Cu alloy is given in Fig. 4.

#### 3.2 The Effect of Growth Velocity and Cu Content on the Microstructure

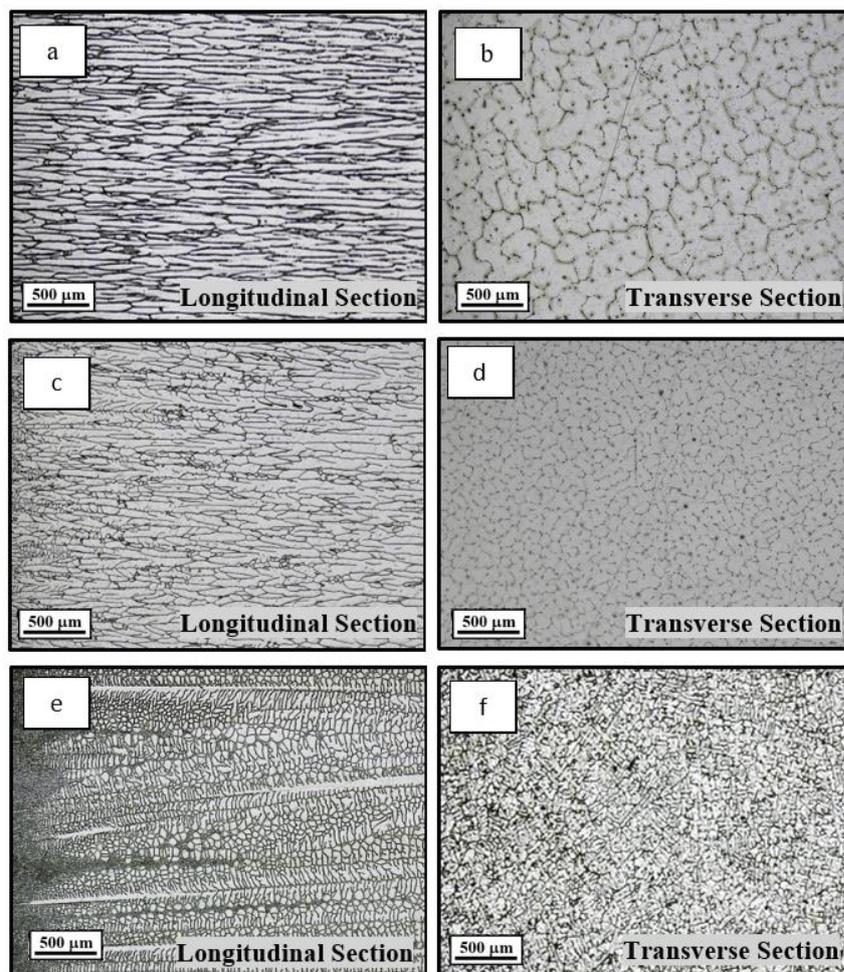
The Al-Mn-xCu samples were directionally solidified under a constant temperature gradient  $G$  ( $7.1 \text{ K/mm}$ ) and different growth velocities  $V$  (from  $8.3$  to  $978 \mu\text{m/s}$ ). Microstructural properties of the grown samples were analyzed, and the effect of the growth velocity on the primary dendritic spacing ( $\lambda$ ) was examined (Fig. 5 and Fig 6). In this work, while a linear relationship was observed between growth rate and primary dendritic spacing for 5Cu composition (Fig. 6c), the cell-dendritic transitions were observed for 0.5Cu (Fig. 6a) and 1.5Cu (Fig. 6b) compositions in the Al-1.9Mn-xCu alloy with increasing growth velocity.



**Figure 3** The chemical composition analysis and growth microstructures of the Al-Mn-xCu alloys, (a) Al-1.9Mn-0.5Cu, (b) Al-1.9Mn-1.5Cu, (c) Al-1.9Mn-5Cu



**Figure 4.** Typical images of cellular, cell-dendritic transition and dendritic structures as a function of  $V$ .

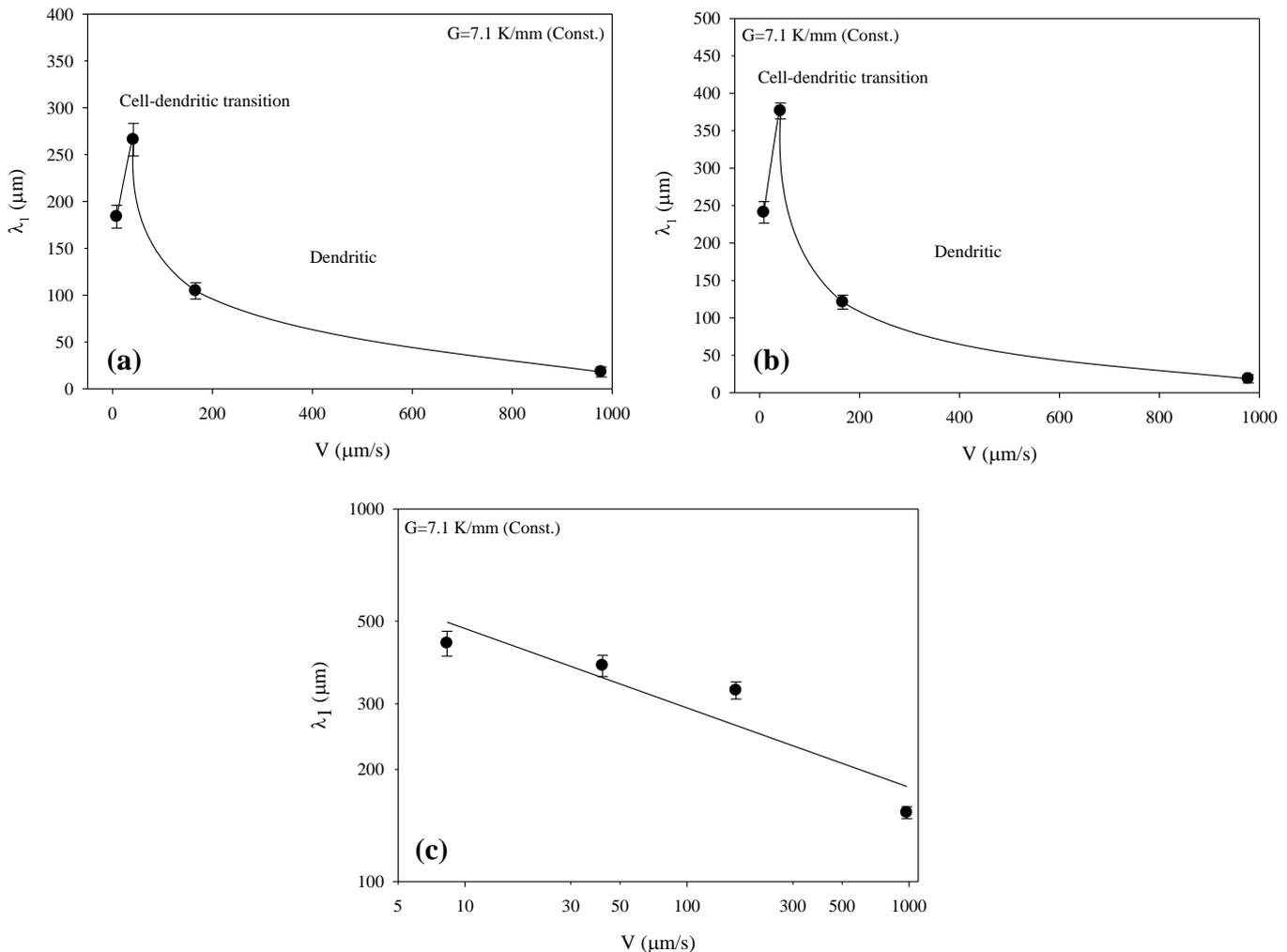


**Figure 5.** Typical optical micrographs of the directionally solidified Al-1.9Mn-xCu alloys at  $V=166.3$   $\mu\text{m/s}$  and  $G$  (7.1 K/mm); (a-b) Al-1.9Mn-0.5Cu, (c-d) Al-1.9Mn-1.5Cu, (e-f) Al-1.9Mn-5Cu

From the experimental results for Al-1.9Mn-5Cu alloy, the relationship between primary dendritic spacing and the growth velocity can be written as:

$$\lambda = k_1 V^{-a} \quad (2)$$

where  $k_1$  is the proportionality constant and  $a$  is the exponent value of  $V$ . As can be seen from Fig. 6, while the value of  $\lambda$  decreases with increasing growth velocity, it decreases with increasing Cu content.



**Figure 6.** Variation of  $\lambda_1$  with  $V$  for directionally solidified Al-1.9Mn-xCu alloys (a) Al-1.9Mn-0.5Cu (b) Al-1.9Mn-1.5Cu (c) Al-1.9Mn-5Cu.

The exponent values of  $V$  relating to  $\lambda$  were obtained to be  $\lambda_1 = k_2 V^{-0.21}$  ( $k_2 = 779.83 \mu\text{m}^{1.21} \text{s}^{-0.21}$ ,  $r^2 = -0.942$ ) for Al-1.9Mn-5Cu alloy. The exponent value (0.21) is close to theoretical value of 0.25 predicted by the Hunt (Hunt, 1979), Kurz-Fisher (Kurz and Fisher, 1982), and Trivedi (Trivedi, 1984) models for binary dendritic growth. However, no comprehensive theoretical model has been proposed for the dendritic solidification of ternary or multicomponent alloys so far.

On the other hand, our exponent value (0.21) is also in good agreement with the values of 0.30, 0.24, 0.25, 0.24 and 0.28 obtained by Fan et al. (Fan et al., 2011), Lapin et al. (Lapin et al., 2011) or Kaya et al. (Kaya et al., 2009, Kaya et al., 2007) for Al-based alloys produced under similar solidification conditions.

As shown in Fig. 6, the  $\lambda$  values increase with the increasing Cu content for the same growth velocities. The  $\lambda$  values were measured to be 184, 241 and 436  $\mu\text{m}$  at a constant growth velocity (8.3  $\mu\text{m/s}$ ) for 0.5Cu, 1.5Cu and 5Cu containing alloys, respectively.

#### 4. CONCLUSIONS

In this work, the effect of growth velocity and Cu content on the microstructure properties of Al-1.9Mn-xCu (x=0.5, 1.5, 5 wt.%) Cu alloys have been investigated. As a result of work, while a linear relationship was observed between growth velocity and primary dendritic arm spacing for Al-1.9Mn-5Cu, the cell-dendritic transitions were observed for Al-1.9Mn-0.5Cu and Al-1.9Mn-1.5Cu alloys at growth rate of 41.6 $\mu\text{m/s}$ . While the value of  $\lambda$  decreases with increasing growth velocity, it decreases with increasing Cu content.

The experimental relationship between  $\lambda_1$  and V was obtained to be:  $\lambda_1=k_2V^{-0.21}$ , ( $k_2=779.83 \mu\text{m}^{1.21} \text{s}^{-0.21}$ ,  $r^2=-0.942$ ) for Al-1.9Mn-5Cu alloy.

#### ACKNOWLEDGEMENTS

This research was financially supported by the Scientific and Technical Research Council of Turkey (TUBİTAK) under contract no. 212T130. The authors are grateful to TUBİTAK.

#### REFERENCES

- [1] R. Trivedi, W. Kurz, Dendritic growth, *Int. Mat. Rev.* 39 (1994) 49–74.
- [2] U. Büyük, N. Maraşlı, H. Kaya, E. Çadırlı, K. Keşlioğlu, Directional solidification of Al-Cu-Ag alloy, *Applied Physics A: Materials Science and Processing*, 95 (2009) 923–932.
- [3] U. Büyük, S. Engin, N. Maraşlı, Microstructural characterization of unidirectional solidified eutectic Al-Si-Ni alloy”, *Materials Characterization*, 62 (2011) 844-851.
- [4] U. Büyük, Physical and mechanical properties of Al-Si-Ni eutectic alloy”, *Metals and Materials International*, 18(6) (2012) 933–938.
- [5] U. Büyük, S. Engin, N. Marasli, directional solidification of Zn–Al–Cu eutectic alloy by the vertical Bridgman method, *Journal of Mining and Metallurgy, Section B: Metallurgy*, 51 (1) (2015) 67–72.
- [6] J. Davis (Ed.), *Alloying: Understanding the Basics* (first ed.), ASM International, Ohio (2001).
- [7] J. Davis (Ed.), *Metals Handbook Desk Edition* (second ed.), ASM International, Ohio (1998).
- [8] P.J. Macken, A.A. Smith, CDA Publication 31, second ed., (1966) 224–238.
- [9] J.M. Langham, A.W.O. Webb *AFS Trans.*, 70 (1962) 686–703.
- [10] S. Ganesan, C.L. Chan and D.R. Poirier, Permeability for flow parallel to primary dendrite arms. *Materials Science and Engineering A*. 151 (1992) 97–105.
- [11] M.S. Bhat, D.R. Poirier and J.C. Heinrich, Permeability for cross flow through columnar-dendritic alloys. *Metallurgical and Materials Transactions B*. 26 (1995) 1049–1056.
- [12] U. Büyük, H. Kaya and E. Çadırlı, Microstructure evolution and mechanical properties of Al-1.94Mn-0.5Cu alloy produced by directional solidification, *IIER International Conference on Applied Physics and Mathematics (ICAPM)*, Page 7-10, Proceedings of The IIER International Conference, Kiev, Ukraine, 17–18 July 2017.
- [13] J. D. Hunt, *Solidification and casting of metals*, The Metal Society, London, (1979) 3–9.
- [14] W. Kurz, D.J. Fisher, Dendritic growth and limit of stability tip radius and spacing, *Acta Metall.* 29 (1981) 11–20.
- [15] R. Trivedi, Interdendritic spacing: part II. A. comparison of theory and experiment, *Met. Trans.* A 15 (1984) 977–982.
- [16] J. Fan, X. Li, Y. Su, R. Chen, J. Guo, H. Fu, Directional solidification of Ti–49 at.%Al alloy, *App. Phys. A* 105 (2011) 239–248.
- [17] J. Lapin, Z. Gabalcová, Solidification behaviour of TiAl-based alloys studied by directional solidification technique, *Intermetallics* 19 (2011) 797–804.

- [18] H. Kaya, E. Çadırlı, M. Gündüz, Directional cellular growth of Al-2wt%Li bulk samples, *Applied Physics A* 94 (2009) 155–165.
- [19] H. Kaya, E. Çadırlı, M. Gündüz, Dendritic growth in an aluminum-silicon alloy, *J. Mater. Eng. Perf.* 16 (2007) 12–21.