

Effect of Processing Temperature on Pure Al/SiC Metal Matrix Composites Produced by The Lanxide Process

Anne Zulfia

Jurusan Metalurgi, Fakultas Teknik Universitas Indonesia

Kampus baru UI, Depok 16424

E-mail : anne @eng.ui.ac.id

Abstrak

Metal matrix composite dari Al murni dengan penguatan serbuk SiC berhasil diproses melalui proses Lanxide dengan suhu yang berbeda selama 8 jam. Proses ini sederhana tanpa adanya tekanan luar, tetapi gas N₂ dialirkan selama proses. Cairan Al murni telah merembes ke preform yang mengandung 5% volume fraction Mg pada suhu 750, 800, 850, 900 and 1000°C, tetapi infiltrasi tidak terjadi pada preform yang mengandung 3Vf% Mg. Pori terbentuk dibagian atas dari preform terutama pada temperatur yang tinggi (900 dan 1000°C) selama 8 jam. Pori tersebut tidak terbentuk pada temperatur proses yang rendah. Analisa dari MMC yang diperoleh akan diuraikan dalam paper ini dan efek dari temperatur akan diselidiki dan hasilnya akan sepenuhnya di analisa.

Abstract

Pure Al/SiC metal matrix composites (MMC) have been successful produced by the Lanxide process with different processing temperature for 8 hours. The process is simply without aid externally pressure, However, N₂ oxygen free was flushed during the process. Molten pure Al has fully infiltrated to the preform with 5Vf% Mg at 750, 800, 850, 900 and 1000°C, but infiltration did not occur to the preform with 3Vf% Mg. Voids exist above the infiltrated preform in the systems with higher temperature (900°C and 1000°C) for 8 hours. Such voids were not formed in the systems with low temperature. This paper described the investigation into the production of pure Al matrix composites, the effect of processing temperature will be investigated and the resulting will be fully characterised.

1. Introduction :

1.1. Metal Matrix Composites

Metal Matrix Composites (MMC's) have been researched and developed in recent years with the intention of providing cost effective, high performance (high strength, stiffness, wear resistance, etc) materials. Of the many potential metal matrix systems, aluminium alloy matrix composites have been the object of much research, primarily due to the lightweight, low cost and ease of fabrication of aluminium [1]. Within the class of aluminium alloy matrix composites there are two distinct MMC's systems, namely: (i). *Discontinuously reinforced composites, with the reinforcement taking the form of particle, a platelet, a whisker, or a chopped fibre.* (ii). *Continuous fibre reinforced composites, with the*

reinforcement being a unidirectional or a multidirectional array continuous fibres.

Both solid and liquid state processes have fabricated discontinuously reinforced aluminium alloys. Solid-state processes include powder metallurgy routes wherein the metal and ceramic powders are mixed and then hot-pressed [2]. Liquid state processes include compo casting (blending of ceramic powder and molten aluminium, agitating and casting) and pressurized liquid metal infiltration [3-5]. The solid state processes have been most successful to date, but are costly. Liquid metal processes have the potential to be more economic, however, the non-wetting nature of many ceramics by molten aluminium, which results in poor ceramic/metal interfaces and incomplete infiltration, is an obstacle [6,7].

Continuous fibre composites have also been produced by both solid and liquid state methods. The primary solid-state process is diffusion bonding where reinforcing fibres are sandwiched between sheets of aluminium alloy to form a tape. The composite is then formed through two hot-pressing operations, the first to consolidate the tape and the second to laminate several tapes together. Liquid phase processing of continuous fibre MMC's involves liquid-metal infiltration. The infiltration can either be pressure assisted, such as with squeeze casting or vacuum assisted [7,8].

The attraction of liquid infiltration routes is that the perform may be conveniently shaped or machined to the desired shape of the composites, resulting in a near net shape component after liquid metal infiltration. Furthermore, the method is fast compared to powder metallurgy. However, liquid metal infiltration typically involves the evacuation of the preform prior to infiltration. The evacuation steps means that vacuum chamber is necessary for the preform. In most cases, the infiltration is carried out with the help of gas (isostatic) pressure, i.e. pressure casting or pressure infiltration. The pressure application means that compressor and a pressure chamber and a compressor makes the equipment for liquid metal infiltration quite expensive.

1.2. LANXIDE process

The process was first developed in 1986 in the Lanxide Corporation of Delaware, USA, for the production of Ceramic Matrix Composites (CMC's). The original version of the process involved the oxidation of a bulk molten metal by a gas via a directed growth process to produce an interconnected ceramic reaction product that may contain several percent of residual metal. There have been many studies published on this version of the process (see, for example, [9-19]).

More recent developments of the process have involved adapting it to produce metal matrix composites by pressureless infiltration of molten metal into preforms consisting of reinforcing particulates (SiC or Al₂O₃). Infiltration is

reported as occurring spontaneously without the aid of any externally applied pressure or vacuum apparatus hence providing cost effective processing. It is claimed that pore free composites with high structural integrity can be produced [20]. Lanxide type processes, whether used for the production of CMC's or MMC's depend on the presence of dopant elements. For CMC's production the dopant must have a high vapor pressure at the reaction temperature (e.g. Mg, Li, Na) [1]. Magnesium is also used as a dopant for the production of MMC's. Previous experimentation identified that there exist two requirements for spontaneous infiltration to occur namely (i) that Mg should be added to the particle reinforcement and (ii) that the atmosphere contain nitrogen and be nominally oxygen-free [20]. However, meeting these two requirements does not ensure infiltration. The correct combination of the various process variables, such as the composition of materials, the process temperature, the process time, and the nitrogen content of the atmosphere, must be employed.

Originally the process was used to produce ceramic matrix composites (CMC's). In this case the reaction product is formed initially on the exposed surface of a pool of the molten metal and then grows outward, fed by transport of additional metal through microscopic channels in the ceramic product of the oxidation reaction between the parent metal and the gas phase oxidant [9,21]. A crucial feature of this process is the need to include one or more dopants to stimulate the unusual oxidation behavior of the parent metal. Typical dopant concentrations range from less than one to a few percent by weight. Also it is frequently found that this oxidation phenomenon occurs within a limited temperature interval, located above the melting point of the parent metal and not necessary extending to the melting point of the reaction product [21]. Two dopant types are recognized; initiators which allow the metallic transport mechanism to begin and accelerators which increase the metallic transport kinetics and the growth rate of

ceramics [22]. The initiator dopants are required to initiate and maintain the growth process and it is this feature that distinguishes it from reaction bonding processes for ceramic production. Growth is limited by the presence of inert (barrier) powder which may be use to define the shape of desired product [9].

Once started under appropriate conditions, the Lanxide process appears to grow composites of uniform microstructure as long as (i) molten metal and oxidant are available to sustain the process, and (ii) the process temperature is maintained. Both the microstructure and the rate of growth are independent of time (and the thickness of the reaction product), and the rate of growth depends upon the metal, oxidant, dopants and processing temperature employed.

The Lanxide process uses raw metals and oxidants of commodity level purity, and in some cases it operates at moderate temperatures (e.g. 1400K or less in the case of Al₂O₃/Al). These factors are economic significance, as is the absence of shrinkage due to densification. Lanxide materials exhibit little or no ductility owing to their characteristic interconnected ceramic skeleton, but they can be produced with a variety of densities, tensile and compressive strength, hardness, elastic modulus, fracture toughness, thermal conductivities, and thermal expansion coefficients. An example is the excellent high temperature strength of Al₂O₃/Al Lanxide materials as compared to sintered Al₂O₃ [23].

2. Experimental Procedure

2.1. Sample Preparation

Samples were prepared using pure aluminium casting (99.8 wt%, and microstructure is shown in fig. 2.1). The chemical composition of this as cast has been analysed by Inductively Coupled Plasma (ICP) method as indicated in Table 2.1). Magnesium metal powder was used as an external dopant in order to promote the wetting system between pure Al and SiC ceramic particles. The aluminium was sliced into blocks 30 x 15 x 10 mm in size (≅14 gram mass). and washed in deionised

water using Kery KS 100 Ultrasonic Cleaner. The prepared block was placed on top of a bed of SiC powder (Sigma Aldrich) mixed with Mg dopant in an alumina crucible (fig. 2.2)

Table 2.1 : Chemical analysis of pure Al checked by ICP method.

| ELEMENT | AMOUNT (%-wt) |
|---------|---------------|
| Cu | <0.02% |
| Zn | 0.03% |
| Fe | 0.04% |
| Mn | <0.02% |
| Ni | <0.02% |
| Ti | <0.02% |
| Cr | <0.02% |
| Mg | <0.02% |
| Si | 0.03% |
| Na | <0.001% |

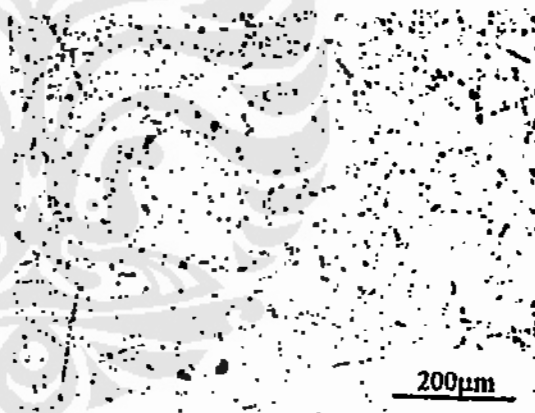


Fig 2.1. Microstructure of pure Al as casted showing a few impurities and porosities.

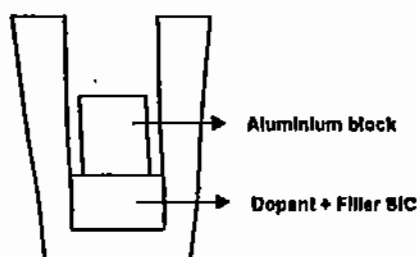


Fig 2.2. Schematic diagram of prepared sample.



Fig 2.3 SEM micrograph of SiC shape.

The SiC powder particle size distribution was measured using a COULTER LS., and the average particles size of SiC is $63.3\mu\text{m}$. The particle shape is shown in figure 2.3.

2.2. Firing

Prepared samples were placed into a tube furnace (Lenton Thermal Design LTD Serial No. 4831,UK). Before firing the furnace was cleaned for one night at temperature of $1100\text{ }^\circ\text{C}$ in the air atmosphere. Then nitrogen gas, nominally oxygen free was flushed for 2 hours. After the furnace was exactly cleaned, the prepared sample was placed into the furnace. To prevent unwanted gases from entering the furnace chamber, all experiments were conducted under a slight positive pressure that was achieved by bubbling the exit gas. The furnace was ramped to temperature at a rate of $200\text{ }^\circ\text{C}/\text{hour}$, to the soaking temperature of $750, 800, 850, 900$ and $1000\text{ }^\circ\text{C}$ for 8 hours. After soaking the samples were furnace cooled

2.3. Material Characterization

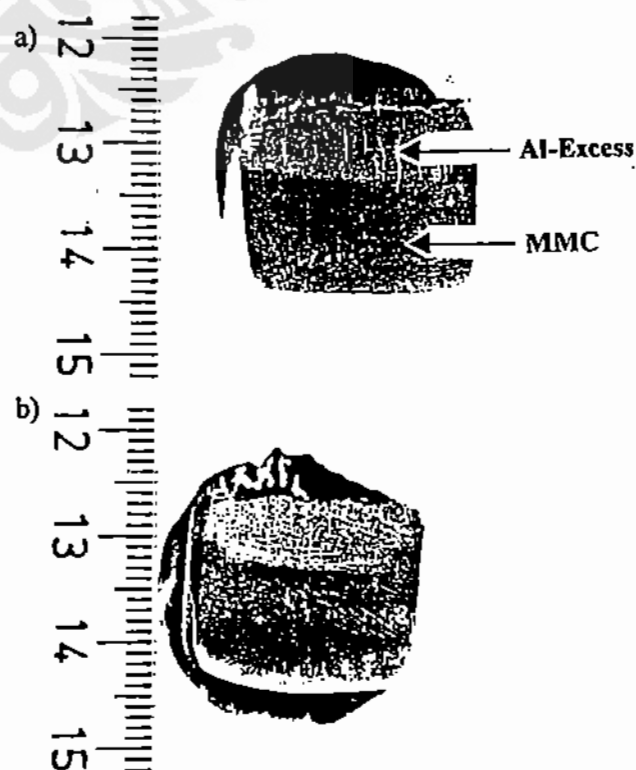
Fired samples were cut transverse to the growth direction of infiltration with diamond saw. The samples were mounted in conductive bakelite using hot mounting equipment and then ground sequentially from 120#, 280#, 400#, 800#, and 1200#. After grinding the samples were polished using $6\text{ }\mu\text{m}$, $1\text{ }\mu\text{m}$ and $0.25\text{ }\mu\text{m}$ diamond paste to $0.25\text{ }\mu\text{m}$ and SILCO finish. Between polishing steps, each specimen

was rinsed with teepol, ethyl alcohol and ultrasonically cleaned for 3 to 5 minutes. The microstructure was examined using both optical (Polyvar Metallurgy) and Scanning Electron Microscopy (CAMSCAN SEM). For SEM samples were carbon coated and they all were analyzed by backscattered imaging. Energy dispersive X-ray Spectroscopy (EDS), Link Analytical was also used in characterizing the microstructure.

The qualitative phase analysis of both starting material and fired specimens were undertaken by XRD using $\text{CoK}\alpha$ radiation at 40 kV and 30 mA (Philips 1710 Diffractometer). The samples were scanned from $20^\circ - 90^\circ$ at a speed of $0.33^\circ/\text{sec}$. The data was collected using the Siemens SIE software.

3. Results and Discussion

The products of Lanxide (pressureless metal infiltration process) into loose beds of SiC containing 5 V% Mg at temperatures of $750, 800, 850, 900$ and 1000°C are shown in figures 3.1.a-e. It seems that full infiltration was achieved at these temperatures. However, infiltration at these temperatures gave two layer products as shown in figures 3.1.a-c, e.



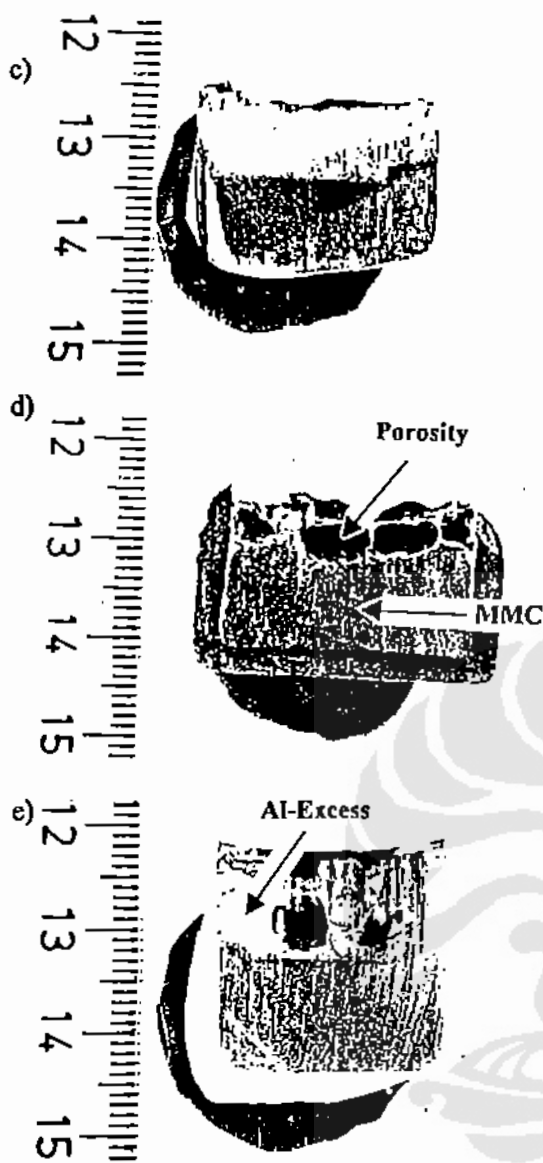


Fig 3.1 Macrograph of molten pure Al infiltrated front into SiC preform after pressureless infiltration for 8 hours. (A:750, B:800, C:850, D:900 and E:1000°C)

Porosity or voids were formed at 900 and 1000°C particularly in the upper layer of the samples. The porosity arises because of shrinkage during solidification of molten aluminium at high temperatures. The MMC product was formed on the bottom layer where molten aluminium has fully infiltrated into SiC preform. The distances of molten Al infiltrated front into preform tend to be increased with increasing temperature and it is found that the maximum molten Al infiltrated into SiC preform occurred at 900°C (Fig.3.2). It's

seemed that infiltration distance at 850 and 1000°C was decreased. The use of a magnesium and a nitrogen atmosphere in conjunction with an appropriate process temperature and time resulted full infiltration.

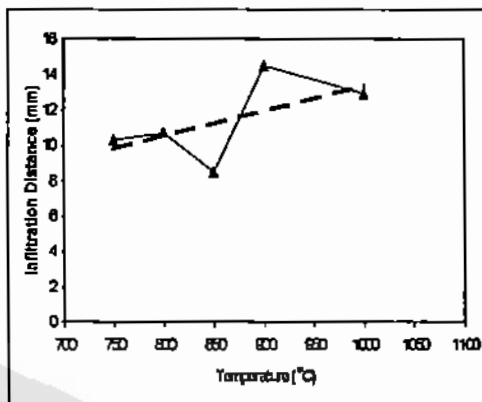
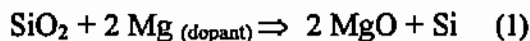


Fig 3.2 Graph showing infiltration distance increase with higher temperature

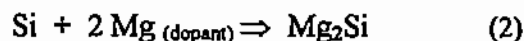
Aghajanian *et al* [20] reported that alloys with higher Mg contents led to infiltrate at lower temperatures, and that under a given set of process conditions there is a critical level of Mg required to induce infiltration. For instance, at 900°C the molten aluminium with 3-wt% of Mg, there is no infiltration occurred, but full infiltration occurred with 5-wt% of Mg. It is found that molten pure aluminium has fully infiltrated to the preform containing 5 wt% of Mg at temperatures of 750, 800, 850, and 1000 °C, even Al still exist on the top layer of the preform. Aghajanian *et al* [20] also demonstrated that the combination of Mg in the alloy and a nitrogenous atmosphere lead to the spontaneous infiltration of aluminium alloys into ceramic fillers. The wetting of ceramic materials resulting from addition of Mg to an aluminium alloy is also well documented [2,3,7]. Several mechanisms are generally discussed when the role of Mg is considered. The addition of Mg can enhance the wetting of a solid surface in three ways, namely : (i) by reducing the surface tension of the alloy, (ii) by decreasing the solid-liquid interfacial energy, and (iii) by promoting a chemical reaction at the solid-liquid interface. Mc

Coy *et al* [3] stated that Mg is effective in reducing the surface tension of the melt and inducing interfacial reactions. According to Aghajanian *et al* [20], this alone does not induce spontaneous infiltration, but a nitrogen atmosphere may cause a further reduction in the surface tension, thus promoting wetting. The results of the present work, demonstrated that the combination of Mg and nitrogen atmosphere has contributed in spontaneous infiltration of pure aluminium into a loose bed ceramic particles (SiC), because this combination has promoted wetting was evident. Additionally, the reactivity of Mg induces interfacial reaction with solid ceramic surfaces.

Microstructure of the infiltration product has been analyzed both top layer (aluminium excess) and bottom layer (MMC's product). Although the top layer is essentially aluminium microstructure, it has been significantly changed compared to the initial aluminium block (compare figs. 3.3a,b,c,d (at the top) with fig. 2.1). The reaction product was analysed by EDS for the range of temperatures investigated, and the Chinese script on the top layer contained of Mg and Si.(fig. 3.4) or Mg₂Si. This precipitate was produced after infiltration at 750, 800, 850 and 1000 °C. No AlN phases were observed in the infiltration products. The Chinese script of Mg₂Si was apparently coarse with increasing temperature. These results simply that Mg and SiO₂ from SiC react to form Mg₂Si in the bottom layer. Then this precipitate diffuses back into the molten aluminium source to give an aluminium layer containing Mg₂Si., as indicated by EDS (Fig. 3.4) and this Mg₂Si was precipitated in the aluminium matrix (see figures 3.3a,b,c,d(top)). There are two possible sources for providing Si to form Mg₂Si in loose bed infiltrated with pure aluminium. One source is the SiO₂ film present on the surface of SiC particles, which could react with Mg in the melt as follows:



The other source is from dissolution of SiC as follows :



These reaction products were formed during the infiltration process at these temperatures.

The microstructure in the bottom layer showed that preform has been fully infiltrated by pure molten aluminium giving a metal matrix composite comprised of SiC particles in an aluminium matrix. Some porosity is formed within the clusters of SiC particles. The microstructures of metal matrix composites are shown in Figs. 3.3a,b,c,d(at the bottom).

The MMC product (bottom layer), generally showed a rather uniform distribution of particles in the matrix (Fig. 3.5) indicating that particles were not disturbed during infiltration. XRD analysis indicated that Al, SiC, Al₄C₃ and Mg₂Si are all present in the MMC product (Fig.3.6). Carotenuto *et al* (24) have explained the formation of Al₄C₃ phase as follows. When the surface of the SiC contacts with liquid aluminium, it slowly dissolves, producing silicon and carbon ions:



Because of high diffusivity and low solubility of carbon in liquid aluminium, the matrix is rapidly saturated with carbon and then Al₄C₃ precipitation starts according to the reaction:



This precipitate Al₄C₃ formed at the Al/SiC interface during production. EDS showed that the interface between aluminium matrix and SiC particles contains aluminium and silicon, here silicon from the SiC particle dissolved to the aluminium matrix to form AlSi precipitate (see figure 3.7 The white phase of AlSi attributable to Fe containing alloy in pure aluminium (0.04±0.01 wt%), while the gray phase of AlSi was formed attributable to no further reaction of Fe. Because of the high solubility of Si in Al, the dissolution of SiC in liquid aluminium is to be expected. The microstructures of SiC/Al specimens

showed the evidence of SiC dissolution in the appearance of Si phase in aluminium (figure 3.7).

The previous work was carried out by Aghajanian *et al* and Oh *et al* [20,25], reported that magnesium significantly improve wettability, suggest that the promotion of interfacial reaction is the most active mechanism for enhancing the wetting of a solid ceramic surface with a molten aluminium alloy. Interaction between the matrix and SiC is higher in Mg containing aluminium alloys than pure aluminium. Several studies have been conducted in reaction of SiC with aluminium alloy on the production of metal matrix composites. Since the surface characteristic of solid substrate play an important role in wetting, gas-solid reaction of ceramic phase should be considered carefully. The wetting mechanism of solid ceramic surface with a molten aluminium alloy has been examined and the Washburn equation, which is often used to measure pore size distributions by

the mercury porosimetry technique [26], provides the pressure, P , required to infiltrate a pore of given radius. It can be expressed as :

$$P = (2\gamma_{LV} \cos \phi) / r$$

Where γ_{LV} is the liquid-vapor surface energy (surface tension), ϕ is the liquid-solid contact angle, and r is the pore or capillary radius. Thus, by changing both the surface tensions of the molten alloy and the liquid-solid contact angle (through interfacial reactions), magnesium additions to an aluminium alloy can affect the pressure required for infiltration. In this study the addition of 5-wt% Mg as an external dopant into SiC particles was significantly promoted the wetting of pure aluminium. It is proven that infiltration at 750, 800, 850, 900 and 1000 °C led to full infiltration of the preform.

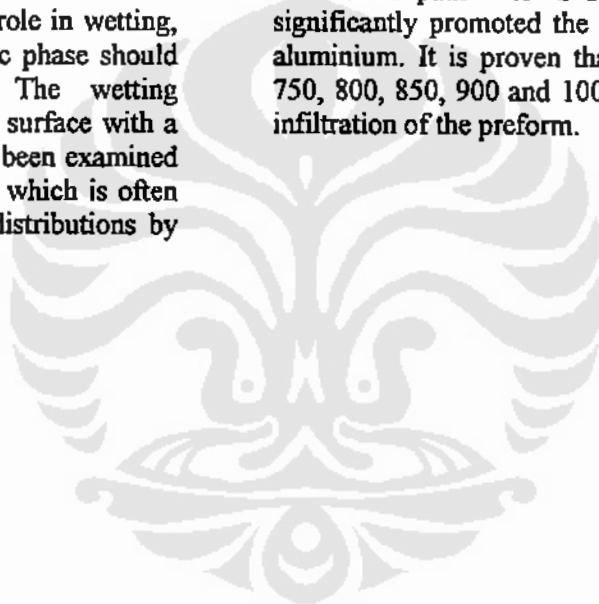
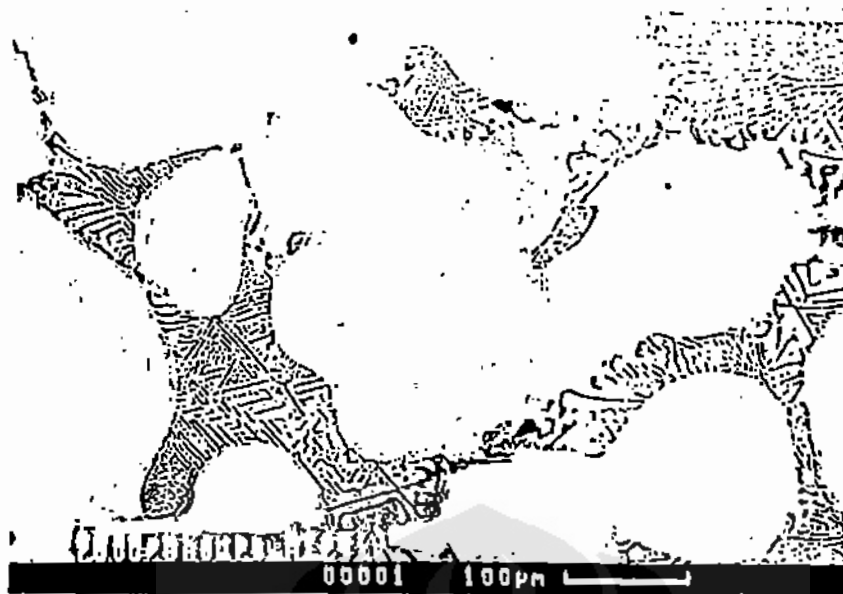
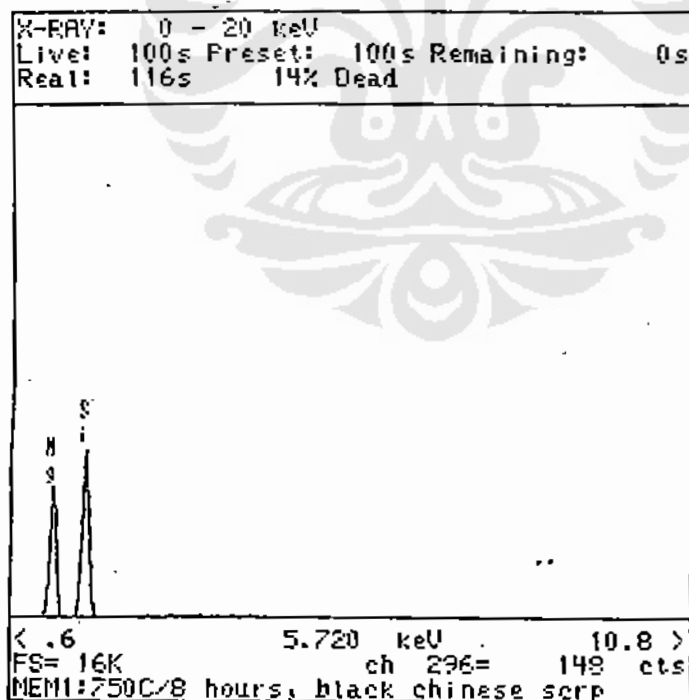




Figure 3.3. Micrographs of the microstructure change after pressureless infiltration at different processing temperature for 8 hours. A)750°C ; B)800°C ; C)850°C, D)1000 °C. (*) Top layer, (**) Botom layer.



(a)



(b)

Figure 3.4. a) SEM micrographs (back-scattered image) showing a Chinese script phase on the top layer after pressureless infiltration at 750°C for 8 hours and b) EDS of this phase contained Mg and Si.

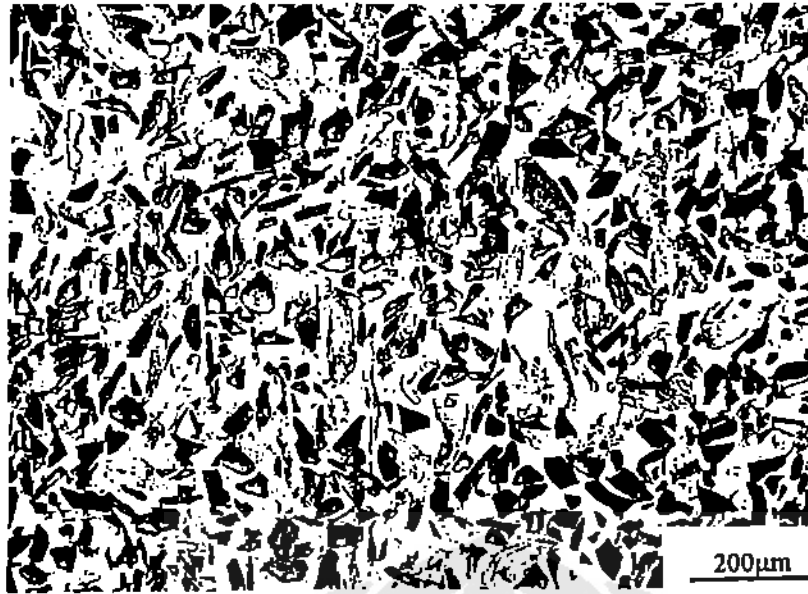


Figure 3.5 : Photomicrograph showing the microstructure of metal matrix composites after pressureless infiltration at 900°C for 8 hours. SiC particles appear slightly uniform distribution in the Al matrix.

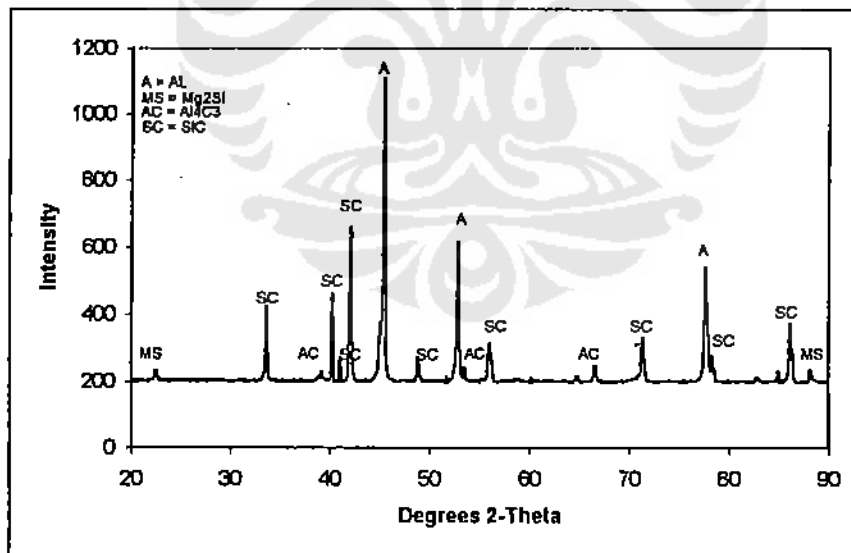
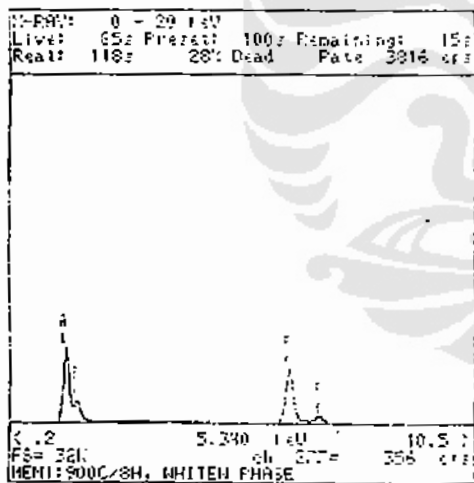


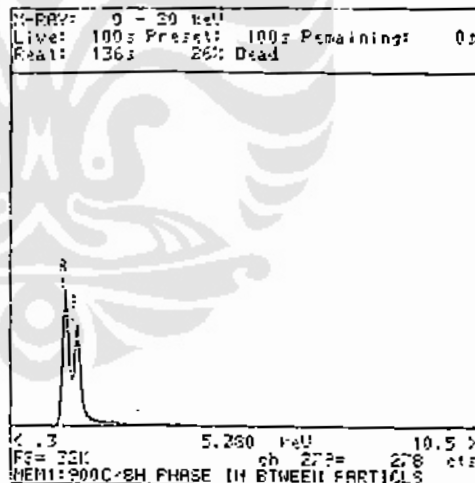
Figure 3.6 : X-Ray Diffraction Patterns for aluminium infiltrated to SiC performed at 900°C for 8 hours



(a)



(b)



(c)

Figure 3.7. a) SEM micrographs (back scattered image) of MMC on the bottom layer after pressureless infiltration at 9000°C for 8 hours, b) EDS of white phase A contained Al, Si and Fe and c) EDS of grey phase B contained Al and Si

4. Conclusion

1. Pressureless infiltration of pure Al into loose beds of reinforcing ceramics at temperature of 750, 800, 850, 900 and 1000°C for 8 hours generated full infiltration, even infiltration at 900 and 1000°C gave porosity at the top layer.
2. The combination of 5wt% Mg as an external dopant and pure N₂ have promoted wetting between pure Al and ceramics

5. Acknowledgement

The author would like to thank DR. R.J. Hand for his direction and technicians at Dept of Eng Materials, The University of Sheffield, UK for their help during my first year of my PhD research at this university.

References

1. F. Delannay, L. Froyen, A. Deruyttere, *J. Mater. Sci*, **22**, 1987, pp. 1-16.
2. S. Das, T.K. Dan, S.V. Prasad and P.K. Rohatgi, *J. Mater. Sci. Lett*, **5**, 1986, pp. 562-564
3. J.W. Mc Coy, C.J ones and F.E. Warner, *SAMPE Q*, **19** (2), 1988, pp. 37-50
4. G.P. Martins, D.L. Olsen and G.R. Edward, *Metall. Trans. B*, **19**, 1988, pp.95-101
5. A. Mortensen and J.A. Cornie, *Metall. Trans. A*, Vol **18**, 1987, pp. 1160-63.
6. D.J. Lloyd, *J. Mat. Sci*, **19**, 1984, pp. 2488-2492
7. J.A. Cornie, A. Mortensen and Mc.Flemming, *A Research Summary, Proceeding of the 6th International Conference on Composites Materials, Part 2*, 1987, pp. 2297-23119
8. T.W. Clyne and J.F. Masson, *Metall Trans*
9. X. Gu and R.J. Hand, *J. Eur. Ceram. Soc*, **15**, 1995, pp.823-831.
10. T. Watari, K. Mori, T. Torikai and O. Matsuda, *J. Am. Ceram. Soc*, **77**(10), 1994, pp.2599-602.
11. E. Manor, H. Ni and C.G. Levi, *J. Am. Ceram. Soc*, **76**(7), 1993, pp. 1777-87.
12. M.K. Aghajanian, N.H. Mac Millan, C.R. Kennedy, S.J. Luszcz, and R. Roy, *J. Mater. Sci*, **24**, 1989, pp. 658-670.
13. M. Sindel, N. Travitzky, and N. Glaussen, *J. Am. Ceram. Soc*, **73**(9), 1990, pp. 2615-18.
14. E. Breval, *J. Am. Ceram. Soc*, **73**(9), 1990, pp. 2610-14.
15. O. Salas, H. Ni, V. Jayaram, K.C. Ulach, L.G. Levi, and R. Mehrabian, *Journal Material and Research*, Vol. **6**, No. **9**, 1991, pp.1964-81.
16. M.S. Newkirk, A.W. Urquhart, H.R. Zwicker, and E. Breval, *Journal Mater Res*, **1**(1), 1986, pp. 81- 89
17. A.W. Urquhart, *J. Mater. Sci. Eng* . A144, 1991, pp. 75-82.
18. S. Pickard, E. Manor, H. Ni, A.G. Evans and R. Mehrabian, *Acta Metal Mater*. Vol. **40**, No.1, 1992, pp. 177-84.
19. E. Breval, M.K. Aghajanian, J.P. Biel, and A.S. Nagelberg, *J. Mater. Sci. Lett*, **14**, 1995, pp. 28-30.
20. M.K. Aghajanian, M.A. Rocazella, J.T. Burke and S.D. Keck, *J. Mater. Sci*, **26**, 1991, pp. 447-454. *A* . **18**, 1987, pp.1519-1530
21. M.S. Newkirk, A.W. Urquhart, and H.R. Zwicklen, *J.Mater Res 1* (1), 1986, pp. 81-89
22. W.E. Lee and W.M. Rainforth, *Ceramic Microstructures Property Control by Processing*, Published by Chapman & Hall, First Edition, 1994, ISBN. 0412 431408
23. M.K. Aghajanian, N.H. Mc Millan, C.R. Kennedy, S.J. Luszcz and R. Roy, *J. Mat. Sci*, **24**, 1989, pp.658-670
24. G. Carotenuto, A. Galo, and L. Nicolais, *J. Mater. Sci*. **29**, 1994, pp. 4967-4974
25. S.Y. Oh, J.A. Cornie, and K.C. Russel, *Ceram Eng Sci. Proc*. **8**, (7-8) , 1987, pp.912-936
26. A. Lane, N. Shah and W.C. Conner, *J. Colloid Interface Sci*, **109**, 1986, p.235