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Influences of Al_2O_3 grain size on high-temperature oxidation of nano-Ni/Al_2O_3 composites

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ABSTRACT

Two 5 vol% Ni/Al_2O_3 composites with the difference in Al_2O_3 grain size were fabricated by pulsed electric current sintering technique to investigate the influence of Al_2O_3 grain size on oxidation behavior of the composites. Average Al_2O_3 grain sizes of two fabricated composites were 1.1 μm and 0.5 μm after sintering. Oxidation tests were conducted at temperatures ranging from 1100 to 1350 °C for 1–48 h in air. A thin $NiAl_2O_4$ layer was observed in exposed surface of samples after oxidation. An oxidized zone that consisted of Al_2O_3 matrix and $NiAl_2O_4$ grains was defined. Growth of the oxidized zone obeyed the parabolic law. Influences of Al_2O_3 grain size on high-temperature oxidation of the composites were discussed.

1. Introduction

Alumina (Al_2O_3) has been well-known as a high-performance structural material and widely applied in industries because of its advantages such as excellent heat resistance, high mechanical strength and wear resistance. However, similar to other ceramics, Al_2O_3 materials have inherently strong covalent and ionic bonds that prohibit substantial dislocation motion or plastic deformation. They fail to alleviate stress concentrations that occur in front of crack tips. Hence, Al_2O_3 materials are easily fractured as a consequence of crack propagation resulting from a slight surface flaw or internal flaw. Al_2O_3 materials thus exhibit poor toughness that restricts their application. In order to increase their toughness, dispersion of non-oxide phases such as Ni, NiAl, Co and SiC in Al_2O_3 matrix were proposed [1–4]. On the other hand, the presence of the non-oxide phases gives the materials the self-healing function that occurs at high temperatures in the air [2,5–7].

Ando and his co-workers reported self-healing function that can be achieved on ceramic-based composites such as SiC/mullite, SiC/Al_2O_3 [7–11]. When non-oxide dispersoids in the matrix were oxidized at high temperatures, the oxidation product filled up the cracks and involved the reduction of stress concentrations at crack tips. As a result, the mechanical strength was recovered up to the level of as-polished samples. Furthermore, this mechanism improves their performance with longer lifetime and more reliability at high temperatures.

Similarly, nano-Ni dispersed Al_2O_3 composites also have the self-healing function at high temperatures in the air [12–14]. Maruoka et al. reported the recovery of mechanical strength for nano-Ni/Al_2O_3 that was able to achieve when the fraction of surface crack disappearance is over 50% [14]. The study indicated that the crack disappearance effectiveness depended on the formation of $NiAl_2O_4$-oxidation product that mostly referred to the diffusion of Ni^{2+} along grain boundaries of the Al_2O_3 matrix. The conclusion implies that finer grain size of the matrix could give a positive effect on the crack-healing function.

While self-healing effectiveness mostly depends on the diffusion of Ni^{2+} and Al^{3+} cations, the oxidation resistance of materials depends on diffusion of O^{2-} ions along grain boundaries. At high temperatures, oxygen can penetrate the oxide matrix that causes the oxidation of dispersed Ni particles within the matrix. The region consisting of oxidized nickel particles develops as increasing oxidation temperature and duration. Nanko et al. reported the oxidation behavior of Ni/Al_2O_3 composites at temperatures ranging from 1200 to 1300 °C [15], in which the growth of oxidized region was attributed to obedience of the parabolic law. It

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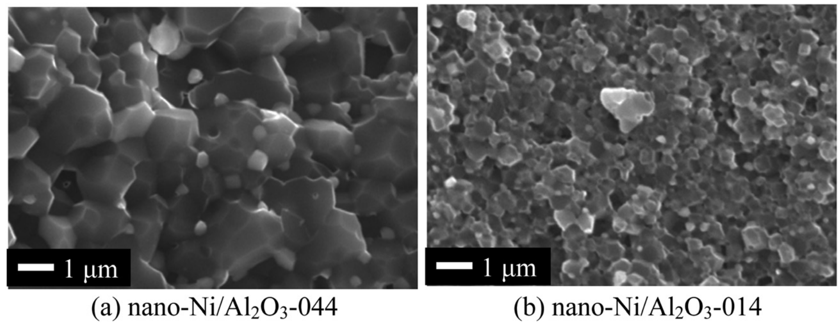

Fig. 1. SEM images of fractured surface of as-sintered samples: (a) Ni/Al_2O_3-044 and (b) Ni/Al_2O_3-014.

implied that the mass transport in the oxidized region was the rate-controlling process, which was the diffusions of cations and oxide ions along grain boundaries of the Al_2O_3 matrix. Maruoka et al. reported the influence of Si-doping of nano-Ni dispersed Al_2O_3 composites on high-temperature oxidation [16]. The study concluded that the growth rate of the oxidized zone was decreased effectively at 1200 °C by doping 0.1 mol% Si in Ni/Al_2O_3. Doping of Si was attributed to the change to grain boundary diffusion in polycrystalline Al_2O_3. The grain boundary diffusion in polycrystalline Al_2O_3 could be also decreased by either dopant of Y, SrO, $LuO_{1.5}$ or ZrO_2 [17,18]. Since the grain boundary diffusion is an important factor responsible for the oxidation resistance of the composite, influences of Al_2O_3 grain size on oxidation resistance of Ni/Al_2O_3 are in need of investigation for high-temperature applications.

In this study, the influences of the different structure of Al_2O_3 matrix on oxidation resistance of 5 vol% Ni/Al_2O_3 composites are discussed. The oxidation resistance of the composites was evaluated through the growth rate of oxidized zone after heat treatment in air at temperatures ranging from 1100 to 1350 °C for 1–48 h.

2. Experimental procedures

In order to fabricate two types of 5 vol% Ni dispersed Al_2O_3 composites with the difference in Al_2O_3 grain structure, two different types of Al_2O_3 powder (Sumitomo Chemical Co. Ltd, AA-04, $d = 0.44\,\mu m$ and Taimei Chemicals Co. Ltd, TM-DAR, $d = 0.14\,\mu m$) were used as starting materials. Aqueous slurries containing $Ni(NO_3)_2 \cdot 6H_2O$ (Kojundo Chemical Laboratory Co. Ltd, purity 99.9%) and the Al_2O_3 powders were prepared by milling in a plastic bottle with Al_2O_3 balls for 24 h. The composite that was prepared from Al_2O_3 with average particle size of 0.44 μm is referred to as Ni/Al_2O_3-044, hereafter. Similarly, Ni/Al_2O_3-014 is referred to as the composite that was prepared from the Al_2O_3 with average particle size of 0.14 μm. After drying at 400 °C in a boiling flask, the powder mixtures were milled by using an alumina mortar for 15 min. These powder mixtures were heat-treated at 600 °C for 12 h in a stream of the Ar–1% H_2 gas mixture to reduce NiO to Ni metallic phase. The powder mixtures were consolidated in a graphite die by pulsed electric current sintering (PECS) at 1400 °C for Ni/Al_2O_3-044 and 1200 °C for Ni/Al_2O_3-014 under 50 MPa uniaxial pressure in a vacuum for 5 min holding time. The relative density of all the consolidated samples used in this study attained at least 99% of the theoretical density. Fig. 1 shows the fractured surfaces of the as-sintered samples. Ni particles, that could be visible as the bright contrast dots, were homogeneously dispersed in the Al_2O_3 matrix. The average particle size of these Ni particles was approximately 300 nm while the average Al_2O_3 grain size of Ni/Al_2O_3-044 was 1.1 μm, and that of Ni/Al_2O_3-014 was 0.5 μm.

High-temperature oxidation tests were conducted at temperatures ranging from 1100 to 1350 °C for 1 h up to 48 h in the air with a heating rate of 400 °C/h. The tested samples were put on alumina balls (3 mm in diameter) in an alumina crucible and exposed in the air at the investigated conditions. Phase identification of the samples was carried out by X-ray diffraction (XRD). Oxidation evolution of heat-treated samples was evaluated via the growth rate of oxidized zone observed on cross-sectioned surface by scanning electron microscope (SEM).

3. Results

Fig. 2 shows XRD patterns obtained from sample surface before and after oxidation tests. Before oxidation, only two dominant phases of Ni and Al_2O_3 are detected on the sample surface (Fig. 2a). After oxidation at 1200 °C for 1 h, $NiAl_2O_4$ phase was identified as shown in Fig. 2b. On the other hand, the intensity of signals from Ni phase was decreased. The peak intensity of $NiAl_2O_4$ phase which was formed after oxidation at 1200 °C for 24 h was significantly increased and replaced for the existence of Ni-metallic phase. In addition, nickel oxide phase could not be identified on oxidized samples. This phenomenon has been reported by Trumble et al. [19] that NiO did not coexist with Al_2O_3. It implies that Ni particles reacted with Al_2O_3 matrix as the following equilibrium:

$$2Ni + 2Al_2O_3 + O_2 = 2NiAl_2O_4 \qquad (1)$$

Fig. 3 shows the SEM images of cross-section surface of Ni/Al_2O_3-014 and Ni/Al_2O_3-044 after oxidation at 1200 °C for 24 h.

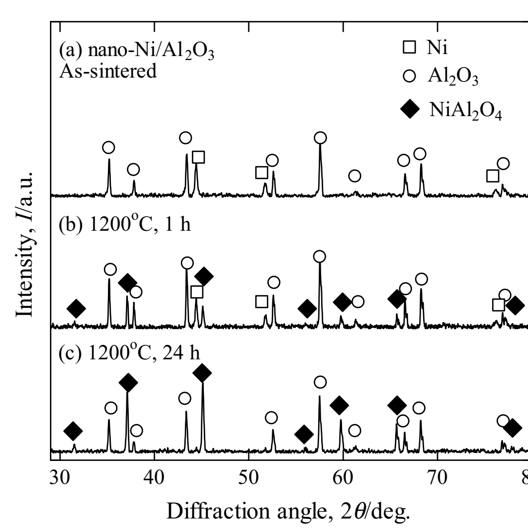

Fig. 2. XRD patterns of nano-Ni/Al_2O_3: (a) before oxidation, (b) oxidized at 1200 °C for 1 h and (c) oxidized at 1200 °C for 24 h in air.