

COMPLEX CHARACTERIZATION OF IRREGULAR EUTECTIC STRUCTURE

KASSAB AL-OMARI¹ – ARNOLD RÓNAFÖLDI² – ZSOLT VERES³

Abstract: Al-12.6Si alloy was solidified in a vertical Bridgman-type furnace equipped with a rotating magnetic inductor to induce the flow in the melt. The eutectic alloy was solidified unidirectionally by a growth rate of $v = 0.1$ mm/s, by a temperature gradient of $G = 6$ K/mm and with or without magnetic stirring. The interlamellar distances, the length, and orientation of Si lamellas were investigated by using new measuring methods.

Keywords: Eutectic solidification, Irregular eutectic, Rotation Magnetic Field RMF, interlamellar distances (λ .)

INTRODUCTION

The investigation of pattern formation in Al alloys has been the topic of several research studies, [1]. The formation of microstructures during solidification plays an important role in determining the properties of metal products [2]. Depending on the applied processing parameters and on the physical parameters of the material, a wide variety of solidified microstructures are observed in the course of the casting, welding, and other solidification processes.

The melt flow is an important parameter that affects the microstructure developing during solidification. The melt flow can be categorized into natural and forced convection. The contraction (or expansion) when the densities of the solid and liquid phases are different and the presence of the gravitational force during solidification are the two main reasons that cause the natural convection. The forced convection is a convection which is due to externally applied conditions. The melt flow can be controlled by a rotating magnetic field (RMF). The melt flow induced by RMF leads to macrosegregation in radial and axial directions, which leads to a change of phase ratio and distribution [3] [4] [5].

The combination of the Al-Si component improves the mechanical properties by which the wide range of application of the Al-Si eutectic alloy can be explained. The solidification of Al-Si eutectic alloys generally takes place at 12.6 w% of Si and at

¹ Institute of Physical Metallurgy, Metal forming & Nanotechnology, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
kassabalomari1992@gmail.com

² Institute of Physical Metallurgy, Metal forming & Nanotechnology, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary

³ Institute of Physical Metallurgy, Metal forming & Nanotechnology, University of Miskolc
H-3515 Miskolc-Egyetemváros, Hungary
vezso3838@gmail.com

an equilibrium temperature of 577 °C. The structure of Al-Si eutectic alloy shows two phases – they are as follows: Al solid solution phase as a matrix and a lamella-like Si particle. [6] [7]

The first theoretical explanation of the regular structure was developed by Jackson and Hunt in 1966. They simplified the problem of diffusion ahead of the solidification front by assuming a planar interface solidification and identical undercooling of melt in front of the solid phases which grow together from the melt of eutectic composition [8]. Many other explanations are based on the Jackson and Hunt model which developed new approaches in order to explain the relationship between the different parameters like the growth rate of the solid phase(v), undercooling of the melt (ΔT), temperature gradient (G) and the microstructural parameters such as the interlamellar distances λ and lamella length. [9] [10] [11] [12].

Experimentally, the Al-Si eutectic alloy has an irregular eutectic structure; owing to the growth of lamella with an undefined orientation and with different length, the microstructural measurements became a complex task for researchers.

Usually, the characterization of the eutectic structure means comparing two places in different samples and give a “fine/finer” or “coarse/coarser” description (Figure 1) [13]. These descriptions which are out of any numerical indication, are not enough to characterize the eutectic structure.

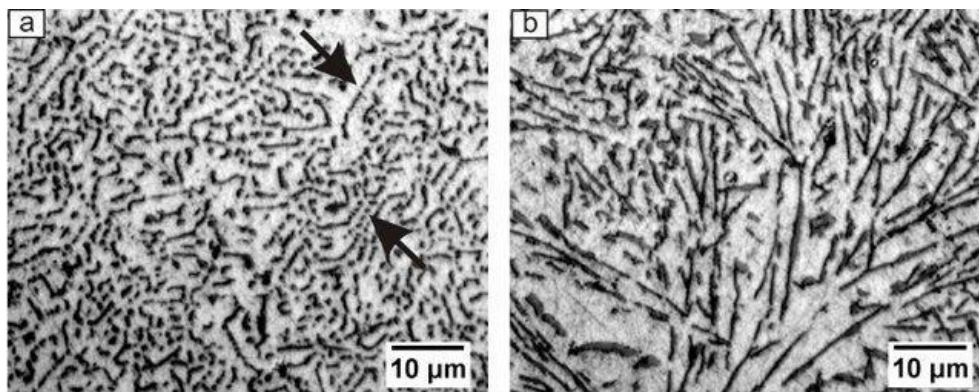
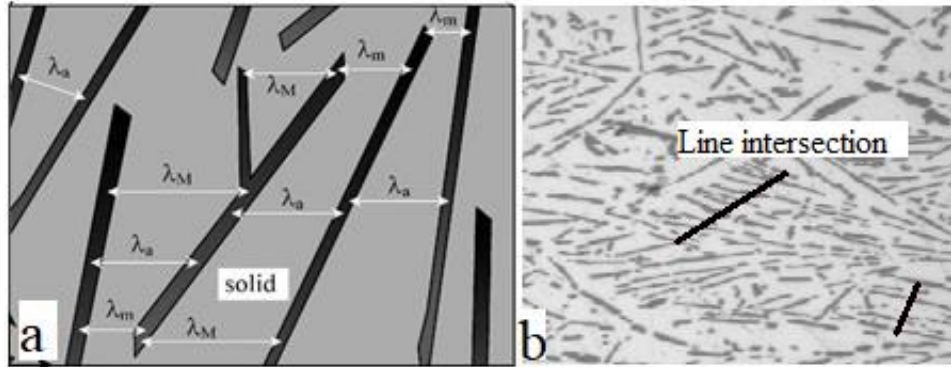


Figure 1

(a) Fine, and (b) coarse Al-Si eutectic structure [13]

On the other side, direct measurements were performed to characterize the interlamellar distances [14], on the transverse section (perpendicular to the growth direction) or on the longitudinal section (parallel to the growth direction) (Figure 2a).

Among others, Clapham and Smith consider the line intercept method to measure the interlamellar distances, where the measurements are done by draw line in a “Wheatsheaf” formation of the eutectic lamellae, with their axes lying perpendicular to the quenched interface. Each line crosses several lamellae nearly at right angles (Figure 2b) [15] [16] [17].

**Figure 2**

(a) Direct measurements (λ_m : minimum, λ_M : maximum, λ_a : average interlamellar distances) (b) line intercept method ($M = 500$)

There are a lot of Si eutectic lamellas with different growing directions in the eutectic Al alloys therefore the measurements of the interlamellar distances become very complicated. The two above methods (direct measurement and line intercept method) try to represent the eutectic structure with a limited number of measurements on the selected parts of the sample. Owing to this fact, the results of these methods highly depend on the examiner and the human factor.

Some researchers don't believe in the previous method and use the "Fiber spacing method" as an alternative method to measure the interlamellar distances [18] [19]. Originally, the aim of this method was to measure the distance between the primary dendritic arms on primary phases in the dendritic structures where the number of particles (N) and the area (A) were taken into account to calculate the spacing (λ). [$\lambda = (A/N)^{0.5}$]. However, two valuable factors are missing from the fiber spacing method namely the particular area for each component (phase ratio) and the eutectic size factor.

Based on the previous reasons, the present paper suggests a new measuring method for measuring the interlamellar distances. For a better characterization of the eutectic structure, the measurement of further parameters of Si lamellae is suggested by us.

1. COMPLEX CHARACTERIZATION OF AN IRREGULAR EUTECTIC

Characterizing the irregular eutectic structure has been a big challenge for researchers because every eutectic lamella has a different length, thickness, and angle which represent the inclination between the axis of the lamellae and the direction of the solidification movement.

The distance between the lamellar spans a wide range as well, therefore we suggest to use the following three kinds of parameters to describe the irregular eutectic structure: the average interlamellar distance, the length and the angle of lamella.

By means of the image analysis software, we use the “Specific perimeter method” to measure the average interlamellar distance (λ), see *Equation (1)*

$$\lambda = 2 * \frac{A_p * (1 - A_f)}{N * P_o} \quad (1)$$

Where:

- P_o : is the average perimeter of the silicon lamella in the investigated microscopic photos (μm); it can be measured by image analysis software.
- N : Number of the eutectic lamellae.
- A_p : Area of the microscopic photo (μm^2).
- A_f : Area fraction of the eutectic lamellae in the eutectic structure.

Near the average lamellae distance, we measure the length of lamellae, which is equal to the Ferret diameter. The Ferret diameter can be defined as the distance between two parallel tangential lines restricting the object perpendicular to that direction (*Figure 3a*). The new image processing techniques allow us to measure the Ferrets diameter for each object (every eutectic Si lamella) and analyze them. The measured Ferret diameters were divided into ranges and the percentage distribution of lamellas belonging to the given ranges were investigated.

In addition, the angle measurement has been performed as well. The angle measurements describe the orientation between the axis of the eutectic silicon lamella and the axis of direction of solidification front movement (*Figure 3b*). A percentage relation between the above-mentioned size ranges and the angle distribution in that range was created.

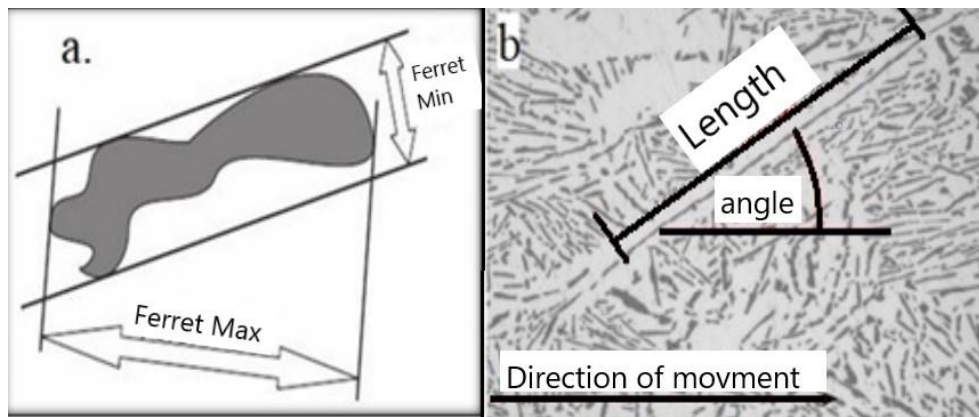


Figure 3

(a) Description of Ferret diameter. (b) The different measured parameters in Al-Si eutectic alloy ($M = 500$)

2. VALIDATION OF THE SPECIFIC PERIMETER METHOD

The new method has to be validated; a sample of Al-12.6 Si alloy was used for this purpose. This composition is the eutectic composition of the Al-Si alloys which contains only eutectic structure in equilibrium.

The validation process shall be performed on a different structure therefore the eutectic sample has been solidified under the effect of forced melt flow. Rotating Magnetic Field (RMF) was used for creating a magnetic stirring during solidification. Different flow velocities were used by changing the magnetic induction intensity from 0 mT (without stirring) to 150 mT.

Under the influence of magnetic stirring, a finer eutectic structure develops and the Si composition distribution changes; this phenomenon is called “macrosegregation”. Due to the macrosegregation, the primary phase α and the primary silicon solidify alongside with the eutectic structure, but our research concern only the investigation of the eutectic structure.

The solidified samples were sectioned longitudinally and prepared by grinding, polishing, and etching in HF acid. The specimens were studied by Light Microscopy and Image J image analysis software.

The microscopic photos were taken by using the mosaic photo technique; The mosaic acquisition is a processing system to make large expanses of the target area on the object sample close to the resolution limit of light microscopy. To take a mosaic photo, a larger image is created from a series of smaller images individually collected orderly across a sample.

2.1. Interlamellar spaces

The present work recommends the specific perimeter method as a valid method for characterizing the interlamellar distances. This recommendation is based on several tens or more of validity tests. The validity of the specific perimeter method was tested on different Al-Si eutectic structure and compared with the Direct measurement method. The result shows a promising method because the error percentage does not exceed 2% in some cases, which makes the invented method a precise and fast measurement method. *Table 1* shows an example of some validation tests for random eutectic structures in the Al-Si eutectic specimen.

Table 1

Validation tests of the special Perimeter method

Number of tests	Magnetic induction (mT)	Surrounding structure	Interlamellar distance [μm]		Standard deviation
			Specific Perimeter method	Real hand Measurements	
1	0	Eutectic structure	3.64	3.7	0.04
2	0	Eutectic structure	3.8	3.5	0.2
3	0	Eutectic structure	3.45	3.6	0.1
4	150	Eutectic structure	3.35	3.3	0.03
5	150	Primary Al- Phase	2.26	2.4	0.09
6	150	Primary Si phase	4.32	4.4	0.05

It proves to be a complex task to measure the interlamellar distances owing to the formation of α primary phase and primary silicon in the stirred microstructure. Three different appearance of eutectic structure grow depending on the surrounding phase i.e. it has been found that a fine eutectic structure forms near the primary α phase, while a coarse eutectic structure grows near the primary silicon, and lastly the well-known eutectic structure forms where there are no primary structures nearby. (Figure 4)

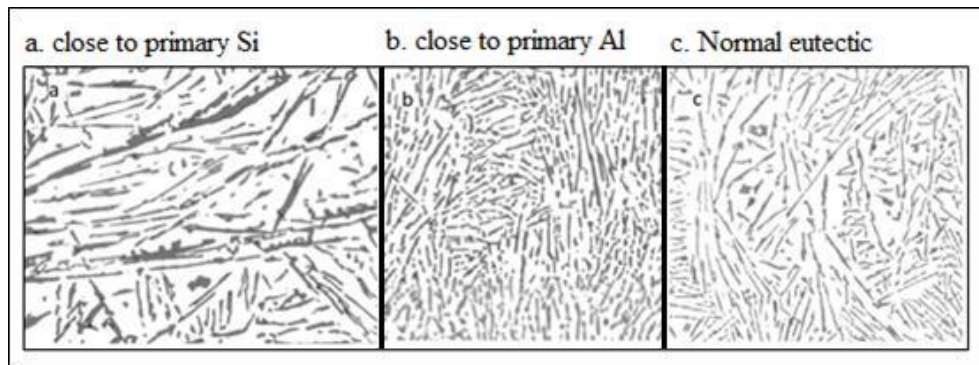


Figure 4

*Different structures of eutectic type, ($M = 500$).
($v = 0.1 \text{ mm/s}$, $G = 6 \text{ K/mm}$, magnetic intensity = 150 mT)*

Owing to the aforementioned facts, it is not recommended to characterize the interlamellar distances without considering the place of eutectic structure; the average value was calculated for each type and given in Table (2). The process of studying the effect of primary phases on the eutectic structure will be described in our following paper.

Table 2
Measurements of the interlamellar distances

Eutectic group	Distance (μm) ± 0.2
Without RMF	3.7
With RMF main eutectic structure	3.5
With RMF close to primary Si	4.06
With RMF near to primary Al	1.99

2.2. Length of lamellae

As mentioned before, the length measurements were performed by measuring the Ferret diameter of each Si eutectic lamellae. An image analysis software has been used to measure them.

The length measurements were analysed by categorizing the size of the eutectic Si lamella into different ranges starting from (0–10) and ending with (290–300) μm . In this way, we can investigate the size at which the effect of RMF will be the largest one. *Figure 5* shows the size ranges percentage both in the stirred and non-stirred parts.

The fine eutectic structure range (i.e. $< 10\mu\text{m}$) has the largest percentage both in the stirred and non-stirred parts. However, 95% of fine eutectic can be observed in the stirred part while 75% of it can be observed in the non-stirred specimen. The mechanisms of size reduction due to the magnetic fields applied during solidification are not yet well understood, but some authors state that they are due to the fragmentation of the primary arms of dendrites that act as nuclei [20], [21]; it can be true for large eutectic Si lamellas as well. Others state that they are due to the pressure differences caused by the oscillation of liquid that causes cavitation and consequently some nucleation of embryos [22], [23]. Yet, others claim that there is an undercooling effect due to the magnetic intensity [22]. There is some general agreement that the mass transportation of the cooled liquid from the mould perimeter to the hotter regions increases the solidification rate [22]. The mass transportation provides an opportunity to solute undercooling [20], [21].

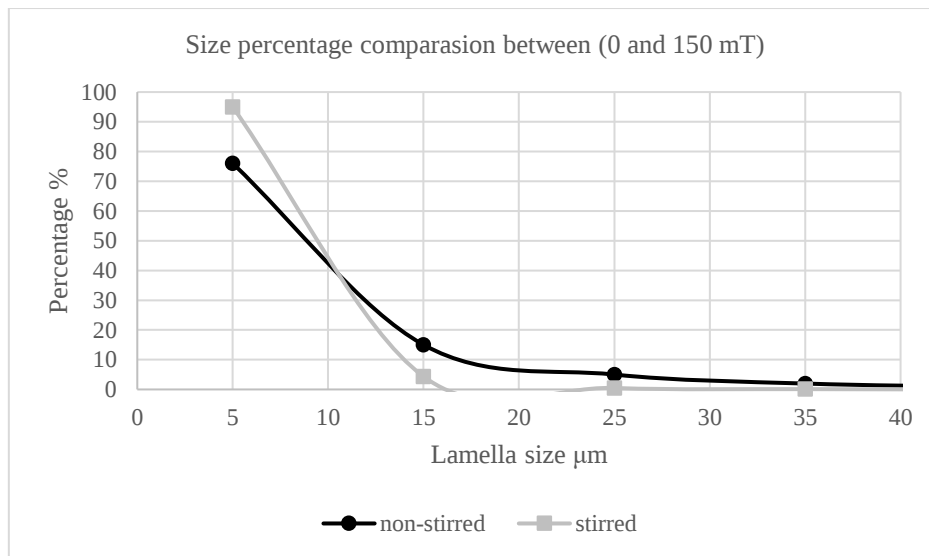


Figure 5
Si lamella size percentage of Al-Si eutectic sample with 0 and 150 mT

2.3. Angle measurements

The angle of the lamella describes the inclination degree between the axis of the eutectic silicon lamellas and the direction of the solidification movement.

The size measurements show the large effect of RMF on fine eutectic Silicon. Therefore, an angle distribution curve was created for calculating the effect of RMF on the angle of fine eutectic Si lamellas (*Figure 6*).

When the solidification process takes place without the effect of the forced melt flow, 60% of the fine eutectic lamella tends to orient in a 45° from the sample axis.

Figure 6 shows a decreasing at 45° angle range percentage when RMF is switched on. This percentage will reduce to 35% by applying 150 mT. It means that the orientations of Si lamellas are more diverse when using RMF than those without RMF.

These large orientation differences suggest that the Si lamellas do not have an epitaxial growth in case of using RMF at 50 Hz as most of the Si lamellas without RMF seem to have.

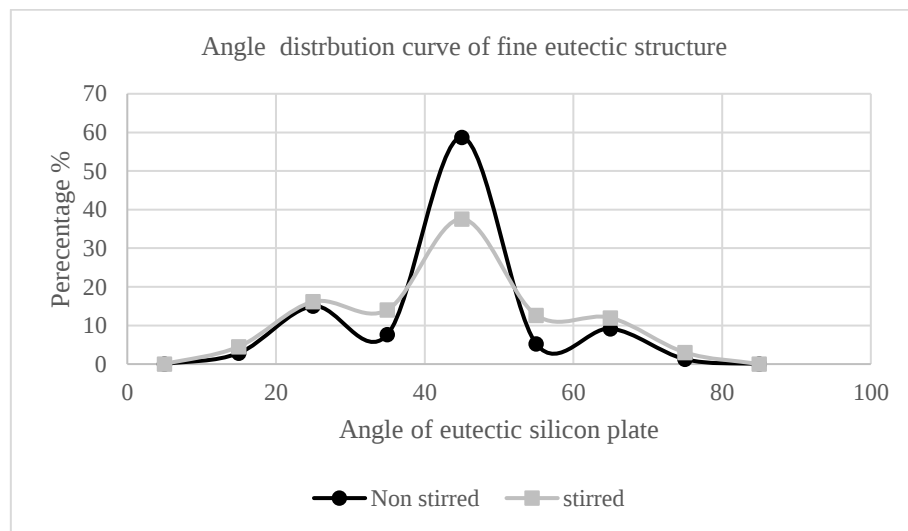


Figure 6

The angle distribution curve of fine eutectic Si lamella ($<1\mu\text{m}$) of Al-Si eutectic specimen

CONCLUSION

A newly developed complex characterizing method was used to describe the irregular eutectic structure. Three parameters were characterized.

- The average interlamellar distance λ was characterized by using the specific perimeter method which proves its validity when it is compared with the other methods used recently.
- The length of the eutectic Si lamellas was characterized by using an image analysis software in order to measure the Ferret diameter of every Si lamella.
- The angle of the eutectic Si lamellae, where it was measured by determining the inclination degree between the axis of the Si lamella and the direction of the solidification front movement.

When Al-Si eutectic alloy is solidified under the effect of magnetic stirring induced by RMF, the microstructure will respond by reducing the length of the eutectic Si lamella. It results in more disorientations in the eutectic silicon lamella, and different eutectic appearances which means different interlamellar distances.

Based on our investigation, the used characterization method is good enough to characterize the different parameters in an irregular eutectic structure because it is a fast measurement, considers every single lamella and an accurate measurement with an error percentage which does not exceed 0.2%.

ACKNOWLEDGMENT

The described article was carried out as part of the EFOP-3.6.1-16- 2016-00011 *Younger and Renewing University – Innovative Knowledge City –institutional development of the University of Miskolc aiming at intelligent specialization* project implemented in the framework of the Szechenyi 2020 program. The realization of this project is supported by the European Union, co-financed by the European Social Fund.

REFERENCES

- [1] Karma, A., Rappel, W.-J. (1998). Quantitative phase-field modeling of dendritic growth in two and three dimensions. *Phys. Rev. E.*, Vol. 57, No. 4, pp. 4323–4349.
- [2] Dantzig, J., Rappaz, M. (2009). *Solidification*. Switzerland, EPFL Press.
- [3] Tiller, W., Jackson, K., Rutter J., Chalmers, B. (1953). The redistribution of solute atoms during the solidification of metals. *Acta Metallurgica*, Vol. 1, No. July, pp. 428–437.
- [4] Svéda, M., Sycheva, A., Kovács, J., Rónaföldi, A., Roósz, A. (2014). The effect of rotating magnetic field on the solidified structure of Sn–Cd peritectic alloys. *Materials Science Forum*, Vols. 790–791, pp. 414–419.
- [5] Rónaföldi, A., Kovács, J., Roósz, A. (2007). Investigation and visualization of melt flow under rotating magnetic field. *Transactions of the Indian Institute of Metals*, Vols. 2–3, pp. 213–218.
- [6] Fredriksson, H., Åkerlind, U. (2012). *Solidification and Crystallization Processing in Metals*. Chichester, John Wiley & Sons Ltd.
- [7] Stefanescu, D. (2002). *Science and Engineering of Casting Solidification*. New York, Kluwer Academic/Plenum Publishers.
- [8] Jackson, K., Hunt, J. (1966). *Trans. Met. Soc. AIME*, Vol. 236, p. 1129.
- [9] Fisher, D., Kurz, W. (1980). A theory of branching limited growth of irregular eutectics. *Acta Metallurgica*, Vol. 28, No. 6, pp. 777–794.

-
- [10] Magnin, P., Kurz, W. (1987). An analytical model of irregular eutectic growth and its application to Fe-C. *Acta Metallurgica*, Vol. 35, No. 5, pp. 1119–1128.
- [11] Magnin, P., Mason, J. T., Trivedi, R. (1991). Growth of irregular eutectics and the Al-Si system. *Acta Metallurgica et Materialia*, Vol. 39, No. 4, pp. 469–480.
- [12] Steen, H. A. H., Hellawell, A. (1975). The growth of eutectic silicon – contribution to undercooling. *Acta Metallurgica*, Vol. 23, pp. 529–535.
- [13] Timpel, M., Wanderka, N., Kumar, G. V., Banhart, J. (2011). Microstructural investigation of Sr-modified Al-15 wt%Si alloys in the range from micrometer to atomic scale. *Ultramicroscopy*, Vol. 111, pp. 695–700.
- [14] Gündüz, M., Kaya, H., Çadırlı, E., Özmen, A. (2004). Interflake spacings and undercoolings in Al-Si irregular eutectic alloy. *Material Science and Engineering*, Vol. A3690, pp. 215–229.
- [15] Clapham, L., Smith, R. W. (1989). Partial modification in unidirectionally solidified Al-Si eutectic alloys. *Acta Metall.*, Vol. 37, No. 1, pp. 303–311.
- [16] Flood, S., Hunt, J. (1981). Modification of Al-Si eutectic alloys with Na. *Metal Science*, Vol. 15, No. 7, pp. 287–294.
- [17] Hogan, L., Song, H. (1987). Interparticle Spacings and Undercoolings in Al-Si Eutectic Microstructures. *Metallurgical Transactions A*, Vol. 18A, pp. 707–713.
- [18] Glenister, S. M. D., Elliott, R. (1981). Strontium modification of Al-12.7wt-%Si alloys. *Metal Science*, Vol. 15, No. 4, pp. 181–184.
- [19] Kobayashi, K. F., Hogan, L. M. (1985). The crystal growth of silicon in Al-Si alloys. *Journal of Materials Science*, Vol. 20, pp. 1961–1975.
- [20] Yasuda, H., Nakatsuka, N., Nagira, T., Sugiyama, A., Yoshiya, M., Uesugi, K. et al. (2009). X-ray imaging study on grain refinement due to dendrite fragmentation. In: *EPM 2009, 6th International Conference on Electromagnetic Processing of Materials*, Dresden, Germany.
- [21] Sugiura, K., Iwai, K. (2005). Refining mechanism of solidified structure of alloy by electromagnetic refining process. *ISIJ international*, Vol. 45, No. 7, pp. 962–966.
- [22] Jie, D., Jianzhong, C., Wenjiang, D. (2006). Theoretical discussion of the effect of a low-frequency electromagnetic vibrating field on the as-cast microstructures of DC Al–Zn–Mg–Cu–Zr ingots. *Cryst Growth*, Vol. 2, No. 295, pp. 179–187.
- [23] Hernandez, F. R., Sokolowski, J. (2006). Comparison among chemical and electromagnetic stirring and vibration melt treatments for Al–Si hypereutectic alloys. *Journal of alloys and compounds*, Vols. 1–2, No. 426, pp. 205–212.

- [24] Jackson, K., Hunt, J. (1966). Lamellar and rod eutectic growth. *Transactions of the Metallurgical Society of AIME*, Vol. 236 (8), p. 1129.
- [25] Kurz, W., Giovanola, B., Trivedi, R. (1986). Theory of microstructural development during rapid solidification. *Acta Metallurgica*, Vol. 34, No. 5, pp. 823–830.