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# EFFECT OF SINTERING TEMPERATURE AND 5 WT.% $Al(NO_3)_3$ ADDITIVE TO METAL MATRIX COMPOSITES CHARACTERISTICS BY POWDER METALLURGY TECHNIQUE

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## ABSTRACT

In this work, Al/SiC metal matrix composites have been successfully produced by powder metallurgy process. The surface arrangement of SiC powder were performed by oxide metal coating process using an extra addition of $Al(NO_3)_3$ 5 wt.%. The coating process was promoted to enhance the wetting at the matrix (Al) and reinforcement (SiC) interface. The mechanism of powder metallurgy processes include: wet mixing, cold compaction at 120 MPa, and sintering under high purity argon atmosphere and heated at a rate of 15 $^0$C/min. Sintering were carried out in an electrical tube furnace at different sinter temperatures (450, 500, 550, and 600 $^0$C) for 20 wt.% SiC and holding time for 1 hour. The testing of substrate includes density and porosity using the Archimedes principle. For the microstructure characterization was investigated to evaluate the homogeneity and interaction the constituent. Results from the investigation show the increasing (decreasing) tendency of density and porosity by the increasing of sintering temperature. The SEM analysis shows relative homogeneity of distribution of Al and SiC particle. Thus, the XRD analysis reveals that the influence of sintering temperature was confirmed by the XRD pattern.

***Keywords:*** Metal matrix composite, powder metallurgy, wettability, inert gas, Al/SiC, wet mixing

## ABSTRAK

Dalam penelitian ini, komposit matriks logam Al/SiC telah diproduksi melalui teknik metalurgi serbuk. Perekayasaan permukaan serbuk SiC dilakukan dengan proses pelapisan oksida logam menggunakan aditif $Al(NO_3)_3$ 5 % berat. Proses pelapisan dilakukan untuk menaikkan tingkat kebasahan (*wettability*) serbuk pada *interface* matriks (Al) dan penguat (SiC). Proses mekanisme metalurgi serbuk, antara lain: *wet mixing*, penekanan dingin pada 120 MPa, dan sintering dalam kondisi atmosfir gas argon dan dipanaskan dengan *heating rate* pada 15$^0$C/min. Proses sintering dilakukan dalam tungku elektrik tabung pada suhu sinter yang berbeda (450, 500, 550, and 600$^0$C) untuk komposisi penguat 20 % berat SiC dan waktu tahan selama 1 jam. Pengujian substrat meliputi densitas dan porositas menggunakan prinsip Archimedes. Pengujian mikrostruktur dilakukan untuk mengevaluasi homogenitas dan interaksi antar penyusun. Hasil pengujian menunjukkan kecenderungan pola peningkatan nilai densitas atau penuruanan nilai porositas dengan meningkatnya suhu sintering. Analisa SEM menunjukkan distribusi partikel Al dan SiC yang relatif homogen. Sedangkan uji analisa XRD menunjukkan bahwa pengaruh suhu sintering telah diperlihatkan melalui pola-pola puncak XRD.

**Kata Kunci**: Komposit matriks logam, metalurgi serbuk, tingkat kebasahan, gas inert, Al/SiC, *wet mixing*

## I. INTRODUCTION

In recent years, the interest in materials which have light weight, high resistances, and hardness, are continuously developed to displace traditional engineering materials. Especially in metal matrix composites (MMC), aluminium matrix composite reinforced with silicon carbide are attractive for aerospace, automobile, and ceramic industries applications. The major methods to produce the aluminium metal matrix composites are stir casting, powder metallurgy, liquid metal infiltration, squeeze casting, rheocasting (compocasting), and spray deposition. The wettability is a complex phenomenon that depends on several factors, such as: geometry of interface, process temperature, and soaks time, which is determines the quality of composites [1].

Powder metallurgy (P/M) is one of the processing techniques adopted for silicon carbide-reinforced aluminium alloy-based composites. Because of the relatively lower temperatures (below the melting point) involved in P/M processing, the undesirable interfacial reactions and the development of detrimental intermetallic phases are negligible in Al/SiC composites, as compared to the cast composites. Unlike the liquid metallurgy route, the P/M technique renders a homogeneous distribution of the reinforcement in the matrix [1].

The Al/SiC composite includes in the discontinuities metal matrix composites (DMMC) type. This type has an isotropic reinforcement orientation; it means all of the strength orientation is equal. P/M method is commonly used for the DMMC type [2]. In this process, fine powder of the matrix and reinforce were blended and fed into the mould of the desired shape. The Blending process can be carried out in dry or liquid suspension. Pressure is applied to further powder compaction (cold compaction). The powder was formed into green body; the density at this phase has approximately 80%. Then, the green body is heated to the temperature, which is below the matrix melting point but high enough to develop significant solid state interface diffusion; it was promoted to enhance the density. The formation of liquid bridge at interfacial phase (new phase) acting as a binder agent. This phase has a significant effect in the wettability of matrixes and reinforce interface.

In this work Al/$SiC_p$ composites were prepared by powder metallurgy synthesis using aluminium (Al) fine powder as a matrix and SiC powder as reinforce. Aluminium and $SiC_p$ compact (green body) were sealed in rigid steel die with certain shape and size. The mixing samples were compacted using uniaxial hydraulic press at 120 MPa pressure. The sinter process that carried out in solid state condition was sintered by transparent silica tube under pure argon atmosphere. Since, the aluminium has the melting point about 660 $^0$C, therefore the variation of sinter temperature was limited until 600 $^0$C; and the holding time was 1 hour for each variation. The objective was to investigate the physical and microstructural behavior of these Al/$SiC_p$ composites. The study involved the wettability of reinforcement powder by coating process using an extra addition of $Al(NO_3)_3$.

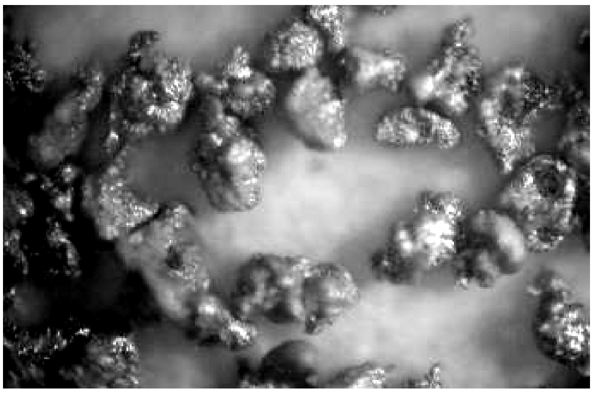
## II. EXPERIMENTAL PROCEDURE

In this work, samples were prepared using Al 2124 aluminium alloy 92.5 wt.% as raw material. The chemical composition of Al 2124 which has been analyzed by X-Ray Fluorescence (XRF) method is shown in Table 1. The powder is nominally 100 - 200 mesh and have an average particle size of 85 μm. The reinforcement was prepared using SiC powder with an average of $SiC_p$ size of 56 μm. The optical microscope of Al 2124 and $SiC_p$ as raw material is shown in Figure 1.

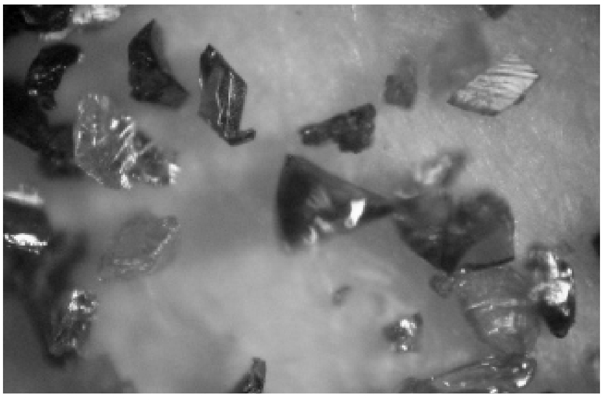
The $Al(NO_3)_3$ were used as an external dopant (dispersant) in order to promote the wetting system between pure Al and $SiC_p$ ceramics particle interface.

**Table 1 Chemical composition of Al 2124 powder**

| Element | Si | Fe | Cu | Mn | Mg | Zn | Cr | Ti | other |
|---|---|---|---|---|---|---|---|---|---|
| Wt % | 0.2 | 0.3 | 4.4 | 0.6 | 1.5 | 0.01 | 0.1 | 0.15 | 0.2 |



(a)



(b)

**Fig.1. Optical microscope photograph: (a) aluminium 2124 and (b) silicon carbide powder.**

At the first stage, $SiC_p$ were coated in magnetic stirrer using $Al(NO_3)_3$ 5 wt.% additive acting as a dispersant for 30 minute in ethanol medium (control agent). Then, the $SiC_p$ were dried in the furnace at 200 $^0$C for 2 h and then at 400 $^0$C for 2 h more. $SiC_p$ were used in this work at 20 wt.%. Al 2124 and $SiC_p$ were blended in magnetic stirrer for 1.5 h by wet mixing process. These powder mixtures were mixed until homogeny in ethanol medium. Then, Al and $SiC_p$ powder were placed in cylindrical steel die with die-wall lubricant to produce cylindrical samples in hydraulic press. The die wall was lubricated with stearat acid powder about 0.2 – 1 wt.%. This was followed by cold compaction at pressure of 120 MPa. Then, the compact body was ejected and heated in argon atmosphere circulating furnace at different temperatures (450, 500, 550, and 600 $^0$C) for 1 hour holding time, respectively. The synthesis – consolidation procedure flowchart and the experimental set-up for sintering process are shown in Figure 2 and Figure 3, respectively.

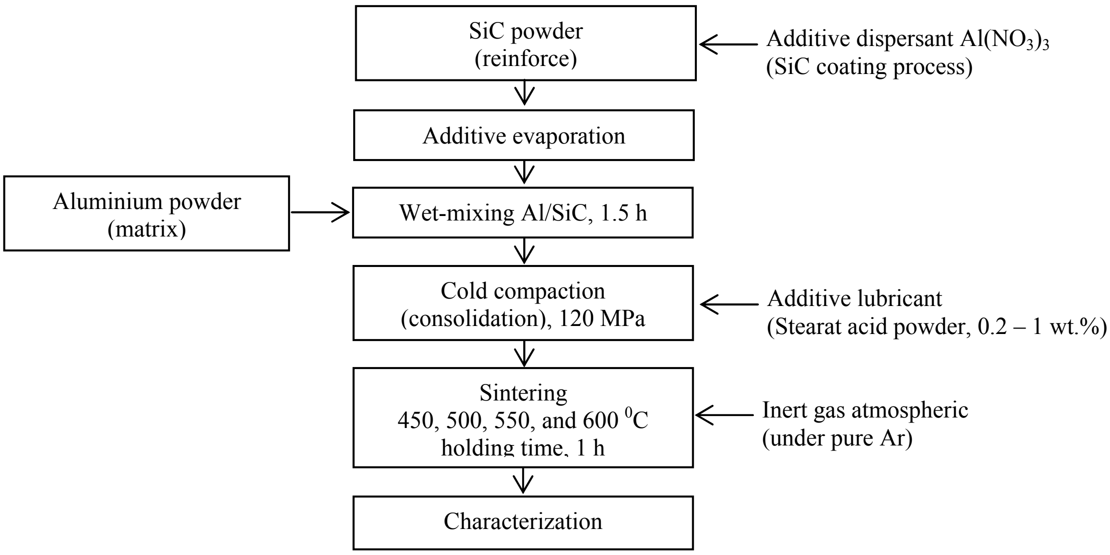



**Figure 2. Process diagram for production of Al2124/SiC$_P$ composites.**

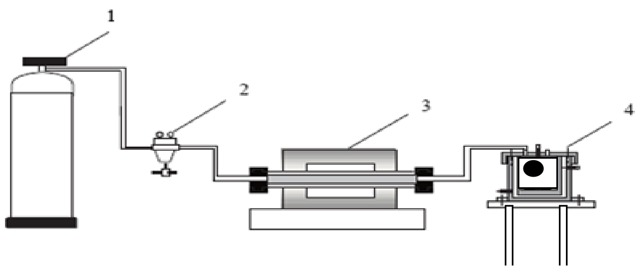



**Figure 3. Experimental set-up: (1) Argon gas, (2) Manometer (argon regulator), (3) Tube furnace, and (4) Temperature control**

## III. RESULTS AND DISCUSSION

Figure 4 shows the graphic of sintering temperature to relative density of composite. It shows that the composite density depends on the amount of volume fraction from each constituent; the density of Al is 2.73 g/cm$^3$ and SiC$_p$ is 3.11 g/cm$^3$. The density of soft Al and hard SiC$_p$ powder during the compaction carried out, lead to reduce of the composites density, which is affected by the mixing method. During sinter process, the soft particle Al occurred in plastics phase; whiles the hard particle

SiC is still in elastics phase. Therefore, at this stage the SiC powder were not yet sintered, because the melting point of SiC is higher than Al.

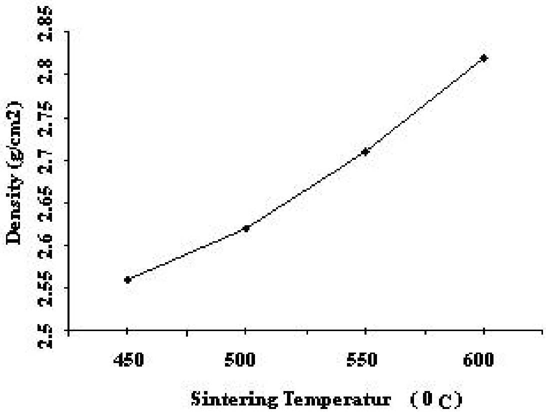



**Figure 4. Effect of sintering temperature on density.**

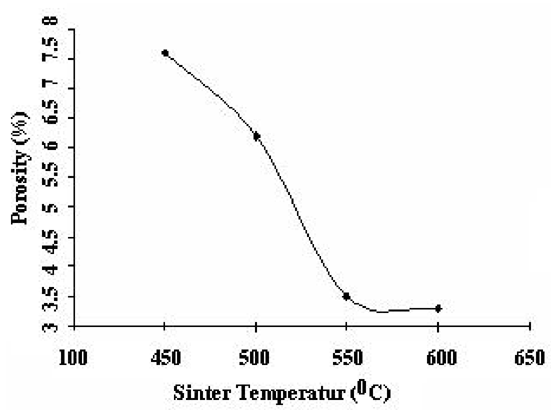



**Figure 5. Effect of sintering temperature on porosity.**

As shown in Figure 4, the increasing of sinter density as the sintering temperature increase. However, this phenomenon can be perceived within porosity of substrate that decreases as the sintering temperature increase (Figure 5). At this stage, the gases and lubricant which are trapped in the constituent particles were expanded (generated) within the porosity known as the "degassing process" [3, 4]. This phenomenon is induced by the increasing of atom diffusion; therefore the particle transport phase will substitute (freer) the pore of composite. The porosity reductions in phase material transport process will obtain the "liquid bridge" as a binder agent.

The density in P/M processing route is also affected by pressure process [4-6]. The mechanical forces bond of constituent particle after compaction (pressure) process is obtained by the adhesive-cohesive forces bond. The other possible of cohesive mechanism include: the interlocking (roughness of particle interface), electrostatic, and van der Walls forces bond. The interlocking is the phenomenon when the homogeneity of particle interface obtained, therefore the composite become compacted (solid state reaction). For the electrostatic forces bond is obtained when the compaction and friction of particle carried out. While, van der Walls forces bond is obtained when the dipole fluctuation (forces bond effect) within the reinforce and matrix powder takes place. The pressure applications addressed to the powder obtain three possibilities of bond types, namely: spherical-spherical type (if the pressure application is below the matrix-reinforcement yield strength), spherical-planar type (if the pressure application is between the matrix-reinforcement yield strength), and planar-planar type (if the pressure application is above the matrix-reinforcement yield strength).

The atomic diffusion to the particle surface barrier, after sintering process, is critically important to the increasing of the cohesive of particle. The atomic diffusion, which is obtained by the formation of liquid bridge at the pores, is located around at the interfacial of particles, and it reduces the porosity of composite. Generally, the particles show the increasing of shrinkage of composite as the compaction increase. Therefore, the composite porosity decrease as the sintering temperature increases as shown in Figure 5. Other phenomenon of porosity obtains when the metal (matrix) shrinkage (consolidation or gas trapping) carried out; which affected to the mechanical strength of composite. Meanwhile, the volume fraction of reinforce will pursue the liquid flow at dendrite phase; and it will prevent the metal particle movement at metastable (metasolid) state.

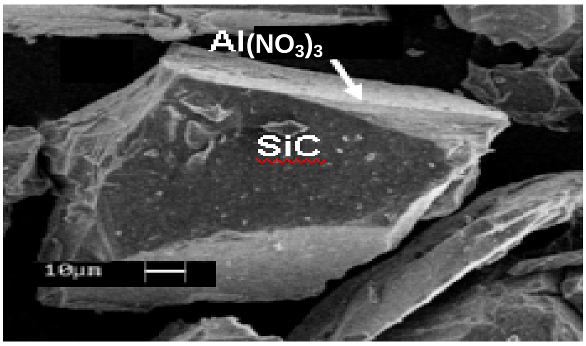



**Fig.6. SEM Micrograph of SiC particle after coated with $Al(NO_3)_3$**

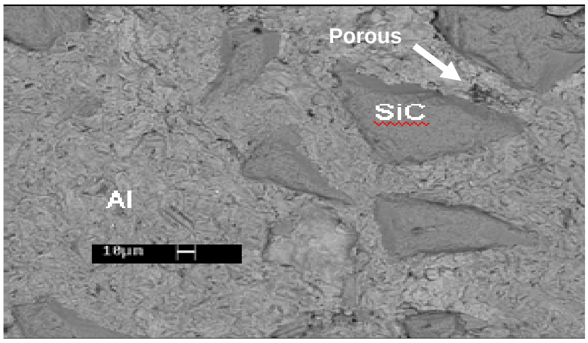



**Fig.7. SEM Micrograph of Al/SiC sample after coated with $Al(NO_3)_3$; compacted at 120 MPa and sintered at 600 $^0$C for 1 h**

The microstructure of $SiC_p$ after $Al(NO_3)_3$ coated processes is shown in Figure 6. The thickness of SiC surface after coating process is relative homogeny. For the other surface of $SiC_p$ which is not fully coated will have the less wettability to the matrix. It affected to the low density in the composites. Other phenomenon of the decreasing of density is the formation of pore when compactions take place. The existence of metal oxide (dispersant) at $SiC_p$ surface affects to the bond quality of composite [5] (as evidenced by the SEM photomicrograph in Figure 6). It also affects to interface and adhesive reactivity of SiC. The dispersant (metal oxide), which tends to forming the inert phase, decrease the adhesive of particle interface, if it compared to the metastable phase.

However, the differences in coefficient of thermal expansion (CTE) among the matrix (Al) and reinforce (SiC) can strongly influence the internal tension in matrix and perfection of the interface [6]. It will also influence to the microstructure properties of matrix, especially the increasing of dislocation density (point defect) (Figure 7). Therefore, the interface characteristic of the constituent is critically determines the quality of composite, which affected by the matrix and fabrication technique. In Figure 7 shows the reinforcement distribution of substrate as sintered at 600$^0$C for 1 h, which the distributions of particle is not fully homogeny. It occurred, when the sintering process take place; yet the local movement and low wetting of reinforce particle is obtained at interfacial tension of particle, therefore the reinforcement is tends to become the cluster or lump.

Figure 8 shows a SEM photomicrograph taken in a section of Al/$SiC_p$ at low sintering temperature (450 $^0$C), at the particle interface shows $Al_4C_3$ phase (the black spot).

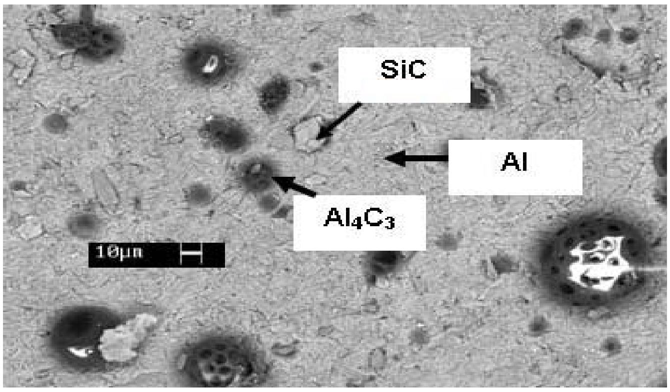



**Figure 8. SEM Micrograph of the Al/SiC sample with $Al_4C_3$ phase after sintered at 450 $^0$C for 1 h**

The existing of aluminium carbide ($Al_4C_3$) phase and the reinforce particle have the order size about 10 – 15 µm. Since, the aluminium carbide is unwanted in fabrication of composite. From Figure 8, it can be explained the formation of $Al_4C_3$ phase as follows:

When the interfacial reaction of the $SiC_p$ to the liquid aluminium, it slowly dissolves, then obtained the silicon and carbon ions: $SiC \rightarrow Si^{4+} + C^{4-}$. Because the high diffusivity and low solubility of carbon in liquid aluminium phase, therefore the matrix is rapidly saturated in carbon and then $Al_4C_3$ precipitation is begin, according to the reaction: $4Al + 3SiC \rightarrow Al_4C_3 + 3Si$. Carbon and Aluminium reaction obtained the $Al_4C_3$, and it produces an adverse effect on physical and mechanical properties of the reinforcement. Since, the unwanted $Al_4C_3$ have the hygroscopic properties that affect to the substrate local corrosion. The formations of brittle phase at reinforce affects to the less contribution as tension transmitter in matrix.

The sintering mechanism include: necking formation of particle, growth of new grain, and shrinkage of substrate. The atomic diffusion carried out at particle interfacial when the heat transfer takes place by sintering process. At this process, the activation energy generated (transfer) to the atoms. So, it causes the atom vibration and the bonding become weakness and the atoms moved to the new position (other lattice), the process called the "diffusion". The process at this phase conducted only at local interface as known the "solid sintering". Then, the extended solid solution formation at interface of crystalline substrate, called the "liquid bridge". Within the liquid bridge, therefore the compaction ability of substrate will expose increasingly.

Sintering is the diffusion phenomenon of particle interfaces in atomic scale; it depends on the surface reactivity in entire particles interaction. For Al/$SiC_p$ fabrication, the interface of matrix (Al) and reinforcement (SiC) which has the inert properties is acting as the barrier in the diffusion process. The particle surface arrangement of SiC by metal oxide $Al(NO_3)_3$ is expected act as a binder agent (Figure 6). So, the diffusion process could be perceived by sintering densities (Figure 7).

In Figure 9, an XRD pattern for Al/$SiC_p$ substrate obtained under condition at 600 $^0$C for 1 h. The $Al_2O_3$ phase, which is affected by $SiC_p$ coating process, is a stable phase and influence to the low wetting of SiC [5]. It will also affect to the low adhesives of substrate (reinforcement and matrix). The $SiO_2$ phase is the phase when SiC coating processes carried out. The contributions of metal oxide coating within the electroless platting and sintering is influence to the increasing of wettability aspect of Al and SiC particle (as evidenced by the SEM photomicrograph in Figure 7).

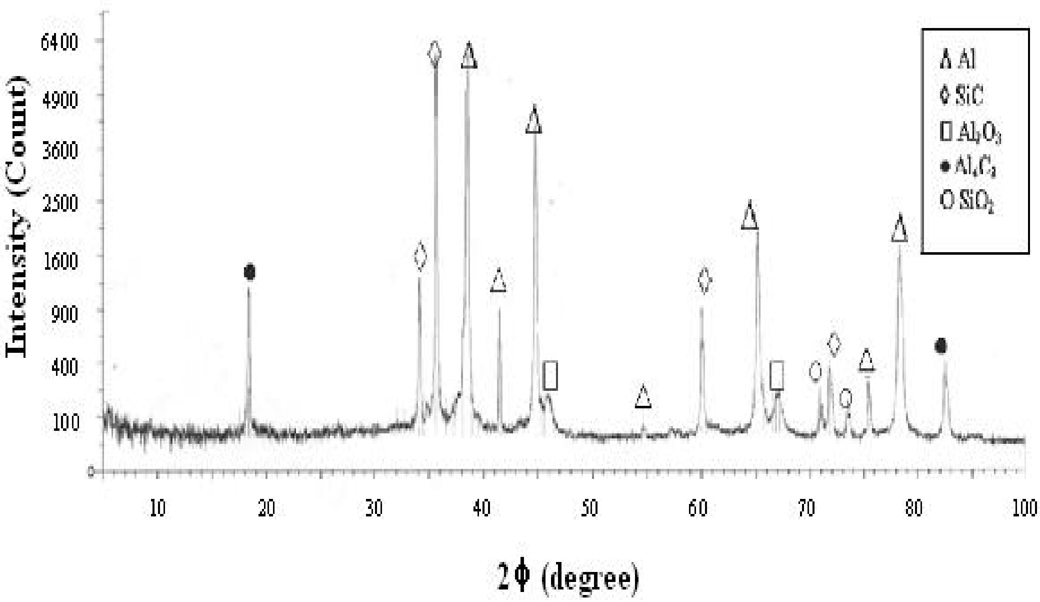



**Figure 9. XRD pattern of the Al/SiC sample compacted at 120 MPa and sintered at 600 $^0$C for 1 h**

## CONCLUSIONS

Results from this investigation show the characteristics of composite in the fabrication of Al/$SiC_p$ by solid state processing (powder metallurgy). Generally, the composite has the main problem in low wettability and adhesives of matrix and reinforce. To increase the wettability of SiC particle, the process arrangement is within the metal oxide coating method using electroless platting. The composite process of SiC by aluminium alloys is investigated under argon atmosphere. The assess of density and porosity of Al/SiC composite show the best compacting pressure and sintering temperature at 120 MPa and 600 $^0$C for 1 h, respectively.

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