Light Metals 2014

LIGHT-METAL MATRIX (NANO)-COMPOSITES

Microstructure-Property Relationships II: Modeling and Advanced Characterization

SESSION CHAIR

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UNDERSTANDING THE ROLE OF NANODISPERSIONS ON THE PROPERTIES OF A390 HYPER-EUTECTIC Al-Si CAST ALLOY

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Keywords: Nanodispersion, semisolid processing, hypereutectic Al-Si cast alloys, microstructure and strength

Abstract

In this work a series of castings of hypereutectic aluminium-silicon samples (A390) were cast from different pouring temperatures (liquid, liquid-solid, and solid-liquid temperatures) with and without adding Al_2O_3 nanoparticles in liquid and semisolid states, with mechanical stirring. The microstructure features and the tensile strength properties were evaluated and analyzed. The results obtained in this work show that the introduction of Al_2O_3 nanodispersions together with the stirring effect induces a refining role on the Si particles associated with an increase in the tensile strength and ductility of the alloy. This work also illustrates the significance of optimizing type and amount of nanoparticles, addition temperature and pouring temperature to maximize the effect obtained by these parameters.

Introduction

Hypereutectic Al-Si alloys are used in various automotive and aerospace industries due to some outstanding properties that they possess, including high castability, abrasion resistance and excellent strength to weight ratio. Despite those characteristics, the mechanical properties of the hypereutectic Al-Si alloys are remarkably inferior because of the presence of primary silicon particles, which crystallize, in coarse and irregular shapes. These alloys solidify over a wide temperature range, during which the primary silicon forms and grows to large particles. The size and shapes (morphology) of the Primary Si Particles (PSPs) has been identified to be the main cause for fatigue failure and wear of engine parts made for hyper-eutectic Al-Si alloys [1].

The mechanical properties of hypereutectic Al-Si alloys are highly affected by the morphology, size, and distribution of both PSPs and eutectic silicon. The morphology of silicon particles are dependent on the solidification rate; as under normal casting conditions PSPs are very coarse and show star-like and other irregular shapes. Therefore in order to improve the mechanical properties of the hypereutectic Al-Si alloys, size; distribution; and the morphology of PSPs and eutectic silicon should be controlled. The most important known technique for modification of the microstructure of the hyper eutectic Al-Si alloy (A390) is to add phosphorous (P) as a refining agent. Gruzleski et al. [2] discussed this technique commonly used in the foundry, and showed that refinement of the primary silicon is achieved by the addition of phosphorous to the melt, though no effect was observed on the eutectic silicon, as both coarse and unmodified eutectic silicon were still present, after adding P, surrounding the refined primary

silicon. Though sodium (Na) and strontium (Sr) are used to achieve refinement of the eutectic silicon, unfortunately a combined effect does not happen at the same time due to chemical incompatibility of phosphorous with the other modifying chemicals such as strontium and sodium; this has been explained by Gruzleski et al. [2] to result from the formation of strontium phosphide or sodium phosphide upon the addition of strontium or sodium to phosphorous pre-refined alloys. A simultaneous effect of refining primary silicon and modifying eutectic silicon would be very beneficial for the properties of the hypereutectic Al-Si alloys.

Zhao and Wu [3] prepared cast A390 in the semi-solid state and studied its cast and T6 heat treated condition where they found that the semi-solid casting results fine and uniform microstructure where the average size of primary Si spheroids was found to be 20-30 μm, combined with improved tensile strength, ductility, hardness and wear resistance. Tebib et al. [4] have developed a novel rheo-forming process to investigate semi-solid processing of hypereutectic A390 alloys using a combination of the swirl enthalpy equilibration device and isothermal holding, they managed to improve the process-ability of semi-solid slurries for A390 alloy and they reported evidence of enhancement of the microstructure. They have also shown that Primary silicon in the semi solid microstructure can be refined by phosphorous additions, but refinement of primary silicon and modification of eutectic silicon cannot be achieved simultaneously by phosphorous and strontium additions.   Recently, Rosso [5] presented a review on routes and properties of ceramic reinforced metal composites where it was shown that adding reinforcing particles to the liquid phase in casting processes usually results in poor properties obviously due to the defects arising from high melting temperatures at which the reinforcement is usually added. The advantages associated with using rheo-casting was also described from which it could be concluded that this method exhibits several advantages over the first method such as lower porosity content, good distribution of reinforcements, and less harmful interfacial reactions. Moreover, the enhanced viscosity of the semi-solid processing would serve to improve the ceramic particle/melt wettability and entrap or capture the reinforcement material physically. It is difficult to obtain uniform dispersion of the ceramic particles in liquid metals due to poor wettability in the metal matrix, and the large surface-to-volume ratio of the added particles. These problems induce agglomeration and clustering, as presented by Rosso [7]. Evidence of significant enhancement in strength and other properties of hyper-eutectic Al-Si cast alloys by incorporating nanoparticles have been recently presented by the author and others [6,7]. However, the combined effect of semi-

solid parameters and nanodispersion need further elaboration. Hence, the aim of this work is to study the combined effect of semi-solid casting temperature and nanodispersion addition.

Experimental Procedure

A series of six cast hypereutectic Al Si alloy based on A390 (the analysis of which is given in Table (1) were prepared for this work. The samples were prepared in an electrical resistance furnace that was designed and constructed for this research work for preparing the nanodispersed alloys. It consists of a lift out graphite crucible of max. 8 kg capacity and is operated by a heating system equipped with a control unit with a thermocouple for controlling the temperature up to 1200 ^{0}C. An external stirring mechanism of variable height and driven by a motor of 3000 rpm was attached to the system and allowed to the melt through an opening in the top of the furnace. The stirrer was made of a stainless steel rod with four pitched blades weld to its lower end. The design of the impeller was to promote axial flow. A metallic mould made of cast iron with ten cylindrical mould cavities of diameter 20 mm and 150 mm height was used for casting the test specimens. The materials used for reinforcement were Al_2O_3 ceramic nanoparticles (prepared at the Central Metallurgical Research and Development Institute, CMRDI) with average particle size of 50 nm, the description of which is given in reference [7].

Table 1: Chemical composition (in wt. %) of the base alloy.

Chemical composition (in wt. %)							
Alloy	Si	Mg	Fe	Cu	Ti	Zn	Mn	Al
A390	17	0.45	0.5	4.5	0.2	0.1	0.1	Bal.

A charge of 3Kg of the base alloy was introduced to the crucible and heated up to above the melting temperature (730 ^{0}C). After reaching the liquid state the melt was degassed with either argon or hexacholorethane degasser tablet, to get rid of gases. After degassing the melt was brought down to a specific temperature, for each casting; 610 ^{0}C, 640 ^{0}C and 660 ^{0}C, respectively and poured. 1 and 2% Al_2O_3 nanoparticles were prepared in packages of aluminium foil and preheated to about 600 ^{0}C, added once at at 700 ^{0}C and another at 660 ^{0}C, again the melt was brought down to 660 ^{0}C, 640 ^{0}C, and 620 ^{0}C respectively and poured. The packets were added to the melt through the opening in the top of the furnace one packet after the other, simultaneously with mechanical stirring for 1 min at 1000 rpm. Table (2) summarizes the fabrication conditions of the composites prepared in this investigation. Cast samples were poured in the prepared mould without additions and with additions of the different investigated percentage additions. The prepared cast samples were hence used for further investigation.

The microstructure examination was carried out using optical metallurgical microscope OLYMPUS BX41M LED equipped with a high resolution digital camera for investigating the microstructure. The morphology of the primary phases and the eutectic structure was further investigated using a JSM-7000F FEG-SEM using back scattered electrons (BSE) technique. A sample was prepared by grinding followed by polishing and micro-polishing using a vibration-polishing machine. Some selected samples for SEM examination were prepared by ion polishing using argon ions using a Jeol SM09010 cross-section polisher. The tensile test was done on an electronic controlled universal testing machine AMSLER type.

Table 2: Manufacturing conditions and tensile properties of the investigated samples

Samples Group Number	Condition
1	Poured at 610 ^{0}C
2	Poured at 640 ^{0}C
3	Poured at 660 ^{0}C
4	2% Nano- Al_2O_3 added at 700 and poured at 660 ^{0}C
5	2%Nano- Al_2O_3 added at 700 and poured at 640 ^{0}C
6	1%Nano- Al_2O_3 added at 660 and poured at 620 ^{0}C

Results and Discussion

The data obtained in this work are shown in Fig. (1), from which it can be seen that the highest tensile strength for the monolithic alloy was achieved at pouring temperature of 660 ^{0}C which is exactly the solidus temperature, whereas pouring at lower temperatures in the semi-solid region produced less enhancement effects on the tensile strength, though it led to refining of the Si particles. It is also seen from Fig. (1) that the addition of the Al_2O_3 nanoparticles at liquid temperatures (700 ^{0}C) led to an enhancement in average tensile strength of the castings mounting to about 4% when pouring from the semi-solid temperature at 640 ^{0}C. Whereas, adding the nanoparticles at the semisolid stage didn't produce a similar enhancement effect on the tensile strength. The combined strengthening effect with increase in ductility suggests a microstructural refinement effect.
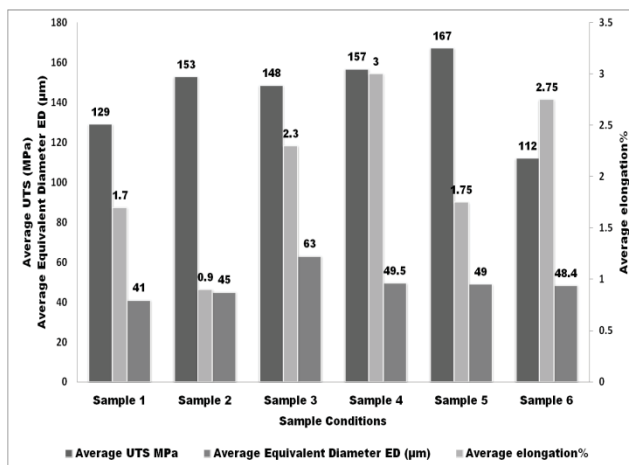

Fig. (1) UTS, elongation% and average equivalent diameter of investigated samples

Figures (2 and 3) show a comparison between the microstructures of the two samples: monolithic poured at 660 ^{0}C without Al_2O_3 nano-additions, and poured at 660 ^{0}C with Al_2O_3 nano-additions, respectively. It is seen from the microstructure graphs in Figs. 2 and 3 that the microstructure is the typical microstructure of hyper eutectic Al-Si alloy, which consists of primary silicon particles, Al-Si eutectic lamellae and aluminium dendrites. The primary silicon particles have a variety of shapes (cubic, polygonal,

elongated and star like flakes). The Al-Si eutectic lamellae presume a needle like structure in all figures which is the typical structure of unmodified Al-Si eutectic phase. The figures also show that the alloys treated with Al_2O_3 tend to show a modification in the shape of the Si particles where they tend to be polygonal shape, unlike the untreated alloy which showed presence of star like and elongated Si particles. It can be seen that the addition of the nanoparticles at 700 ^{0}C did not result significant refining of the Si particles as measured by their equivalent diameter (average perimeter of particles/π).

Also, the monolithic sample poured from 660 ^{0}C showed the occurrence of dendritic and nondendritic Al phase (Fig. 2.b). These nondendritic structures changed to clustered Al phase (Fig. 3.a) and equiaxed structures (Fig. 3.b) after adding the nanoparticles.
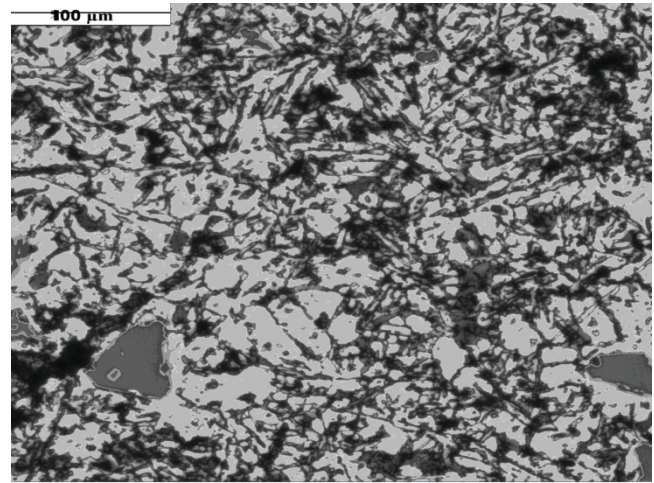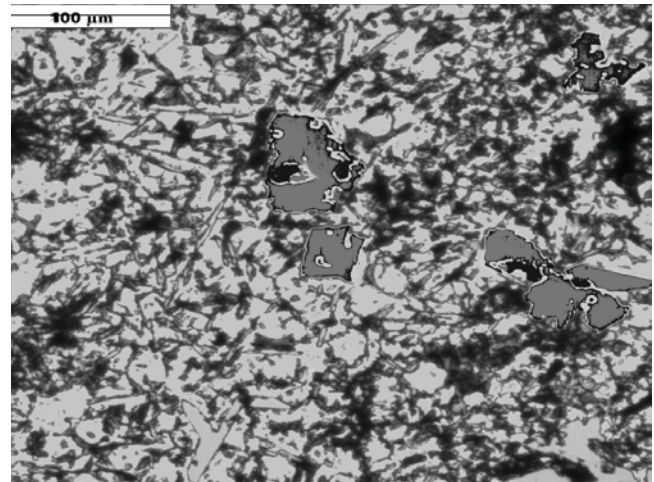

a)
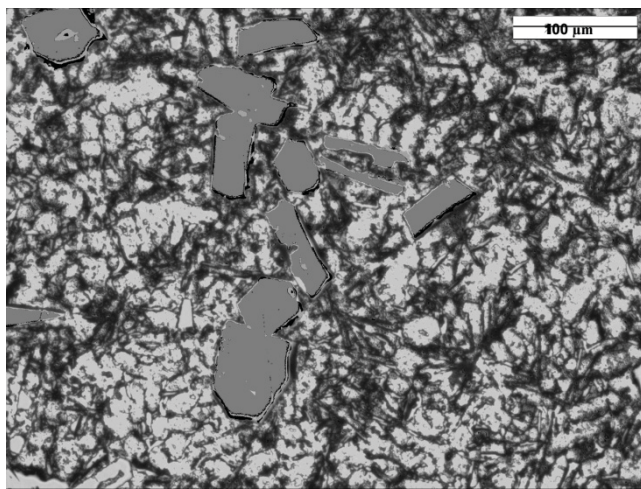
a)
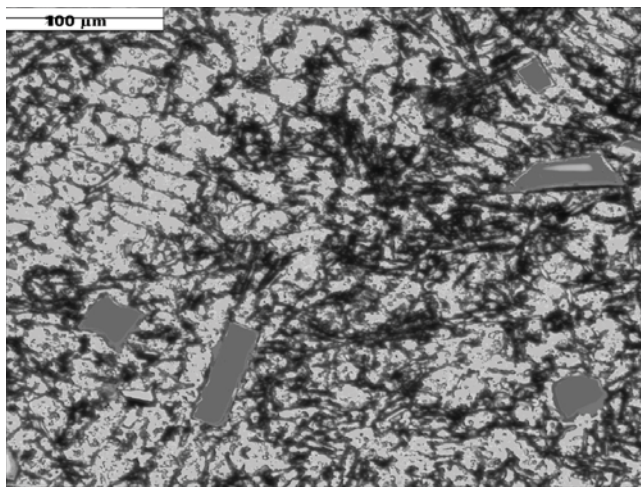
b)

Fig. (2) Sample poured at 660 ^{0}C without nano-additions.

b)

Fig. (3) Sample poured at 660 ^{0}C with 2% Al_2O_3 nano-additions made at 700 ^{0}C.

Figure 4 shows, alternatively the microstructure obtained for the condition where nano-additions were made at 660 ^{0}C and poured at 620 ^{0}C, from which it can be seen that the Si particles have become more refined and polygonal shaped. The complete absence of the dendritic Al structure is also seen from Fig. 4. However, it is noticed from Figs. 3 and 4 that the addition of nanoparticles promotes the formation of α- Al phase. This has been also observed and recorded before [6] and has been attributed to the improved nucleation of Si particles and depletion of Si element near the Si particles [6]. The observed refinement in primary Si particles and modification in eutectic Si after adding nanoparticles has been reported before [6]. However, the authors [6] have attributed this effect to heterogeneous nucleation initiated by the γ- Al_2O_3 nanoparticles. The effect on modifying the eutectic Si has been attributed to result from the effect of the pushed γ- Al_2O_3, during solidification into the remaining melt, on restricting growth of the particles.
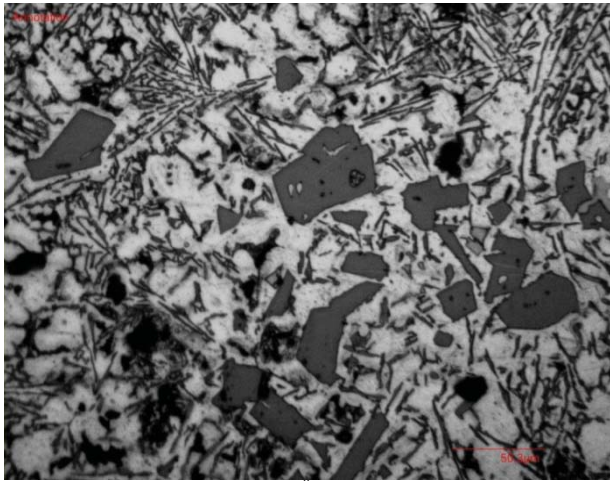
Fig. (4) Sample poured at 620 ^{0}C with 1% Al_2O_3 nano-additions made at 660 ^{0}C.

Figure (5) shows a typical BSE image obtained for the alloy containing 1% Al_2O_3 cast in semi-solid state after ion polishing. The EDX analysis of the various micro-constituents and precipitates were identified by BSE as shown in the figures confirming the presence of: primary silicon phase, α-Al dendrites, and the eutectic structure mainly consisting of binary Al-Si eutectic. The figure also demonstrate the presence of the traditional intermetallic compounds that form in hypereutectic Al-Si alloys that are likely to be; $CuAl_2$, $Cu_2Mg_8Si_6Al_5$ Al5FeSi.

Fig. (5) BSE image of 1% Al_2O_3 cast in semi-solid state showing the main microstructure constituents appearing in the as-cast state.

The previous results show that both size and morphology of the Si particles affect the strength of the hypereutectic Al-Si alloy. The size and morphology of the primary silicon is controlled by increasing the number of nuclei through increasing the frequency of nucleation, accordingly, the beneficial effect of γ- Al_2O_3 nanoparticles on the refinement of primary Si [6] is explained. The increase in the frequency of nucleation may result either from the provision of nucleation sites or increased subcooling. However, the role of cooling rate which is a function of pouring temperature (among many other parameters) is shown to have an effect on the overall structure of the alloy as well as the size and the morphology of the Si particles. It can be seen from figures (2) and (3) that pouring at the solidus temperature 660 ^{0}C

has promoted the formation of crystalline α- Al nondendritic and dendritic shaped structures. Pouring at lower temperatures (Fig. 4 (640 ^{0}C) has lessened the appearance of those α-Al nondendritic and dendritic shaped structures. The strength of the hypereutectic Al-Si alloy depends strongly on the morphology of the Si particles and on the shape of the α-Al, as well as the morphology of the eutectic Si. It has been stated [6] that the cracks easily initiate inside the brittle primary Si or eutectic Si, and then propagate through boundaries with α-Al phase. Thus the enhancement in ductility associated with pouring at 660 ^{0}C with nanoparticles addition may be attributed to the change in the Si morphology (according to restricted nucleation and growth theory of modification), as well as to the crystalline appearance of the α-Al phase. The enhancement in the elongation % may be explained by the Si refining as well as the crystalline shapes of the α-Al phase which has been promoted by pouring from the solidus temperature.

It is known that Solidification of hypereutectic Al-Si begins with the precipitation of primary silicon from the melt as temperature falls under the liquidus [8]. Casting at lower semi-solid temperatures promote conditions for short solidification times leading to fine Si particles due to restricting their growth, but the refined particles may be subject to agglomeration. Casting at higher near solidus temperatures promote conditions favourable for the solidification of the α- Al nondendritic and dendritic phase whilst not restricting the growth of the Si particles. The data obtained in this work has shown that adding the nanoparticles at the higher liquidus temperatures (700 ^{0}C) has resulted enhancement in the tensile strength relative to the condition when the nanoparticles were added at 660 ^{0}C. However, the effect of nanoparticles addition on refining the Si particles was more evident in the case when nano-additions were made at 660 ^{0}C compared to higher (700 ^{0}C) or lower (640 ^{0}C) temperatures. The merit of using controlled thermal-rate treatment and low temperature pouring has been described and discussed [9] and it has been shown that the control of the pouring rate temperature results significant enhancement in the tensile properties of Al-15% Si alloy that has been attributed to the size and morphology of the Si particles.

Though great efforts are done by many researchers to control casting parameters of hyper-eutectic Al-Si alloys to refine the size of the primary Si particles, and modify the eutectic Si, achieving both effects, simultaneously, is only possible by the proper control of melting and pouring temperatures and cooling rates. The significant refining effect of P on the primary Si by forming AlP, which acts as a nucleation site for the primary Si, has been shown to be impotent in achieving a simultaneous refining and modifying effect. [2]. Recently, adding nanoparticles to the melt has proven in this work as well as others [6, 7] to result significant refining effect and improvement in mechanical properties and service performance. The occurrence of more than one acting mechanism during the solidification of the alloy (Si precipitation, eutectic solidification, and α-Al formation) give rise to counteracting effects. To optimise the effect of all acting mechanisms it may be suggested (according to the results of this work) to add the nanoparticles at the liquid temperature and pour at near solidus temperature. The effect of the semi-solid processing on the solidification and microstructure of hyper-eutectic Al-Si alloy was investigated [8] and it was found that structure improvement for the rheo-cast hyper-eutectic Al-Si alloy occurs for fraction of solid less than 0.05. However, the

challenges facing the techniques adopting refining the structure of hypereutectic Al-Si alloys still remain to be nanoparticles agglomeration and increase in porosity associated with adding nanoparticles and mechanical stirring.

The role of nanoparticles in refining the Si particles could be attributed to increasing the nucleation sites [6] or inducing a thermal sub-cooling effect. Though this work aimed at identifying and understanding the real effective mechanism, the results obtained till now by the authors [7] including this work suggest the likelihood of a thermal sub-cooling effect, this will be further investigated in the next stage of this work. This belief is based on the following:
1) The primary Si refining effect of P, which has been attributed to the formation of AlP which causes refinement of the primary Si particles, leaves the eutectic Si unmodified [2],
2) The refining effect of the nanoparticles becomes more pronounced when the addition is made at lower temperatures (660 ^{0}C) than above liquidus temperatures (700 ^{0}C) (according to this work and others [8]), may dispute the suggestion that the nanoparticles act as heterogeneous nucleation sites,
3) The effect of nanoparticles on refining and modifying both primary Si particles and the eutectic Si simultaneously suggest that the nanodispersions result an effect extending to all existing solidification mechanisms that occur during the cooling of this alloy.
4) Simultaneous refining and modifying effects have been reported to occur by other techniques than adding modifiers or nanodispersions, as ultrasonic and thermal treatment methods have been reported to refine the primary Si particles and modify the eutectic Si [8-10].

Conclusions

This work aimed at providing more experimental data which would help in understanding the role of nanodispersions in refining and modifying the structures of hyper-eutectic Al-Si cast alloys. The availability of such data would create a platform for researchers and industry to optimise the manufacturing conditions for producing a nanodispersed Al-Si alloy with higher strength values and homogeneous structures. This work has shown that adding Al_2O_3 nanoparticles at 660^{0}C with mechanical stirring and pouring at 640^{0}C provides the optimum manufacturing conditions for this alloy.

Acknowledgement

References

1. H. Ye, "An Overview of the Development of Al-Si-Alloy Based Material for Engine Applications", *Journal of Materials Engineering and Performance JMEPEG*, 12 (2003), 288-297.
2. J. E Gruzleski. and B. M. Closset, "The treatment of Liquid Alumnuim-Silicon Alloys", *AFS* Des Plaines Illinois, (1990), 107-126.
3. J. ZHAO, S. Wu, "Microstructure and mechanical properties of rheo-diecasted A390 alloy", *Trans. Nonferrous Met. Soc. China,* 20 (2010), 754-757.
4. M. Tebib, J. B. Morin, F. Ajersch, X. Chen, " Semi-solid processing of hypereutectic A390 alloys using novel rheoforming process", *Transactions of Nonferrous Metals Society of China*, 20 (2010), 1743-1748.
5. M. Rosso, Ceramic and metal matrix composites, Routes and properties, *Journal of Materials Processing Technology,* 175 (2006), 364–375.
6. H. Choi, H. Konishi, X. Li, " Al_2O_3 nanoparticles induced simultaneous refinement and modification of primary and eutectic Si particles in hypereutectic Al–20Si alloy", *Materials Science and Engineering, A* 541 (2012), 159–165.
7. I. El Mahallawi, Y. Shash, R. M. Rashad, M.H. Abdelaziz, J. Mayer, A. Schwedt, " Hardness and wear behaviour of semi-solid cast A390 alloy reinforced with Al_2O_3 and TiO2 nanoparticles", accepted for publication in *AJSE*, May (2013).
8. M. Ramadan and N.. Fathy, "Microstructure of Hyper-Eutectic Al–Si alloy Processed in Semi-Solid State", *International Journal of Research in Engineering and Technology (IJRET)*, Vol. 1 No. 4 (2012), 238-241.
9. Q. Wang, S. Zhang, Z. Zhang, X. Yan, and H. Geng, "Study of Melt Thermal-Rate Treatment and Low- Temperature Pouring on Al-15% Si Alloy", *JOM*, Vol 65 No. 8 (2013), 958- 966.
10. W. Khalifa, S. El-Hadad, and Y. Tsunekawa, Microstructure and Wear Behaviour of Solidification Sonoprocessed B390 Hypereutectic Al-Si Alloy, *Metallurgical and Materials Transactions A,* published online 16 October (2013).