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PREPARATION OF γ-Al_2O_3 NANOCRYSTALLITES BY SOL-GEL AUTO COMBUSTION PROCESS AND PRODUCTION OF Al-Al_2O_3 ALUMINUM MATRIX COMPOSITES

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The γ-Al_2O_3 nanocrystallites were synthesized successfully by combination of sol-gel auto-combustion process and ultrasonic method. The overall process involves three steps: formation of homogeneous sol at 50 °C; formation of dried gel at 90 °C, and experiments revealed that Al_2O_3 dried gel derived from glycine (as fuel) and nitrates sol exhibits self propagating combustion at 400 °C that it is ignited in air. After auto-combustion process, the Al_2O_3 nanopowders were calcined at 1100 °C. Also, aluminum alloy matrix composites reinforced with 0.75 and 1.5 vol.% γ-Al_2O_3 nanoparticles were fabricated via stir casting method. Fabrication was performed at 850 °C temperatures. The resulting composites were tested for their nanostructure and present phases. Based on the obtained results, optimum amount of reinforcement and casting temperature were determined by evaluating the density, hardness and compression tests of the composites.

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Keywords: γ-Al_2O_3, Auto-Combustion, Ultrasonic, Aluminum Matrix Composite.

1. Introduction

Alumina ceramic (Al_2O_3) is a hard refractory ceramic, which has been used in high temperature structural and substrate application because of its good strenght and low thermal expansion coefficient. Nevertheless, like other monolithic ceramics, Al_2O_3 is apt to suffer from low ductility and low fracture toughness. Therefore, metals such as aluminum and cobalt or alloys are added to ceramics to improve their toughness [1–5]. However, nanoscale γ-alumina (γ-Al_2O_3) powder is difficult to obtain, because of two reason; First, γ-Al_2O_3 is an a stable phase after calcining at high temperature, which easily prompt the grain growth of powder, and make it difficult to get nanoscale particles, secondly, γ-Al_2O_3 particles tend to aggregate during dehydration process in wet chemistry method. Therefore, it is necessary to develop new methods to overcome this problem [6].

The sol-gel method, in particular, is one of the most useful and attractive method for the synthesis of nanosized materials because of its advantages such as; good stoichiometric control and production of ultrafine powder with narrow size distribution in a relatively short processing time at a very low temperature [7]. Metal oxides powders with nanostructure particles have been synthesized by various methods [8–12].

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There are many methods for fabrication of particulate reinforced metal matrix composites (MMCs) such as powder metallurgy [11], squeeze casting [13, 14], compocasting [15–18]. For the metal matrix composites, molten metal mixing is a cost effective method while powder metallurgy is costly, and squeeze casting provide good in filtration quality of chopped performs [19, 20].

In this research, the synthesis of γ-Al_2O_3 nanostructure was studied by combination of sol-gel auto-combustion and ultrasonic methods using aluminum nitrate and glycine as a precursors. Moreover, γ-Al_2O_3 nanopowders were incorporated into the molten A356 aluminum alloy via stir casting method at various conditions.

2. Experimental

Aluminum nitrate [$Al(NO_3)_3.9H_2O$], glycine, NH_4OH and Al powder were obtained from Merck with analytical grade. All materials were used without further purification. Deionized water was used for all experiments. Aluminum (A356) was obtained from Kian Alloy Company (Kashan–Iran) as metal matrix of composites. The chemical analysis of Al alloy is given in Table 1.

Table 1. Chemical analysis of Al (A356).

Element	*Al*	*Si*	*Mg*	*Fe*	*Cu*	*Ni*	*Zn*	*Mn*	*Ti*
Mass Percent	91.73	7.23	0.38	0.32	0.18	0.05	0.05	0.02	0.01

Nanoperticles of Al_2O_3 were prepared by combination of sol-gel auto-combustion and ultrasonic methods. Appropriate amounts of analytical grade $Al(NO_3)_3.9H_2O$ and glycine were taken and a small amount of ammonium hydroxide was added carefully to the solution to change the pH value to 7. The mole ratio of nitrate to glycine is equal 1:3. During this procedure, the solution was continuously stirred for 8h and kept at 60 °C. The resultant gel is poured in a platinum crucible and heated at about 400 °C. Schematic flow chart of sol-gel processing that was applied in this study is shown in Fig. 1.

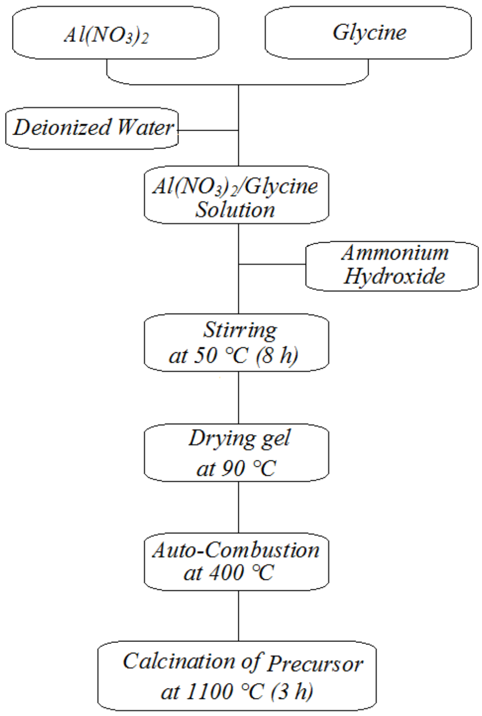

Fig. 1. Flow chart of sol-gel processing.

In this research, the reinforcement phases used to improve strength of A356 aluminum alloy matrix were nano γ-Al_2O_3 as ceramic reinforcing particles. Therefore, cast alloy (A356 Al) was selected. The γ-alumina powders were mixed homogeneously with 0.75 and 1.5 vol.% and Al powder at room temperature. Then, the mixture was placed in stainless steel mold and 20 MPa of pressure was used to form the Al_2O_3 performs.

For casting Al-Al_2O_3 composite, a resistance furnace equipped with a stirring system was used. After smelting the aluminum ingots, an amount given of keryolite was added to the molten Al alloy and the stirring was established for a few minutes. Al_2O_3 nanopowders with 0.75 and 1.5 vol.% were wrapped into the aluminum foils and added to the melt Al alloy to produce Al-Al_2O_3 composite. The processing temperatures have been chosen at 850 °C. Stirring was continued for another 10 minutes for homogenous dispersion and to prevent agglomeration of particulates. A metallic mold is used for casting. Finally, specimens fabricated in six various conditions were prepared for subsequent microstructural and mechanical analyses. The morphology of the γ-Al_2O_3 nanopowders and Al-Al_2O_3 composites were analyzed by scanning electron microscopy (SEM, Oxford CAMSCAN-MV2300), X-ray diffractometer (Model: XPERT-MPD, Phylips) using Cu K_α radiation (λ=1.05406Å) with operated at 40 kV and current of 40 mA and specimens were polished and etched using Keller solution.

The bulk density of samples was determined by the Archimedes method. Theoretical density was estimated by using mixture law. Hardness test were made with a load of 306.56 N and a punch diameter of 2.5 mm. Data of hardness were calculated using at least ten indentations on two polished specimens. The compressive strength test was conducted in air at room temperature (Instron Universal Testing Machine-1195 machine) according to ASTM-B557.

3. Result and iscussion

3.1. XRD and Nanostructural Analyses

The XRD pattern of the Al_2O_3 nanoparticles is shown in Fig. 2. The particle size of the samples has been determined from XRD peaks, employing Scherrer's equation:

$$D = \frac{k\lambda}{\beta cos\theta}$$

Where D is the crystallite size, θ is the Bragg angle, λ is the wavelength of X-ray radiation (Cu K_α) and β is the full width at half maximum (FWHM) of the most intense diffraction peak. The results of XRD show the formation of single phase of γ-Al_2O_3 nanostructure. A typical XRD plot is shown in Fig.2. The diffraction angle and relative intensity of the characteristic peak of the samples are well consistent with those of the standard JCPDS 10-0173.

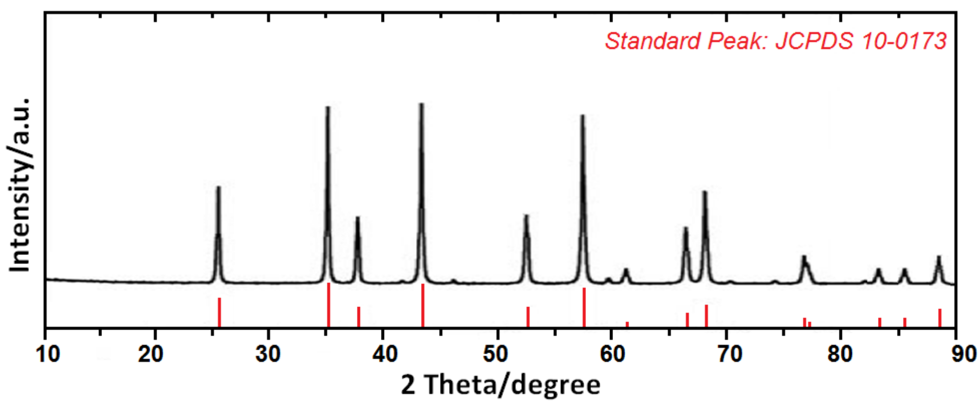

Fig.2. The XRD pattern of standard peak and γ–Al_2O_3 nanoparticles.

The morphology of alumina was investigated with using SEM. The SEM image of γ-Al_2O_3 nanoparticles is shown in Fig. 3. The mean diameter size of the prepared γ-Al_2O_3 is found to be 31 nm.

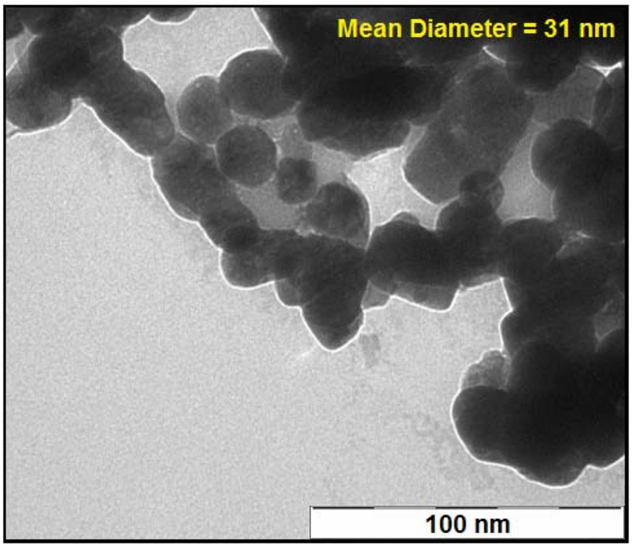

Fig.3. The particle size of γ–Al_2O_3 nanoparticles determine by SEM.

The morphology of the Al-γ-Al_2O_3 is shown in Fig. 4. The black matrix is aluminum; the white spots represent γ-Al_2O_3 nanoparticles and the grey area is silicon-rich interdendritic Al-Si eutectic. The phases are considered by arrow on SEM images. It should be noted that γ-Al_2O_3 nanoparticles were well dispersed in the aluminum matrix and just a partial agglomeration in composites with 0.75 and 1.5 vol.% of γ-Al_2O_3 can be considered in Fig.4.

Similar nanostructure were observed for 0.75 vol.% Al_2O_3 composite. All results from XRD and SEM analyses confirm feasibility of stir casting method to produce this kind of composites with well distribution of reinforcement.

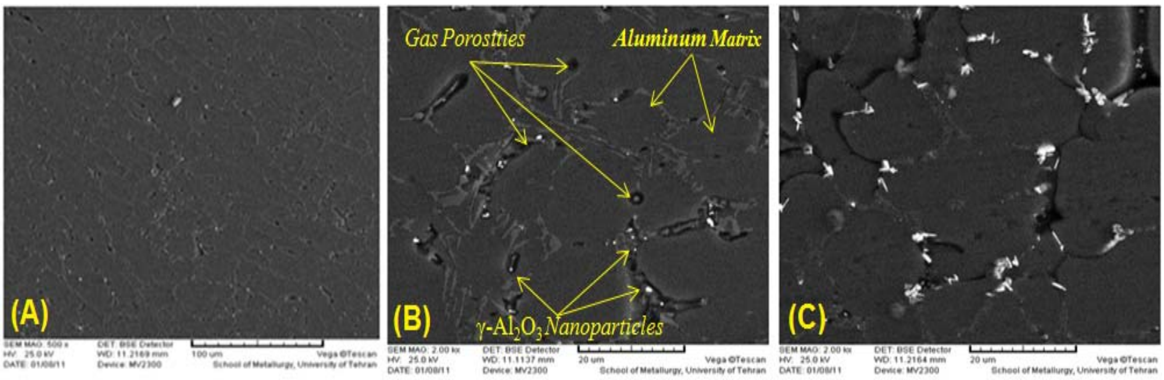

Fig. 4. SEM images of Al-Al_2O_3 composite speciments with 1.5 vol.% Al_2O_3 at 850 °C and different magnifications: (A) 500 kx, (B) 2.00 kx and (C) 20.00 kx.

3.2. Density Measurements

The effects of volume percent of γ-Al_2O_3 nanoparticles on density of Al-Al_2O_3 composites produced by stir casting method is given in Table 2. According to the mixture law, by increasing the volume fraction of Al_2O_3 particle in aluminum, density of sampler should be increased, because the density of Al_2O_3 is more than aluminum. It should be noted that temperature plays an important effect to increase the wet ability of particles.

Table 2. The comparison experimental and theoretical densities of Al-Al_2O_3 composites containing 0.75 and 1.5 vol.% γ-Al_2O_3.

Volume percent of γ-Al_2O_3	*Experimental Density ($g.cm^{-3}$)*	*Theoretical Density ($g.cm^{-3}$)*
0.75	2.65	2.66
1.5	2.65	2.68

3.3. Mechanical Properties

3.3.1. Hardness Test

The effects of the volume percent of γ-Al_2O_3 nanoparticles on hardness properties of the prepared composites with 0.75 and 1.5 vol.% is determined 58 and 64 BHN at 850 °C, respectively.

It is considered that hardness of Al_2O_3-reinforced composite is more than that of unreinforced alloy. It can be attributed to the higher hardness of ceramic particles compared to aluminum matrix alloy. Also, increasing the volume fraction of γ-Al_2O_3 nanoparticles at 850 °C is increased hardness values because of the number and total surface area of Al_2O_3 particles increase with increasing Al_2O_3 particle volume fraction [21, 22].

3.3.2. Compression Test

The effects of the volume percent of γ-Al_2O_3 nanopowder on the compression strength of Al-Al_2O_3 composites were determined with 0, 0.75 and 1.5 vol.% Al_2O_3 700, 819 and 875 MPa at 850 °C, respectively. As demonstrated, with increasing the volume percent of reinforcing particles, the value of the compressive strength compared with unreinforced matrix alloy is increased at 850 °C.

The initial enhancement seems to be due to work-hardening behavior. This could be related to effects of elastic properties of ceramic particles and inhibition response of plastic deformation of matrix by them; ceramic particles can only deform elastically while aluminum matrix can deform plastically. So if the boundary is assumed to be strong, ceramic particles prevent plastic deformation of the matrix and this leads to higher work-hardening rate [22].

4. Conclusion

Nanocrystalline γ-Al_2O_3 powder was successfully synthesized by a combination of sol-gel auto combustion process and ultrasonic irradiation method. The well γ-Al_2O_3 nanopowder was prepared when pH=7. The particles have been calcined at 1100 °C for 3h. Then the products were placed in ultrasonic bath of n-propanol at room temperature for 15 minutes. The grain sizes of the nanopowder are found to be about 31 nm. The SEM results showed that grains were regular sphere-shaped nanoparticles. The XRD data shows Al_2O_3 nanoparticle have γ structure. A356 Aluminum reinforced with γ-Al_2O_3 nanocomposites was successfully fabricated via stir casting method. Reinforced particles were well distributed in the matrix of composites. However, partial agglomeration was observed in composites with increasing of Al_2O_3 content. Mechanical properties such as hardness and compressive strenght improved. Also, composite containing 1.5 vol.% γ-Al_2O_3 fabricated at 850 °C showed improved hardness and compressive strenght of the alloy by 42 and 25%, respectively, in comparison with other specimens. Therefore, it can be concluded that composite with 1.5 vol.% Al_2O_3 content, cast at 850 °C, represent maximum mechanical properties and it can be considered at the optimum fabrication conditions.

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