



Microstructure and mechanical properties of Al-Al₂O₃ micro and nano composites fabricated by stir casting

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ABSTRACT

Aluminum matrix composites (AMCs) reinforced with micro and nano-sized Al₂O₃ particles are widely used for high performance applications such as automotive, military, aerospace and electricity industries because of their improved physical and mechanical properties. In this study, in order to improve the wettability and distribution of reinforcement particles within the matrix, a novel three step mixing method was used. The process included heat treatment of micro and nano Al₂O₃ particles, injection of heat-treated particles within the molten A356 aluminum alloy by inert argon gas and stirring the melt at different speeds. The influence of various processing parameters such as heat treatment of particles, injection process, stirring speed, reinforcement particle size and weight percentage of reinforcement particles on the microstructure and mechanical properties of composites was investigated. The matrix grain size, morphology and distribution of Al₂O₃ nanoparticles were recognized by scanning electron microscopy (SEM), optical microscope (OM) equipped with image analyzer, energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD). Also, the hardness and compression strength of samples was investigated. The results showed the poor incorporation of nano particles in the aluminum melt prepared by the common condition. However, the use of heat-treated particles, injection of particles and the stirring system improved the wettability and distribution of the nano particles within the aluminum melt. In addition, it was revealed that the amount of hardness, compressive strength and porosity increased as weight percentage of nano Al₂O₃ particles increased.

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1. Introduction

Aluminum matrix composites (AMCs) reinforced with micro and nano-sized Al₂O₃ particles are an interested group of advanced materials, which are widely used for high performance applications such as automotive, military, aerospace and electricity industries [1]. The composites possess improved physical and mechanical properties such as superior strength to weight ratio, good ductility, high strength and high modulus [2], low thermal expansion coefficient, excellent wear resistance, excellent corrosion resistance [3], high temperature creep resistance and better fatigue strength.

The production methods of these composites can be categorized into three types: solid state methods, semi-solid state methods and liquid state methods. The solid state methods can be divided into powder metallurgy, mechanical alloying and diffusion bonding methods. The method generally involves the fabrication of particulate-reinforced MMCs from blended elemental powders by a number of steps prior to final consolidation [4]. It allows essentially a wide range of materials to be used as the matrix and

reinforcement [5]. Also, separation effects and intermetallic phase formations are less for these processes [6]. However, in this case manufacturing route is relatively complex, lengthy, expensive, and energy consuming [5]. The semi-solid process (SSP), which consists of the mixing of ceramic and matrix containing both solid and liquid phases, can take the form of various routes such as compocasting and thixoforging. Semi-solid process allows very large sized components to be fabricated, and is able to sustain high productivity rates [7]. Although, the process is restricted to longer freezing range alloys and, further, has the same limitations as the fully liquid mixing methods [5].

Liquid metallurgy technique is the most economical of all the available routes for metal-matrix composite production and generally can be classified into four categories: pressure infiltration, stir casting, spray deposition and in situ processing [4]. Compared to other routes, melt stirring process has some important advantages, e.g., the wide selection of materials, better matrix-particle bonding, easier control of matrix structure, simple and inexpensive processing, flexibility and applicability to large quantity production and excellent productivity for near-net shaped components [3]. However, there are some problems associated with stir casting of AMCs such as: poor wettability and heterogeneous distribution of the reinforcement material.

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Table 1

Chemical composition of A356 alloy used in experiments (wt.%).

Si	Fe	Mn	Mg	Zn	Ti	Cr	Ni	Pb	Sn	Ca	P	Al
6.104	0.180	0.013	0.425	0.063	0.009	0.001	0.006	0.002	0.002	0.005	0.002	Bal.

Poor wettability of reinforcement in the melt means that the molten matrix cannot wet the surface of reinforcement particles. Therefore, when the reinforcement particles are added into the molten matrix, they float on the melt surface. This is due to the surface tension, very large specific surface area and high interfacial energy of reinforcement particles, presence of oxide films on the melt surface and presence of a gas layer on the ceramic particle surface. Mechanical stirring can usually be applied in order to mix the particles into the melt, but when stirring stops, the particles tend to return to the surface. There are some methods to improve the wettability of the reinforcement particles within the molten matrix alloy; for example heat treatment of the particles before dispersion into the melt caused removal of the adsorbed gases from the particle surface [8].

Another problem is distributing of reinforcement particles uniformly in molten matrix. When the particles were wetted in the metal melt, the particles will tend to sink or float to the molten melt due to the density differences between the reinforcement particles and the matrix alloy melt, so that the dispersion of the ceramic particles is not uniform and the particles have high tendency for agglomeration and clustering. In addition to mechanical stirring, there are some other techniques for introducing particles into the matrix. One of them is injection of the particles with an inert carrier gas into the melt. It has been reported that the technique is helpful in improving the distribution of the reinforcement particles within the melt [9].

Wettability and distribution of reinforcement particles becomes more difficult when the particle size decreases to the nano scales. This is due to the increasing surface area and surface energy of nano particles which cause an increasing tendency for agglomeration of reinforcement particles. Moreover, several structural defects such as porosity, particle clusters, oxide inclusions and interfacial

reactions arise from the unsatisfactory casting technology [10]. Therefore, it is strongly required to develop a novel AMC fabrication route which can improve the incorporation and distribution of nano particles within the molten matrix.

In this study, in order to improve the wettability and distribution of micron and nano-sized Al_2O_3 particles within the A356 aluminum alloy and to avoid agglomeration and separation of particles, a novel three step mixing method was used.

2. Experimental procedure

Table 1 shows the chemical composition of A356 aluminum alloy as determined by Optical Emission Spectroscopy (OES). This alloy was used as the matrix of the composites investigated in this study. Also Al_2O_3 particles with two different sizes of 20 μm and 50 nm were chosen as the reinforcement particles.

Fig. 1 shows the schematic of designed equipment that was used in this study. Aluminum melting process was performed in a graphite crucible placed in a resistance furnace. While the graphite crucible was fixed in the middle of furnace, a hole was created in the bottom of the crucible for bottom pouring of the composite slurry. The hole was closed during the melting, injection and stirring process with a stainless steel stopper. Also, a K-type thermocouple and a high frequency stainless steel stirrer system were placed on the top of the furnace. Injection of the reinforcement particles into the melt is carried out using a stainless steel injection tube and inert argon gas. In this part of equipment the reinforcement powder is placed in a chamber and injected to the melt by pressure of the inert gas. This chamber also has the ability to heat treat the particles in an inert atmosphere before the injection process started.

Initially, calculated amount of the A356 aluminum alloy was charged into the graphite crucible and heated up to 700 °C for

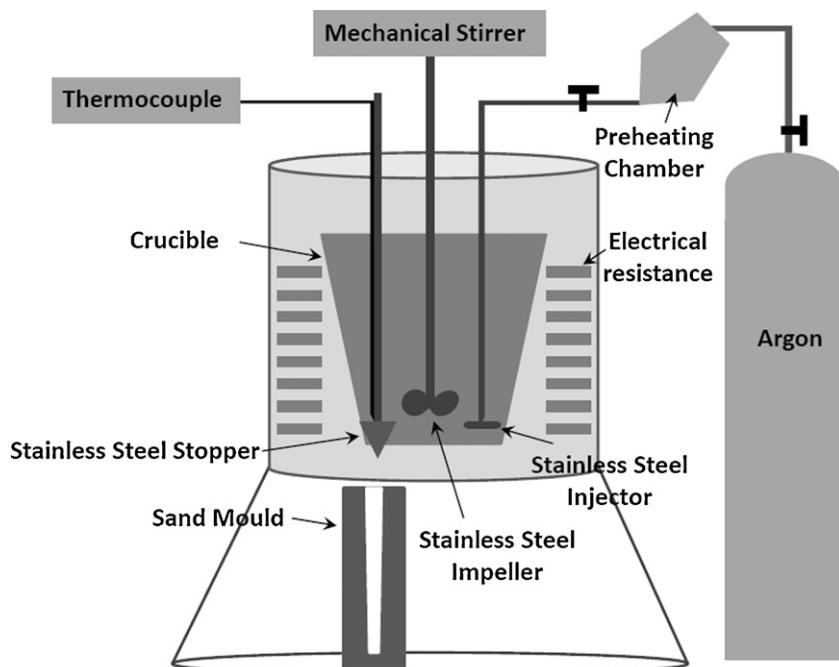


Fig. 1. Schematic of designed equipment.

Table 2
Characteristics of fabricated samples.

Sample	Number
Cast A356 aluminum alloy	A356
Al-5% Al ₂ O ₃ (20 μm) untreated	S5m.P1
Al-5% Al ₂ O ₃ (20 μm) stir cast at 200 rpm	S5m.P2
Al-5% Al ₂ O ₃ (20 μm) stir cast at 200 rpm by injection process	S5m.P3
Al-5% Al ₂ O ₃ (20 μm) stir cast at 200 rpm by injection process of heat treated powders	S5m.P4
Al-5% Al ₂ O ₃ (20 μm) stir cast at 300 rpm by injection process of heat treated powders	S5m.P5 (S5m)
Al-5% Al ₂ O ₃ (20 μm) stir cast at 450 rpm by injection process of heat treated powders	S5m.P6
Al-1% Al ₂ O ₃ (20 μm) stir cast at 300 rpm by injection process of heat treated powders	S1m
Al-3% Al ₂ O ₃ (20 μm) stir cast at 300 rpm by injection process of heat treated powders	S3m
Al-10% Al ₂ O ₃ (20 μm) stir cast at 300 rpm by injection process of heat treated powders	S10m
Al-1% Al ₂ O ₃ (50 nm) stir cast at 300 rpm by injection process of heat treated powders	S1n
Al-2% Al ₂ O ₃ (50 nm) stir cast at 300 rpm by injection process of heat treated powders	S2n
Al-3% Al ₂ O ₃ (50 nm) stir cast at 300 rpm by injection process of heat treated powders	S3n

completely melting of the alloy. A novel three-step mixing method for addition of Al₂O₃ particles to the molten aluminum was presented:

- (i) Heat treatment of reinforcement particles at 1100 °C for 20 min in an inert atmosphere.
- (ii) Injection of heat-treated particles within the melt by inert argon gas.
- (iii) Stirring of the melt before and after incorporation of particles in the aluminum melt at different speed of 200, 300 and 450 rpm. The stirrer was preheated before immersing in the melt and located approximately at the depth of 2/3 of the height of the molten metal from the bottom of the crucible. Stirring process was started 10 min before addition of reinforcement particles in the melt and continued 10 min after that. Then, the stirrer was turned off and the molten mixture was rested for 5 min at 700 °C. Finally, the stopper was picked up and the composite slurry was poured in a preheated cylindrical sand mold located below the furnace. **Table 2** presents characteristics of the fabricated samples used in this investigation.

The matrix grain size, morphology and distribution of Al₂O₃ micron and nanoparticles were recognized by scanning electron microscopy (SEM), optical microscope (OM) equipped with image analyzer, energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD). The density of the samples was measured by the Archimedes's method, while the theoretical densities calculated by taking the densities of A356 aluminum alloy and Al₂O₃ particles were equal to 2.7 and 3.9 g/cm³, respectively. The porosity percentage in the materials was calculated according to the difference between the theoretical and measured density. In addition, compression test was done in the samples with 9 mm diameter and 14 mm height. Compression test was applied at the strain rate of 5 × 10⁻⁴, and each test was repeated three times at room temperature. Also, the Brinell hardness tests of the unreinforced A356 aluminum alloy and fabricated composite materials were determined using a ball with 2.5 mm diameter at a load of 10 kg. The average of 5–10 measurements has been reported as the hardness of the samples.

3. Result and discussion

3.1. Microstructure analysis

Fig. 2 shows the microstructure of composite samples containing 5 weight percent of micron sized Al₂O₃ particles fabricated by different processes. During solidification of A356-Al₂O₃ composites, because of lower thermal conductivity and heat diffusivity of Al₂O₃ particles in comparison with the metal matrix, Al₂O₃ particles are cooled down more slowly than the melt and so the temperature of the particles is somewhat higher than liquid alloy. The hotter particles may heat up their surrounding melt thus, delaying solidification of the surrounding liquid alloy. As a result, nucleation of α-Al phase starts in the liquid at a distance away from the particles, where the temperature is lower. The growth of α-Al nuclei leads to enrichment of Si and other solutes in the remaining melt. Because of Si enrichment in the zones near Al₂O₃ particles, surface of Al₂O₃ particles can act as the suitable substrates for nucleation of Si phase [10]. Therefore, the microstructure of the composites contains primary α-Al dendrites and eutectic silicon, while Al₂O₃ particles are separated at inter-dendritic regions and in eutectic silicon.

Fig. 2a shows microstructure of the fabricated samples without applying stirring process. It is indicated that in this processing route, a few particles were wetted by the aluminum melt. Therefore, only in a special inter-dendritic region particle clustering has been observed. According to **Fig. 2b** stirring the melt has three effects on the microstructure of composite samples: at first it causes to break the dendrite shaped structure and leave the structure in equiaxed form [11]; second, it improves the wettability and incorporation of particles within the melt; and third it causes to disperse the particles more uniformly in the matrix. However, as it can be seen in the figure, stirring the melt is not very useful to improve the incorporation of reinforcement particles in the matrix alloy. As a result, refinement of α-Al grains and improving the distribution of reinforcement particles within the melt are the most important effects of stirring process. **Fig. 2c** shows the microstructure of sample that reinforcement particles were incorporated to the molten metal by injection of particles using inert argon gas. Compared to the previous microstructure more Al₂O₃ particles were observed in the grain boundaries of α-Al grains. Heat treatment of the particles before dispersion into the melt (**Fig. 2d**) removes the adsorbed gases and impurities from the particle surface and improves the wettability and distribution of Al₂O₃ particles in the A356 matrix alloy. It was found that increasing the stirring speed up to 450 rpm caused to decrease the grain size of aluminum matrix (**Fig. 2e** and f). The best distribution of Al₂O₃ in the composites has been found at stirring speed of 300 rpm. While lower stirring speed of 200 rpm was not enough to fully disperse Al₂O₃ particles, the higher stirring speed of 450 rpm caused more wastage of Al₂O₃ particles and more porosity because of more melt turbulence.

In order to study the effects of each process on distribution and wettability of the reinforcement particles in the cast composite samples, a quantitatively analysis was applied. At first, specimens from bottom, middle and top piece of each composite sample were prepared and then pictures from different parts of each specimen were taken. Subsequently, volume percentage of Al₂O₃ particles was calculated using the image analyzer and the average volume percentage (AVP) for each sample was reported according to Eq. (1):

$$\text{AVP} = \frac{\sum_1^n \text{volume percent } i}{n}, \quad (n = 10-20) \quad (1)$$

The results of image analyzing are listed in **Table 3**. Distribution factor (DF) has been defined as the difference between the volume

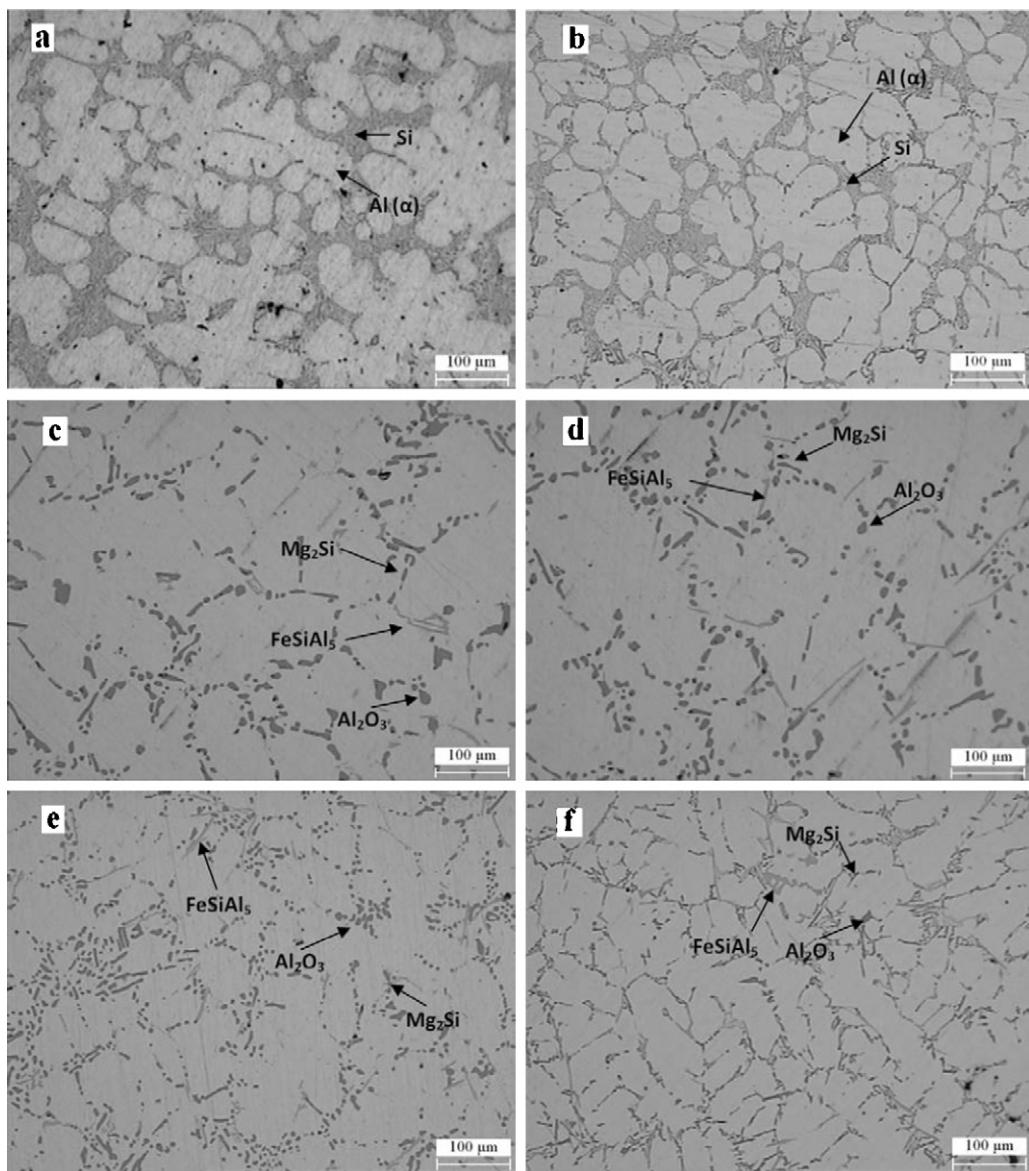


Fig. 2. Typical micrographs of S5m composite sample fabricated by different process: (a) P1; (b) P2; (c) P3; (d) P4; (e) P5; and (f) P6.

percentages of dispersed particles in the different part of samples and calculated according to Eq. (2).

$$DF(\%) = 100 - \frac{|\max(\text{or min})VP - AVP| \times 100}{AVP} \quad (2)$$

As it can be seen, the three-step method has the ability to fabricate composites with homogenous distribution of reinforcement particles and the stirring process has the most effect on the uniform

distribution of particles. Also, wettability factor (WF) is defined by Eq. (3) and the effect of different parameters on the WF has been considered.

$$WF (\%) = 100 - \frac{(VP \text{ of injected } Al_2O_3 \text{ particles} - AVP) \times 100}{VP \text{ of injected } Al_2O_3 \text{ particles}} \quad (3)$$

As it can be seen, when Al_2O_3 particles are added into the molten matrix without applying the three steps of mixing, most of the

Table 3

Grain size and volume percent of Al_2O_3 particle on the surface of samples.

Sample	Average volume percent of particles (AVP)	Distribution factor (DF)	Wettability factor (WF)	Grain size (μm)
S5m.P1	0.53	23	10.75	500
S5m.P2	0.87	69	21.22	100
S5m.P3	3.35	64	81.70	83
S5m.P4	3.56	85	86.82	75
S5m	3.29	93	80.24	55
S5m.P6	2.94	93	71.71	48
S1m	0.82	89	100	52
S3m	2.34	89	88	58
S10m	6.18	92	75.36	71

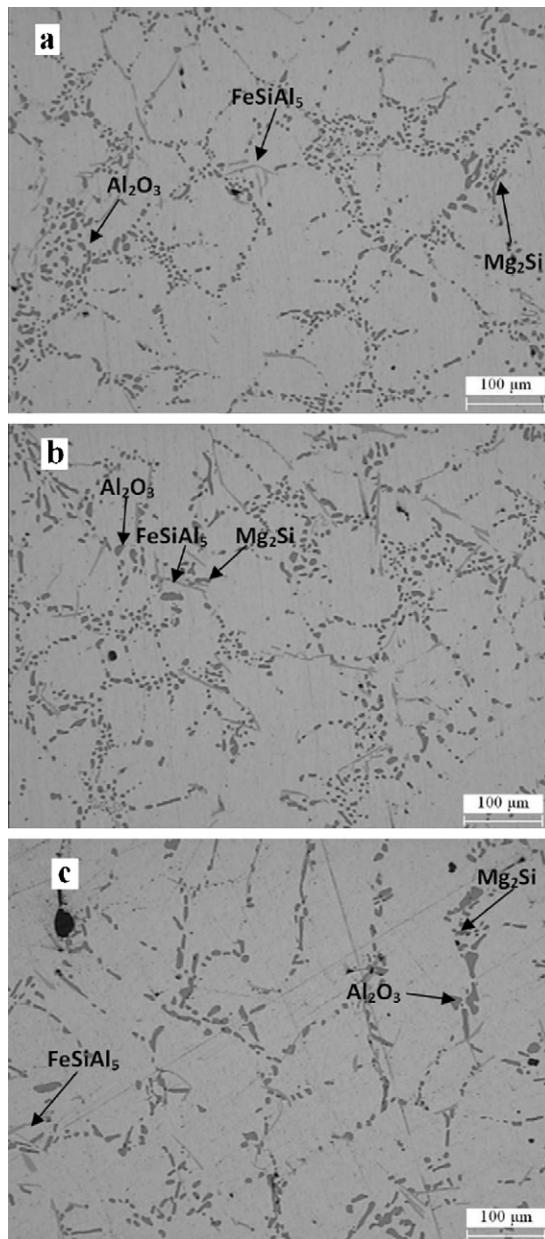


Fig. 3. Typical micrographs of composite samples: (a) S1m; (b) S3m; and (c) S10m.

particles are floating on the melt surface because of poor wetting, while applying the three steps improve the wettability and incorporation of the reinforcement particles, respectively. Introduction of particles into the matrix by injection process has the most important effect on improving the particle wettability. Heat treatment of particles also, can help to improve the particles wettability by breaking the gas layers from the particle surface.

Fig. 3 shows the microstructure of composites containing different percentage of micro-sized Al₂O₃ from 1 to 10 wt.%. The image analysis results presented in Table 3 indicate that the process has only the ability to fabricate samples up to 5 wt.% of micron sized Al₂O₃ reinforcement successfully, and by increasing the reinforcement percentage, wettability of particles in the molten matrix has been decreased. It has been found that the distribution of particles in the different composite samples is uniform and the ununiformity increases by increasing the reinforcement percentage.

SEM micrographs of composite samples with 1, 2 and 3 wt.% of nano Al₂O₃, fabricated under the optimized production conditions,

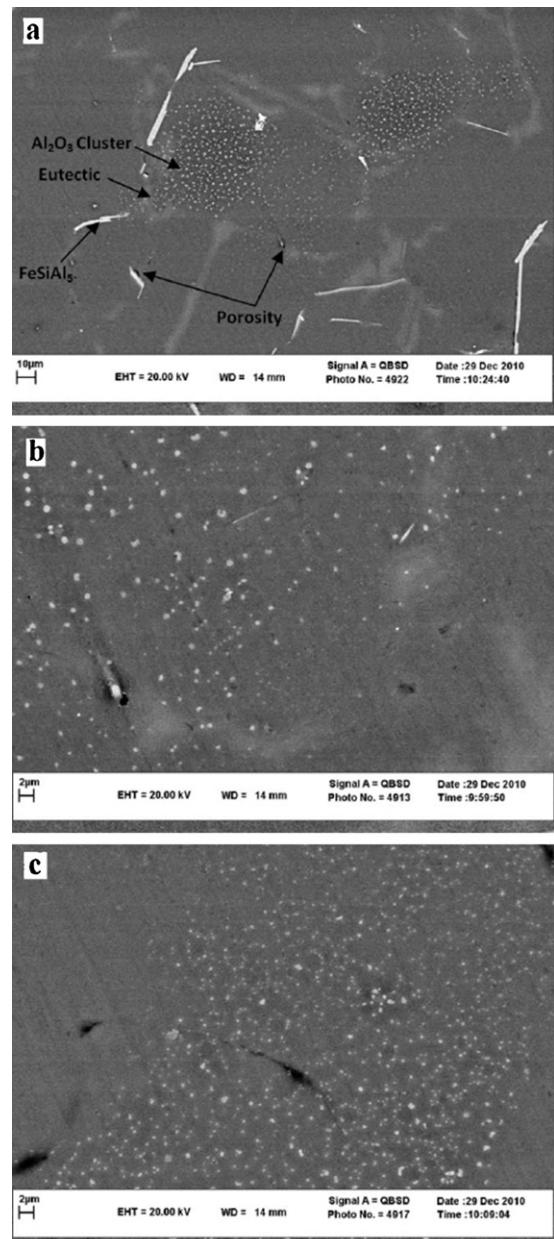


Fig. 4. Typical SEM images of nanocomposite samples: (a) S1n; (b) S2n; and (c) S3n.

are shown in Fig. 4. As it can be seen, particle clustering is more in the case of nano composites because of increasing surface area and surface energy of nano particles. Also, porosities are observed at the matrix–particle interfaces that weaken the bonding between reinforcements and matrix. Microstructure of the composites shows that the Al₂O₃ nano particles have tendency to separate at interdendritic regions where the eutectic silicon is located. EDS analysis and X-ray diffraction of samples were applied for detecting the other phases that were formed in the A356–Al₂O₃ nanocomposites. The bright plate-shaped phases were recognized as the iron intermetallic phase of FeSiAl₅. The formation of this phase is due to the iron-containing impurities which have moved into the melt from the furnace attachments. Other intermetallic compounds such as Mg₂Si and Al₄C₃ were observed at the eutectic phase, however the amount of Al₄C₃ was trivial and it was recognized in the special places.

As a result, comparing with micro composites, particle clustering and agglomeration are more in the case of nanocomposites and

Table 4

Hardness, density and porosity of prepared samples.

Sample	Hardness (HBN)	Theoretical density (g/cm ³)	Measured density (g/cm ³)	Porosity (%)
A356	53.1	2.700	2.67	1.11
S1m	62.4	2.712	2.68	1.18
S3m	64.6	2.736	2.68	2.05
S5m.P4	68.8	2.760	2.69	2.90
S5m	73.5	2.760	2.67	3.26
S5m.P6	72.0	2.760	2.65	3.98
S10	77.1	2.820	2.65	6.02
S1n	67.8	2.712	2.68	1.18
S2n	70.6	2.724	2.68	1.62
S3n	76.3	2.736	2.67	2.41

the three-step mixing method has only the capacity to fabricate samples up to 3 wt.% of nano sized Al₂O₃ reinforcement successfully. Thus, the other treatments, such as the reinforcement surface modification, should be done for improving the wettability of nano particles within the matrix.

3.2. Hardness and density

Table 4 shows the results of density and hardness measurement of composite samples fabricated by different conditions. It is indicated that hardness of samples increases with increasing the weight percentage of Al₂O₃ particles and decreasing the particle size. This is because of hindering of dislocations movement due to increase in the particle concentration and decrease in the grain size of the A356 aluminum alloy.

Also, according to the measured and theoretical densities of composite samples, it is revealed that the amount of porosity in the composite samples increases with increasing weight percentage of Al₂O₃ particles and decreasing the size of particles. The porosity is generally because of some principles such as: (1) increase in surface area in contact with air caused by increasing the weight percent and decreasing the particle size [3]; (2) gas entrapment during stirring; (3) gas injection of particles introduces a quantity of gas into the melt; (4) hydrogen evolution; (5) the pouring distance from the crucible to the mold and (6) shrinkage during solidification [9]. Also, it is revealed that stirring speed of 300 rpm shows the best result in hardness. Moreover, with increasing the stirring speed up to 450 rpm more porosity has been observed due to the more gas entrapment during stirring.

3.3. Compressive strength

Fig. 5 shows the stress-strain curve of compression test related to nanocomposite samples fabricated by 3-step processing method. The compressive strength of composite can primarily be attributed to: the significant grain refinement, the presence of reasonably

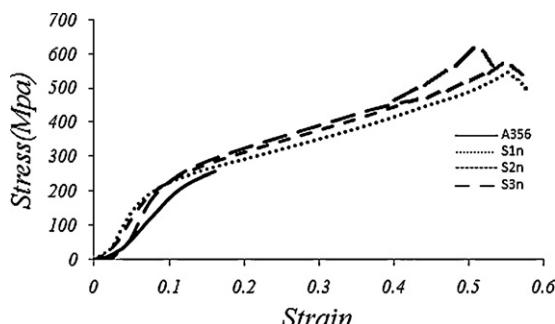


Fig. 5. Stress–strain curve of compression test for nanocomposite samples.

Table 5

Compression strength of composite samples.

Samples	Compressive strength (MPa)
A356	234.26
S1m	250.65
S3m	310.50
S5m	390.55
S10m	453.12
S1n	550.80
S2n	586.39
S3n	610.30

distributed hard particulates, dislocation generation due to elastic modulus mismatch and coefficient of thermal expansion mismatch between the matrix and reinforcement phase, load transfer from matrix to reinforcement phase and Orowan strengthening mechanism [12–14].

Variation of compressive strength for different composite samples is presented in Table 5. First of all, effect of Al₂O₃ content on compressive strength is considered and it is concluded that, by increasing in Al₂O₃ percentage, the compressive strength shows an increasing trend. This confirms the obvious effect of Al₂O₃ particles on strengthening of the composites. The results also show that the compression strength of nanocomposites is greater than that of micro-composites; because nano particles are more effective in strengthening of the composites.

4. Conclusions

In the present research Al-Al₂O₃ micro and nano composites were successfully fabricated in the designed equipment by the novel three step mixing method including heat treatment of nano particles, injection of heat-treated particles within the melt by inert argon gas and stirring the melt and the following results were obtained:

1. The results showed the poor incorporation and heterogeneous distribution of micro particles in the aluminum melt prepared by the common condition, while particle heat treatment and injection of the particles within the melt by inert gas help the particulate reinforcements to wet better in the molten matrix.
2. The microstructure of the composites contained the primary α -Al dendrites and eutectic silicon. While Al₂O₃ particles were separated at inter-dendritic regions and in the eutectic silicon. Also, another intermetallic compounds such as Mg₂Si, FeSiAl₅ and Al₄C₃ were observed at the eutectic phase.
3. Distribution of particles in the different composite samples is uniform and the stirring process has the important effect on improving the reinforcement distribution. The best distribution of reinforcements in composite microstructure has been found at stirring speed of 300 rpm.
4. Wettability of particles within the molten matrix had been decreased by increasing the reinforcement percentage and decreasing the reinforcement size. It has been found that the three-step mixing method has only the ability to fabricate samples up to 5 wt.% of micron sized and 3 wt.% of nano sized Al₂O₃ reinforcement successfully.
5. It was revealed that the amount of hardness and porosity of composite samples increased with increasing the weight percentage of Al₂O₃ particles and decreasing the particle size.
6. By increasing in Al₂O₃ percentage, the compressive strength shows an increasing trend. The results also show that the compression strength of nanocomposites is greater than that of micro-composites.

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