Characteristics of aluminum AC4B with variations of silicon nitride reinforced composite produced by a stir casting process

A A Faisal and A Zulfia
Department of Metallurgy and Materials Engineering, University of Indonesia, Jalan Prof. Dr. Sudjono D Pusponegoro, Depok, 16424, Indonesia

Email: anne@metal.ui.ac.id

Abstract. At present, the combination of metal matrices and ceramic reinforcement as composite is popular for creating materials with good mechanical properties. Aluminum is used as a matrix because of its lightweight, ductile, and low melting point. In this research, the effect of adding silicon nitride on characteristics of Al AC4B is studied. Five sample variations were made based on fraction volume percent (1, 3, 5, 7, 10) which were fabricated through stir casting method. In the fabrication process the magnesium element was added to increase the wettability, TiB grain refiner, and modification with strontium element. The samples made got destructive, tensile, impact, and hardness testing to determine the mechanical properties of the composite. Observation of phases, elements, compounds and microstructure was done to see the distribution of reinforcing particles in the aluminum matrix and the estimated phases formed. The results showed that the optimum tensile strength of variable occurred at the addition of 1% volume of silicon nitride with a strength value of 104.94, and the optimum hardness value was at the addition of 3% Si3N4, which was 44.8 HRB. The phases formed in the composite were Mg2Si, Al2Cu, β-Al5FeSi, and π-Al9FeMg3Si5 phases.

1. Introduction
With the development of an advanced era, efficiency is an important factor in the selection of a material. In the world of transportation, materials with low density and good strength and hardness are indispensable. Low density will help reduce fuel consumption from a transportation. Efficiency can also be created through the process of making material at a low cost with regard to its melting point and formation. The low melting point of material causes the reduction of fabrication costs that results in a more efficiency. Composite is a promising solution as a material that will be used today because it can combine two different material properties. Aluminum is a metal that is often applied as a matrix to a composite [1]. Aluminum has the characteristics of low density, melting point, resilience, and good formation. In this research, the composite used was AC4B aluminum matrix and the micro Si3N4 reinforcement. Mg element was added to increase the wettability. Modifier Sr was also added to change the aluminum micro structure to be more rounded [2]. Besides, TiB grain refiner was used as a nucleating agent to elevate the mechanical properties of aluminum [3]. Addition of Si3N4 would cause a dispersion hardening mechanism to increase the composite hardness. The composite was made through stir casting method. The method was occupied because the process was economical and uncomplicated [4].
2. Methods
The research began with fabrication process of the composite samples which were then tested. AC4B was melted in the tilting melting furnace at 850°C. Si₃N₄ was preheated at 850°C for 1 hour in the muffle furnace then put into molten aluminum. The melt was then stirred using a stirrer for 40 seconds then the degassing process was done with argon gas for 2 minutes. 5wt% Mg, 0.15wt% TiB, and 0.04wt% Sr were added in the melt, stirred again with the stirrer for 40 seconds followed by the degassing process for 2 minutes, and then deslagging in the molten aluminum and Si₃N₄ mixture. The melt was then poured into a mold that had been preheated at 400°C, followed by water cooling process. AC4B / Si₃N₄ composite was made with variations of 1, 3, 5, 7, and 10 v%. The composition of pure AC4B aluminum can be seen in Table 1.

| Table 1. Al AC4B composition determined by OES |
|---------------------------------------------|
| Element  | Al  | Si  | Fe  | Cu  | Mg  |
| %        |     |     |     |     |     |
| Al       | 86.5000 | 7.6500 | 1.01 | 2.51 | 1.05 |

Destructive testing and identification of elements and compounds were carried out to determine the characteristics of the composite samples. Meanwhile, tensile testing was carried out to determine the tensile strength and elongation with the GoTech 27-7000 LA 10 Tensile Test Equipment according to ASTM E8 standard. Hardness testing was applied using the Rockwell B method according to ASTM E18-11 standard using a steel ball indenter with 1/16 inch diameter. Not to mention that impact testing was done using the Charpy method with the GoTech Testing Machine according to the E23-01 standard. Density testing was carried out to determine the effect of adding Si₃N₄ on the composite density by using the Achimedes law and then the amount of porosity in the samples were calculated. Further, microstructural observation was done using an optical microscope according to the ASTM E3-11 standard using ETSA keller. Identification of phases, interface, and distribution of Si₃N₄ particles were carried out using SEM and EDS. Finally, observing the elements contained in the composite was done through OES testing, and XRD was carried out to determine the compounds that could be formed in AC4B / Si₃N₄ composite.

3. Results and Discussion
3.1 Chemical Composition in Al AC4B/Si₃N₄ Composite

| Table 2. AC4B / Si₃N₄ Composite Compositions Determined by Optical Electron Spectroscopy |
|---------------------------------------------|
| Element | AC4B | 1% Si₃N₄ | 3% Si₃N₄ | 5% Si₃N₄ | 7% Si₃N₄ | 10% Si₃N₄ |
|         |      |          |          |          |          |           |
| Al      | 86.5000 | 78.5000 | 82.9333 | 83.3000 | 79.4333 | 81.9667   |
| Si      | 7.6500  | 14.2067  | 6.5333  | 5.3800  | 7.3200  | 6.2900    |
| Fe      | 1.0100  | 0.8817   | 1.4933  | 1.8733  | 1.8617  | 3.8167    |
| Cu      | 2.5100  | 0.7943   | 2.0767  | 2.1267  | 2.1267  | 1.8733    |
| Mn      | 0.2330  | 0.0780   | 0.2043  | 0.3903  | 0.2903  | 0.1950    |
| Mg      | 0.0500  | >2.4     | >2.4    | >2.4    | 5.5100  | 4.7800    |
| Sr.     | <0.0001 | 0.0237   | 0.0078  | 0.0112  | 0.0104  | 0.0078    |

Table 2 shows the comparison of composite compositions with different volume variations of Si₃N₄ fractions. AC4B was aluminum with the main alloy 7-10% Si and 2-4% Cu. When viewed from the test results, there were excessive or less Si and Cu levels in the composite material. Mg element got increased significantly in the composite because it was added to increase the wettability. High Fe element was caused by the SKD61 stirring rod which melted during the stirring process.

3.2 Microstructure of Al AC4B/Si₃N₄ Composite
As shown in Figure 1, Al$_5$FeSi phase has a flat and sharp shape that is hard and brittle, where the phase can be formed due to the presence of Fe element in the composite. The phase is clearly seen in the composition of 10% Si$_3$N$_4$ shown in Figure 1 (f). Eutectic Si phase is also seen which would increase the composite hardness. Then Mg$_2$Si phase is also seen that the binary Mg$_2$Si has a Chinese script form. In addition, π-Al$_5$FeMg$_3$Si$_5$ is also present in microstructure whose property is brittle.

**Figure 1** Microstructure of (a) Al AC4B as Cast and and Al AC4B / Si$_3$N$_4$ Composite with different Volume Fractions (b) 1 vf-%, (c) 3 vf-%, (d) 5 vf-%, (e) 7 vf-%, and (f) 10vf-% at 500X

Al$_5$FeSi phase could be formed through a rapid cooling process with the reaction of L + Al$_8$Fe$_2$Si → Al + Al$_5$FeSi, and this phase had lower mechanical properties than that of Al$_8$Fe$_2$Si [5]. In addition, π-Al$_5$FeMg$_3$Si$_5$ phase could be formed due to the presence of Fe and Mg with a content below 6%. The phase was formed by reaction L → Al + Si + Mg$_2$Si + Al$_5$FeMg$_3$Si$_5$.

### 3.3 XRD
Figure 2 XRD pattern of Al AC4B with 1 vf-% Si₃N₄ Composite

The XRD test results in figure 2 show the presence of MgAl₂O₄ spinel phase which increases contact between AC4B matrix and Si₃N₄. MgO phase could be formed due to the reaction of magnesium with oxygen in the environment. The presence of Al₃Fe and Al₅FeSi phases were due to the presence of Fe content in the composite. π-Al₉FeMg₃Si₅ phase was formed due to the presence of Fe and Mg levels below 6% [6]. Mg₃Si phase which also had hard characteristic was also found in the composite.

3.4 SEM & EDX

Table 3 EDS Testing Result
| No. | B     | N     | O     | Mg    | Al    | Si    | Fe    | Cu    | Phase                                      |
|-----|-------|-------|-------|-------|-------|-------|-------|-------|--------------------------------------------|
| 1   | 0.39  | 46.29 | 16.55 | 24.06 | 12.16 | 0.54  | α-β Si$_3$N$_4$                          |
| 2   | 42.47 | 0.66  | 32.96 | 6.9   | 16.75 | 0.24  | π-Al$_9$FeMg$_5$Si$_5$, MgAl$_2$O$_4$     |
| 3   | 6.98  | 0.12  | 51.09 | 9.11  | 29.82 | 2.63  | Mg$_2$Si                                   |
| 4   | 6.07  | 0.07  | 46.03 | 2.86  | 27.17 | 2.24  | 1.1  | 14.47  | Al$_2$Cu                                   |
| 5   | 5.15  | 50.22 | 0.54  | 42.45 | 0.33  | 0.44  | α Al                                       |
| 6   | 5.32  | 0.05  | 50.33 | 0.48  | 42.22 | 0.34  | 0.02  | 0.49  | α Al                                       |

In addition to the SEM and EDS tests that were carried out at 6 different points, the observation on the fracture formed on the composite was done. Based on SEM results, there was a clustering of Si$_3$N$_4$ particles due to their higher density that caused agglomeration to occur at a certain point because aluminum dendrites would be consolidated first and finally caused Si$_3$N$_4$ particles to be blocked. Uneven distribution of Mg element could also be a cause of this phenomenon. MgAl$_2$O$_4$ is also predicted to occur in the area, but because the AC4B solidification process is higher than Si$_3$N$_4$ it causes the effluent effect and the interface between the matrix and the reinforce is not created [7]. Area number 2 shows the presence of elements of Al, Fe, Mg, and Si which indicate the existence of π-Al$_9$FeMg$_5$Si$_5$ phase. Mg$_2$Si and Al$_2$Cu phases are also detected in the EDS test results shown in areas number 3 and 4.

The fracture formed on the composite is mixed fracture. This is indicated by the cleavage and dimple of the fracture. Fractures with Cleavage form indicate brittle fracture, while dimple shows ductile fracture. Mixed fractures in composites are caused by a combination of two material properties between the reinforce and the matrix. Silicon nitride reinforce have brittle properties and aluminum matrices have ductile properties. The fracture image also shows the presence of cavities or porosity in the material caused by the presence of hydrogen gas that enters the molten aluminum during the casting process [8].

### 3.5 Mechanical Properties of Al AC4B/Si$_3$N$_4$ Composite

Composite density was supposed to increase because of the addition of Si$_3$N$_4$ since Si$_3$N$_4$ density was higher than that of AC4B in accordance with the theoretical density calculation. However, the composite density decreased as shown in Figure 4 (a). The decrease in density value was related to the higher amount of porosity in the addition of Si$_3$N$_4$ in Figure 4 (b) which would affect the composite density. This high porosity affected the fluctuations in the hardness, tensile strength, and toughness values of the composite [9].

![Figure 4](image-url) **Figure 4** Effect of Volume Fraction of Si$_3$N$_4$ on Density and Porosity

The highest tensile strength is obtained by adding 1% volume fraction of Si$_3$N$_4$ which results in a tensile strength of 104.94 MPa as shown in figure 5 (a). The composite elongation decreases proportionally with the addition of Si$_3$N$_4$ as shown in figure 5 (b). The tensile strength increased in the addition of 1% Si$_3$N$_4$ was
caused by a dispersion strengthening mechanism due to differences in the thermal expansion coefficient (CTE) between the AC4B aluminum matrix and Si₃N₄ which caused increased composite dislocation density [10]. Furthermore, fluctuating tensile strength values were due to the porosity of hydrogen gas formed in the composite since hydrogen gas solubility in molten aluminum would increase at high temperatures, while the decreasing elongation could be caused by the presence of the Mg₅Si phase which was brittle and the porosity formed.

![Graphs showing effects of Si₃N₄ volume fraction on tensile strength, elongation, hardness, and wear rate](image)

**Figure 5** Effect of Si₃N₄ Volume Fraction Addition on Tensile Strength, Elongation, Hardness, and Wear Rate

The addition of Si₃N₄ volume fraction increased the hardness of the composite, where the highest hardness occurred at the addition of 3% Si₃N₄ which was 49.5 HRB, while the hardness at 1% volume Si₃N₄ was 44.8 HRB as shown in figure 5 (c). Increased hardness was result of dispersion strengthening mechanism and the formation of Mg₅Si and Al₃Cu phases which were hard. The addition of Si₃N₄ particles caused composite toughness to decrease, indicated by the decreasing impact price according to Figure 5 (d). This showed that the energy that could be absorbed by the composite decreased. The reduced impact price could be caused by the presence of brittle π-Al₆FeMg₅Si₃ phase [11].

4. **Conclusion**

Si₃N₄ particles could increase the hardness and tensile strength of aluminum through a dispersion strengthening mechanism. It was found that the optimum tensile strength was obtained at the addition of 1% volume of Si₃N₄ and optimum hardness at the addition of 3% volume of Si₃N₄. Composite hardness was also influenced by the formation of Mg₅Si and Al₃Cu phases. The fluctuation of composite mechanical properties was due to the presence of porosity formed and Si₃N₄ clustering. This clustering was caused by poor solidity between the reinforcement and the matrix.
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Reference

[1] F. Campbell 2010 Chapter 1: Introduction to Composite Materials. Manuf. Process p 103
[2] C. Limmaneevichit and W. Eidhed 2003 Novel technique for grain refinement in aluminum casting by Al-Ti-B powder injection Mater. Sci. Eng. A, vol 355 pp 174–179
[3] K. T. Kashyap and T. Chandrashekar 2001 Effects and mechanisms of grain refinement in aluminium alloys, Bull. Mater. Sci vol 24 pp 345–353
[4] J. Hashim, L. Looney M.S.J. Hashimi 1999 Metal matrix composites: production by the stir casting method, J. Mater. Process. Technol vol 92–93 pp 1–7
[5] P. Tang et al. 2017 Effect of Al-Ti-C master alloy addition on microstructures and mechanical properties of cast eutectic Al-Si-Fe-Cu alloy pp 147–157
[6] M. Glazoff, A. V Khvan, V. S. Zolotorevsky, N. Belov, and A. Dinsdale 2018 Casting Aluminum Alloys: Their Physical and Mechanical Metallurgy p 180
[7] S. B. Prabu, L. Karunamoorthy, S. Kathiresan, and B. Mohan 2006 Influence of stirring speed and stirring time on distribution of particles in cast metal matrix composite Influence of stirring speed and stirring time on distribution of particles in cast metal matrix composite pp 268-273
[8] Y. L. Liu, S. B. Kang, and H. W. Kim 1999 Complex microstructures in an as-cast Al-Mg-Si alloy vol 41 pp 267-272
[9] M. Kok 2005 Production and mechanical properties of Al2O3 particle-reinforced 2024 aluminium alloy composites vol 161 pp 381-387
[10] P. Sharma, S. Sharma, and D. Khanduja 2015 Journal of Asian Ceramic Societies Production and some properties of Si3N4 reinforced aluminium alloy composites, Integr. Med. Res., vol 3 pp 352–359
[11] M. Salleh, M. Z. Omar, J. Syarif, and M. Abdulrazaq 2014 Thermodynamic Modelling of Al-Si-Cu Alloys for Semisolid Metal Processing, vol 43 pp 791-798