Al-Si Grain Refinement in Semisolid Processes

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The effect of inoculation has been studied on the microstructural evolution in conventional and semisolid-metal (SSM) processing of Al-6%Si alloy. It has been found that small additions of Ti-6Al-4V shift the liquids temperature up and the recalescence decreases. The nucleation event takes place at higher temperatures. Furthermore, the rate at which growth temperature increases is less than that of nucleation temperature and therefore more nuclei form with less potential for growth. In SSM processing refiner increases the α-Al percentage and reduces globule size. Improvement of primary particles' sphericity and globule size reduction are the main advantages of refiner addition.

Keywords: Al-6%Si Alloy; Semisolid Process; Stirring; Grain Refinement; Morphology

Grain refinement is a common way of achieving a proper, uniform and fine grain structure in cast aluminum alloys, since metals and alloys usually solidify with a coarse columnar grain structure under normal casting conditions [1]. The percentage of each zone depends on a multitude of parameters including pouring temperature, cooling rate, temperature gradient in the liquid and the mold material which also influences thermal gradient established within the molten alloy [2]. Grain size and morphology for equiaxed, columnar and dendrites grains are the major metallurgical factors which affect mechanical properties especially in tensile strength of metal products. The integrity of hypoeutectic Al-Si cast products is dependent on the fraction, size and morphology of primary α-Al and this is why in foundry operations a close control over the α-Al formation is of great importance. The quality of the casting is improved by reducing the α-Al grain size, and manipulating its morphology, for example refinement process [3-5]. In the first method, a fine dendritic structure is formed in the casting due to rapid cooling where large undercooling may be achieved before solidification can start. This in turn affects the size of critical nuclei, smaller critical nuclei size, and thus increasing the effective number of nuclei to eventually rendering fine grained structures. In the second method, refinement is accomplished by some means of breaking up newly formed dendrites in the semisolid state. By far the most convenient method for the control of grain size is to introduce particles into the melt which nucleate new crystals during solidification as there are certain restrictions due to mechanics of casting for the two former methods. Several works discuss the mechanisms of grain refining in alloys including Al-Si alloys. The most popular alloy is Al-6wt. %Si. These alloys are featured with excellent casting characteristics, weldability, and pressure tightness. The need for producing near net shape castings with superior properties to that cast conventionally has drawn attentions to new processes such as semisolid-metal (SSM) processing. In this field and amongst different alloying systems, Al-Si alloys have great attention due to their superior formability, better mechanical properties, and higher strength to weight ratio, relative lower melting point, and wider α-Al solidification range. For SSM processes however, the size, distribution and morphology of α-Al particles are quite important since they control die filling and thus the ability to produce thin wall castings. Researches show that, with the rounder and smaller the particles the smoother is die filling and also lesser the segregation of liquid and formation of other defects [6-9]. In the case of refining, according to literatures, there are few works reported on the direct incorporation of refiners during the processing and preparation of SSM slurries and most of the works are related to thixocasting process [10-12]. This work is focused of on the effects of inoculation on the micro-structural evolution of Al-6%Si alloy in two different processes, conventional and rheocasting, using semisolid mechanical stirring.

Experimental part

The objective of these experiments was to study the evolution of as-cast structure due to grain refiner addition in both conventional and semisolid processes. The base alloy with the composition given in table 1 was melted in a resistance furnace. For refining purpose alloy in the form of rod was used. Additions were made at 630-660°C preheated graphite crucibles. The melt is degassed by argon and rested for approximately 20 min before sampling. Two minutes before sampling, the melt was stirred.

For conventional method, for example, static condition, graphite cups were used. The prepared charge was poured in graphite cups of 25 mm inner diameter and 5 mm wall

Table 1
CHEMICAL ANALYSIS OF THE BASE ALLOY

Material	Weight %									
	Si	Cu	Zn	Fe	Mg	Ti	Mn	Ni	Sn	Pb
Before Ti-6Al-4V addition	6.00	1.45	1.54	0.78	0.15	0.10	0.10	0.02	0.001	0.001

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thickness. Cups were held in the melt for approximately 1 min before sampling to ensure uniform temperature distribution across the graphite cups at the beginning of solidification. Each cup with about 50 g of alloy was transferred to testing station and two thermocouples were quickly immersed into the melt near the cup center and wall at 10 mm from bottom. Temperature readings were collected by a data logger system with 5 readings per second sampling rate. For these series of experiments, cooling rates were between 1.5 and 2°Cs^{-1} above liquids temperature. The analysis of thermal data was carried out based on analysis of the Al-6%Si solidification temperature. For clarity of the definitions used for thermal data, they are defined and illustrated on an actual cooling curve in figure 1:

$T_{nuc\ Al}$: Start of primary α-Al dendrites nucleation temperature.

$T_{min\ Al}$: Unsteady state growth temperature, the temperature beyond which the newly nucleated crystals grow to such extent that the latent heat liberated surpasses the heat extracted from the sample.

$T_{g\ Al}$: Recalescence of steady state growth due to release of latent heat of primary α-Al dendrites.

ΔT_{Rec}: Temperature difference between unsteady ($T_{min\ Al}$) and steady ($T_{g\ Al}$) state growth temperatures of primary α-Al particles.

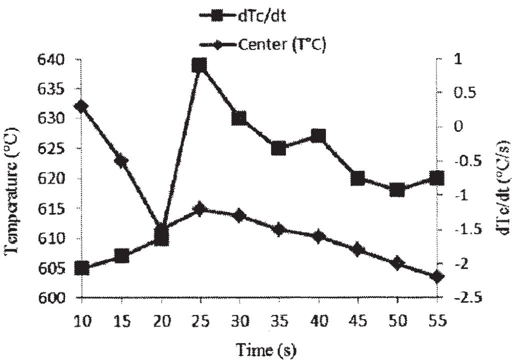

Fig. 1. Cooling curve which shows the solidification in the alloy

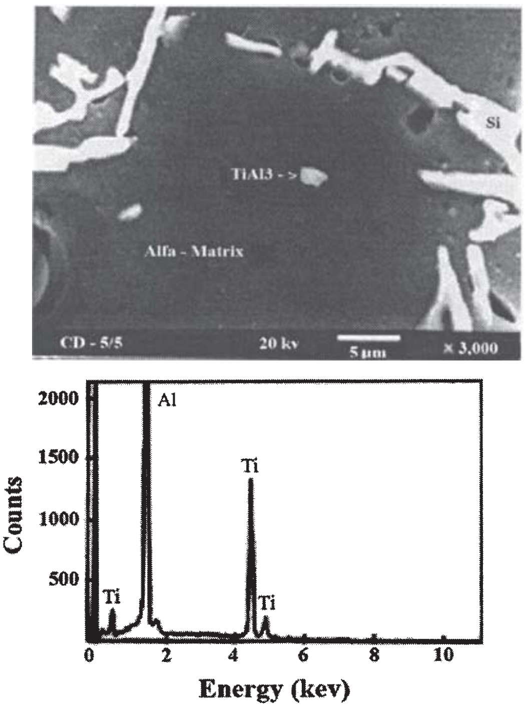

Fig. 2. a) Microstructure of as-cast Al-6% Si alloy refined by Ti-6Al-4V master alloy by scanning electron microscope (SEM). Microstructure consists of aluminum matrix and silicon secondary phase. b) SEM/EDX micro-analyzing shows that TiAl3 compound has been formed

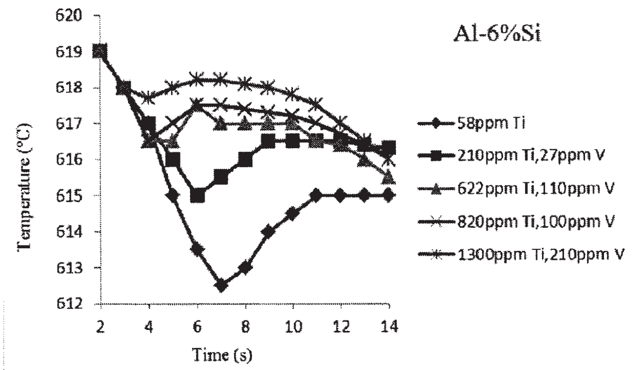

Fig. 3. The initial section of cooling curves for the conventionally cast specimens (central thermocouple)

About 2 kg of the alloy was poured at 660°C into a refractory coated steel mold of about 75mm diameter and 250 mm long. The stirrer motion was 400-1000 rpm mold. After that, the bottom closure of the mold was opened to drain the remaining liquid for a specific period of time. The overall casting procedure did not exceed up to 90s. In all experiments, a 0.8 mm diameter thermocouple was inserted in the mold center. For the purpose of studying the structural evolution, samples were sectioned transversely. Optical microscopy and image analysis were used to characterize the microstructure including grain size, primary α-Al particle size, average circular diameter, number density, sphericity, and area to perimeter ratio, *A/P*; which is proportional to inverse of surface area per unit volume, S_V. The specimen preparation procedure has great implications on the quantitative analysis of the resulting microstructure since proper polishing and etching enhance the contrast between constituents and lead to better detection and identification of different phases. For the reason of having a representative structure, all the data were obtained from image processing of the resulting microstructure between the center and the wall surface of the billet.

Results and discussions

Figure 2(a) shows microstructure after nucleation studied by scanning electron microscope (SEM). Microstructure consists of aluminum matrix and silicon secondary phase. SEM/EDX micro-analyzing in figure 2 (b) shows that $TiAl_3$ compound has been formed. Metastable aluminide phase $TiAl_3$ is formed prior to solidification of Al-Si alloy. Due to its better lattice compatibility with solid aluminum, makes it an efficient nucleation site for aluminum grains upon solidification. The $TiAl_3$ grains dissolve upon solidification of aluminum and most probably responsible for the enhanced nucleation of aluminum grains. The grains are refined in the presence of solute titanium in aluminum alloy. The influence of titanium diffusion, latent heat, and cooling rate on the growth behavior of individual aluminum grains during the phase transformation needed further study.

The effect of Ti-6Al-4V grain refiner addition to the Al-6%Si alloy on the early stages of solidification is illustrated in figure 3 for conventionally cast specimens. The results are from the thermocouple at the center of the graphite cups.

As it is obvious, small additions of Ti-6Al-4V shifted the cooling curves up and to the left with the recalescence decreasing with increasing master alloy content. The nucleation event takes place at higher temperatures and shorter times after pouring. As reported by other researchers, an effective grain refiner should not show any undercooling before the actual growth temperature and therefore the smaller the recalescence, the more effective the grain refiner. In these series of experiments, it is clearly shown that the grain refiner does not remove recalescence completely even at its optimum percentage. Therefore, it could not be rated as a perfect grain refiner for this alloy. With addition of grain refiner, both nucleation and growth temperatures could be increased. The increasing nucleation temperature allows new crystals to form ahead of solidification front, rendering an equiaxed fine grained as-cast structure. Furthermore, the rate at which growth temperature increases is less than nucleation temperature rise. In other words, there are more nuclei with less potential for growth and thus effective grain refinement should be expected. Further addition may reduce grain size even further, but equally may form Ti-based intermetallics which are detrimental to the mechanical properties of the as-cast products as discussed later. Lowering the recalescence has a great influence on growth of primary α-Al particles. Minimum temperature at the beginning of solidification, T_{minAl}, shows the temperature where the rate of latent heat liberation is balanced out with the heat extracted from the sample. In untreated alloy, the existence of recalescence means the heat generated with the commencement of solidification could not be transferred out of the mold completely and therefore the heat balance leads to the appearance of recalescence. However this is not the case for the refined alloy. In refined alloy, nucleation temperature increases and therefore in contrast to the untreated alloy, there are more primaries within the same time interval for the refined alloy. These solid α-Al particles can serve as heat sinks to absorb the heat released from surrounding liquid and therefore leads to lower recalescence compared to the untreated alloy. There could be a second hypothesis to justify the disappearance of recalescence with grain refinement and that is attributed to the lower growth rate in the refined alloys which results in lesser heat being evolved to balance out the heat extracted from the solidifying alloy. Figure 4(a) shows microstructure of Al-Si before refinement. The distribution of the Si phase in the Al-Si alloy not refined by master alloy was homogeneous and no aggregation was found over the section of the samples. The Si phase has mostly coarse crystal. The size of the Si phase was about 10μm. It can be seen that the Al phase was developed obviously into coarse crystal with the average length of up to 35-40μm. The needle-plate like eutectic silicon is distributed arbitrarily in the Al matrix. Figure 4 (b) shows tensile curve of this alloy at the same condition. The maximum strength is about 430MPa and fracture occurred at the elastic limit with 8.9% elongation.

The results of the grain refinement of the Ti-6Al-4V master alloy are presented in table 2. This is related to Average grain size, tensile strength and % elongation of as-cast Al-6%Si refined by mesh 50. Weight percentage of the inoculants varied between of 0.1 to 0.25. Grain size varies between of 6 to 20 μm and the minimum value is related to the 0.15wt%. Tensile strength varies between of 286 to 492 and the maximum value is related to 0.15wt%. The maximum values for elongation obtained with 0.15 -2.5 wt% of grain refiner and is about of 10.2 μm. Figure 4 (a) shows microstructure obtained by mesh 50 with 0.15wt%. Average grain size is about 6 μm and silicon length was reduced to about 11μm which distributed homogeneously in the section of the specimen. As compared with the microstructures without refinement, the large Si phase disappeared, and the size of the Si phase decreased obviously and was about few micrometers. The morphology of the eutectic silicon is changed from needle-plate to a fibrous form, which is under modification. Figure 4 (b) shows that tensile strength and elongation is 492 MPa and 7.25%, respectively.

The results of the grain refinement of the Ti-6Al-4V master alloy with the mesh 140 are presented in table 3. Weight percentage of the inoculants varied from 0.1 to 0.25. Grain size varies from 10 to 20 μm and the minimum value is related to the 0.15wt%. Tensile strength varies between of 384 to 540 and the maximum value is related to 0.15wt%. The maximum values for elongation obtained with 0.1-0.15 wt% of grain refiner and is about of 9.4 μm. Figure6 (a) shows microstructure obtained by mesh 140 with 0.15wt%. Average grain size is about 10μm and silicon length was reduced to about 9 μm which distributed homogeneously in the section of the specimen. After the Al-Si alloy was refined, most of the Si phase distributed homogeneously in the section of the specimen. As compared with the microstructures without refinement, the large Si phase disappeared, and the size of the Si phase decreased obviously and was about few micrometers. The eutectic silicon changes its morphology from a needle-

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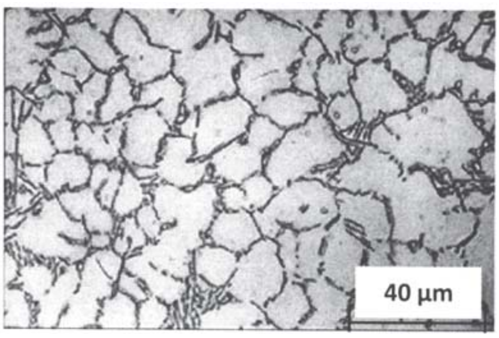

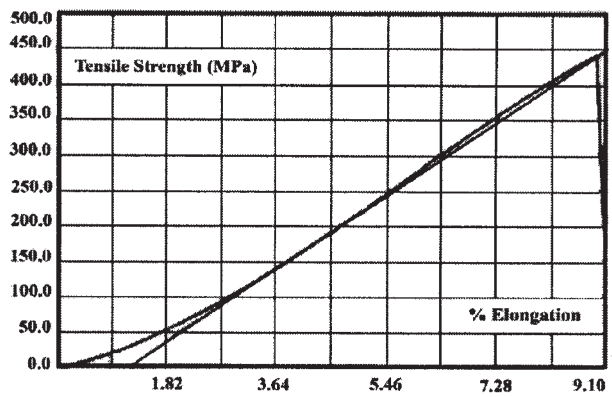

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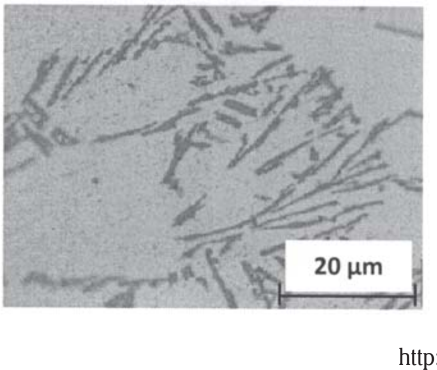

Fig. 4. a) Microstructure of as-cast Al-6%Si alloy before refinement. Coarse Si phase homogeneously distributed without aggregation. The size of the Si phase and the grains are about 10μm and 35-40μm.b) Tensile curve at strain rate of 10-1 s-1 in room temperature. The maximum strength is about 430MPa and fracture occurred at the elastic limit with 8.9% elongation. c) The same microstructure with more magnification. The needle-plate like eutectic silicon is distributed arbitrarily in the Al matrix.

Inoculants wt%	Grain size µm	Tensile Strength MPa	Elongation %
0.1	20	286	9.6
0.15	6	492	7.25
0.2	10	468	9.5
0.25	20	442	10.2

Table 2
AVERAGE GRAIN SIZE, TENSILE STRENGTH AND % ELONGATION OF AS-CAST Al-12.1% Si REFINED BY MESh 50 OF Ti-6Al-4V MASTER ALLOY

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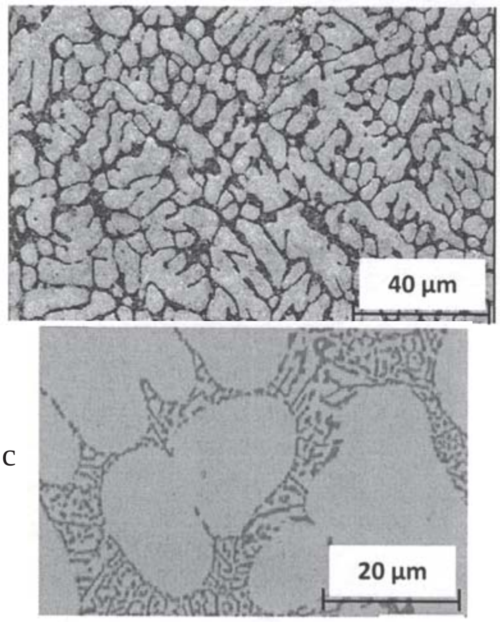

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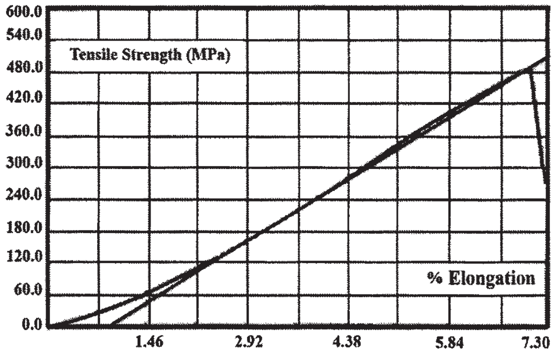

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Fig. 5. a) Microstructure of as-cast Al-6%Si alloy refined by Ti-6Al-4V by mesh 50 with 0.15wt%. Average grain size is about 6 μm and silicon length was reduced to a few micrometers and distributed homogeneously in the section of the specimen. b) Tensile curve at strain rate of 10-1 s-1 in room temperature. The maximum strength is about 492 MPa and fracture occurred at the elastic limit with 7.25%elongation. c) The same microstructure with more magnification. The morphology of the eutectic silicon is changed from needle-plate to a fibrous form

plate to a globular form, which is fully modified. Figure 6 (b) shows that tensile strength and elongation is 540 MPa and 8.82% respectively. Favorite nucleation as a function of weight percentage of inoculants considering the grain size modification and damping effects causes tensile strength increase in this alloy. Mesh size of inoculants has a significant effect on wetting properties and prevents formation of any inactive colony of these particles.

The eutectic encapsulation is one of the major defects in SSM processing due to decreasing of the efficient liquid accessible for deformation. It appears that grain refiner addition has further reduced primary α-Al particles size and dispersed eutectic regions uniformly as an added bonus to the already refined structure resulted from stirring. The percentage of primary α-Al phase is also increased slightly with grain refiner addition which is due to higher nucleation temperature, numerous nuclei, as already discussed for thermal analysis results. By refiner addition, the quantity of active nucleants in the melt is increased and as a result, more α-Al particles can nucleate. These effective nucleation sites lead to more spherical particles and lesser growth ability of the primary particles. On the other hand, as it is reported by the authors, with addition of Ti-6Al-4V, grain growth is restricted due to constitutional supercooling where the growing primary α-Al dendrites reject Ti into the solid-liquid interface as they grow. As the concentration of Ti increases at the interface, it may reach the level where new nucleants of Al_3Ti could form within the interface layer. Because of the efficacy of refiner, the equivalent circular diameter decreases. This parameter has a direct relation with number density, the quantity of primary particles per unit area. More effective nucleation sites lead to better distribution of particles in the sample powered by limited growth leads to smaller particle size. Such increase in number density is in spite of the difficulty in image processing and differentiating between two adjacent α-Al particles especially when they coarsen or there is less eutectic in the structure; it is possible to under estimate α-Al number density. Also as a marginal but critical result, the main criterion of SSM processing, for example, fine primary phase particles, preferably less than 100μm diameter with globular or rosette structure. After the Al-Si alloy was refined by the master alloy, most of the Si phase distributed homogeneously in the section of the specimen. As compared with the microstructures without refinement, the large Si phase disappeared, and the size of the Si phase decreased obviously and was about few micrometers. The grain refiner facilitates the modification of eutectic silicon by baffling the growing of eutectic silicon. In other words, the grain refinement has a definite modification effect on eutectic silicon. In the solidification of Al-Si alloy, Al is precipitated firstly and the eutectic silicon is then precipitated in the succeeding eutectic transformation. The eutectic silicon is precipitated and it grows in the clearance of Al columnar crystal. Therefore Al grain size determines the size of the eutectic silicon. It can be seen that the Al phase, not refined by master alloy, was developed obviously into coarse crystal with the average length of up to 35- 40μm . After refinement, the size of the Al phase reduced obviously to about 10-20μm. Before refinement the needle-plate like eutectic silicon is distributed arbitrarily in the Al matrix. The morphology of the eutectic silicon is changed from needle-plate to a fibrous form with addition of 0.15 wt. % of mesh 50 and finally, the eutectic silicon changes its morphology from a needle-plate to a globular form after adding 0.15 wt. % of mesh 140, which is fully modified. The precipitation of the Si phase resulted in the lack of Si element in matrix and promoted the formation of the Al phases around the primary Si phase. The Hall-Petch plot relation is an empirical law that has been verified experimentally. However, non-linearity can occur in this plot if the grain size is smaller than 10 μm. Initial yield strength has a stronger grain size dependence upon the inverse square-root used by this latter effect. Finer grained metal has wide boundaries which prevent dislocation movement and causes increase in energy level of plastic formation. Non-uniformity in grain size distribution may be occurred by an undesirable

Inoculants wt%	Grain size µm	Tensile Strength MPa	Elongation %
0.1	15	468	9.4
0.15	10	540	8.82
0.2	20	480	8.82
0.25	20	384	8.82

Table 3
AVERAGE GRAIN SIZE, TENSILE STRENGTH AND % ELONGATION OF AS-CAST Al-12.1% Si rEFINED BY MESH 140 OF Ti-6Al-4V MASTER ALLOY

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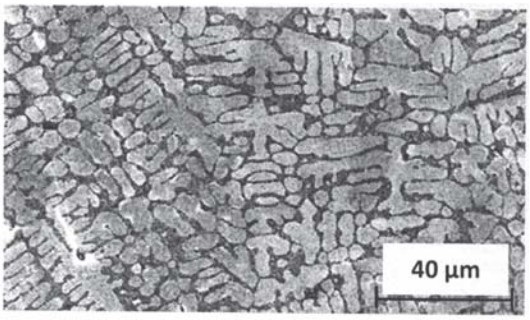

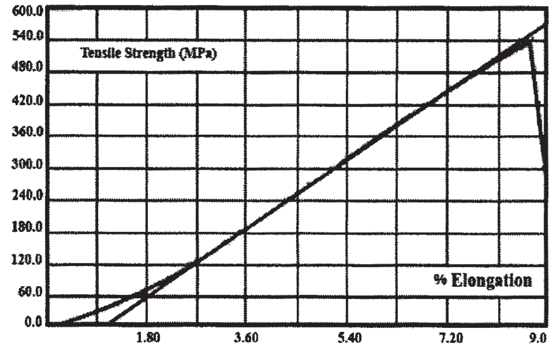

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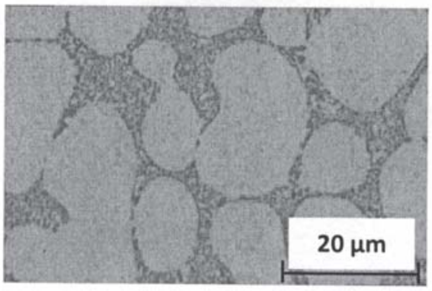

Fig. 6. a) Microstructure of as-cast Al-6%Si alloy refined by Ti-6Al-4V by mesh 140 with 0.15wt%. Average grain size is about 10 µm and silicon length was reduced to a few micrometers and distributed homogeneously in the section of the specimen. b) Tensile curve at strain rate of 10-1 s-1 in room temperature. The maximum strength is about 540 MPa and fracture occurred at the elastic limit with 8.82 % elongation. c) The same microstructure with more magnification. The eutectic silicon changes its morphology from a needle-plate to a globular form, which is higher fully modified

nucleation condition during casting process. In fcc structure alloy with the finer grain size, the shearing bands are formed which have an important role in strain softening. Improvement in both properties of tensile strength and ductility in this alloy is important because they increase fracture toughness and introduce a favorable fracture mechanism.

Conclusions

The effect of Ti-6Al-4V grain refiner addition on the microstructure of Al-6%Si alloy was studied for both conventional and SSM processes. The increasing nucleation temperature allows new crystals to form ahead of solidification front, rendering an equiaxed fine grained as-cast structure. Furthermore, the rate at which growth temperature increased was less than that for nucleation temperature. In other words, there are more nuclei with less potential for growth and thus effective grain refinement is resulted. Before refinement the needle-plate like eutectic silicon is distributed arbitrarily in the Al matrix. The morphology of the eutectic silicon is changed from needle-plate to a fibrous form with addition of 0.15 wt. % of mesh 50 and finally, the eutectic silicon changes its morphology from a needle-plate to a globular form after adding 0.15 wt. % of mesh 140, which is fully modified. With addition of Ti-6Al-4V grain refiner in the SSM processing, the number density of α-Al primaries increases which means smaller globules are formed. The remarkable result is the formation of particles with greater sphericity numbers, closer to 1.

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