

The effect of lanthanum on the solidification curve and microstructure of Al-Mg alloy during eutectic solidification

Shikun Xie^{*}, Rongxi Yi, Xiuyan Guo, Xiaoliang Pan and Xiang Xia

*School of Mechanical and Electrical Engineering, Jinggangshan University,
28 Xueyuan Road, Ji'an 343009, China*

(Received December 26, 2014, Revised July 8, 2015, Accepted July 10, 2015)

Abstract. The influence of rare earth lanthanum (La) on solidification cooling range, microstructure of aluminum-magnesium (Al-Mg) alloy and mechanical properties were investigated. Five kinds of Al-Mg alloys with rare earth content of La (i.e., 0, 0.5, 1.0, 1.5 and 2.0 wt.%) were prepared. Samples were either slowly cooled in furnace or water cooled. Results indicate that the addition of the rare earth (RE) La can significantly influence the solidification range, the resultant microstructure, and tensile strength. RE La can extend the alloy solidification range, increase the solidification time, and also greatly improve the flow performance. The addition of La takes a metamorphism effect on Al-Mg alloy, resulting in that the finer the grain is obtained, the rounder the morphology becomes. RE La can significantly increase the mechanical properties for its metamorphism and reinforcement. When the La content is about 1.5 wt.%, the tensile strength of Al-Mg alloy reaches its maximum value of 314 MPa.

Keywords: Al-Mg alloy; rare earth lanthanum (La); solidification curve; microstructure; extrusion; tensile strength

1. Introduction

The combination of low density, high strength, high fracture toughness, and excellent resistance to stress corrosion and fatigue performance are most desirable properties for those materials in high-strength applications, such as in aerospace, automotive, and mechanical industry (Montalba *et al.* 2015, Chen *et al.* 2007, Su *et al.* 2012, Yu *et al.* 2007). Due to its low density, high specific strength, easy processing and the economic advantages, cast aluminum alloy are widely used in a variety of industries (Liu and Hu 2008, Yi *et al.* 2009, Xie *et al.* 2010). Al-Mg-based alloy is a class of high-strength heat-resistant alloys. The Al-Mg-based alloys are widely used in earlier industrial applications, owing to their high temperature stability and high heat resistance. However, its wide solidification zone usually leads to a pasty solidification and synchronous solidification in the cross section of the cast when the alloy liquid is cooled gradually (Liang *et al.* 2013). As a result, dendritic crystals solidified firstly block the flow of liquid metal, resulting in a poor fluidity of the alloy liquid. The growth of alloy dendritic crystal will cause dendritic segregation with a lower melting temperature (Bai *et al.* 2012). The presence of such a dendritic segregation will

*Corresponding Author, Professor, E-mail: xskun@163.com

bring about a significant decrease in both technological properties (e.g., hot cracking tendency) and mechanical properties (i.e., toughness and plastic) of Al-Mg alloy. This considerably limits the applications of Al-Mg-based alloy (Si *et al.* 2006, Li *et al.* 2006 and Yi *et al.* 2010).

It is known that the rare earth can play an important role in the mechanical properties of alloys (Ma *et al.* 2009, Zhang *et al.* 2012, Gao *et al.* 2012). For example, lanthanum (La) can take a metamorphism effect on Al-Mg alloy. This work investigated the influence of rare earth La on the solidification cooling range, the metallographic microstructure, and mechanical properties of Al-Mg alloy. The main purpose of this work was to improve the quality of Al-Mg alloy castings and to enhance the properties of the alloy by alloying La in Al-Mg alloy.

2. Experimental procedures

2.1 Experimental equipments

The self-developed experimental equipment shown in Fig. 1 was employed in Al-Mg alloy ramp vibration pouring experiment, which consists of vibrating apparatus, ramp guide slot, cooling water system, casting mold, etc. Casting with a certain amount of mechanical vibration can further refine the alloy microstructure, and the grains are more rounded in the whole with its number increasing and its size reducing (Yi *et al.* 2010). The simplified schematic of the mechanical vibrating apparatus which is controlled by a RF-500TB DC motor is shown in Fig. 2. It consists of a vibration motor attached to the vibrating vessel via a vessel holding table supported by a group of springs. The vibrating vessel shall be fixed by bolts on the vessel holding table of the vibrating apparatus.

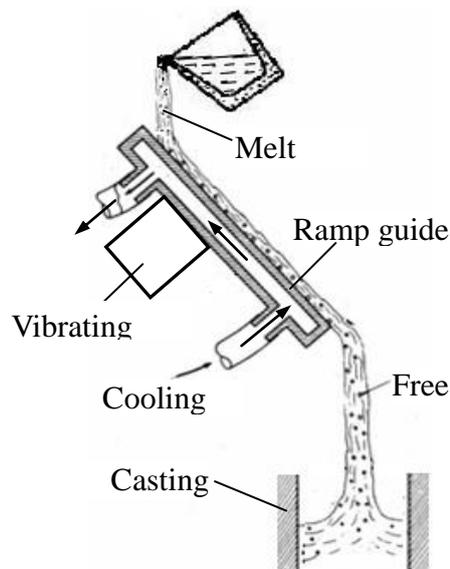


Fig. 1 Ramp vibration pouring device

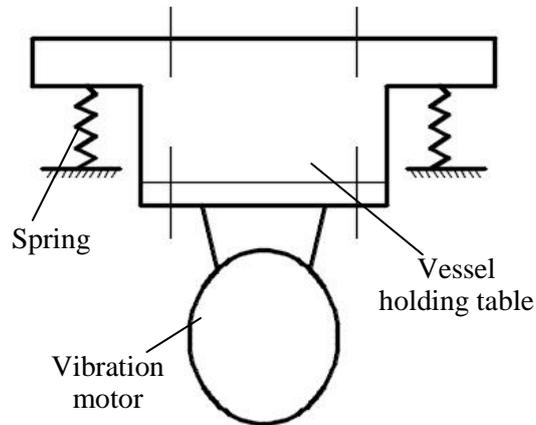


Fig. 2 Diagram of vibrating apparatus

2.2 Experimental

Starting from aluminum ingot, magnesium ingot, and rare earth La (all with high purity greater than 99.9 wt.%), five Al-Mg alloys with different rare earth La contents (i.e., 0%, 0.5 wt.%, 1.0 wt.%, 1.5 wt.%, and 2.0 wt.%) were prepared respectively. The mixed materials of each alloy were put in graphite crucible and loaded into a vacuum box-type resistance SX2-4-10 furnace with graphite crucibles to make the Al-Mg-based alloys. The working parameters are shown as below: voltage of 220 V, power of 5 kw, maximum working temperature of 1200°C, and normal operating temperature of 950°C. Then, the alloys were evacuated the box resistance furnace with a vacuum of about -0.08 MPa. Afterwards, smelt the mixed materials by heating them to 750°C and incubating for 2 hours. Then, the as-prepared alloy was poured into a graphite mold. A part of the prepared alloy with the graphite mold was put into a tube furnace at 700°C, and was slowly cooled down with furnace cooling. A KSW-8D-13 resistance furnace temperature controller and A K series 101 type thermocouple were used to control the furnace temperature. The range of the KSW-8D-13 temperature controller and thermocouples were from 0°C to 1200°C with their operating error more than $\pm 1^\circ\text{C}$. Meanwhile, the temperature change with time during the solidification process was recorded in real time by a data acquisition module. As such, the solidification cooling curve was drawn from the recorded data.

In order to test the impact of different rare earth La content on the microstructure of Al-Mg alloy, another part of the prepared alloy was cooled to 700°C and was directly poured into a ramp copper mold. Afterwards, the alloy in the ramp copper molds was cooled to room temperature in the air.

In the experiment of mechanical testing, the mechanical properties of the alloys in ramp copper molds were found not stable enough to evaluate the mechanical properties. Therefore, a part of samples were extruded on a 200T hydraulic machine after heated to 410°C and kept for one hour. The extruded parts were made into standard specimens for mechanical testing, and a series of tensile tests were carried out on a microcomputer control electronic WDW-200KN universal testing machine.

Table 1 The solidification temperature parameters for the Al-Mg alloys containing different La contents

La content (wt.%)	Start temperature (°C)	End temperature (°C)	Temperature range (°C)	The growth rate
0	652.5	628.5	24	--
0.5	651.5	627.3	24.2	0.83%
1.0	649.7	625.2	24.5	1.24%
1.5	648.9	618.7	30.2	23.27%
2.0	647.7	616.8	30.9	2.32%

Samples were taken from the casts and extrusion parts in the same positions and were ground up to 2000 grits SiC paper then polished for metallographic microstructural observations. All the prepared metallographic samples were etched in 0.5 vol.% HF solution.

3. Results

3.1 Effects of La on solidification curves

Fig. 3 shows the solidification curves of aluminum-magnesium alloys with different amount of La, i.e. at 0 wt.%, 0.5 wt.%, 1.0 wt.%, 1.5 wt.%, and 2.0 wt.%. As shown in Fig. 3, when La content is at 0 wt.%, the solidification of the alloy begins at 652.5°C and finishes at 628.5°C. While the addition of La increases to 0.5 wt.%, the alloy begins to solidify at 651.5°C, and ends solidification at 627.3°C. When the La content increases to 1.0 wt.%, 1.5 wt.%, and 2.0 wt.%, respectively, the alloy starts to solidify at 649.7°C, 648.9°C, and 647.7°C, and ends solidification at 625.2°C, 618.7°C, 616.8°C, respectively. Therefore, the addition of rare earth La could significantly alter the solidification range of aluminum-magnesium alloy.

In general, a wide two-phase region temperature of alloy is helpful in the solidification processes of semi-liquid metals during cooling (Liang *et al.* 2013). The solidification data of these Al-Mg alloys containing La contents were summarized in Table 1. As shown in Table 1, without rare earth La, the two-phase region in the Al-Mg alloy is from 652.5°C to 628.5°C, resulting in the solid-liquid interval temperature is about 24°C. The two-phase region in the aluminum-magnesium alloy is broadened with the addition of La. When the La content reaches about 2.0 wt.%, the two-phase region temperature of the alloy is from 647.7°C to 616.8°C, and the temperature range is 30.9°C. The growth rate of temperature range reaches its maximum value of 23.27% at 1.5 wt.% addition of La. It is concluded that the highest growth rate of temperature range and largest two-phase region temperature is the Al-Mg alloy containing 1.5 wt.% La and 2.0 wt.% La, respectively. But when La content is more than 1.5%, the growth rate of temperature range is not obvious. In addition, the solidification time of the alloy increases with the doping of rare earth La.

3.2 Effects of La on cast microstructure

Fig. 4 shows the metallographic microstructure of the cast Al-Mg alloys at 700°C slope copper water-cooled casting with La content of at 0%, 0.5 wt.%, 1.0 wt.%, 1.5 wt.%, and 2.0 wt.%. As seen from Fig. 4(a), when the addition of La is 0%, the grain size of the alloy is coarse with developed dendrite, and the grain boundary is not evident. However, the micro-alloying of rare

earth La into the Al-Mg alloy significantly altered the microstructure. Apparently, the crystallization of dendrites was frustrated. The number of the primary α -Al phase grains is increased with the addition of La. The grain becomes small, round and uniform distributed, and shows equiaxed and rose shape, and finally, its dendrites are degenerated completely.

By comparing the cast microstructure in Fig. 4, it can be seen that with increasing the content of La in Al-Mg alloy, the grain of α -Al phase becomes smaller and smaller, rounder and rounder. When the La content of is about 1.5 wt.%, the microstructure of the alloy reaches the best combination of refined microstructure and round-shaped grains. This is favorable for alloy shrinkage compensation during the late process of alloy solidification, for increasing fluidity of alloy and hence improving the casting quality, especially to form those products with complex shape and/or specific requirements.

3.3 Effects of La on extrusion microstructure

Fig. 5 shows the metallographic microstructure of Al-Mg alloys with La content at 0.5 wt.%, 1.0 wt.%, 1.5 wt.%, and 2.0 wt.% extruded at 410°C. As seen from Fig. 5, a lot of large isolated and discontinuous reticular phases were crushed into fine small granular strengthen-ing phases by extrusion. And, the α -phase grains show a flat strip shape along the extrusion direction. The internal micro-defects, micro-cracks and porosity were welded by high pressure at high temperature.

3.4 Effects of La on tensile strength

Fig. 6 shows the tensile strength of the hot-extruded Al-Mg alloys samples at 410°C with La content. In order to ensure the stability and reliability of the experimental data, all the data in Fig. 6 were the average values for 10 tests. In general, the addition of rare earth La can significantly improve the tensile strength of Al-Mg alloy. With no La addition, the tensile strength of the base alloy is only about 260 MPa. After adding rare earth La, its tensile strength is greater than 300 MPa. When the La content is about 1.5 wt.%, the tensile strength of Al-Mg alloy reaches its maximum value of 314 MPa.

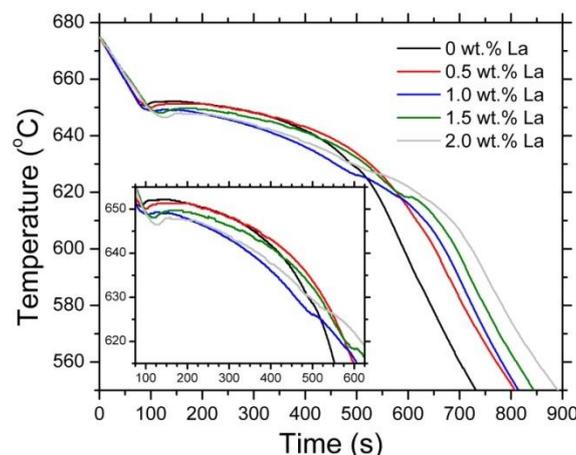
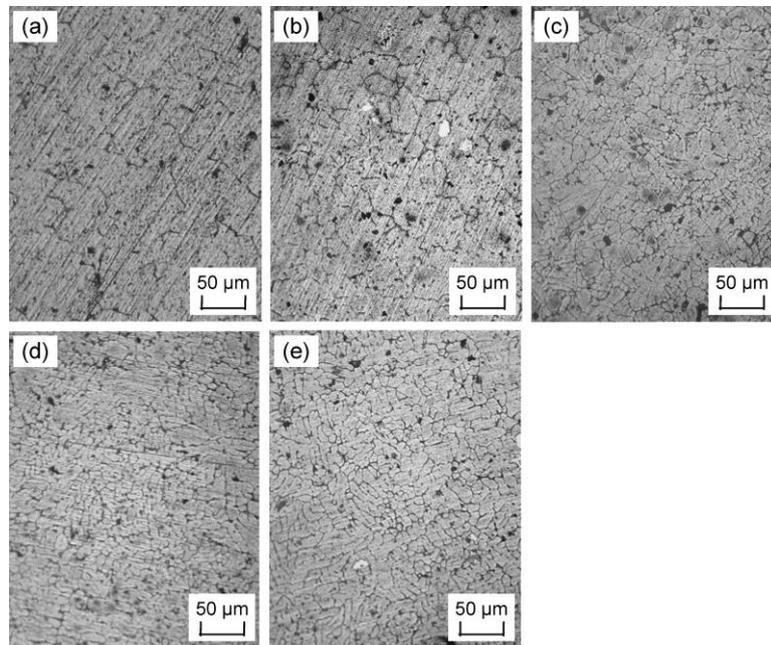
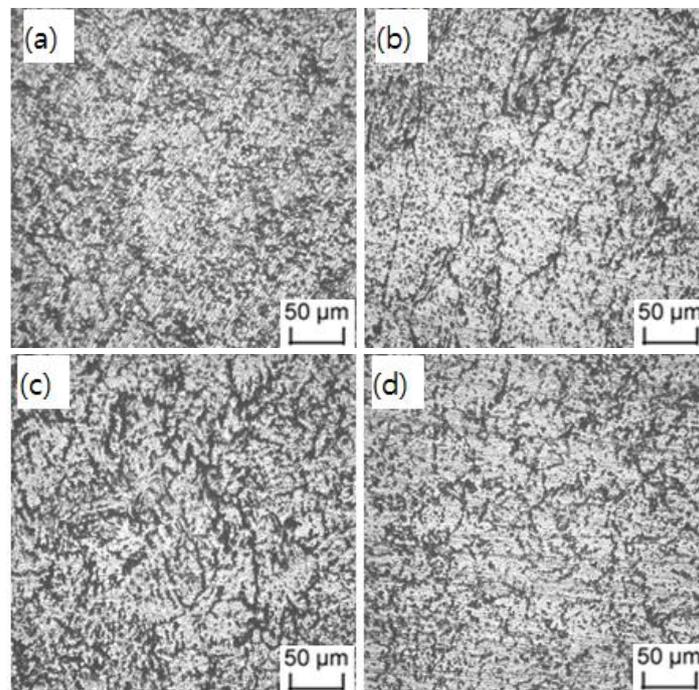


Fig. 3 Solidification curves of Al-Mg alloy with different La contents



(a) 0%, (b) 0.5 wt.%, (c) 1.0 wt.%, (d) 1.5 wt., and (e) 2.0 wt.%

Fig. 4 Optical microstructure of the cast Al-Mg alloys with different La content



(a) 0.5 wt.%, (b) 1.0 wt.%, (c) 1.5 wt., and (d) 2.0 wt.%

Fig. 5 Optical microstructure of the extruded Al-Mg alloys with different La content

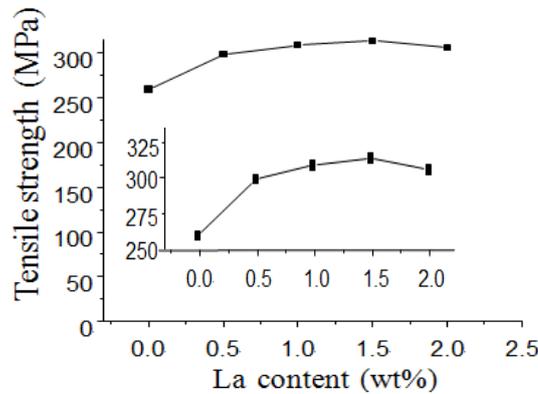


Fig. 6 Tensile strength of Al-Mg alloys with different La content

4. Discussion

The atomic radius of the rare earth La is 0.1877 nm, with a relative atomic radius of 17% compared with the element Mg (with a radius of 0.1602 nm) and electronegativity of 1.10 (Chen *et al.* 2014). The solubility of La in Al-Mg alloy is 0.5 wt.%. The rod-shaped compounds LaAl₄ is one of the main products during the alloy solidification (Gao *et al.* 2014). Because the phase LaAl₄ is a high temperature phase, it can still remain stable at 420 °C (Fan *et al.* 2005). As can be seen from Fig. 7, at the beginning, the influence of La addition in alloy on solidification temperature range is not evident before the La content in the Al-Mg alloy is less than 1.0 wt.%. This is mainly owing to that most of La is dissolved into the alloy to form solid solution phase. Hence, the addition of La on the solidification behavior of the Al-Mg alloy is not distinct. However, when the La content is enhanced up to 1.5 wt.%, the alloying of La broadens the solidification temperature range of the Al-Mg alloy. The main reason is that after the amount of dissolved the rare earth La reaches a saturation status, the atoms of La congregates at the outside of the grains. As such, the grain growth is inhibited, making the alloy solidification process slow. This could also be supported by the solidification time of abscissa in Fig. 3.

It is also shown from the cast microstructure of Al-Mg alloy in Fig. 4 that after the La content of is raised up to 1.0 wt.%, the pronounced metamorphism took place in microstructure, such as the refined grains, more grain numbers, and altered grain morphology from dendritic to round shape. All these microstructural manipulations are beneficial to improve the mechanical properties of the alloy.

In addition, compared with the dendrites, fine equiaxed crystals and rose-shaped grains will greatly increase the liquidity in the solidification process. It helps alloy feeding during alloy solidification, thus improving the resistance of castings to the thermal cracking and reducing the defects of porosity and segregation. This improves the mechanical properties of Al-Mg alloy.

The strengthening effects of rare earth La on Al-5Mg alloy can be analyzed from the following three aspects:

Firstly, from the point of view of the electronegativity properties, the difference of electronegativity between Al and La is much higher than that between Mg and La. So typically, there is a reaction preferentially between Al and La, and its generation is high melting

intermetallic strengthening phase. Strengthening phase with high melting point can be used as the nucleation core of molten metal solidification and crystallization. So it can increase the nucleation rate.

Secondly, on one hand, under rapid solidification of metal type casting crystallization, the solid solubility of rare earth La is lower in Al-Mg alloy for its larger atomic radius. On the other hand, rare earth La is easy to gather at the solidification front due to its slower diffusion. All these inhibit grain growth.

Thirdly, at the solidification front, the enrichment of rare earth La will cause the composition super-cooling, which will activate the nucleation agent of the solidification front, and increase the nucleation rate and thus refine the microstructure. La can effectively reduce the surface tension of the solid-liquid surface. It decreases the liquid metal critical nucleation radius, and refines the grain of alloy.

5. Conclusions

- As the rare earth La is added into Al-Mg alloy, their solid-liquid diphasic region is broadened. While the La content reaches about 1.5 wt.% in the alloy, two-phase region temperature of the alloy changes from 648.9°C to 618.7°C, and the solidification temperature range is 30.2°C.
- With the addition of the rare earth La, the metamorphism occurs in the microstructure of Al-Mg alloy, and the grains of alloy are refined.
- While La is alloyed, the solidification time of Al-Mg alloy will be prolonged.
- When the La content in the Al-Mg alloy is about 1.5 wt.%, the alloy achieves the best effect on the solidification range, microstructure and tensile strength with 314 MPa.

Acknowledgments

The authors are grateful to financial support from the National Natural Science Foundation of China (Grant No. 51165010) and the Natural Science Foundation of Jiangxi Province, China (Grant No. 20114BAB206018) as well as partial support from the floor plan of Jiangxi science and technology (Grant No. KJLD14066). The authors would also like to thank Dr. Laichang Zhang, School of Engineering, Edith Cowan University, Australia, and Dr. Peng Jiang, Department of Chemical Engineering, University of Florida, USA for their valuable discussions.

References

- Bai, J., Sun, Y.S., Xue, F. and Qiang, J. (2012), "Microstructures and creep properties of Mg-4Al-(1-4) La alloys produced by different casting techniques", *Mater. Sci. Eng. A.*, **552**, 472-480.
- Chen, Z.Y., Shu, Q. and Chen, Y.Y. (2007), "The study of microstructure and mechanical properties of high strength and toughness casting Al-Cu alloy", *Mater. Technol.*, **15**(5), 718-722.
- Chen, Y., Pan, Y., Lu, T., Tao, S.W. and Wu, J.L. (2014), "Effects of combinative addition of lanthanum and boron on grain refinement of Al-Si casting alloys", *Mater. Design.*, **64**, 423-426.
- Fan, C.H., Chen, G., Yan, H.G. and Chen, Z.H. (2005), "Effect of rare earth in magnesium and magnesium alloys", *Mater. Rev.*, **19**(7), 61-64.

- Gao, Y., Zhang, W., Li, B., Yuan, L. and Chen, X.N. (2012), "Effect of RE (Y) on diffusion coefficients of tungsten and molybdenum atoms", *Rare. Metal. Mat. Eng.*, **41**(12), 2154-2156.
- Gao, H.X., Zhang, X.Y., Fan, L., Huang, X. and Gong, X.P. (2014), "Effect of rare earth element La and solid solution treatment on microstructures and mechanical properties of Al-Mg alloy", *Foundry.*, **63**(3), 265-268.
- Liu, Z. and Hu, Y.M. (2008), "Effect of RE grain-refiner on microstructure of semi-solid ZL101 Al alloy", *Foundry.*, **57**(6), 596-599.
- Li, Q.F., Wang, L.B., Yu, B.Y., Wang, F. and Yu, H.P. (2006), "Effect of processing parameters on mechanical properties and hot tearing tendency of indirect squeeze casting Al-Cu based alloy", *Foundry.*, **55**(12), 1271-1275.
- Liang, Y., Wei, S.J. and Ling, M.X. (2013), "Phase equilibria of the Al-Cr-Pr ternary system at 773K", *Int. J. Mater. Res.*, **104**(12), 1233.
- Montalba, C., Ramam, K., Eskin, D.G., Ruiz-Navas, E.M. and Prat, O. (2015), "Fabrication of a novel hybrid AlMg5/SiC/PLZT metal matrix composite produced by hot extrusion", *Mater. Design.*, **69**, 213-218.
- Ma, J.L. and Wen, J.B. (2009), "The effects of lanthanum on microstructure and electrochemical properties of Al-Zn-In based sacrificial anode alloys", *Corros. Sci.*, **51**, 2115-2119.
- Su, L.H., Lu, C., He, L.Z., Zhang, L.C., Guagliardo, P., Tieu, A.K., Samarin, S.N., Williams, J.F. and Li, H.J. (2012), "Study of vacancy-type defects by positron annihilation in ultrafine-grained aluminum severely deformed at room and cryogenic temperature", *Acta. Mater.*, **60**(10), 4218-4228.
- Si, N.C., Wu, Q. and Li, G.Q. (2006), "Effect of Ti on thermal fatigue property of Al-4.5%Cu alloy", *Foundry.*, **55**(7), 731-734.
- Xie, S.K., Yi, R.X., Deng, X.C. and Ai, Y.P. (2010), "Preparation process for semi-solid slurry of Al-4.5Cu alloy", *Hot. Work. Technol.*, **39**(5), 21-23.
- Yu, P., Zhang, L.C., Zhang, W.Y., Das, J., Kim, K.B. and Eckert, J. (2007), "Interfacial reaction during the fabrication of Ni60Nb40 metallic glass particles-reinforced Al based MMCs", *Mater. Sci. Eng. A.*, **444**(1-2), 206-213.
- Yi, R.X., Xie, S.K., Zheng, X.Q. and Ai, Y.P. (2009), "Effect of RE on performance of semi-solid AlSi7 alloy", *Key. Eng. Mater.*, **426**(1), 581-584.
- Yi, R.X., Xie, S.K., Wu, W.T., Zheng, X.Q. and Pan, X.L. (2010), "Influence of vibration on microstructure of the aluminum-copper alloy of rare earth Ce", *Ord. Mater. Sci. Eng.*, **33**(1), 75-77.
- Zhang, W.Q., Li, A.J., Chen, H.Y., Lan, B.Y., Pan, K., Zhang, T.R., Fang, M.X., Liu, S.Y. and Zhang, W. (2012), "The effect of rare earth metals on the microstructure and electrochemical corrosion behavior of lead calcium grid alloys in sulfuric acid solution", *J. Power. Sources.*, **203**, 145-152.