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FABRICATION AND CHARACTERIZATION OF ALUMINUM- GRAPHITE COMPOSITES

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ABSTRACT

In the present study, a proposed technique called “ex-situ and in-situ powder metallurgy” has been developed to produce aluminum–graphite (Al/Gr) composites. In order to avoid any interfacial reactions between the graphite and the aluminum, an isostatic pressing of material powders followed by hot extrusion techniques was used. Five weight percentages of graphite flakes were mixed with Al powder using a mechanical mixing stirrer. The thermal characteristics and microstructures of the composite were investigated. The effects of graphite content and SiC formation on structure characteristics and properties of composites were investigated. The results showed that some graphite flakes have reacted with silicon present in commercial aluminum powder and formed silicon carbide (SiC) at temperatures above 252 ◦C. SiC has great effect on composite characteristics. The results also showed refinement and uniform distribution of graphite and SiC particles within the aluminum matrix.

فى هذا البحث تم تطوير طريقة تصنيع المواد المؤتلفة من الومنيوم و رقائق الجرافيت. ولتفادى التفاعل الكميائى لرقائق الجرافيت مع الومنيوم استخدمت طريقة الضغط على البارد والبثق على الساخن. وتم فحص البنية المجهرية بواسطة الماسح المجهرى الإلكتروني. كما تم قياس الخصائص الحرارية للمواد المؤتلفة بواسطة مسعر المسح التباينى (DSC) وكذلك تحليل العناصر كميائيا بواسطة محلل الانبعاثات الطيفية مع الاشعة السينية (EDS) . وأظهرت نتائج الاختبارات ان جزء من رقائق الجرافيت تفاعل مع حبيبات السيليكا الموجودة فى بودرة الالومنيوم ذات النقاوة التجارية مكونا كربيدات السيليكون وذالك عند درجة حرارة ٢٥٢ درجة مئوية . كما وجد ان رقائق الجرافيت و كربيدات السيليكون الموزعة بانتظام في الالومنيوم ادى الى تحسن ملحوظ فى خصائصها الميكانيكية .

***Keywords*:** Composite; graphite flakes; cold pressing; hot extrusion; tensile properties.

1. INTRODUCTION

Recently metal matrix composites (MMCs) are increasingly used for critical structural and wear resistant applications because of their excellent strength to weight ratio [1] and interesting physical properties. Normally MMCs are reinforced with continuous fibers, discontinuous particles or whiskers. However, particle-reinforced MMCs possess some distinct advantages over fiber-reinforced composites in terms of low cost and isotropic mechanical property.

Aluminum alloys have a great diversity of industrial applications because of their low density and good workability, but the use of these alloys is limited due to their relatively low yield stress. Recently, the interest to increase aluminum strength for applications in the aerospace and aeronautic industries has motivated the study of aluminum matrix composites. One of their most important characteristics of aluminum matrix composites is its high specific stiffness while maintaining a low density [2–5]. Self-lubricating materials offer many improvements over the materials to which lubricant needs to be applied periodically. Among these materials, considerable work has been done on aluminum alloy–graphite particulate composites (Al/Gr MMCs). The processes used to synthesize the Al/Gr composites can be classified into three main categories: (i) liquid phase; (ii) solid phase; and (iii) two phase (solid–liquid) routes. It has been documented that the production method has a strong influence on the mechanical and tribological properties of the composite via its effects on the matrix grain size, porosity and distribution of graphite particles [6] and the interfacial properties of the Al/Gr couple [7].

The main reasons to produce Al/Gr composite are to increase the strength, stiffness and wear resistance of aluminum or aluminum alloys, but this is usually achieved at the expense of other properties such as ductility. Aluminum and aluminum alloys can be strengthened by dispersing hard particles like carbides, oxides or nitrides into the aluminum matrix by using solid or liquid state techniques [8]. The reinforcement can be done by adding continuous or discontinuous fibers, particles, or whiskers. The last three are usually ceramic materials such as alumina, silicon carbide, or silicon nitride.

The composites so developed are called ex-situ MMCs. It is reported that agglomeration of reinforcement particulates may occur during processing of most ex-situ composites leading to inferior mechanical strength and toughness. Therefore, uniform distribution of reinforcement particulates in the matrices of MMCs is essential to achieve effective load bearing capacity of the composite. Another production route that is attracting a number of researchers is the in-situ process. In this route, particles are obtained in the matrix due to chemical reaction or diffusion, which usually occur under isothermal conditions [9-14].

The interfaces in this in-situ particle/matrix are clean and free of impurities [15]. Moreover, the sizes of in-situ formed reinforcements are finer and the distribution is more uniform compared to ex-situ composite, resulting in better mechanical properties of the in-situ composites. However, in most of the cases it has been found that due to the high initiation temperature of the in-situ reaction, formation of the reinforcements within the matrix necessitates high processing temperature [16]. But processing the composites at high temperatures involves the risk of oxidation of the matrix and may also cause agglomeration and coarsening of the reinforcements, which would cause adverse influence on the mechanical properties [17]. Powder metallurgy (P/M) is important processing technique for MMCs that tends to offer homogeneity of both composition and microstructure of the matrix alloy together with more control of processing temperatures and reinforcement distributions [18, 19].

In the present study, a new method namely, “ex-situ / in-situ powder metallurgy”, is developed and described for consolidating the aluminum–graphite particle composites. In this method, the cold compact powders are followed by hot extrusion were combined into an integrated net shape forming process.

2. EXPRIMENTAL WORK

Aluminum powder (Aluminum Powder Company, Anglesey, UK, 99.4% pure, 150 µm size) was used as the matrix, it has the chemical composition of 0.2% Si, 0.15% Fe, 0.1% Cu, 0.1% Mg, 0.05% Mn and the rest is Al. Five weight percentages of natural graphite flakes were used (1, 2, 3, 4 and 5 wt %). The graphite flakes have an average thickness at flake middle of 500 µm. The graphite flakes were treated in nitric acid (68 wt. %) for 10 h at 120 0C to produce graphite (Grs). The graphite flakes were washed several times with distilled water until the washings show no acidity. Finally, they were added to the dimethylformamide (DMF) in order to retain uniform distribution. Then the aluminum powder was mixed with the Grs–DMF solution using mechanical stirrer for 30 min. Finally, the mixed powders were dried at 120 0C. Consolidated bulk products (40mm of diameter Ø) were prepared by pressing the mixed powder at 400MPa for 20 min under uniaxial load. Pressed samples were next hot extruded at 500 0C into a rod of 10 mm of diameter using indirect extrusion with an extrusion ratio of 16. For comparison, samples of aluminum powder without any graphite were fabricated under the same processing condition. All tested samples were machined from the middle portion of the as-fabricated materials.

The thermal characteristics of the composite were studied under argon gas atmosphere with the aid of a differential scanning calorimeter (DSC) model Mettler, TA400. The flow rate of argon gas was maintained at 80 cc/min throughout the test procedure. During test the green compact samples were heated up to 700 0C at heating rate of 3 0C /min and then cooled to room temperature with same rate. X-ray diffractometer with Cu Kα radiation was used to assess the formation of phase’s transformation and to measure the crystallite size of powders using the Scherer’s formula[16] B.D. Cullity, Elements of X-ray diffraction (2nd ed.), Addison-Wesley, California, USA (1978) p. 102..

A Scanning Electron Microscope (JEOL, JSM-840A) along with the energy dispersion spectroscopy (EDS) was used to study the microstructural architecture and composition of the different phases of the composite samples. Tension tests were carried out in extrusion direction at room temperature with initial strain rate of 0.0083 s-1. Fig.1. Shows flowchart of fabrication process of Aluminum-Graphite (Al-Gr) composites.

3. RESULTS AND DISSCUSSTIONS

3.1. Characteristics of Aluminum-Graphite composites and the in-situ reaction

To better examine the reaction condition of carbon flakes and Al powders, 4 wt. % carbon flakes were added to Al powders. Fig. 2 shows the DSC plot obtained during heating and cooling of the green compact from room temperature to 750 0C and from 750 0C to room temperature at a heating/cooling rate of 3 0C /min under argon gas atmosphere. From the DSC heating curve it is evident that the total amount of heat absorbed during melting of Al-Gr is significantly the same as heat liberated during solidification. The small endothermic peak observed at 252.5 0C in the heating curve is attributed to the formation of silicon carbide (SiC). It is worth while to mention that presence of graphite flakes and silicon carbide in the composite samples has been established by conducting X-ray diffraction studies on the composite samples.

Figure 2 shows the differential scanning calorimeter (DSC) pattern representing heat flow against temperature of Al-Gr composite system. An endothermic peak appeared at 252.5 0C corresponds to formation of silicon carbide (SiC). At 604 0C an exothermic sharp peak appeared due to aluminum carbide formation (Al_4C_3), which is an intensely exothermic reaction. However this aluminum carbide (Al_4C_3) deteriorates the composite properties and should be avoided. The final endothermic peak at 655.6 0C corresponds to temperature approaching the melting of aluminum. Based on shown DSC result, SiC particles are found to be synthesized in-situ in the aluminum alloy matrix at relatively lower temperature (252.5 0C) for this compact and hot extruded composite.

Figure 3 illustrates the XRD diffraction pattern of the Al-Gr powder mixture before compacting. Presence of graphite and aluminum elements is indicated in the XRD pattern, indicating no reaction product in powder sample before compact and extrusion.

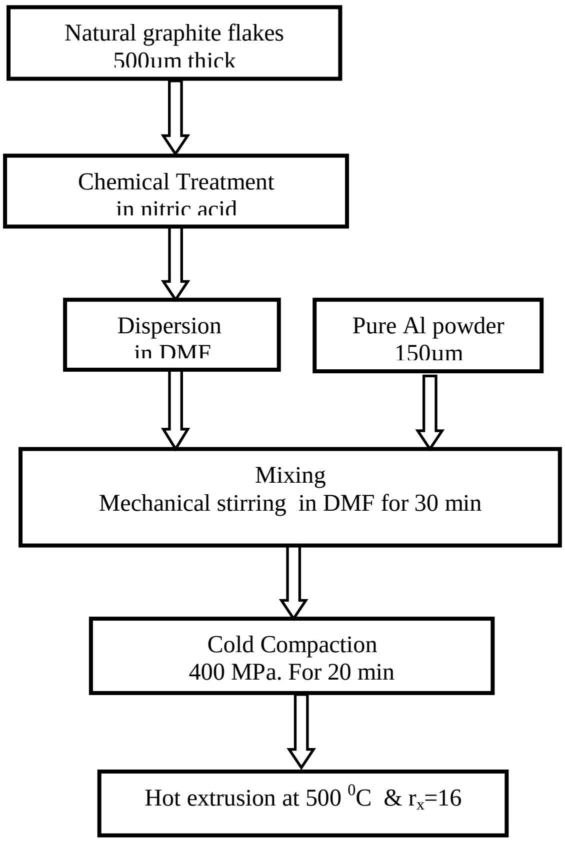

Fig. 1 Flowchart of fabrication process of Aluminum-Graphite composites

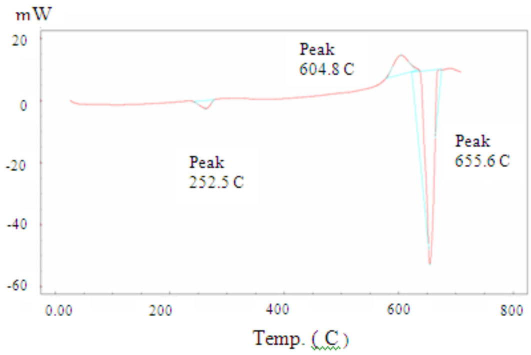

Fig. 2 DSC heating and cooling curve of the green compact and hot extruded composite of Al-Gr.

Figure 4 shows XRD patterns of composites with three different graphite contents after compact and extrusion. The XRD pattern is different; SiC phase appeared in addition to both Al and carbon. There is a good matching of position and intensities compared to SiC standard spectrum. It should be noted that SiC peak has not be seen in 1.0 wt.% Gr composites, suggesting that reaction of carbon and Si did not take place in 1.0 wt.% Gr. The appearance of SiC phase for 2, 3 and 4.0 wt. % C can be formed during hot extrusion process. This is also in good agreement with endothermic peak at 252.5 ◦C observed in DSC of figure 2 and shown in SEM micrographs of Figure 6. The Presence of SiC as indicated in the XRD pattern confirms the feasibility of the in-situ reaction of silicon and carbon. The initiation temperature of this endothermic reaction with conventional micron sized SiC powder is usually carried out at relatively high temperature (500 ◦C or 600◦C) [20, 21]. The low temperature of 252.5 ◦C observed in the present study is attributed to application of hot extrusion. The die pressure of extrusion enhanced the overall kinetic conditions and reaction rate of the in-situ reaction process at low temperature.

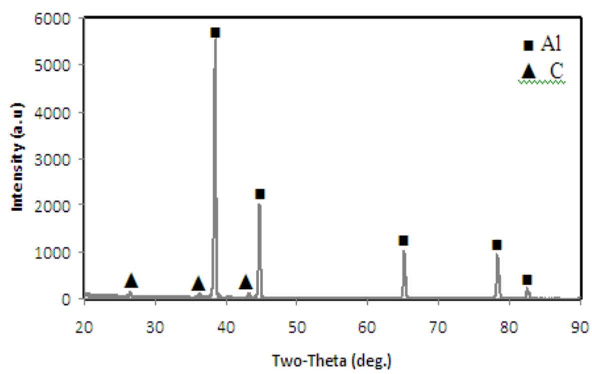

Fig. 3 XRD patterns of Al-Gr powders

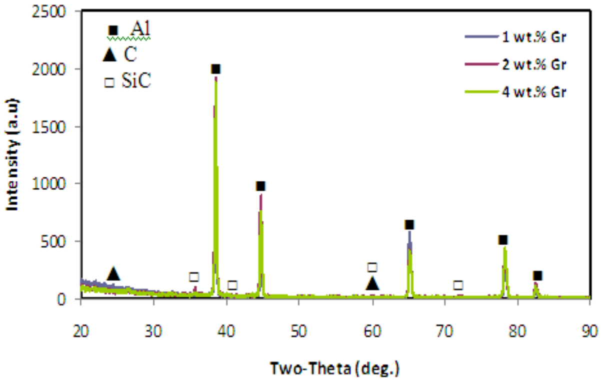

Fig. 4 XRD patterns Al-Gr composite

Representative SEM micrographs of the composite with 3wt. % C prepared by compact and hot extrusion held at 500 ◦C are illustrated in Fig. 5. The microstructural feature of the composite reveals the presence of graphite and fine particles of SiC (less than 1 μm) that are uniformly distributed in aluminum matrix. XRD and EDS results indicate that both SiC and C are present in the aluminum matrix. The formation of SiC occurred mainly after hot extrusion process at relatively low temperature and lead to further improvement in mechanical properties.

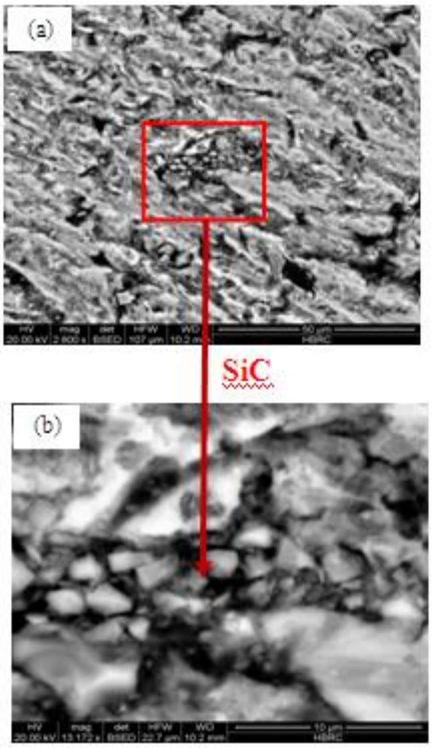

Fig. 5 SEM micrographs of the in-situ formation of SiC in composites with magnification a) 2000x and b) 13172x

Figure 6 shows SEM micrographs of the mixed powders of Al–2 wt. % Gr and Al–4 wt. % Gr composites respectively after hot extrusion. Structures showed homogeneous dense compacts. The dark regions represent the pores or voids, which were left behind by evacuation of graphite particles from surfaces during the polishing process. It can be seen that the graphite particles have been distributed uniformly within the matrix, which is due to dispersion action of graphite in dimethyl-formamide (DMF) solution.

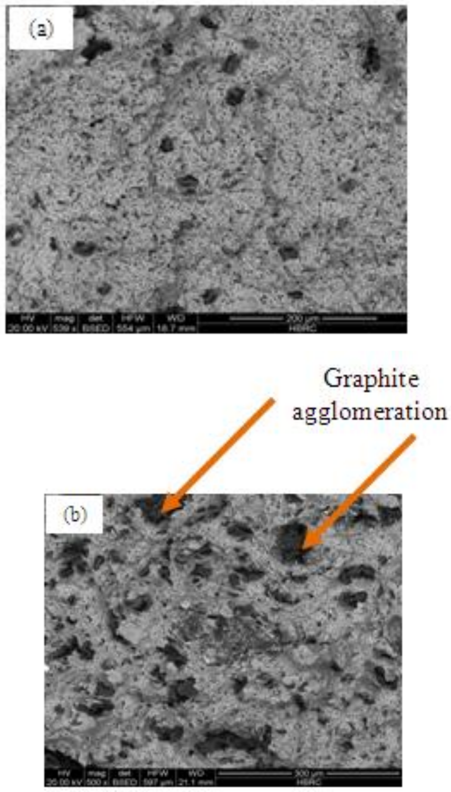

Fig. 6 SEM micrograph of Al-Gr composites after hot extrusion (a) 2.0 wt. % Gr; (b) 4.0 wt. % Gr

In order to verify the composition of Al-Gr composite, EDS analysis was used. The typical result is shown in Fig. 7. It seems that the composite was protected well during fabrication since the oxidation level is quite low. Fig. 7 shows the morphology and distribution of graphite and SiC in Al matrix. It can be seen that SiC particles are well distributed and dispersed. To determine the chemical composition of the reinforcements in the Al matrix, energy dispersion spectrum (EDS) was used. Because the detection zone of EDS beam is bigger than the average size of C and SiC, the EDS peaks for C and SiC particles (Fig. 7a) will inevitably include compositional information of Al matrix. However, through subtracting the compositional information of matrix (Fig. 7b&c), it is evident that (C, O) and (Si, O) peaks, respectively correspond only to composition of graphite and Si particles.

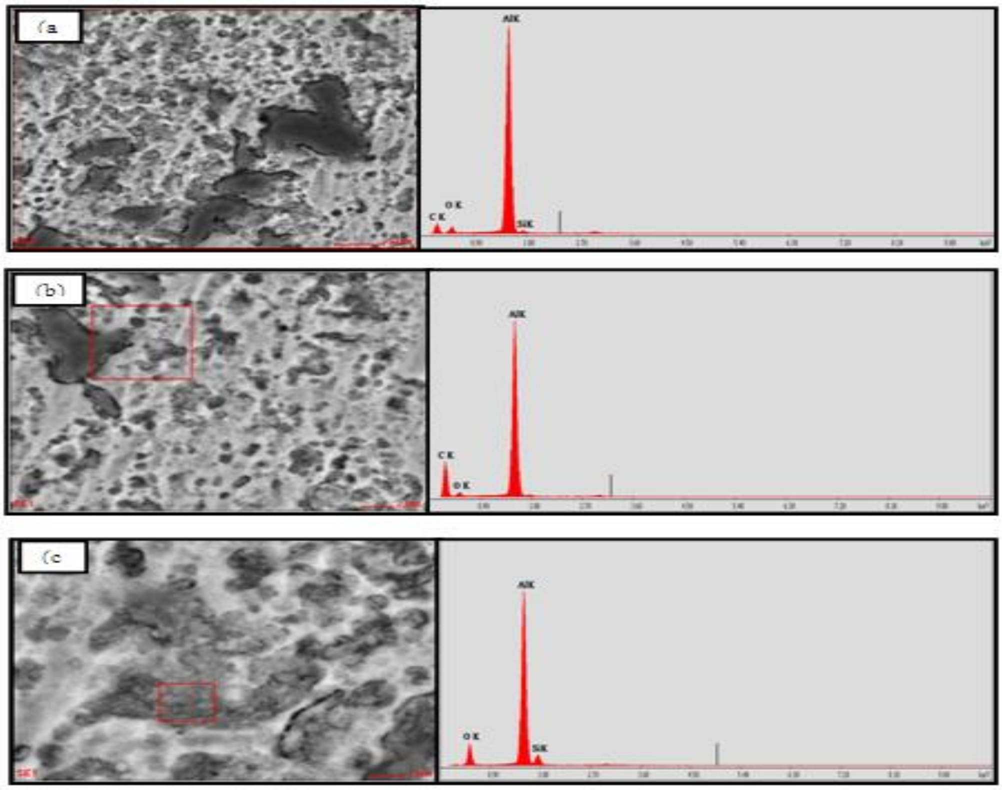

Fig. 7 Distribution and dispersion of graphite and SiC particles in Al matrix and spectrum of EDS. (a) C and SiC in Al; (b) C in Al; (c) Si in AL

3.2. Mechanical properties

The stress–strain curves obtained from tensile tests of Al- Gr composites pulled in extrusion directions are shown in Figure 8. For each type of composite material, the 0.2% proof stress, the ultimate tensile strength (UTS), and Young's modulus were determined from these curves as indicated in Table 1. The tensile strength and Young's modulus are increased with increasing the graphite contents. The tensile strength of Al- Gr composite with 3 wt. % Gr is measured as 140 MPa, which is approximately 1.15 times higher than that of monolithic Al. Young's modulus of Al- Gr composite with 3 wt. % Gr is measured as 86 GPa, which is almost 1.23 times higher than that of monolithic Al, which is measured as 70 GPa. The yield strength increased from 121 to 140 MPa by addition of 3 wt. % Gr. The improved tensile properties indicated that both graphite reinforcement and nanosized particles of SiC particles have great effect of structure characteristics. From the fracture surfaces of the composites containing 2 and 4 wt% Graphite shown in Fig. 9a and 9b, it was found that spherical dimples occurred in both composites. Smaller dimple sizes were found in the matrix containing 4% Gr. Some SiC particles cleavage was also observed in structures. Some pulled out SiC particles were found in the composites instead of their cleavage. The SiC particles were uniformly distributed in the matrices, and little pores were found in the composites.

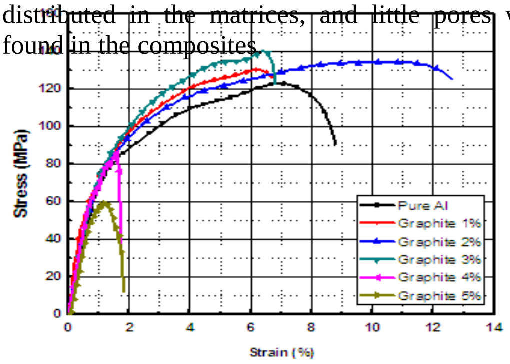

Fig. 8 Stress–strain curves of Al-Gr composites obtained by tensile test

The presence of the larger amount of graphite seemed to make the dispersion of the graphite in the aluminum matrix more difficult, as apparent in 4 Gr% sample compared to 2 Gr% where some clusters are visible. The shallow dimple morphologies of the composite are sign of good ductility. Improvements in the strength of Al- Gr are attributable to nanosized SiC particles and to sufficient load transfer from matrix to graphite through the interface

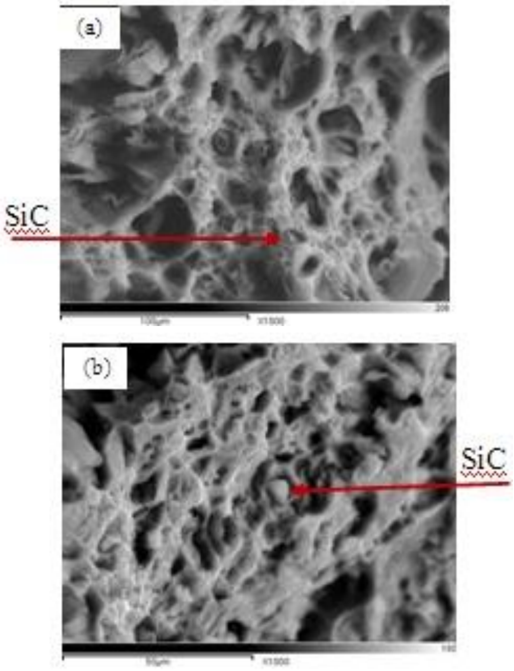

Fig. 9 SEM micrographs of the composites fracture surfaces with magnification 1000x; (a) 2.0 wt. % Gr and (b) 4.0 wt. % Gr.

4. CONCLUSIONS

The following conclusions can be drawn from this investigation:

1. Aluminum matrix composites reinforced with well dispersed graphite can be prepared by cold compaction followed by hot extrusion.
2. Nanosized SiC particles are synthesized in-situ in Al matrix. During hot extrusion, the particles disperse and reach a relatively homogeneous distribution in the matrix.
3. The SiC particles are formed by the in-situ reaction between silicon present in commercial purity aluminum and graphite. SiC particles have greatly improved the tensile properties of fabricated composites.
4. Die extrusion pressure results in refinement, dispersion and lowering initiation temperature of the SiC formation.
5. The SiC particles were refined and uniformly distributed in the matrices as a result of hot extrusions and little pores were found in the composites This significantly improves mechanical properties.

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