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THE EFFECT OF SOL-GEL TECHNIQUE ON THE ALUMINIUM – SiCp COMPOSITE

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ABSTRACT

Due to the low reactivity between the silicon carbide particulate (SiC_p) and Aluminium (Al) interface bonding strength, SiC_p is treated by applying a coating on its surface. The objective of this paper is to study the effect applying ceramic sol coating, which is silica sol via sol gel technique onto SiC_p. The various concentrations applied are 2 wt%, 4 wt% and 10wt% of silica sol. Aluminium metal matrix composite (Al-MMC) reinforced with 10% and 20% volume fraction of SiC_p produced by powder metallurgy technique with sintering temperature of 550°C was studied. The surface morphology of Al-MMC is observed using SEM to examine the particle distribution, porosity and flow pattern. While the degradation of SiC_p due to interfacial reaction between the particulate and Al matrix is examined by x-ray diffraction (XRD). SiC_p degradation is evaluated by the ratio of the SiC to the silicon (Si) XRD peak. From microstructure analysis, the specimens coated with 2 wt% and 4 wt% silica sol coating showed the best result which has uniform reinforcement distribution, lower porosity and better flow pattern. For XRD analysis, the best coating was obtained by 4 wt% silica sol concentration that has greater reaction between Al and SiC .

Keywords: *Sol Gel, Metal Matrix Composites, and Silicon Carbide Particulate, cold pressing*

1.0 INTRODUCTION

Metal matrix composite (MMC) is of great interest especially in the automotive and aerospace industries due to their relatively high specific strength, good dimensional stability and excellent wear resistance. Aluminium (Al) reinforced with silicon carbide particulate (SiC_p) is being developed for various applications. Their attractive properties are such as high modulus, high specific stiffness, high temperature strength, good workability and isotropy. However, their high specific mechanical properties are very much due to the high SiC_p-Al interface bonding strength.

In order to obtain low reactivity between the SiC_p-Al interfaces, SiC_p is treated by applying coating on its surface. These coatings are obtained by sol gel technology, Which is a simple and inexpensive process. It is shown that the mechanical properties of the composite materials are improved. The high-temperature degradation of SiC is the problem faced by MMC. The reaction between liquid aluminium and SiC is $4\mathrm{Al} + 3\mathrm{SiC} \longrightarrow \mathrm{Al}_4\mathrm{C}_3 + 3\mathrm{Si}$. Additionally, the reinforcement phase can distribute uniformly throughout the matrix phase. The effect of sol concentration is studied.

Fabrication of Al-MMC is done by powder metallurgy (P/M) process. P/M gives better properties compared to other conventional MMC processing technique. In order to optimise the P/M process, the effect of processing parameter are studied. In this project, the volume fraction of reinforcement phase is varied.

2.0 EXPERIMENTAL PROCEDURE

The material needed to fabricate Al-SiC_p MMC is Aluminium powder and silicon carbide powder. Eight specimens, which consist of both Al-10 vol% SiCp and Al-20 vol% SiCp, are prepared. All these specimens are either treated with various concentration of silica sol or untreated. The work methodology is shown in Figure 1.

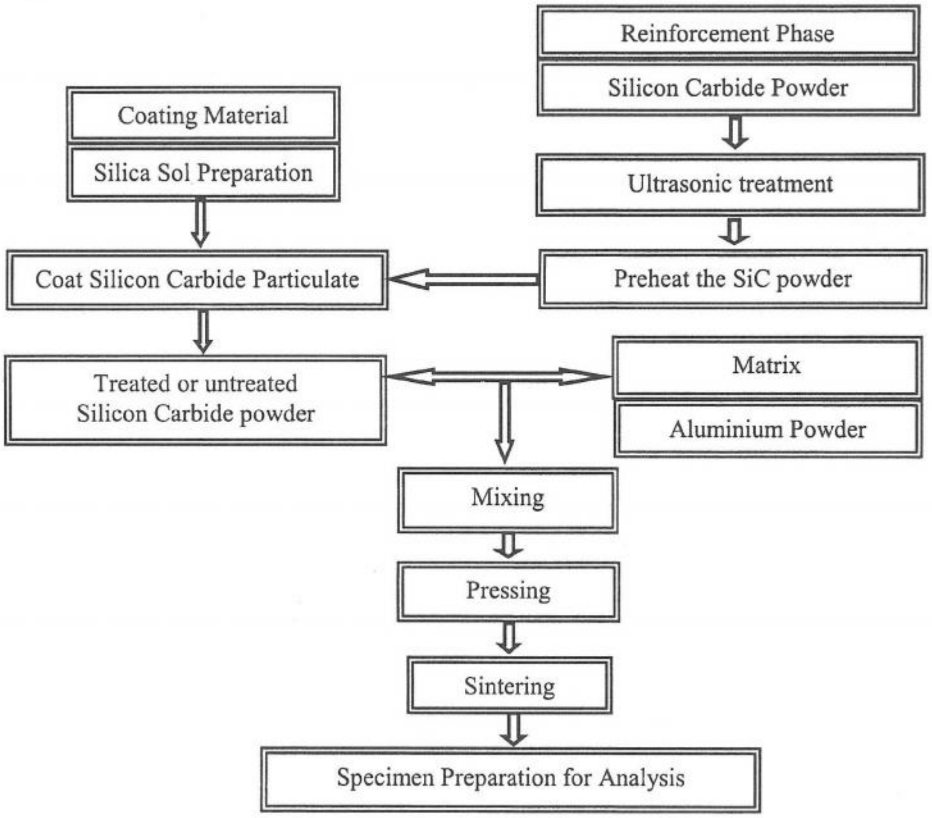

Figure 1: Flow chart of work methodology

Table 1: Details of the specimens prepared

No.	Specimen	Vol% of SiC_p	Silica Sol Concentration
1	A2	10%	Nil.
2	B2	20%	Nil.
3	C2	10%	2 wt%
4	D2	20%	2 wt%
5	E2	10%	4 wt%
6	F2	20%	4 wt%
7	G2	10%	10 wt%
8	H2	20%	10 wt%

The size of the pure aluminium powder used is in the ranges from 63 to 90 μm. The aluminium powder should be free from oxide. The size of the silicon carbide particulate is kept between 12 to 18 μm. The SiC_p powder had already been sieved by using sieving machine into the size of 400 mesh by the supplier. In order to obtain clean SiC_p powder, ultrasonic treatment is done by using ultrasonic machine (Branson 5200). SiC_p powder, which is added to acetone, is vibrated until the SiC_p is cleaned. The vibration process is repeated several times until the acetone poured away is thoroughly clean and clear. Before coating the particulate with silica sol, the particulate surface should be cleaned from oxide or any other impurities. This can be achieved by preheating the SiC_p in furnace for 2 hours at $800\,^{\circ}C$.

In this work, silica sol (SiO_2) is used to coat the SiC_p. As a starting material for silica sol preparation, tetraethylorthosilicate or TEOS [Si $(OC_2H_5OH)_4$] is used. Other than this, ethanol-95% (C_2H_5OH) distilled water and acid hydrochloric (as the catalyst) is used. The compositions of silica sols coating are varied in terms of concentration. Three concentrations of silica sol are prepared, which are 2 wt%, 4 wt% and 10 wt%.

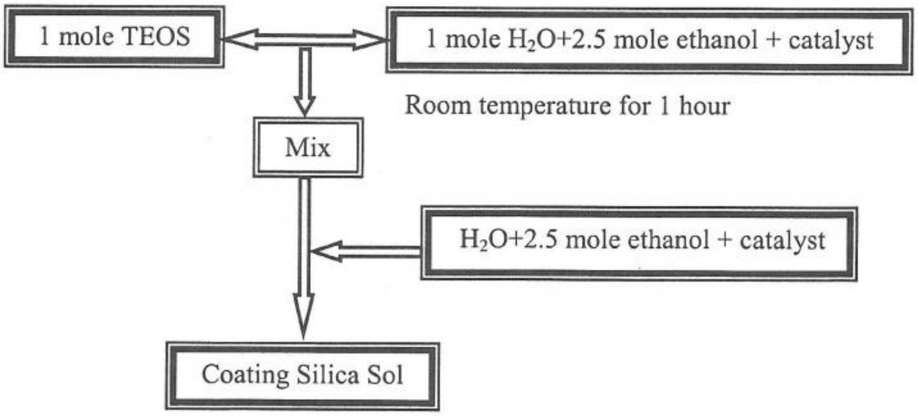

Figure 2: Overall process of preparing silica sol [3]

Silica coating samples were prepared by partially hydrolyzing TEOS (FLUKA), with one mole of water per one mole of TEOS in half of the total amount of ethanol required. A small amount of catalyst (HCl for obtaining acidic pH) was added into the water. After mixing for about one hour in room temperature, the rest of the ethanol and water with catalyst was added into this mixture, as shown in Figure 2. Using magnetic stirrer with the highest velocity possible, the mixing is done. Then, stiring the final mixture at room temperature for 1 hour by using magnetic stirrer. The colloidal silica sol produced is diluted to obtain the required concentration, which is 2 wt%, 4 wt% and 10 wt% silica sol. The final SiC_p concentration in the sol is equal to 6g of SiC_p in 20 ml of the colloidal silica sol. Disperse the required amount of SiC_p into the correct amount of colloidal silica sol and mix in magnetic stirrer for 30 minutes. The treated SiC_p are dried at vacuum furnace at 40 for 10 hours. During the particulate dispersion and drying, the silica sol is deposited on the particulate surface. After drying, the particulate mass is broken by mortar and pestle. Therefore, coated SiC_p is obtained [5]. After silica sol is applied on SiC_p, the SiC powder is mixed with aluminium powder. Before the mixing process, the container and the zirconia ball needed to be in a clean condition. This can be achieved by cleaning it with a cleaner. The parameter for mixing 10 vol% and 20 vol% of SiC_p is 6 hours of mixing time and speed of 55rpm. The weight ratio of the ball to the composite powder is 3:5. After mixing, the powders is poured carefully into an airtight bottle. This is to prevent contamination of the powder. After mixing, the powder annealing process is done in a furnace with continuous inert gas flowing inside it. This process is done at $500\,°C$ for 1.5 hours. The purpose of powder annealing is to increase the powder compressibility [2]. It has been established in previous works done by G. O'Donnell that annealing procedure softened the aluminium powder and reduced the absorbed H_2O content.

Subsequently, Al-SiC_p MMC specimens are cold compacted using Herzog Pressing Machine into a capsule size before sintering at $550\,°C$. The specimens are then cooled and sectioned. The sections are then ground to form a planar surface for x-ray diffraction (XRD) analysis. XRD testing is the quantitative evaluation for the extent of SiCp degradation. Polished samples are then prepared for microstructure analysis by using scanning electron microscope (SEM).

3.0 RESULTS AND DISCUSSION

3.1 Reinforcement Distribution

The reinforcement distribution is fairly uniform for all the top surfaces of the samples. For specimens with 2 wt% and 4 wt% of SiO_2 coating, the SiC distribution is more uniform compared to untreated specimens. Figure 3a shows the SEM micrograph of the uncoated specimen. There seems to be clustering of SiC_p. For Figure 3b (2 wt% SiO_2 coating), there is no clustering of SiC_p.

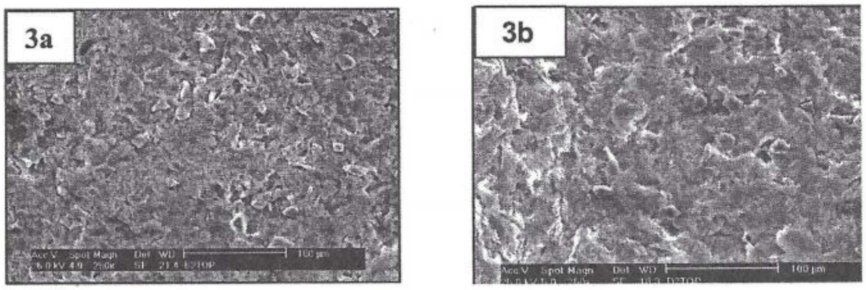

Figure 3: SEM micrograph (250×) showing the top surface of specimens (a) B2 (b) D2

For the cross sections of the specimens, specimens with 2wt% SiO_2 coating (Figure 4a) show good distribution of SiC_p compared to uncoated specimen (Figure 4b). There was higher percentage of SiC_p clustering in specimens with 20% of reinforcement compared to specimens with 10% of reinforcement (Figure 5a and Figure 5b).

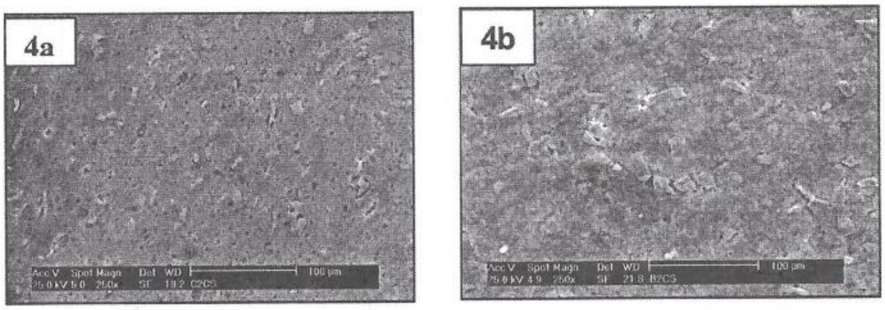

Figure 4: SEM micrograph (250×) showing the cross section of specimens (a) C2 (b) B2

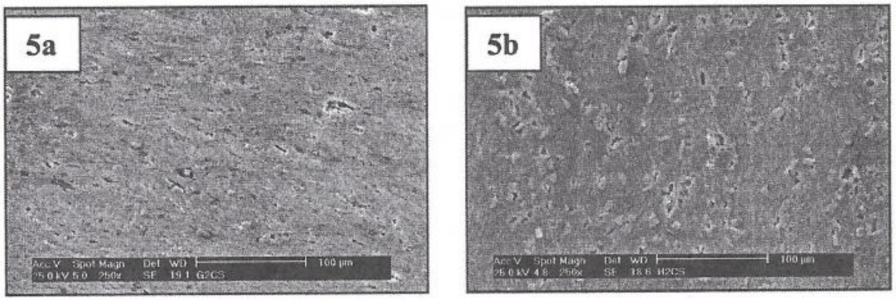

Figure 5: SEM micrograph (250×) showing the cross section of specimens (a) G2 (b) H2

Observation at the bottom surfaces of the specimens showed that clustering appears for samples with higher percentage of reinforcement and the coated specimen showed better particulate reinforcement compared to uncoated specimen. (Figures 6a and 6b)

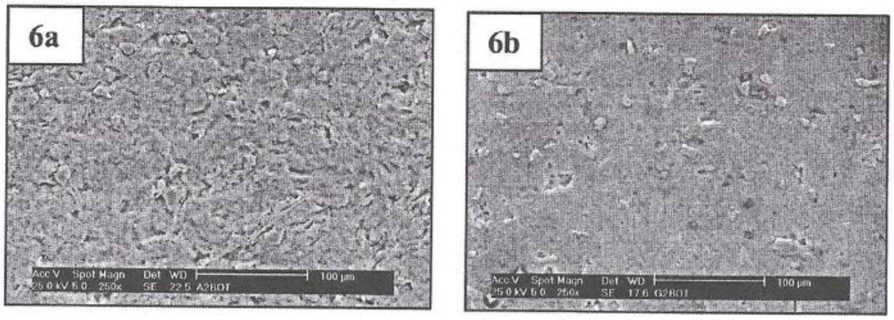

Figure 6: SEM micrograph (250 ×) showing the bottom surface of specimens (a) A2 (b) G2

The reason specimens with coated reinforcement have better reinforcement distribution is that the silica particles of the sol have been adsorbed onto the surface of the particulate. Their repulsive charges have caused the particulate separation and therefore uniform dispersion. The uniformity is maintained when blended with the aluminium and compacted. There is a reduction in the concentration of agglomerates by the dispersing treatment. Thus, it provides an increased number of SiC_p that are free to flow with the aluminium matrix during the compaction. [5]

The reason specimens with higher percentage of SiC_p showed higher clustering because when the SiC volume fraction increased, the available space between SiC particles decreased. Thus, 20 volume percent SiC_p-Al showed increasing propensity of interaction between SiC particles and less homogenous particle distribution. [2]

3.2 Porosity

The micrographs showed that the porosity problem still exist. Most of the porosity occurs near by the SiC_p. The porosity that exists in the specimens is further detailed with the high magnification of the micrographs. Figure 7a, (untreated particulate) shows that the SiC_p is not surrounded by the aluminium matrix. This is evident by the uniform holes seen adjacent to the larger SiC particles. Theoretically, this condition is not good. The force loaded on the specimen cannot be transferred from the matrix to the reinforcement phase. In the composite, the deformable matrix powder could be partially shielded by SiC_p from the total applied pressure or force. Figure 7b shows that specimen C2 (2 wt% SiO_2 on SiC_p) is being uniformly surrounded by the aluminium matrix. The load or stress can be transferred better from the matrix to the reinforcement. There exist a good interface between the matrix and the reinforcement phase. The same goes to specimen F2 (4 wt% SiO_2 on SiC_p) and specimen H2 (10 wt% SiO_2 on SiC_p).

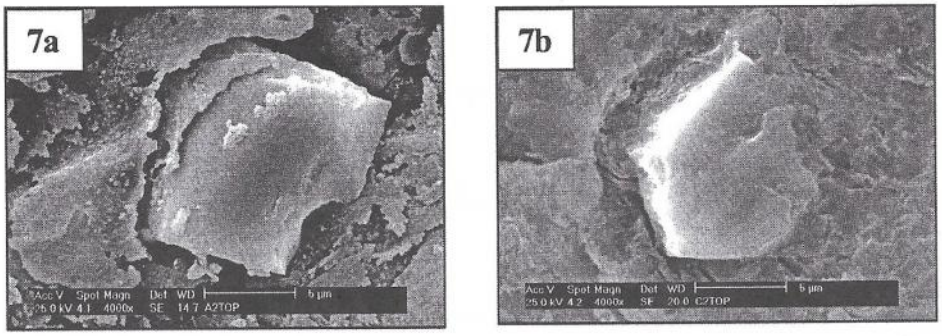

Figure 7: SEM micrograph (4000 ×) showing the top surface of specimens (a) A2 (b) C2

For the cross section of the specimens, the coated specimens (2 wt%, 4 wt% or 10 wt% of SiO_2) showed that the particulates are uniformly surrounded by the matrix phase. The uncoated specimen is surrounded by holes. The best result is obtained by specimens with 2 wt% SiO_2 coating. (Figure 8a and 8b)

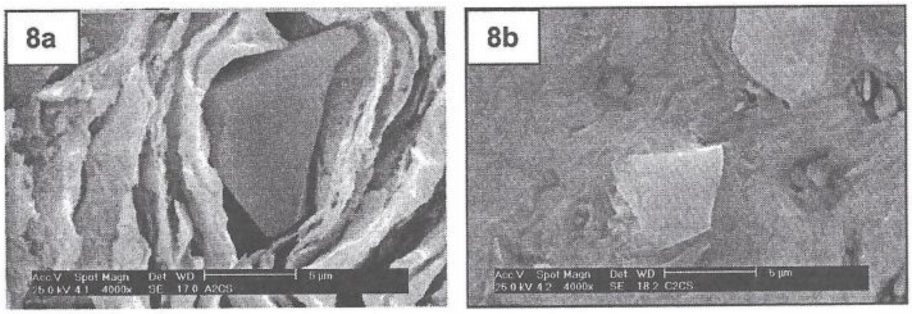

Figure 8: SEM micrograph (4000 ×) showing the cross section of specimens (a) A2 (b) C2

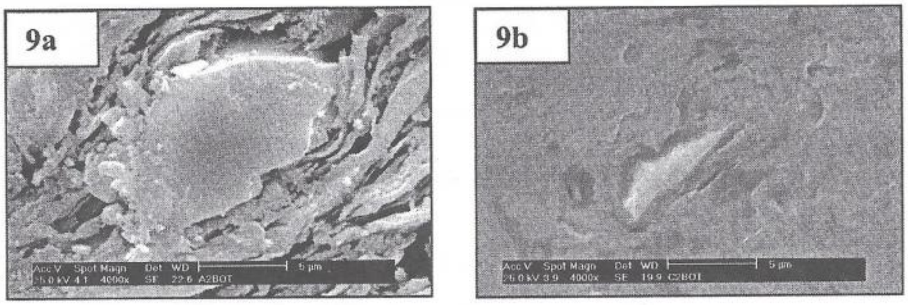

Figure 9: SEM micrograph (4000 ×) showing the bottom surface of specimens (a) A2 (b) C2

Figure 9a shows the micrograph of bottom surface of uncoated specimen. It is very obvious that the particulate is "spoiled" or fractured. Smaller sizes of the particles are seen adjacent to the larger SiC particle. This specimen does not have a good interface. It may be due to interaction between the matrix and the reinforcement phase. However, this condition does not occur in the coated SiC_p (Figure 9b). All the other coated specimens show good interface between the matrix phase and the reinforcement phase.

3.3 Flow Pattern

All the micrographs show that the aluminium matrix flowed around the reinforcement phase. However, the micrographs of cross sections show an obvious pattern where the direction of flow followed the direction of the press. The matrix flows from higher pressure or higher force region to lower pressure or lower force region. According to G. O'Donnell [2] SiC_p surface on the fracture surface were surrounded by the matrix material. The flow of matrix around the reinforcement reduced the properties of the materials.

Pressure is an important parameter that affects the flow. Higher pressure shows lower effect of flow. Higher energy is needed to reduce that effect of flow. The top surfaces have less effect of flow if compared to the cross section area. (Figure 10a and Figure 10b). Other than this, the volume fraction of the reinforcement also influences the flow pattern. The higher the volume fraction, the more prominent is the flow pattern.

However, for the micrographs presented, most of the specimens have a clear flow since all of them are fabricated by using cold pressing method. There is less observation of flow pattern for hot pressed samples. Therefore, temperature also has a control over the flow pattern of the aluminium matrix.

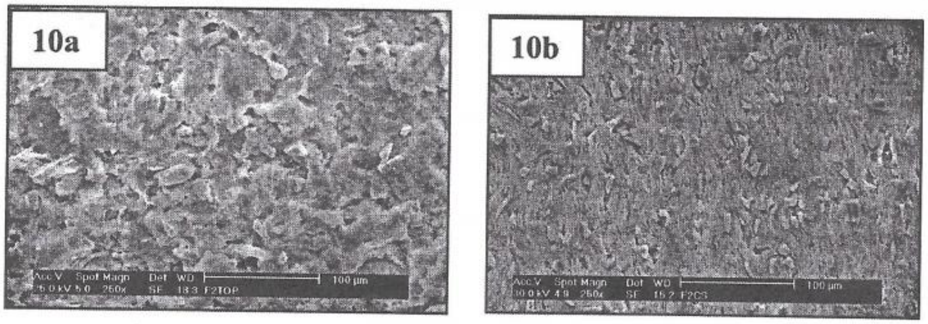

Figure 10: SEM micrograph (250×) showing the (a) top surface of specimen F2 (b) cross section of specimen F2

3.4 Reaction Product

Reaction between aluminium and SiC_p will form a reaction product. The reaction is such as follows, $4Al + 3SiC \rightarrow Al_4C_3 + 3Si$. The higher the volume of aluminium, the higher is the formation of intermetallic reaction product (Al_4C_3) [3]. When the volume fraction of SiC increased, the total intermetallic reaction product is reduced due to less aluminium.

According to Shin *at el* [4], intermetallic particles are formed during consolidation. The volume fraction will increase as the consolidation temperature increased. Their distribution was inhomogeneous and amount was related to the consolidation process. Sintering process is the process that will form more intermetallic particles. Therefore, consolidation temperature had to be kept as low as possible to reduce this type of problem.

The existence and amount of the reaction product cannot be made clear by only observing the micrographs. To get a more accurate result about the reaction product or SiC_p degradation, XRD results will give a good explanation. According to the research done by E. Nyberg [1] in Riso National Laboratory, the degradation of SiC_p can be evaluated by the ratio of the SiC to silicon XRD peak. The lower the SiC/Si ratio, the greater the reaction between the aluminium and SiC.

In this study, the XRD peaks obtained were SiC peak, $Al_4Si_4C_7$ peak and SiO_2 peak. $Al_4Si_4C_7$ (Aluminium Silicon Carbide) is a compound formed by the mixtures of Al_4C_3, SiO_2 and C. The reaction is $Al_4C_3 + SiO_2 + C \rightarrow Al_4Si_4C_7$. The amount of Al_4C_3 present can be calculated by using the ratio of $Al_4Si_4C_7$ to SiO_2. The severity of the degradation of SiC_p can be evaluated by dividing SiC intensity to Al_4C_3 relative intensity. Similar to the work done by E. Nyberg, the higher the ratio of SiC to Al_4C_3, the better is the protection of SiC_p. This is because less reaction product is produced.

The result in Figure 11 showed that the best protection is obtained when the coating concentration is 4 wt% SiO_2 sol. Both specimens E2 (Al-10 vol% SiC treated with 4 wt% SiO_2 sol) and F2 (Al-20 vol% SiC treated with 4 wt% SiO_2 sol) showed the best result. From the graph, 20 vol% SiC is higher than for 10 vol% SiC. This is because when the volume percent of SiC increased, the volume percent for Al matrix is lower. The lower the amount of the Al matrixs; the less is the formation of the intermetallic reaction product and also the SiC degradation. Figure 12 shows the XRD result for one of the specimens. All the specimens have the same peak and only varied in terms of the intensity.

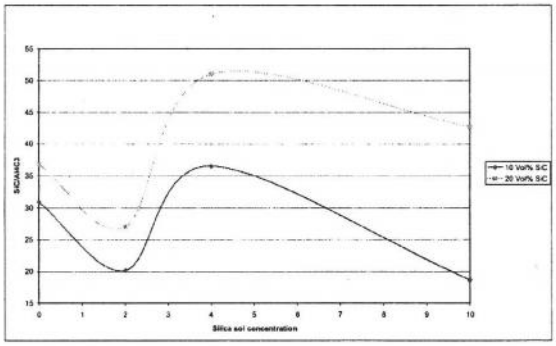

Figure 11: Graph for optimum coating concentration

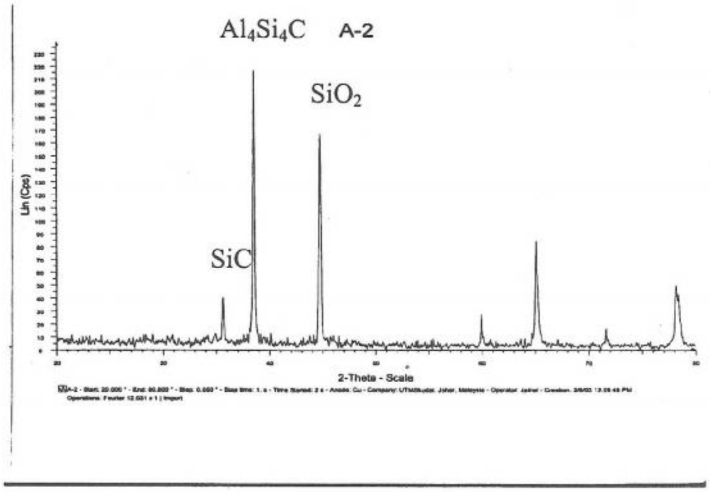

Figure 12: XRD result for specimen A2

4.0 CONCLUSION

By observing the micrographs obtained by Scanning Electron Microscope, 2 wt% and 4 wt% of silica coating shows good result in terms of SiC_p distribution, porosity and aluminium flow. As the result from X-ray diffraction analysis, the best coating is coating with 4 wt% silica sol concentration. Less reaction and degradation is observed for this type of silica sol concentration. The results showed the top surface has higher pressure than the bottom surface. The reinforcement distribution is better in the top surface and the porosity problem is less. Specimens with higher percentage of SiC_p or lower concentration of aluminium show better protection from the environment. This is because intermetallic reaction product was formed duc to reaction between aluminium and the environment.

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