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Fabrication of AA6061/Al_2O_{3p} composites from elemental and alloy powders

K. B. LEE, H. S. SIM, Y.-S. KIM
School of Metallurgical and Materials Engineering, Kookmin University, Seoul 136-702, Korea
E-mail: kblee@kmu.kookmin.ac.kr

J. H. HAN
Division of Materials Science and Technology, Korea Institute of Science and Technology, P.O. Box 131, Cheongryang, Seoul, 130-650, Korea

H. KWON
School of Metallurgical and Materials Engineering, Kookmin University, Seoul 136-702; Center for Advanced Aerospace Materials, Postech, Pohang 790-784, Korea

The tensile properties and microstructures of AA6061/Al_2O_{3p} composites fabricated by the pressureless infiltration method under a nitrogen atmosphere were examined. Since the spontaneous infiltration of molten metal into elemental powders bed as well as alloy powders bed occurred at 700°C for 1 hour under a nitrogen atmosphere, it was possible to fabricate 6061 Al matrix composite reinforced with Al_2O_{3p} irrespective of the type of metal powders. Both $MgAl_2O_4$ and MgO were formed at interfaces between Al_2O_3 and the matrix. In addition, $MgAl_2O_4$ was formed at within the matrix by *in situ* reaction during composite fabrication. Fine AlN was formed by *in situ* reaction in both composites. A significant strengthening in the composites occurred due to the formation of *in situ* AlN particle and addition of Al_2O_3 particles, as compared to the commercial alloy, while tensile properties in the both elemental and alloy powders composites showed similar trend.

1. Introduction

Metal matrix composites (MMCs) reinforced with ceramic phase have become of great interest because of the combined effects of metallic and ceramic materials relative to the corresponding monolithic alloys. Thus various fabrication methods have been developed, such as powder metallurgy, stir casting, pressure infiltration method, etc. Furthermore, in recent years there has been a growing interest in the development of technologies for the *in situ* production of MMCs, such as Lanxide's PRIMEX^{TM} process, Martin Marietta's XD^{TM} process, self-propagating high-temperature synthesis (SHS) and reactive-gas injection [1–15].

The PRIMEX process is an innovative technique for fabricating MMCs by the spontaneous infiltration of molten Al alloy containing Mg into a ceramic filler or preform under a nitrogen atmosphere without the aid of vacuum or externally applied pressure [5–15]. For the pressureless infiltration in this process, both nitrogen and Mg should be needed. Previous studies have investigated that the spontaneous infiltration of molten Al alloy to provide the matrix of Al composites in this process is influenced by Mg content, additional alloying elements, nitrogen concentration in the gas atmosphere, infiltration temperature and time, etc. [5–10]. In particular, at least 1 wt%, and preferably 3 wt% Mg is required.

The PRIMEX process has many advantages relative to conventional MMC fabrication techniques. One of them is easy control of the volume fraction of reinforcement and thus it is possible to fabricate MMC with uniform loading of reinforcement from 10 to 80 volume percent. There are two approaches for fabricate MMC of the lower volume fraction. One is known as the PRIMEX CAST^{TM} process. At first, in this method, MMC of the higher volume fraction is produced by a novel technology involving the pressureless infiltration of aluminum alloy into a mass of ceramic reinforcement particles. Then, this concentrate is loaded to molten aluminum alloy and the melt is mechanically stirred to prepare the MMC diluted to the appropriate level. Following complete dispersion, the melt can be processed by various casting methods. The other is to fabricate MMC using preform (or bed) of the lower volume fraction of reinforcement. That is, reinforcement is mixed with alloy powder to prepare preform (or bed) containing the appropriate volume fraction and then an Al alloy ingot is placed on this preform (or bed). If such an assembly is heated to a required temperature, it is possible to fabricate MMC of lower volume fraction. In addition, it can be used elemental powders instead of alloy powders. Because cost of the alloy powders is higher than that of elemental powders, elemental powders are more economical than alloy powders. Therefore, in this

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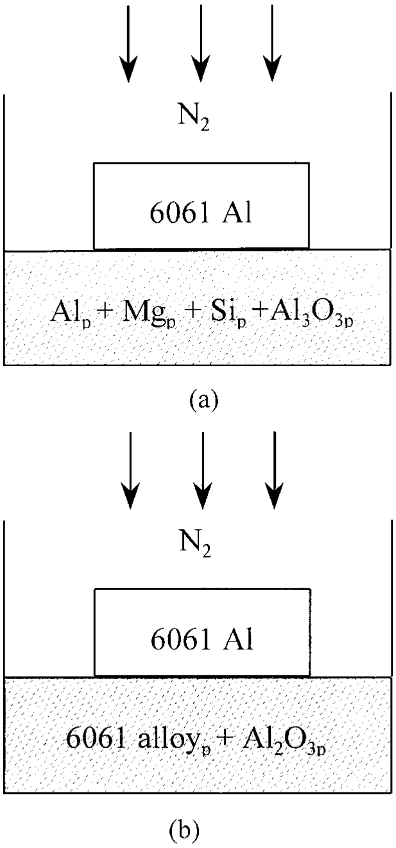

Figure 1 Schematic of the infiltration experiment: (a) elemental and (b) alloy powders composite.

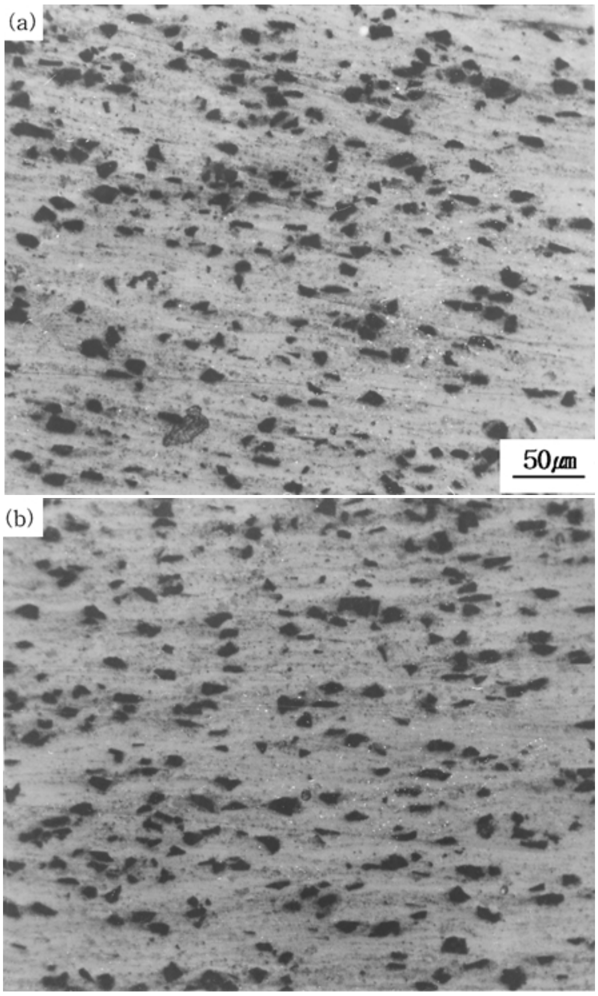

Figure 2 Optical micrographs showing the distribution of Al_2O_3 particles: (a) elemental and (b) alloy powders composite.

study, AA6061/Al_2O_{3p} composites were fabricated by the pressureless infiltration method using both the alloy and elemental powders, and the tensile properties and interfacial reaction were investigated.

2. Experimental procedure

In the present study, two types of metal powder with the composition of AA6061 were used to prepare composites. Fig. 1 shows the schematic arrangement employed for both composites fabrication. The average size of the Al, Mg, Si, and Al_2O_3 particles was about 50, 13, 20, and 10 μm, respectively. In order to prepare the elemental powder mixture, Al, Mg (1.2 wt%), Si (0.8 wt%), and Al_2O_3 (20 vol%) powders were blended by roll mixing in an alumina jar. The alloy powder mixture was composed of Al_2O_{3p} and atomized 6061 alloy powder

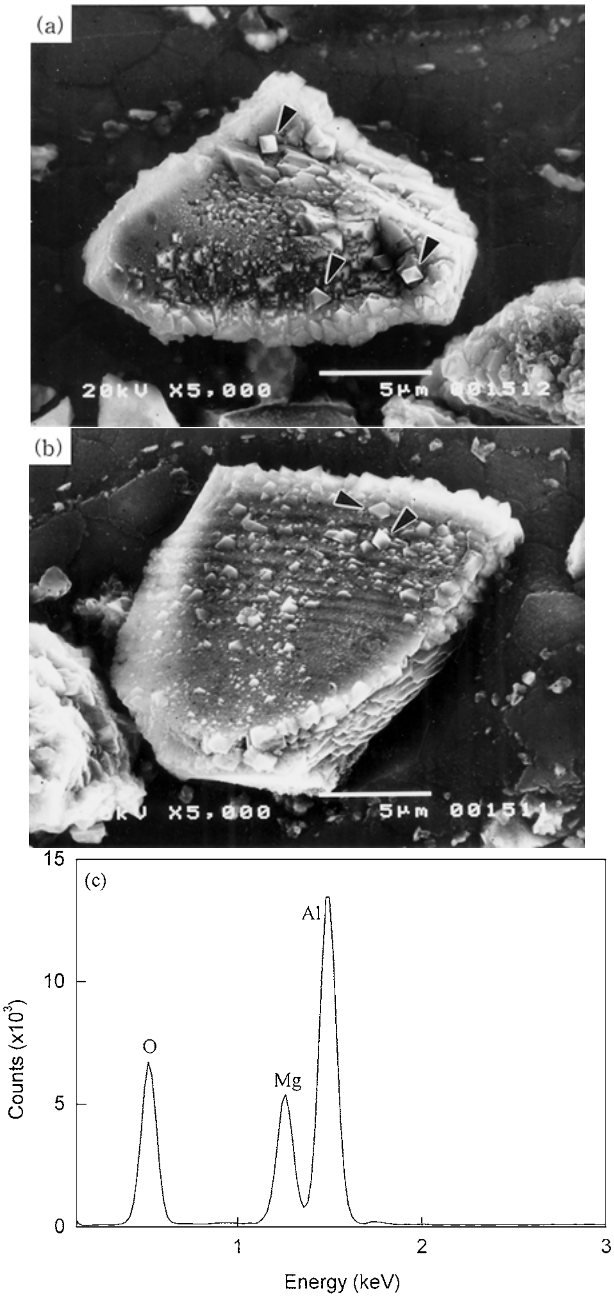

Figure 3 SEM micrographs and EDS profile showing the $MgAl_2O_4$ reaction products, after dissolving away the Al alloy matrix: (a) elemental and (b) alloy powders composite.

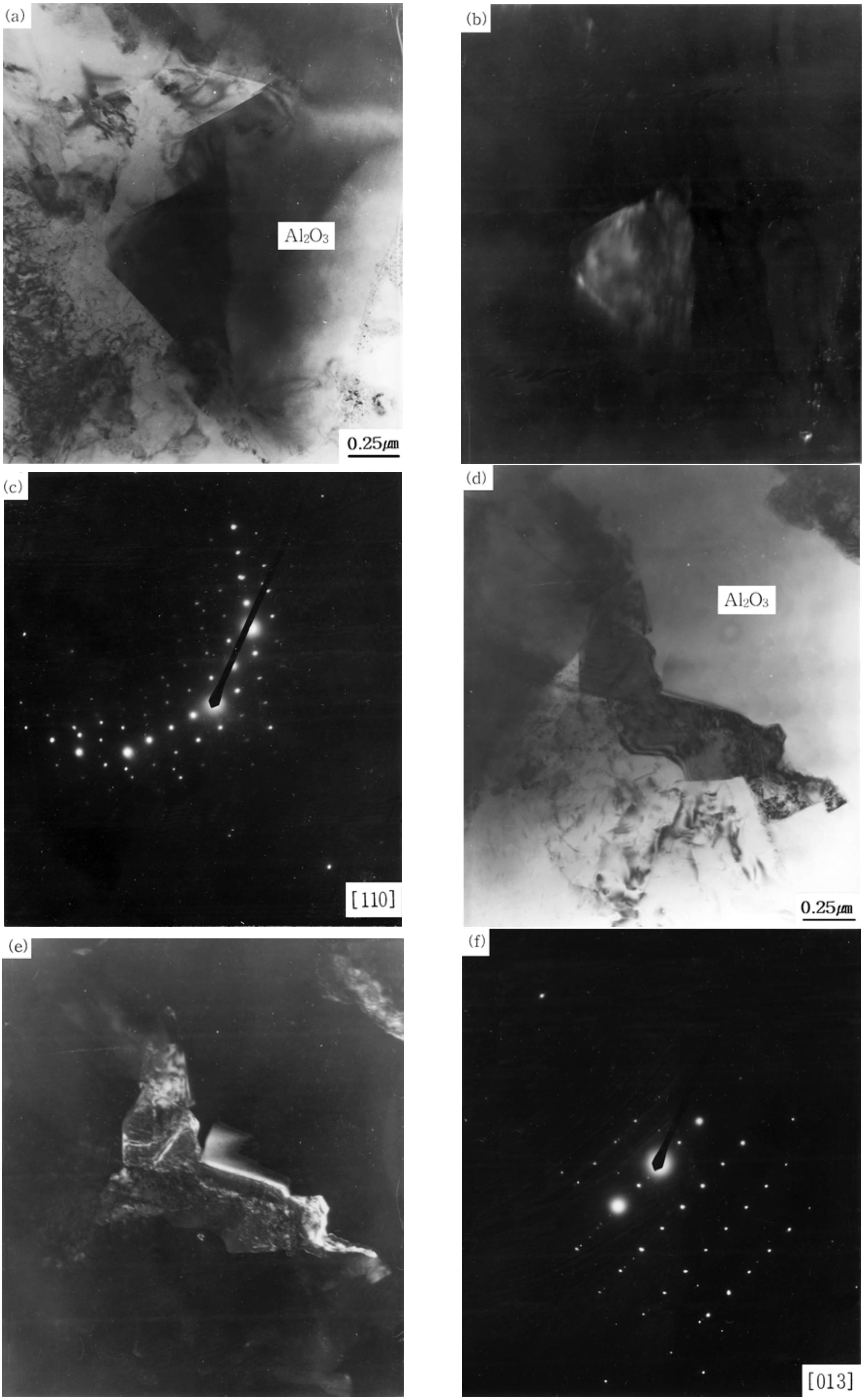

Figure 4 TEM micrographs showing reaction products on the surface of Al_2O_{3p}, $MgAl_2O_4$: (a) BF, (b) DF, and (c) SADP in the elemental powders composite, and (d) BF, (e) DF, and (f) in the alloy powders composite, respectively.

which composition was 1.0 wt% Mg, 0.76 wt% Si, and balance Al. Each powder mixture was put into a crucible and a 6061 Al ingot was placed on this powder bed. This assembly was heated to 700°C, and held for 1 hour under a flowing nitrogen atmosphere in the retort furnace (5000 cc/min). Then the assembly was cooled to about 600°C under the nitrogen atmosphere, in order to inhibit oxidation during solidification, and was removed from the furnace.

Both of these ingots were extruded at 450°C into a bar 16 mm in diameter (extrusion ratio 22:1). Tensile specimens having a gauge length of 25 mm and 2 mm thickness were machined from the extruded bars, parallel to the extrusion direction. Those specimens were heat treated to the T6 condition (529°C−2 h solution treatment, water quenching and 177°C, 6–8 h aging). Tensile testing was performed at room temperature, using a cross-head speed of 1 mm/min. Average tensile data were obtained from at least five tests for each condition.

The resulting microstructures and reaction products were investigated using scanning electron microscopy (SEM), and transmission electron microscopy (TEM). For the SEM analysis of the reaction products, specimens were prepared by dissolving away the metal matrix in a solution of methanol bromine and examined using a JEM 1210LV SEM equipped with an energy dispersive X-ray spectrometer (EDS) operated at 20 kV. Thin foils for TEM analysis were ground mechanically

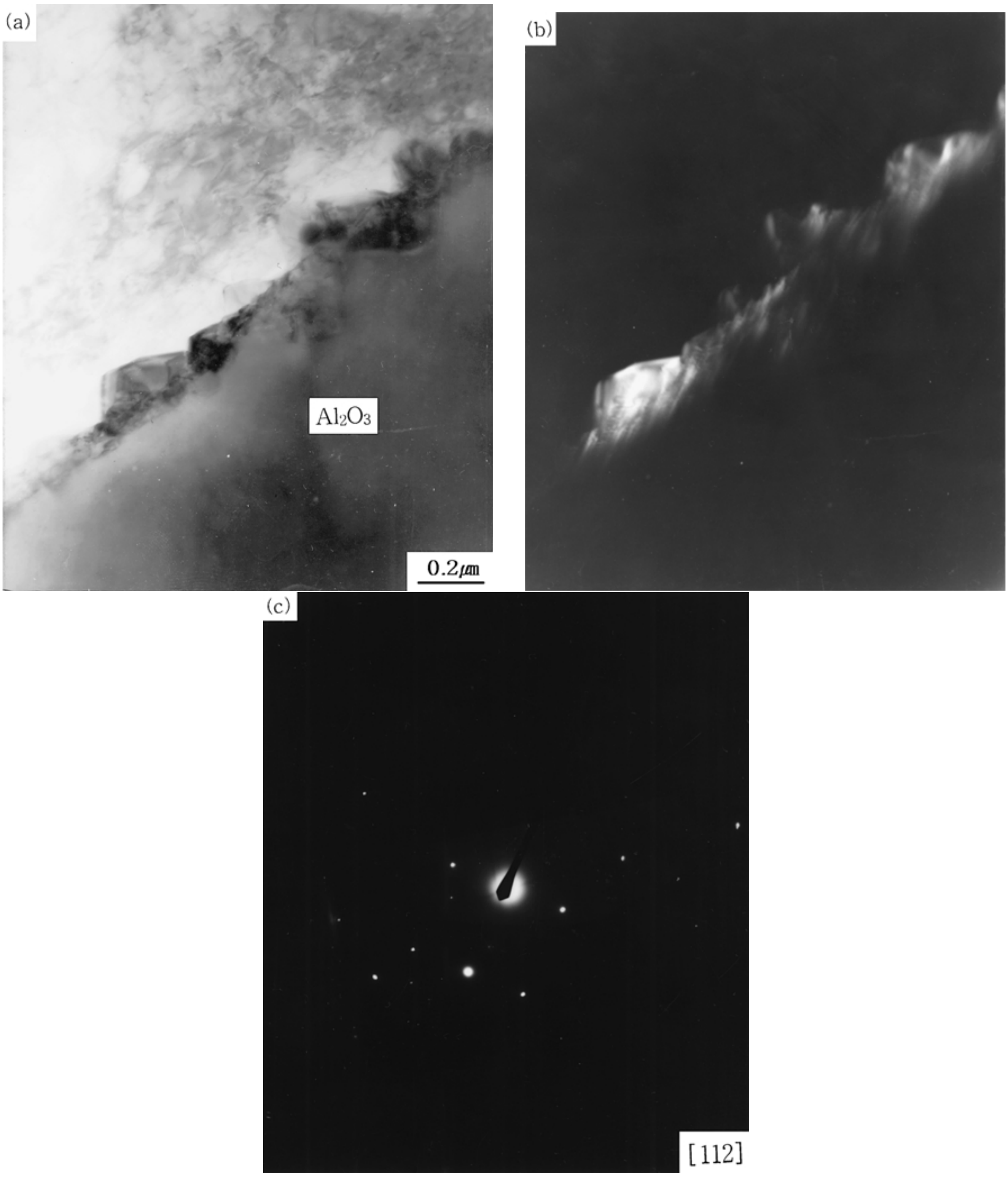

Figure 5 TEM micrographs showing reaction product on the surface of Al_2O_{3p}, MgO: (a) BF, (b) DF, and (c) SADP in the elemental powders composite, respectively.

to a thickness of about 60 μm and then punched to into 3 mm diameter discs. Finally, the discs were thinned using dimpling and ion milling (5 kV, at a tilt angle of 4–6°). All samples were examined by JEM 1210 TEM coupled to an EDS system operated at 120 kV.

3. Results and discussion

3.1. Microstructural analysis

Since the spontaneous infiltration of molten metal into two types of the powder bed occurred at 700°C for 1 hour under a nitrogen atmosphere, it was possible to fabricate 6061 Al matrix composite reinforced with Al_2O_{3p}. Fig. 2 shows the relatively uniform distribution of the Al_2O_3 particles in the both composites. There is no agglomeration and segregation of the Al_2O_3 particles as well as microstructural difference. Mg is indispensable for the spontaneous infiltration of molten Al in the pressureless state. In addition, the effect of Mg on wetting was greatly enhanced when Mg was freshly added to the reinforcement bed, such as elemental powder bed, as opposed to when alloyed into the infiltrating metal, such as alloy powder bed [10, 16]. Therefore, it was expected that the extent of infiltration in the case of elemental powder bed would be better than that of alloy powder bed. However, there was no appreciable difference in infiltration phenomenon of the both composites. The spontaneous infiltration behavior was explained in detail in previous papers [11–15].

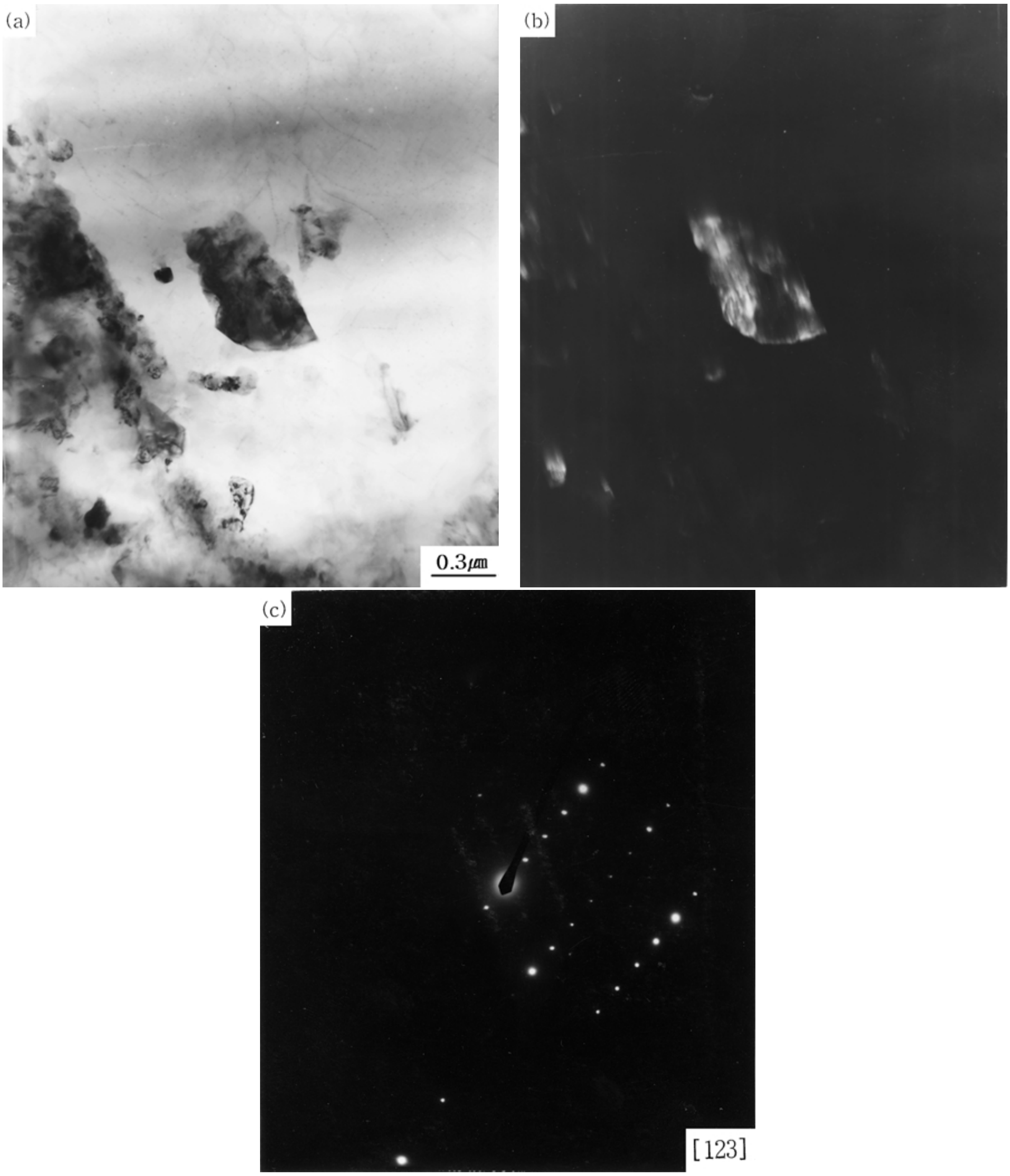

Figure 6 TEM micrographs showing reaction product in the matrix, $MgAl_2O_4$: (a) BF, (b) DF, and (c) SADP in the alloy powders composite, respectively.

Fig. 3 shows the reaction products and EDS analysis observed by SEM in both composites, after Al alloy matrix was dissolved away with a solution of methanol bromine. Reaction product of the various sizes was formed on the Al_2O_3 particles. From the EDS analysis, composition of reaction products of the polyhedron morphology with relatively larger size is consistent with the stoichiometry of $MgAl_2O_4$ (arrows). In addition, the reaction products of very fine size were significantly formed on the surface of Al_2O_3 particle. These products could not clearly identify with EDS analysis due to its large probe size. Therefore, these reaction products were investigated by TEM observation.

Fig. 4 shows the bright, dark field images, and selected area diffraction patterns of the reaction product observed in the both elemental and alloy powders composite, respectively. The reaction product was identified as the $MgAl_2O_4$ of cubic structure with the measured lattice parameter, $a = 8.069$ and 8.108 nm (theoretical value, 8.0831 nm, space group: *Fd3m*) [17]. This was formed by interfacial reaction between Al alloy matrix and Al_2O_3 particle. Fig. 5 shows the bright, dark field image, and selected area diffraction pattern of the reaction product observed on the surface Al_2O_3 particle. The reaction product was identified as the MgO of cubic structure with the measured lattice parameter, $a = 4.218$ (theoretical value, 4.213 nm, space group: *Fm3m*) [17]. Al_2O_3 is thermodynamically stable in contact with the pure Al melt. When the alloy contains Mg, however, MgO or $MgAl_2O_4$ can be formed as reaction product. There were many reports that the $MgAl_2O_4$ was the main reaction product at the interface Al/Al_2O_3 composites with a Mg content lower than 4 wt%, while MgO was the main reaction product in composites with a Mg content higher than 4 wt% [18–20]. On the other hand, Molins *et al.* [21] and Zhong *et al.* [22] reported that MgO as well as $MgAl_2O_4$ were formed as interfacial reaction products in the Al_2O_3/Al-Mg (2 to 3 wt%) composites. In the previous study on Al-3 wt% Mg/Al_2O_3 composite fabricated by the same method, MgO as well as $MgAl_2O_4$ were formed as interfacial reaction products [11]. Therefore, it is evident that the interfacial reaction are different in the various systems and are caused by the different fabrication condition, alloy compositions, or types and volume fractions of the reinforcements.

Fig. 6 is TEM micrographs of reaction product observed within the matrix of composite. According to SADP analysis, it was identified as $MgAl_2O_4$. This is not formed by interfacial reaction between the Al alloy and Al_2O_3 particles but formed by *in situ* reaction during composite fabrication. Oxide films on Al, Mg, and Si particles in the powder mixture (or alloy powders) can provide a source of oxygen for formation of $MgAl_2O_4$, in addition to oxygen as an impurity in the furnace. Furthermore, this $MgAl_2O_4$ formed *in situ* have been also observed in all the composite system which was not reinforce with Al_2O_3, such as SiC, BN, Si_3N_4, and B_4C fabricated by the same pressureless infiltration method [11–15].

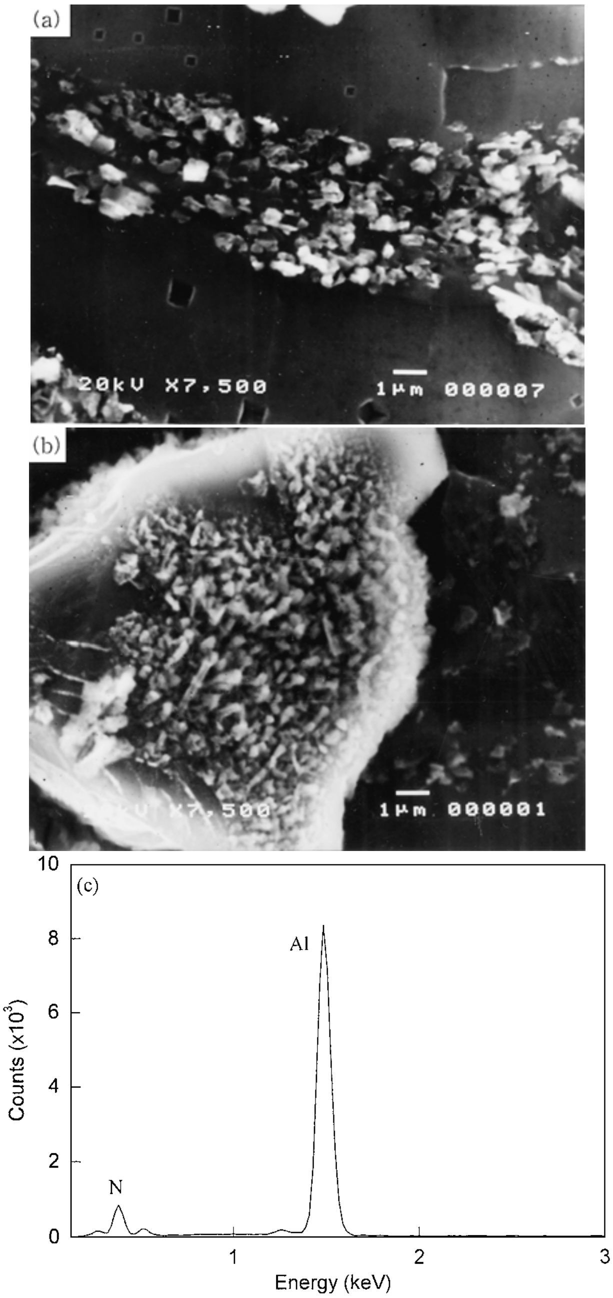

Figure 7 SEM micrographs and EDS profile showing the AlN reaction products, after dissolving away the Al alloy matrix: (a) elemental and (b) alloy powders composite.

Fig. 7 shows SEM micrographs and EDS profile of another reaction product observed both within the matrix and on the surface of Al_2O_3 in the composite. According to the analytical results obtained from EDS analysis (Fig. 7c), it was identified as AlN. Fig. 8 shows bright field and dark field images, and selected area diffraction patterns (SADP) of the reaction product observed by TEM in both composites, respectively. This reaction product was identified as AlN with a hexagonal structure and lattice parameters, $a = 3.117$ nm and $c = 4.989$ nm in the elemental powders composite and $a = 3.115$ nm and $c = 4.985$ nm in the alloy powders composite (theoretical value $a = 3.1114$ nm and $c = 4.9792$ nm, space group: $P6_3mc$) [17]. Formation of AlN is related to fabrication method of MMC (e.g. pressureless infiltration method) used in the present study. It is suggested that Mg vapor (serving as an infiltration enhancer precursor) react with nitrogen to form Mg_3N_2 coating (which serves as infiltration enhancer) around the particles in the preform or filler [10]. This Mg_3N_2

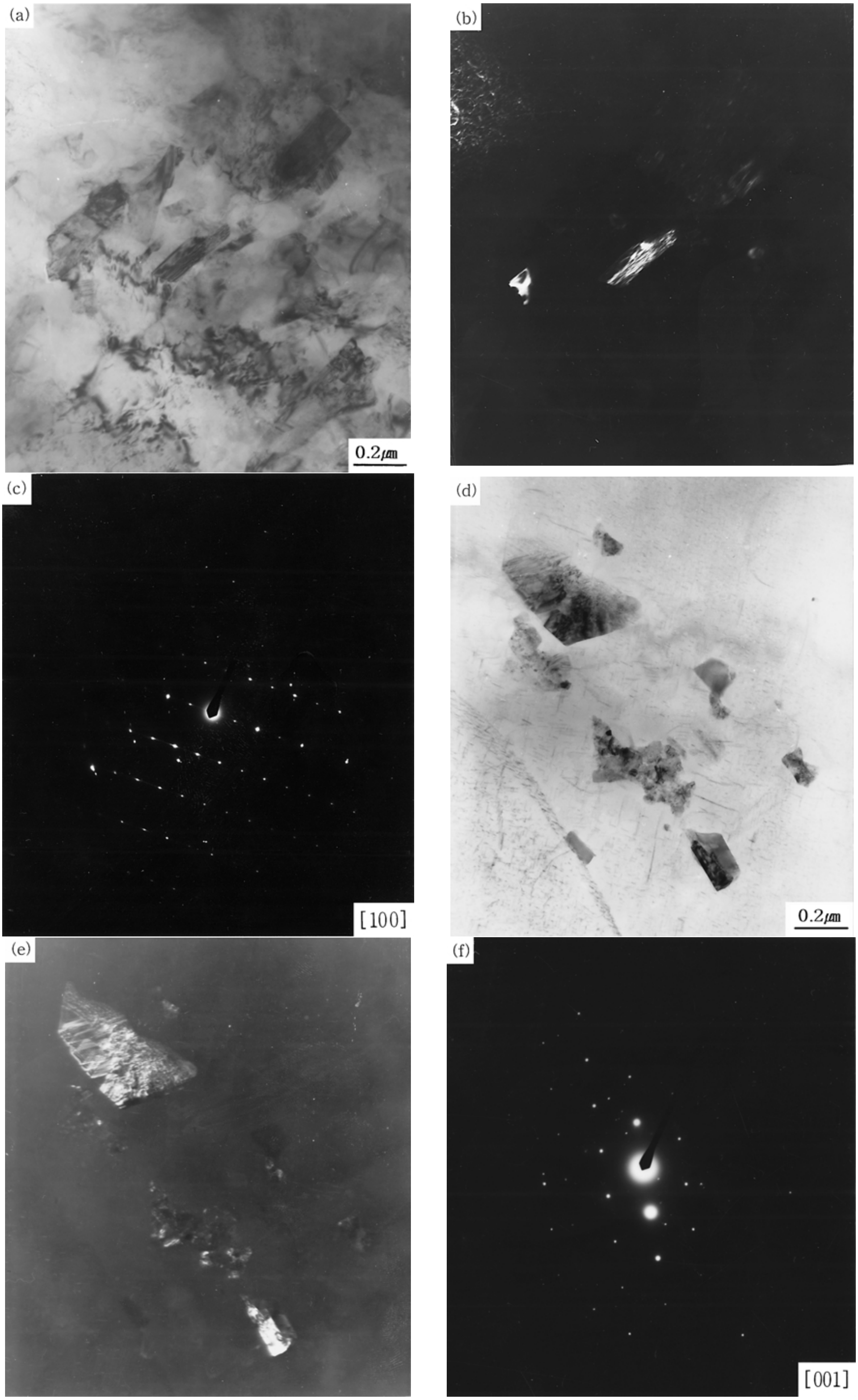

Figure 8 TEM micrographs showing reaction product, AlN: (a) BF, (b) DF, and (c) SADP in the elemental powders composite, and (d) BF, (e) DF, and (f) in the alloy powders composite, respectively.

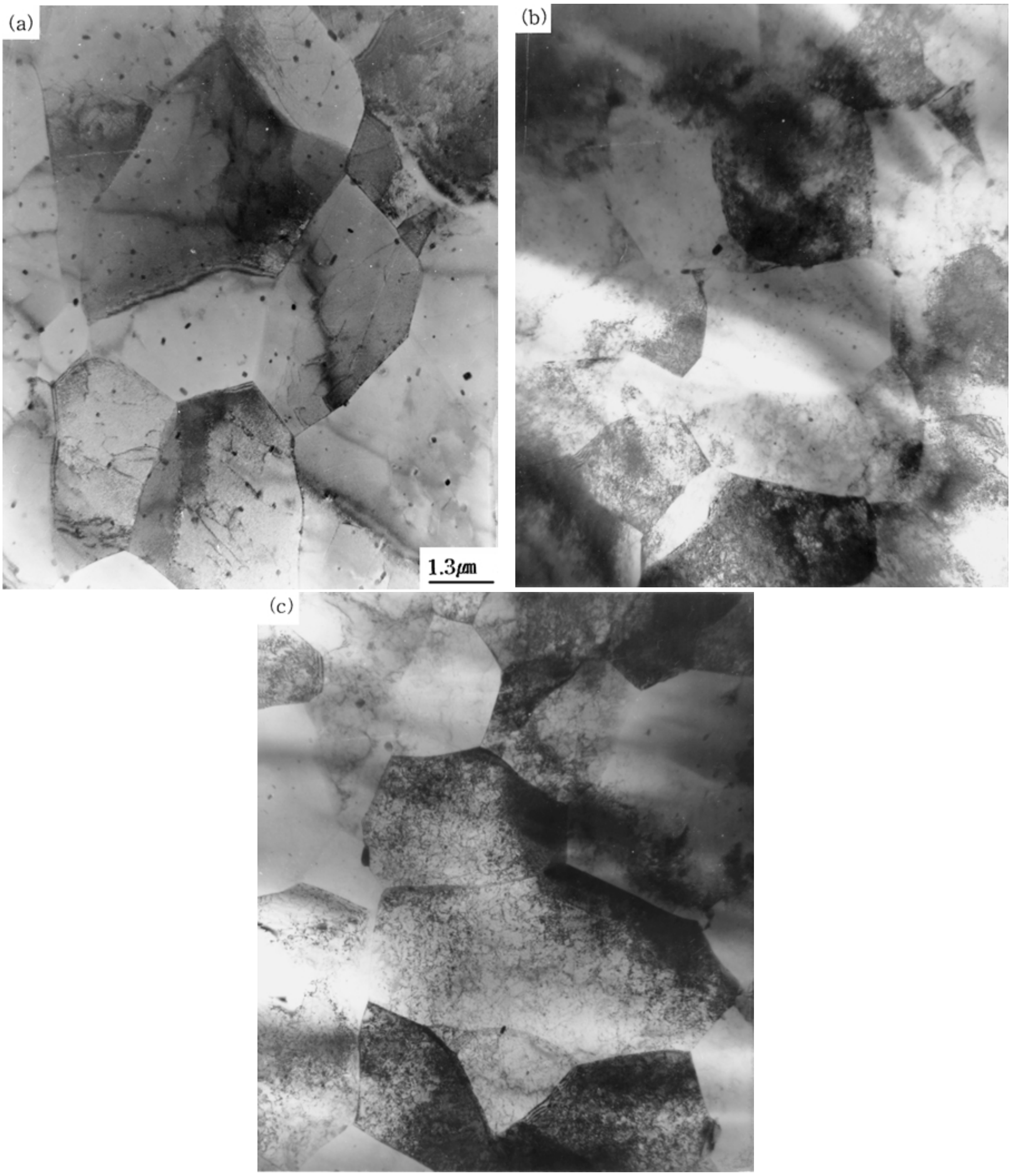

Figure 9 TEM micrographs showing the subgrain structure: (a) commercial alloy (b) elemental, and (c) alloy powders composite, respectively.

may lead to a spontaneous infiltration of molten Al alloy via great enhancement of wetting between molten alloy and reinforcement. In addition, this Mg_3N_2 reacts with molten Al and thus AlN is formed as a result of the *in situ* reaction ($Mg_3N_2 + 2Al = 2AlN + 3\ Mg$). Lee and Kwon indirectly demonstrated that the formation of Mg_3N_2 is necessary to induce spontaneous infiltration [11–15]. The AlN was also observed in previous studies of all composite systems, such as Al/SiC, Al/BN, Al/Si_3N_4, Al/B_4C etc, fabricated by the same pressureless infiltration method [11–15].

TABLE I Tensile properties of commercial alloy and composites

	UTS (MPa)		YS (MPa)		El (Pct)	
Material designations	ST	T6	ST	T6	ST	T6
Commercial AA6061	220	312	104	271	15.1	6.8
AA6061 (Elemental powders)/ SiC composite	270	417	184	367	7.2	4.8
AA6061 (Alloy powders)/ SiC composite	276	404	181	351	8.8	4.1

ST: solution-treated condition UTS: ultimate tensile strength,
YS: 0.2% offset yield strength El: elongation

3.2. Tensile properties

Table I shows the tensile properties of commercial 6061 Al alloy, elemental powders, and alloy powders composites in solution-treated (529°C, 2 hours) and T6 condition (177°C, 8–10 hours). The strength values in the composites were higher than those of the commercial alloy. In addition, the strengths of elemental powders composite were slightly increased relative to alloy

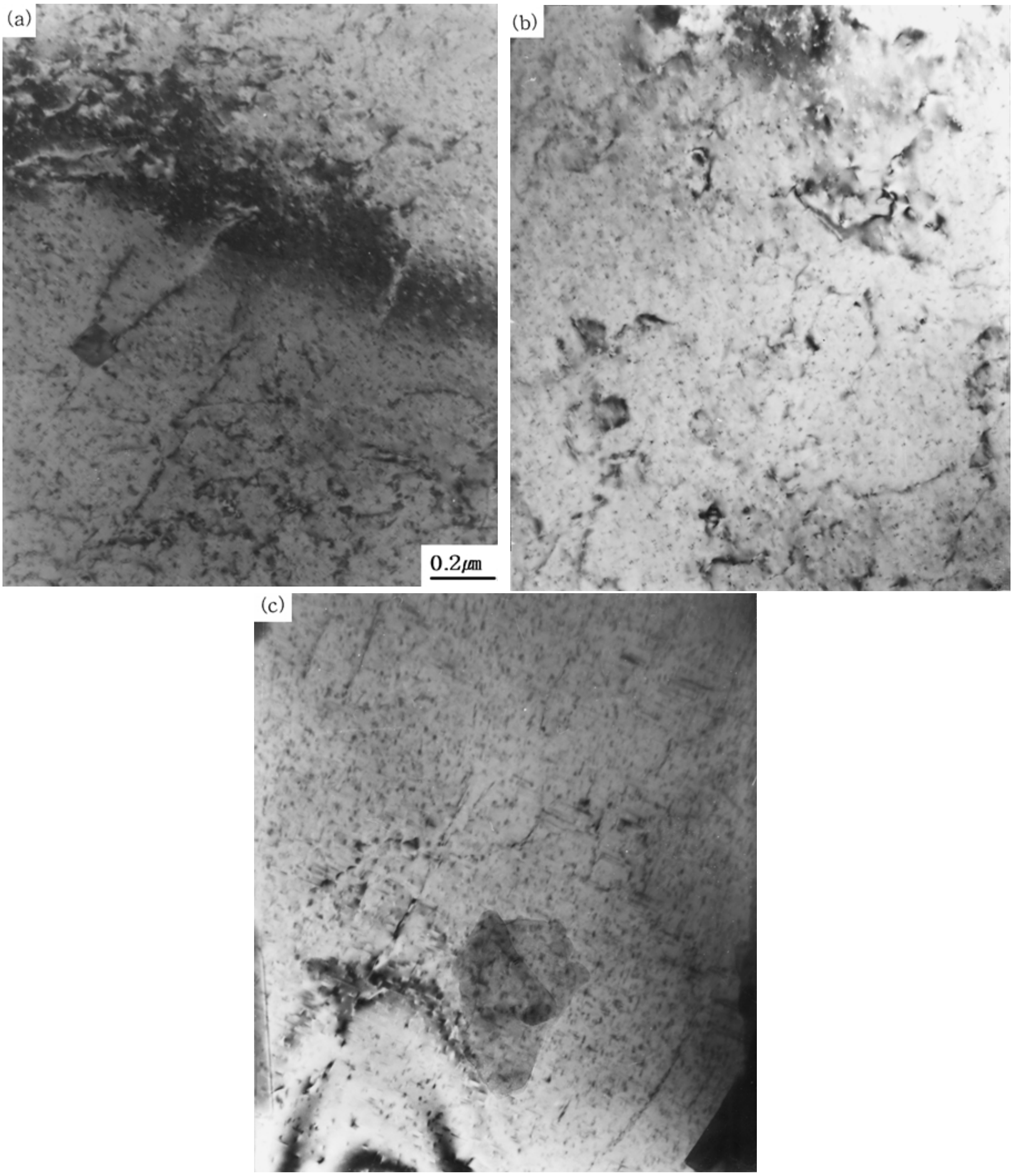

Figure 10 TEM micrographs showing the Mg_2Si precipitates in the T6 condition: (a) commercial alloy (b) elemental, and (c) alloy powders composite, respectively.

powders composite. Several theories and mechanisms have been suggested to explain the strengthening of MMCs. However, generally the strength of the composites does not depend on a unique mechanism but several mechanisms may be act simultaneously [23].

In the present study, it is believed that strength enhancement in the composites is contributed by both reinforcement artificially added (Al_2O_3) and AlN *in situ* formed. As previously mentioned, when the MMCs were fabricated by pressureless infiltration technique, AlN is formed as a result of the *in situ* reaction ($Mg_3N_2 + 2Al = 2AlN + 3Mg$). Furthermore, these fine AlN particles intimately contact with the matrix and also show clean interface (Fig. 7). Actually, when control alloys, such as AA5052, 5083, 6061 and 7075 without artificial reinforcement, were fabricated by the same process, AlN was formed by *in situ* reaction. Thus, these control alloys exhibited considerable strengthening, as compared to the monolithic commercial Al alloys. For Instance, the tensile strength in the control AA6061 was 55–70 MPa greater than the commercial AA6061 [11].

Fig. 9 shows the subgrain structures observed by TEM in both the commercial alloy and composites in solution-treated condition. Although grain size was not constant, the subgrain size of the composite was smaller than that of the commercial alloy. In addition, the composites showed recovery structure with high dislocation density, while the commercial alloy exhibited nearly recrystallized grain structure. It is well known that in particle-reinforced MMCs, the grain size is generally small compared to an unreinforced material of similar composition, and it has been shown that particle-stimulated nucleation (PSN), coupled with particle pinning during normal grain growth, is responsible for this small grain size [24, 25].

In addition, since aging kinetics was accelerated by the addition of Al_2O_3, the time to peak strength decreased from 8 hours in the commercial alloy to 6 hours in the composites. Discontinuously reinforced MMCs with precipitation hardenable matrix are known to age considerably faster than the unreinforecd alloys. This acceleration was attributed to the thermal mismatch between the reinforcement and the matrix, which generates upon cooling from solutionizing temperature a high dislocation density (Fig. 9). High matrix dislocations reduce the incubation time for heterogeneous nucleation of the strengthening precipitates and concomitantly increase solute diffusivity [26–28]. Fig. 10 is TEM micrographs showing Mg_2Si precipitates in the commercial and composites in the T6 condition, respectively.

4. Conclusions

The tensile properties and microstructures of AA6061/Al_2O_{3p} composites fabricated by the pressureless infiltration method under a nitrogen atmosphere were examined. Since the spontaneous infiltration of molten metal into elemental powders bed as well as alloy powders bed occurred at 700°C for 1 hour under a nitrogen atmosphere, it was possible to fabricate 6061 Al matrix composite reinforced with Al_2O_{3p} irrespective of the type of metal powders. Both $MgAl_2O_4$ and MgO were formed at interfaces between Al_2O_3 and the matrix. In addition, $MgAl_2O_4$ was formed at within the matrix by *in situ* reaction during composite fabrication. Fine AlN was formed by *in situ* reaction in both composites. A significant strengthening in the composites occurred due to the formation of *in situ* AlN particle and addition of Al_2O_3 particles, as compared to the commercial alloy, while tensile properties in the both elemental and alloy powders composites showed similar trend.

Acknowledgment

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