MICROSTRUCTURE EVOLUTION OF AL–AL2O3 MICRO AND NANO COMPOSITES FABRICATED BY A MODIFIED STIR CASTING ROUTE

H. Beygi[1]*,H.R. Ezatpour[1], S. A. Sajjadi[1], S. M. Zebarjad[1]
[1]Department of Materials Science, Engineering Faculty, Ferdowsi University of Mashhad, Mashhad, Iran
* Corresponding author(Sajjadi@um.ac.ir)

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Abstract
A modified stir casting method is applied to fabricate Al-Al2O3 micro and nano composites. The method consisted of heat treatment of reinforcement particles, addition of 1wt.% magnesium as the wetting agent, injection of heat treated particles within the melt by inert argon gas and finally stirring the melt. All the processes are performed in a designed furnace and attached equipment. A novel measurement method was presented in this study to quantitatively study the wettability and distribution of the particles in the composite samples. Subsequently effects of various process parameters e.g. heat treatment of reinforcement particles, additive wetting agent, injection process, stirring the melt, weight percentage of Al2O3 particles and Al2O3 particle size (micron and nano size) on the wettability and distribution of particles investigated. The results showed the poor incorporation of Al2O3 particles in the aluminum melt prepared by the common condition while the use of heat treated particles and 1wt.% additive Mg significantly increases the wettability of particles and also injection of particles and the stirring process improved distribution of the Al2O3 particles within the aluminum melt.

1 Introduction

Stir casting technique is known as the most economical method for production of metal matrix composite because of its important advantages, e.g., the wide selection of materials, better matrixparticle 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. However there are some problems associates with stir cast producing of AMCs. Poor wettability and heterogeneous distribution of the reinforcement material are two major problems in this method [1-4].

Poor wettability of reinforcement in the melt means that the molten matrix cannot wet the surface of reinforcement particles and so when the reinforcement particles were added into the molten matrix, they were observed to be floating 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 particles surface. Mechanical stirring can usually be applied to mix the particles into the melt, but when stirring stopped, the particles tended to return to the surface, indicates that the particles floated mainly because it has been still difficult for the particles to be wetted by the molten metals because of the gas layers. 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 to removing the adsorbed gases from the particle surface, and adding some surface-active elements such as magnesium, lithium, calcium, titanium or zirconium into the matrix to changed the morphology of the interface from convex to concave [4,8].

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 are not uniform and the particles have high tendency for agglomeration and clustering. In Addition of using the mechanical stirring some other technique for introduction of particles into the matrix, such as Injection of the particles with an inert carrier gas into the melt, are observed to be helpful to improve the distribution of the reinforcement particles within the melt [3,4].

Wettability and distribution of reinforcement particles becomes more difficult when the particle size decreases to the nano scales. This is due to the increasing the surface area and surface energy of nano particles, caused an increasing tendency for agglomeration of reinforcement particles. In addition of poor wettability and agglomeration of nano particles, several structural defects such as porosity, particle clusters, oxide inclusions and interfacial reactions were found to arise from the unsatisfactory casting technology [6]. 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 the present study, effects of various process parameters on the wettability and distribution of particles in the aluminum alloy are investigated. several experiments such as heat treatment of the Al2O3 particles, the use of a 1Wt%mg as the wetting agent, in order to improve wettability of particles, also stirring the melt and injection of particles within the melt by inert Argon gas are introduced to enhancing the distribution. In the next parts, influence of weight percentage of alumina particles from 1 to 10 wt.% and the size of particles (micron and nano size) were investigated.

2 Experimental

Table 1 shows the composition of A356 aluminum alloy that was used as the main matrix material. Also Al2O3 particles with two different sizes of 20µm and 50nm were chosen as the reinforcement particles and magnesium additive used was also in powder form.

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 are carried out using a stainless steel injection tube and inert Argon gas. In this part of equipment the reinforcement powder are placed in a chamber and injected to the melt because of the pressure of inert argon gas. This chamber also has the ability to preheat 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 0C for completely melting of alloy in the crucible. After melting the Alloy and mixing the reinforcement within the matrix, the stirrer was turned off, and the molten mixture was rested for 5 min and at temperature of 700°C. Finally the stopper was picked up and the composite slurry was poured in a preheated cylindrical sand mould, with 20 mm diameter and 400 mm long, that was located below the furnace.

Especial design of experiments was performed to investigate effects of various process parameters, al2O3 weight percentage and Al2O3 particle size. Table 2 presented the corresponding samples fabricated with each experiment.

At first effects of process parameters studied; Stirring the melt at stirring speed of 200-300 rpm and injection of heat-treated particles within the melt by inert argon gas in order to improving distribution of particles in the melt, and heat treatment of reinforcement particles at 1100 °C for 20 min in an inert atmosphere and addition of 1wt.% mg as the wetting agent to enhancing wettability of particles within the molten A356 alloy.

In the next parts, influence of weight percentage of alumina particles from 1 to 10 wt.% and the size of particles (micron and nano size) were investigated.

The matrix grain size, morphology and distribution of Al2O3 particles were recognized by scanning electron microscopy (SEM), optical microscope (OM) equipped with image analyzer, energy dispersive spectroscopy (EDS) and X-ray diffraction (XRD). For studying the effects of each process on distribution and wettability of reinforcement particles in the cast composite samples, a quantitatively analysis was applied. First specimens from bottom, middle and top piece of each composite sample were prepared and after that pictures from different part of each specimen were taken. Subsequently volume percentage of Al2O3 particles were calculated using the image analyzer and the average for each one was reported. Subsequently wettability and distribution of particles in different samples were quantity measured. The density of the samples was measured by the Archimedes method, while the theoretical densities calculated by taking the densities of A356 aluminum alloy and Al2O3 particles equal to 2.7 and 3.9 g/cm3

respectively. Also the porosity percentage in the materials can be calculated according to the difference between the theoretical and measured density. In addition the Brinell hardness tests of the unreinforced A356 aluminum alloy and fabricated composite materials were determined using a ball with 2.5mm diameter at a load of 10 kg. The average of 5-10 measurements has been reported as the hardness of the samples.

3 Results and discussion

Fig 2 shows the microstructure of composite samples fabricated with different processes. In solidification of A356- Al2O3 composites, because of lower thermal conductivity and heat diffusivity of Al2O3 particles from the metal matrix, Al2O3 particles were cooled down more slowly than the melt and so the temperature of the particles was somewhat higher than liquid alloy. The hotter particles may heat up the liquid in their immediate surroundings, and thus delay solidification of the surrounding liquid alloy. As a result nucleation of a-Al phase starts in the liquid at a distance away from the particles, where the temperature was lower. The growth of a-Al nuclei lead to enrichment of Si and other solutes in the remaining melt and because of Si enrichment in a zone near the Al2O3particles, Surface of Al2O3 particles can be act as the suitable substrates for nucleation of Si phase [6]. As a result, the microstructure of this composites containing the primary a-Al dendrites and eutectic silicon, while Al2O3 particles were segregated at inter-dendritic regions and in the eutectic silicon.

Figure 2.a shows the microstructure of fabricated samples without applying stirring process. It is indicated that in this processing route, about any of particles were wetted by the aluminum melt, and only in a special interdendritic region particle clustering has been observed. According to fig 2.b stirring the melt has three effects on the microstructure of composite samples: at first it caused to break the dendrite shaped structure and leave the structure in equaixed form [2]; second, it improved the wettability and incorporation of particles within the melt; and third it caused to disperse the particles more uniformly in the matrix. But as it can be seen in this fig, stirring the melt is not very useful to improve the incorporation of reinforcement particles in the matrix alloy, and so refinement of a-Al grains and improving the distribution of reinforcement particles within the melt are the most important effects of stirring process. Fig 2.c shows the microstructure of sample that reinforcement particles were incorporated to the molten metal by injection of particles using inert Argon gas. Compared to previous microstructure more Al2O3 particle was observed in the grain boundaries of a-Al grains. Also Heat treatment of the particles before dispersion into the melt (fig 2.d) due to removing the adsorbed gases and impurities from the particle surface, caused to improving the wettability and incorporation of Al2O3 particles in the A356 matrix alloy. From Fig 2e, it was found that the use of 1 wt.% magnesium as the wetting agent caused to more particles were incorporated to in the matrix. Using magnesium significantly increases wetting behavior of the particles, however it is revealed that the Mg addition changes eutectic phase shape, and also leads to increases the viscosity of the slurry to the detriment of particles distribution [7].

Figs 2e-h show the microstructure of composites containing different percentage of micron sized Al2O3 from 1 to 10 wt.%. Uniform distribution of Al2O3 particles observed in all samples. It is indicated that more Al2O3 particles are presented in the micrographs of samples containing more Al2O3 weight percentage, however tendency to incorporate the Al2O3 particles into the matrix alloy reduced when weight percentage of Al2O3 particles increases to 10wt.%.

The results of image analyzing were listed in table 3. Distribution Factor (DF) has been defined as the difference between the volume percentages of dispersed particles in the different part of samples. As it can be seen, the stirring and injection process have the most effect on the uniform distribution of particles respectively. Also the effect of different parameters with the Wettability Factor (WF) has been estimated. It can be founded that, when the Al2O3 particles were added into the molten matrix without applying any process, they were observed to be non-wetting and most of the particles were floating on the melt surface. However injection the particles into the melt, heat treatment of particles and in particular addition of Mg as the wetting agent improved the wettability and incorporation of the reinforcement particles within the Al matix.

The image analysis results from Table 3 indicated that, the process has the ability to fabricate samples up to 5-10 wt.% of micron sized Al2O3 reinforcement successfully, and by increasing the reinforcement percentage, wettability of particle on

to the molten matrix had been decreased. It has been found that the distribution of particles in the different composite samples is uniform, and the reinforcement distribution was increased by increasing the reinforcement percentage maybe because of decreasing the particles segregation.

SEM micrographs of Al-3wt.%Al2O3 nanocomposite sample, that was fabricated by P5 process, are shown in fig 3. As it can be seen, particle clustering is more in the case of nano composites because of increasing the surface area and surface energy of nano particles. Also some porosity was observed at the matrix- particle interfaces that were redounded to weak bonding of reinforcements. The microstructure of these composites shows that the Al2O3 nano particles have tendency to segregate at inter-dendritic region where the eutectic silicon is located. EDS analysis of samples were applied for detecting the other phases that were formed in the A356- Al2O3 nanocomposites. The bright plate shape phases were recognized as the iron intermetallic phase of FeSiAl5. The formation of this phase is due to the iron-containing impurities that were gone into the melt from the furnace attachments. Also another intermetallic compounds such as Mg2Si and Al4C3 were observed at the eutectic phase, however the amount of Al4C3 is very small and it was recognized in the special places.

As the result, comparing with micro composites, particle clustering and agglomeration is more in the case of nano composites and the modified stir casting route has only the capacity to fabricate samples up to 3-5 wt.% of nano sized Al2O3 reinforcement successfully.

4 Conclusions

1- A modified stir casting method is applied to fabricate Al-Al2O3 micro and nano composites. The method consisted of heat treatment of reinforcement particles, addition of 1wt.% magnesium as the wetting agent, injection of heat treated particles within the melt by inert argon gas and finally stirring the melt. All the processes are performed in a designed furnace and attached equipments.

2- Microstructure of these composites containing the primary a-Al dendrites and eutectic silicon. While Al2O3 particles were segregated at inter-dendritic regions and in the eutectic silicon. Also another intermetallic compounds such as Mg2Si, FeSiAl5and Al4C3 were observed at the eutectic phase.

3- Stirring the melt has three effects on the microstructure of composite samples: at first it caused to break the dendrite shaped structure and leave the structure in equaixed form; second, it improved the wettability and incorporation of particles within the melt; and third it caused to disperse the particles more uniformly in the matrix.

4- Heat treatment of the particles before dispersion into the melt due to removing the adsorbed gases and impurities from the particle surface, caused to improving the wettability and incorporation of Al2O3 particles in the A356 matrix alloy.

5- Using magnesium significantly increases wetting behavior of the particles, however it is revealed that the Mg addition changes eutectic phase shape, and also leads to increases the viscosity of the slurry to the detriment of particles distribution.

6- Wettability of particles within the molten matrix had been decreased by increasing the reinforcement percentage and decreasing the reinforcement size.

7- The results showed the poor incorporation of Al2O3 particles in the aluminum melt prepared by the common condition. However injection the particles into the melt, heat treatment of particles and in particular addition of Mg as the wetting agent improved the wettability and incorporation of the reinforcement particles within the Al matix.

8- Distribution of particles in the different composite samples is uniform.

9- The modified stir casting route has the ability to fabricate samples up to 5-10 wt.% of micron sized and 3-5 wt.% of nano sized Al2O3 reinforcement successfully.

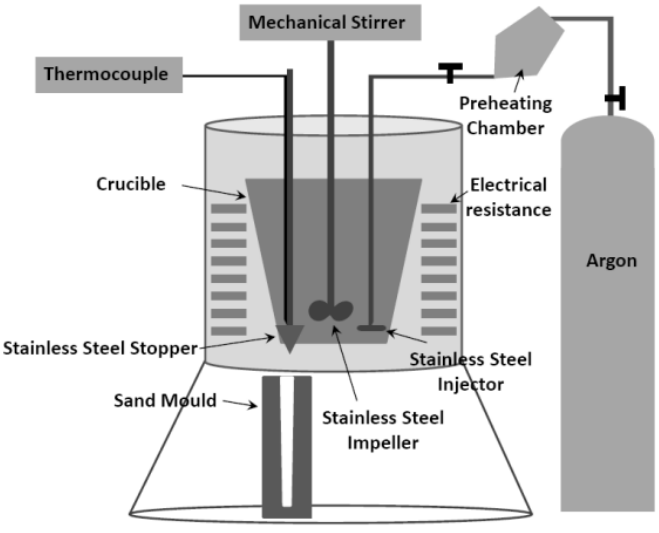

Fig 1- schematic of designed equipment

Table1- Chemical composition of A356 alloy used in experiments

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	93.275

Experiment	Code	Sample	Process
Effect of process parameters	A356	Cast A356 aluminum alloy	
	S5m.P1	Al-5% Al_2O_3 composite	untreated
	S5m.P2	Al-5% Al_2O_3 composite	stir casting
	S5m.P3	Al-5% Al_2O_3 composite	stir casting + injection process
	S5m.P4	Al-5% Al_2O_3 composite	stir casting + injection process+ heat treatment of powders
	S5m.P5	Al-5% Al_2O_3 composite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
Effect of Al2O3 weight percentage	S1m.P5	Al-1% Al_2O_3 composite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
	S3m.P5	Al-3% Al_2O_3 composite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
	S10m.P5	Al-10% Al_2O_3 composite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
Effect of Al2O3 particle size	S1n.P5	Al-1% Al_2O_3nanocomposite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
	S2n.P5	Al-2% Al_2O_3nanocomposite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
	S3n.P5	Al-3% Al_2O_3nanocomposite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive
	S5n.P5	Al-5% Al_2O_3nanocomposite	stir casting + injection process+ heat treatment of powders+ 1wt.% magnesium additive

Table3- Grain Size and volume percent of Al_2O_3 particle on the surface of samples

	Average Volume Percent of Al_2O_3 Particles $AVP = \frac{\sum_1^n volumepercenti}{n}$	**Distribution Factor** $DF(\%) = 100 - \frac{\lvert max(ormin)VP - AVP\rvert \times 100}{AVP}$	**Wettability Factor** $WF(\%) = 100 - \frac{(VPofinjectedAl2O3particles - AVP) \times 100}{VPofinjectedAl2O3particles}$
	0.53	23	10.75
	0.87	69	21.22
	3.35	64	81.70
	3.56	85	86.82
	4.32	85	100
	0.82	86	100
	2.51	83	100
5	7.56	84	92.19

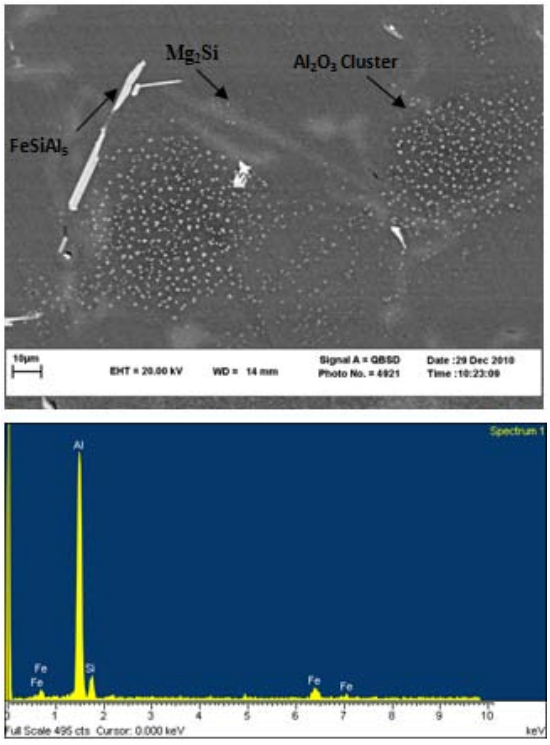

Fig 3- SEM and EDS results of S3n nanocomposite samples

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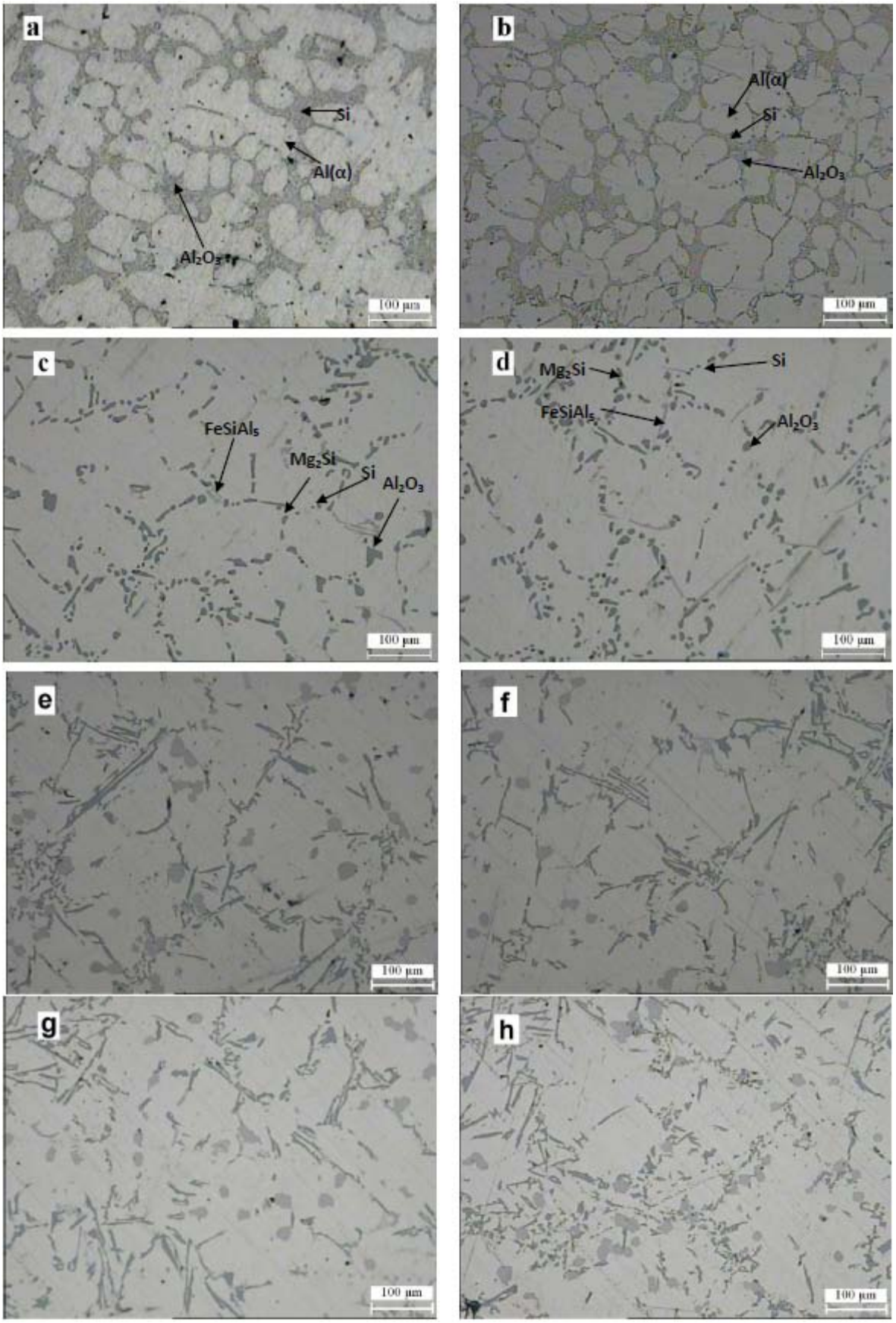

Fig 2- Typical micrographs of (a) S5m.p1; (b) S5m.p2; (c) S5m.p3; (d) S5m.p4; (e) S5m.P5; (f) S1m.P5; (g) S3m.P5; (h) S10m.P5 composite samples.