Journal of Metals, Materials and Minerals. Vol.18 No.2 pp.99-102, 2008

Fabrication of Al/Al_2O_3 Composite by Powder Metallurgy Method from Aluminum and Rice Husk Ash

Chinawad DHADSANADHEP[1*], Tachai LUANGVARANUNT,[1]**
Junko UMEDA[2] and Katsuyoshi KONDOH[2]

[1]Department of Metallurgical Engineering, Faculty of Engineering Chulalongkorn University,
Phyathai Road, Pathumwan, Bangkok 10330, Thailand.
[2]Joining and Welding Research Institute, Osaka University, 11-1 Mihogaoka,
Ibaraki, Osaka, 567-0047 Japan.

Received Sept. 30, 2008
Accepted Nov. 4, 2008

Abstract

Fabrication of Al-4wt.%Cu /Alumina composite was reported. The method of fabrication was powder metallurgy by in-situ reaction to form alumina during fabrication. Starting materials were mixture of aluminum, copper, and silica powders. Silica is in form of rice husk ash. The mixture was cold compacted, and sintered at 650°C for 1 h. Hot forging was carried out to further consolidate the sintered billet, followed by 10-h heat treatment at temperatures between 590 to 650°C. Phase analysis by XRD confirmed that in-situ reaction between silica and aluminum powders occurred during heat treatment at temperatures above 590°C. The products of reaction were silicon and gamma-alumina. Microstructure investigation showed that the reacted areas were the previous locations of silica powder. In some areas, remaining silica was found amid reaction products. Hardness of the fabricated composite made of Al-4wt.%Cu and 5, 10 and 15vol% silica are 16, 23, and 30 HRA, respectively.

Introduction

Aluminum and its alloys are widely used in many applications. They are light weight materials with high specific strength. However, in comparison with other materials such as ferrous alloys, their strength and stiffness are still inferior. Improvement of aluminum alloy can be done by several ways such as new alloy design, heat treatment, or reinforcement with other kinds of material to make composite materials. This research paper focuses on making composite material having pure aluminum as matrix phase, reinforced with alumina, which is produced in-situ during fabrication.

Fabrication method is powder metallurgy with its advantage of homogeneous mixing of constituent materials to obtain the decided composition. Aluminum powders and silica powders are mixed in solid state. The mixture is consolidated and heat treated to induce in-situ reaction between aluminum and silica to produce alumina and silicon as products. Alumina from the reaction will be homogeneously distributed in the matrix due to homogeneous distribution of the mixed silica. To facilitatechemical reaction, high-purity amorphous silica produced by burning of rich husk is chosen as starting materials with aluminum powder for the in-situ reaction. In addition, copper in the amount of 4 wt.% is added to facilitate sintering, by formation of liquid phase.

Previous studies by other researchers showed that in-situ formation of alumina by reaction between silica and aluminum was possible.(1-3) The temperatures of these methods are greater than is the melting temperature of aluminum; hence, they were liquid state processing. For the first time, solid state with partial melting is demonstrated here on the fabrication of aluminum/alumina composite.

Materials and Experimental Procedures

Aluminum powders (99.9% purity, 106 μm mean size, Kojundo Japan), copper powders (99.9% purity, 70 μm mean size, Kojundo Japan) and silica powders (99.7% purity, -250 μm size) were mixed by shaking in a 3x5 cm plastic bag for 20 mins. Rice husk ash silica was obtained from burning of rice husk as reported elsewhere.(4) The

---

*Graduate student, **Corresponding author: Tachai.L@ch la.ac.th

composition of the mixtures are Al-4wt.%Cu / 5vol.%Silica, Al-4wt.%Cu / 10vol.%Silica, and Al-4wt.%Cu / 15vol.%Silica. The mixture was cold compacted in a Ø30-mm die under a pressure of 214 MPa, and subsequently sintered at 650°C for 1 hrs. under flow of Ar gas in a tube furnace, using a heating rate of 5°C/min. The sintered billet was reheated to 600oC for 5 mins under flow of Ar, using a heating rate of 20°C/min. Hot forging was carried out immediately thereafter, under 660 MPa with a strain rate of 17.5 s^{-1}, in a Ø34-mm die. The temperature of the hot-forge die is 315°C. The forged billet was subsequently heat treated at temperatures of 590, 630, and 650°C for 10 hrs. to induce in-situ chemical reaction to form alumina from silica and aluminum. X-ray diffractometer (XRD) was used to investigate the phase change of the materials, and scanning electron microscope (SEM) with energy dispersive analysis (EDS) was used to investigate their microstructure and chemical composition. Hardness of the fabricated composite materials was measured using Rockwell scale A.

Results and Discussion

A typical forged billet is shown in Figure 1. It can be seen that the billet has smooth surfaces with a disc shape conformed to the cavity of the hot-forge die. Density of the composite material after each processing step is summarized in Table 1. After cold compaction and after sintering, the density is lower for billet with greater amount of silica. Due to the porous nature of amorphous silica in rice husk ash, the green density and sinter density is low. Sintered density is lower than green density. During sintering, release of gas previously trapped in porous silica causes slight volume expansion of the billet. After high deformation in hot forging, however, density increases and the variation of density with amount of silica is less. Density of rice husk ash silica in amorphous form is about 2.2 g/cm^3, which is not much different from that of aluminum at 2.7 g/cm^3. Therefore, with addition of silica, density of the composite decreases. Compared with the calculated densities of the three compositions in Table 1, assuming that chemical reaction among constituents did not occur, the density of the forged billet is very close to that of the calculated ones. Due to the porous nature of rice husk ash silica, and difficulty of deformation due to its hard and brittle characteristics, the difference between forged density and calculated density is larger for composition with greater amount of silica.

Figure 1. Forged billet of the Al-4wt.%Cu/15 vol.% Silica.

Table 1. Density of the composite material after cold compaction, sintering, and hot forging.

Composition	Green Density	Sintered Density	Forged Density	Calculated Density
	(g/cm^3)	(g/cm^3)	(g/cm^3)	(g/cm^3)
5vol%RHA	2.43	2.19	2.71	2.75
10vol%RHA	2.24	2.02	2.67	2.72
15vol%RHA	2.12	1.97	2.55	2.69

The phases in the as-sintered, as-forged, and as-heat treated billet were investigated by XRD, and compared in Figure 2. Sintering and hot forging cannot induce chemical reaction between aluminum powders and silica powders. Only after a long-time heat treatment of 10 hrs. gamma-alumina and silicon were formed according to the reaction,

$$4Al(s) + 3SiO_2(amorphous) \rightarrow 2Al_2O_3(s) + 3Si(s).$$

Thermodynamics calculation shows that Gibbs Free Energy of the reaction is -203 kJ/(mol of Al) and -257 kJ/(mol of Al) at 298 and 600 K, respectively. It should be noted that the thermochemical data of SiO_2(l) at 298 and 600 K is selected for that of SiO_2(amorphous) for the calculation. As Gibbs Free Energy of the chemical reaction is negative, the reaction is controlled by transport phenomena, which in this case are diffusion of aluminum and amorphous silica. Reaction was facilitated by a high diffusion rate at high temperature, and liquid phase formed which-according to Al-4wt.%Cu in phase diagram-starts at around 580°C. Long time heating of the forged billet above 580°C as heat treatment after hot forging, therefore, is necessary to accelerate the in-situ reaction to form alumina which is the reinforcement phase.

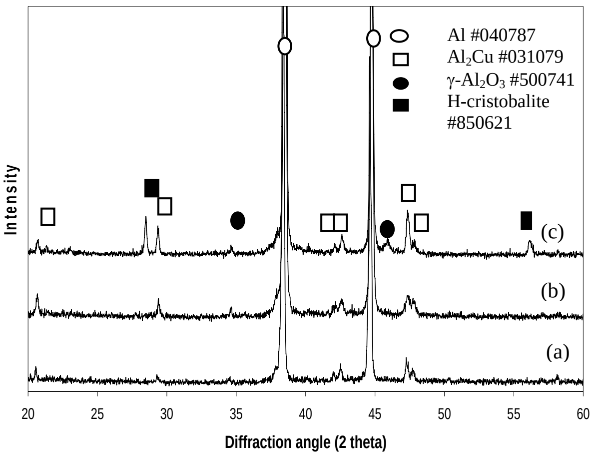
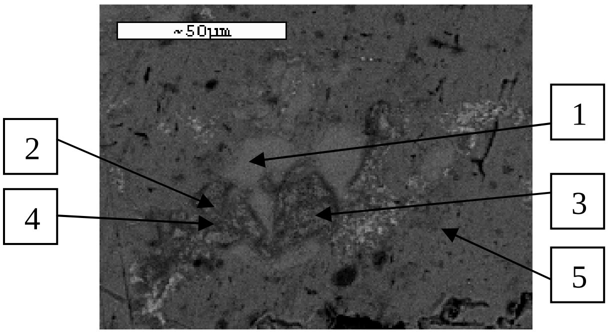


Figure 2. X-Ray diffraction patterns of Al-4wt.%Cu / 15 vol.% Silica (a) as-sintered, (b) as-forged, (c) as-heat treated.

Microstructure of the Al-4wt.%Cu / 15vol% Silica heat treated at 630°C for 10 hrs. is shown in Figure 3. With the XRD result of Figure 2, the following conclusion can be drawn. Area 1, which is silicon rich, is silicon phase. Areas 2 to 4, which are rich in both aluminum and oxygen are gamma-alumina phase. Area 5, which is the matrix, is aluminum phase. The microstructure showed good bonding between gamma-alumina and the matrix, by in-situ reaction to form alumina during fabrication. Copper is distributed in all areas, as precipitate of Al_2Cu compound, which has a size smaller than the resolution of SEM. The products of the chemical reaction, which are silicon and alumina, are in the reacted area, where silica existed formerly. In some areas of the composite material, there are still silica phase existing in the middle of the reacted area, as shown in Figure 4, even after heat treating at 650°C for 10 hrs.

Heat treatment at temperatures of 590, 630, and 650°C for 10 hrs. showed similar microstructure and phases. The amount of porosity in the microstructure increases with increasing heat treatment temperature. The in-situ reaction may occur more violent and at a faster rate when heating at high temperature, and may cause the excessive porosity. Porosity in the microstructure may be caused by gas released from porous silica and the difference of molar volume of reactants and products of reaction. Optimum heat treatment temperature was found to be 590°C, after which porosity in the composite is minimal; and density remained the same as after hot forging. Hardness of the composite materials after heat treatment at 590°C is shown in Figure 5. Hardness of the fabricated composite made of Al-4wt.%Cu and 5, 10 and 15vol% Silica are 16, 23, and 30 HRA, respectively. The hardness increases with the amount of silica, reflecting the reinforcement of the formed alumina in the composite. The matrix material, Al-4wt.%Cu, which corresponds to grade 202AB, was reported to exhibit hardness between 55-60 HRH for T1 heat treatment. The hardness of composite materials in this research thesis is much greater than the value reported for grade 202AB due to reinforcement by alumina.



Element	Weight %	Atomic %	Element	Weight %	Atomic %
Area 1			**Area 3**		
O	1.58	2.74	O	27.94	40.38
Al	6.67	6.86	Al	62.06	53.18
Si	91.25	90.18	Si	6.11	5.03
Cu	0.5	0.22	Cu	3.88	1.41
Area 2			**Area 4**		
O	26.19	38.21	O	23.69	35.63
Al	63.16	54.64	Al	67.34	60.06
Si	6.99	5.81	Si	1.91	1.63
Cu	3.67	1.35	Cu	7.07	2.68
Area 5					
O	5.27	8.72			
Al	90.33	88.65			
Si	1.51	1.42			
Cu	2.9	1.21			

Figure 3. SEM image of reacted area with EDS analysis of chemical composition of each phase.

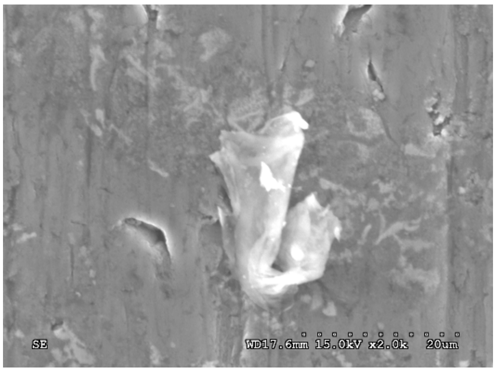


Figure 4. SEM image of remaining silica shown as bright irregular particle surrounded by reacted area (Al-4wt%Cu /15vol%Silica: Sintering 650°C -1h + Hot Forging 600°C + Heat treating 650°C -10hrs).

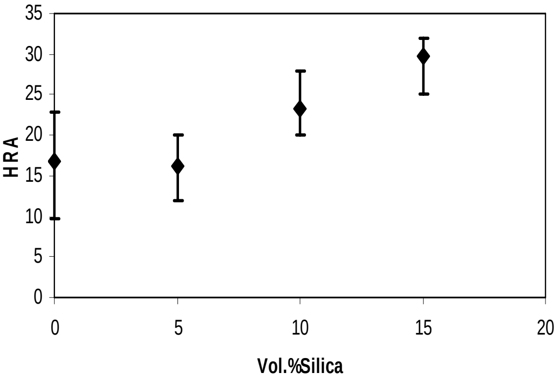


Figure 5. Hardness of the composite after heat treated at 590°C for 10 hrs.

Conclusions

Fabrication of Al-4wt.%Cu / Al_2O_3 composite by in-situ reaction between aluminum powders and silica powders was reported. Powder metallurgy method was utilized, by mixture of aluminum powders, copper powders, and silica powders in form of rice husk ash. Sintering of powder mixture was carried out at 650°C for 1 h, followed by hot forging at 600°C, and finally heat treatment at 590°C for 10 hrs. Chemical reaction between silica and aluminum powder took place during long time heating at temperature above 590°C. The investigation of microstructure showed reacted areas, consisting of gamma-alumina and silicon. In some areas, silica powder remained amid the products of the reaction. Copper was found distributed in all areas, in form of Al_2Cu compound. Hardness values of the fabricated composite made of Al-4wt.%Cu and 5, 10 and 15vol% Silica are 16, 23, and 30 HRA, respectively.

References

1. Liu, W. and Koster, U. 1996. Microstructure and properties of interpenetrating alumina/ aluminium composites made by reaction of SiO_2 glass preforms with molten aluminum. *Mater. Sci. Eng.* **210** : 1-7.

2. Yoshikawa, N., Hattori, A. and Taniguchi, S. 2003. Growth rates and microstructures of reacted layers between molten Al-Fe alloy and SiO_2. *Mater. Sci. Eng.* **342** : 51-57.

3. Li, J.Q. and Xiao, P. 2002. Joining alumina using an alumina/metal composite. *J. Eur. Ceram.* Society, **22** : 1225-1233.

4. Luangvaranunt, T., Jearwun, S. and Kondoh, K. 2007. Removal of adsorbed moisture on iron powders by rice husk ash. *J. Met. Mater. Miner.* **17(1)** : 47-52.