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Influence of alumina powder size on mechanical properties of in-situ coated alumina fiber-reinforced alumina composites

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Abstract

Continuous alumina fiber-reinforced alumina-matrix composites were fabricated by hot-pressing. Fibers were coated by La-monazite, and matrix was prepared using the mean size of 7 nm and/or 180 nm alumina raw powders. It was observed that the powder size of matrix affected the properties of the composites. Composites using the nano-sized powder (7 nm) as matrix showed higher pseudo-ductility, but lower maximum strength and density than those of the composite using the submicron-sized powder (180 nm) as matrix, regardless of the sintering temperature. Woody-type fracture due to fiber pull-out from matrix could be observed for all composites regardless of the size of powder for matrix after sintering at relatively lower temperatures. Nano-sized powder supplies a looser matrix than submicron powder, therefore, it should be corresponded to higher pseudo-ductility of the composites. Combination of the nano-sized and submicron-sized powder as matrix gives composites with highest strength as well as enough ductility.

Keywords: Al_2O_3; Composites; Fibres; Microstructure; Mechanical properties

1. Introduction

All-oxide ceramics matrix composites (CMCs) have received widespread attention and were extensively studied in recent years. This goal was driven by its potential wide applications in high temperature oxidizing environment as damage-tolerant refractory materials since it is inherently oxidation resistant.[1] However, for the engineering materials applied in industry, catastrophic fracture during service should be absolutely avoided. Thus, high toughness of composites is generally required and expected. In the past several years, great efforts for the improvement of toughness of continuous oxide-fiber-reinforced CMCs was mainly concentrated on the development of high strength and high elastic modulus fiber and on the design of the microstructure of fiber/matrix interface. Especially the latter, two approaches have been taken to design microstructure of oxide composites. One is based on the fiber coatings that bond fiber and matrix weakly, if so, fiber debonding and crack deflection can be achieved during fracture.[2] Another relies on the weakness and low stiffness of a porous matrix itself to prevent damage from crack extension into the fibers, even though the interface between fiber and matrix might be strongly bonded.[3] Great progress has been achieved in these researches. On the other hand, further researches to obtain high toughness composites for industrial applications are still necessary, since the fabrication technique of oxide composites is more immature than that of non-oxide composites, such as SiC.[4]

It is well known that properties of composites not only depend on the properties of fiber and fiber/matrix interface, but depend on properties of matrix as well. Therefore, matrix also plays a very important role in the mechanical properties of the composites. It has been reported that the size of powders for matrix could influence the density, thermal conductivity, mechanical properties and sintering properties of ceramics.[5–10] J. C. Goldsby found that high temperature mechanical behavior of alumina ceramics was affected by nanometer-sized alumina powder.[5] G. Li et al. reported that an addition of nano-alumina particles could increase strength of porous alumina ceramics.[6] Effect of alumina powder size on density and thermal conductivity was also reported.[7,8] However, it is still unclear of their role in the continuous fiber reinforced CMCs. In order to clarify how the powder size of matrix effects on

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mechanical properties of composites, in this study, powders with different size, including nanometer-sized alumina powder (7 nm) and submicron-sized alumina powder (180 nm), were applied to form matrix component of composites, and effects of raw powder size on mechanical properties were investigated.

2. Experimental procedures

2.1. Fiber coating

Fabric cloth used in this experiment was a commercially available satin woven αAl_2O_3 fiber cloth (Almax, 99.5 wt.% Al_2O_3, from Mitsui Mining and Material Co. Ltd., Tokyo, Japan). Fiber bundle is consisted of 1000 fibers with diameter of ~10 μm for each fiber. Tensile strength of the as-supplied fiber is 1.65 GPa. Before coating, cloth was heated to 1000 °C for 1 h in air to remove a sizing material, and then cut to 35×35 mm in size. Details of the coating process were mentioned elsewhere,[11] and simply described as follows. $La(NO_3)_3$ and H_3PO_4 solutions with 0.1 molar concentration were prepared by dissolving La $(NO_3)_3{\cdot}6H_2O$ and 85% phosphoric acid in distilled water at room temperature, respectively. First, the fabric cloth was dipped in the $La(NO_3)_3$ solution with slightly stirring and dried at 60 °C for 10 min. After that, the cloth was dipped in H_3PO_4 solution slowly and dried at 110 °C for 5 min. Following that, it was rinsed in distilled water cautiously to remove the excessive $LaPO_4$ and/or other impurities from coating precursor. And then it was dried at 110 °C for 30 min in air to form monazite coating. After 10 cycles of dipping and drying, the deposited coating layer was heated to 500 °C for 1h for further solidification of the coating and removing organic impurities.

2.2. Fabrication of composites and characterization

Alumina matrix was formed by infiltration of a well-dispersed alumina suspension into the open space of the La-monazite coated alumina woven fabric, as mentioned above. Flow chart of the matrix infiltration process is shown in Fig. 1. High purity nano-sized alumina powder (mean size of 7 nm, γ-alumina, 99.99 wt.%, Taimei Chemical, Co. Ltd, Japan) and submicron-sized power (mean size of 180 nm, α-alumina, 99.5 wt.%, Taimei Chemical, Co. Ltd, Japan) were used to prepare two kinds of aqueous alumina suspensions. Both of the suspensions were prepared with low concentration (10 wt.% of solid) and high concentration (30 wt.% of solid) for the formation of matrix. All of the suspensions contained a small amount of polyvinyl alcohol (PVA, Kanto Chemical, Japan), and were stirred for 24 h at room temperature to disperse particle agglomerates completely. All samples in this experiment were infiltrated by the lower-concentration alumina suspension with five infiltration-dry cycles at first to form matrix in the open space among fibers in bundles, because better rheological property of the lower concentration suspension was supposed to be beneficial to infiltrate powder much sufficiently into the fiber bundle. And then the higher-concentration alumina suspensions were utilized to form green matrix on the surface of the cloth by another 3–4 infiltration-dry rounds. Detail of the infiltration process is as follows: monazite-coated cloths were immersed into the 10 wt.% concentration nano-sized powder suspension to form sample A, or immersed into the 10 wt.% submicron-sized powder suspension to form sample B. Both of them were put into a vacuum chamber, which was pumped by rotary pump for 30 min and then dried at 110 °C for 30 min in air. This infiltration process was repeated 5 times. After that, A and

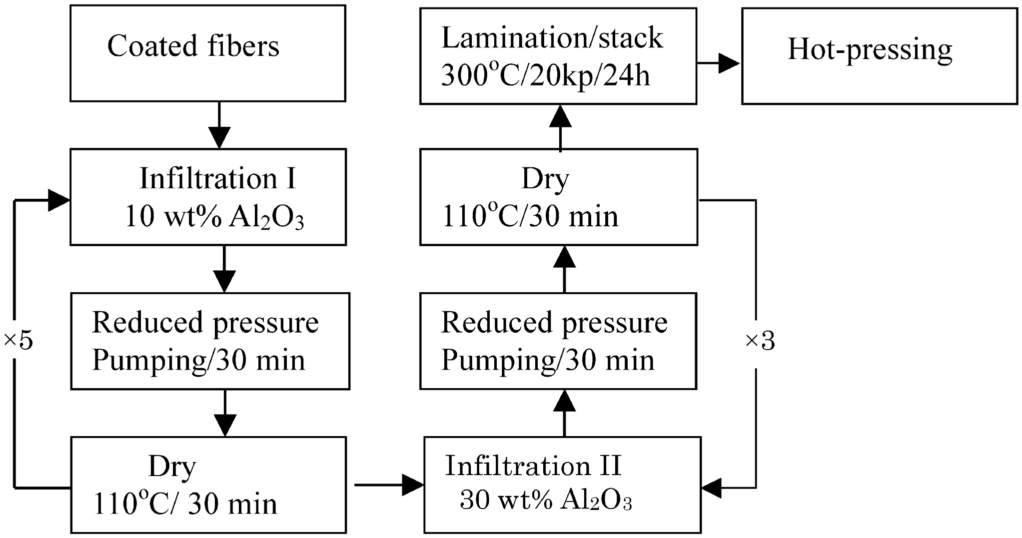

Fig. 1. Flow chart of matrix infiltration for alumina/alumina composites.

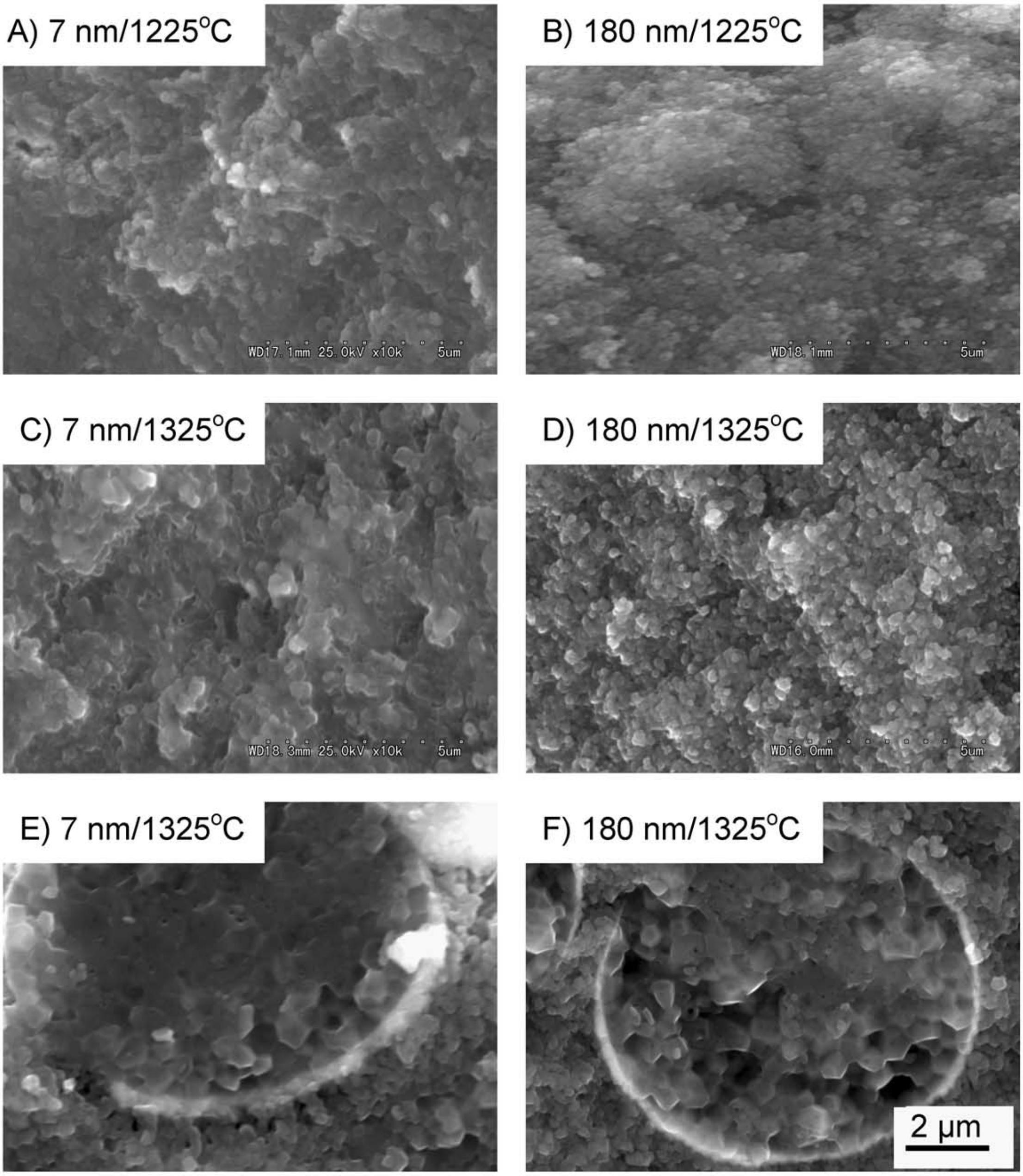

Fig. 2. SEM photographs of fracture surface of alumina matrix in composites, composites sintered at 1225 °C with (A) 7 nm and (B) 180 nm powder as matrix, (C) and (E) 7nm, (D) and (F) 180 nm, sintered at 1325 °C.

Table 1
Properties of hot-pressed alumina/alumina composites

	Matrix powder size (nm)	Sintering temp. (°C)	Bulk density (g/cm^3)	Porosity (%)	Maximum strength (MPa)
Sample A	7	1225	2.62	28.4	85
		1275	2.71	26.6	103
		1325	2.93	21.2	122
Sample B	180	1225	2.76	22.2	102
		1275	3.16	13.2	151
		1325	3.35	7.7	203
Sample C	7 and 180	1225	2.87	18.6	116
		1275	3.24	15.3	155
		1325	3.36	12.6	226

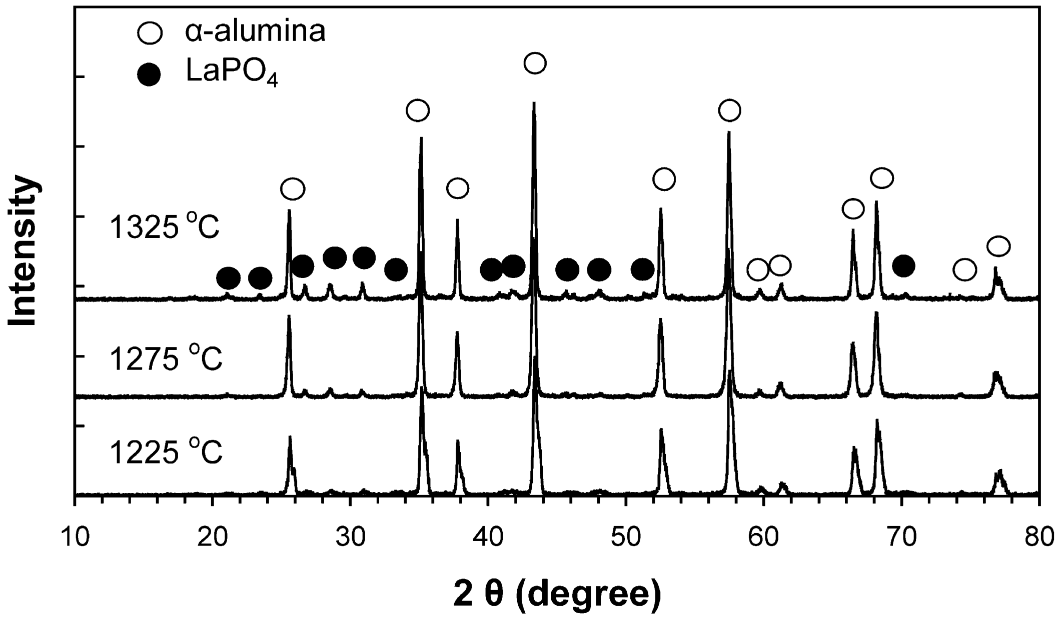

Fig. 3. XRD profiles (Cu-K α radiation) of composites using nano-sized alumina powder as matrix sintered at different temperature.

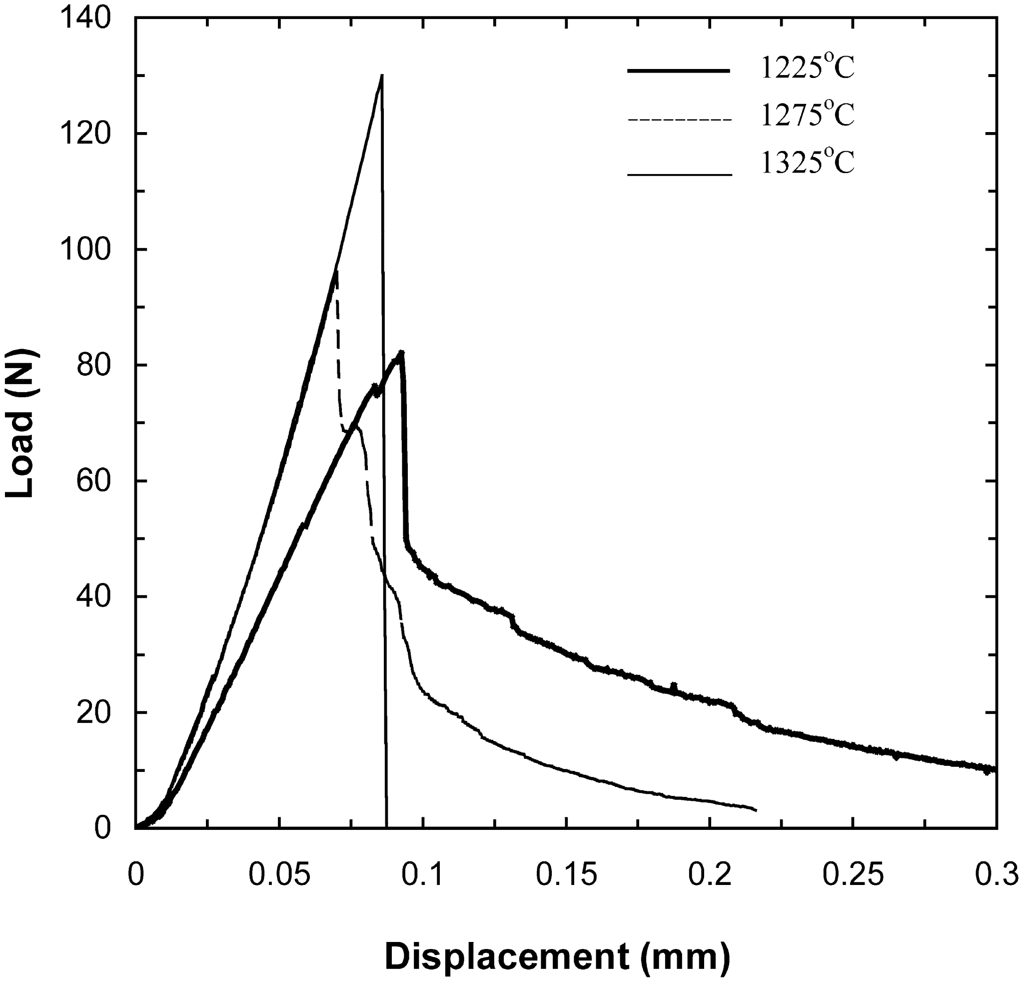

Fig. 4. Load–displacement curves of composites with 7 nm powder as matrix sintered at different temperatures.

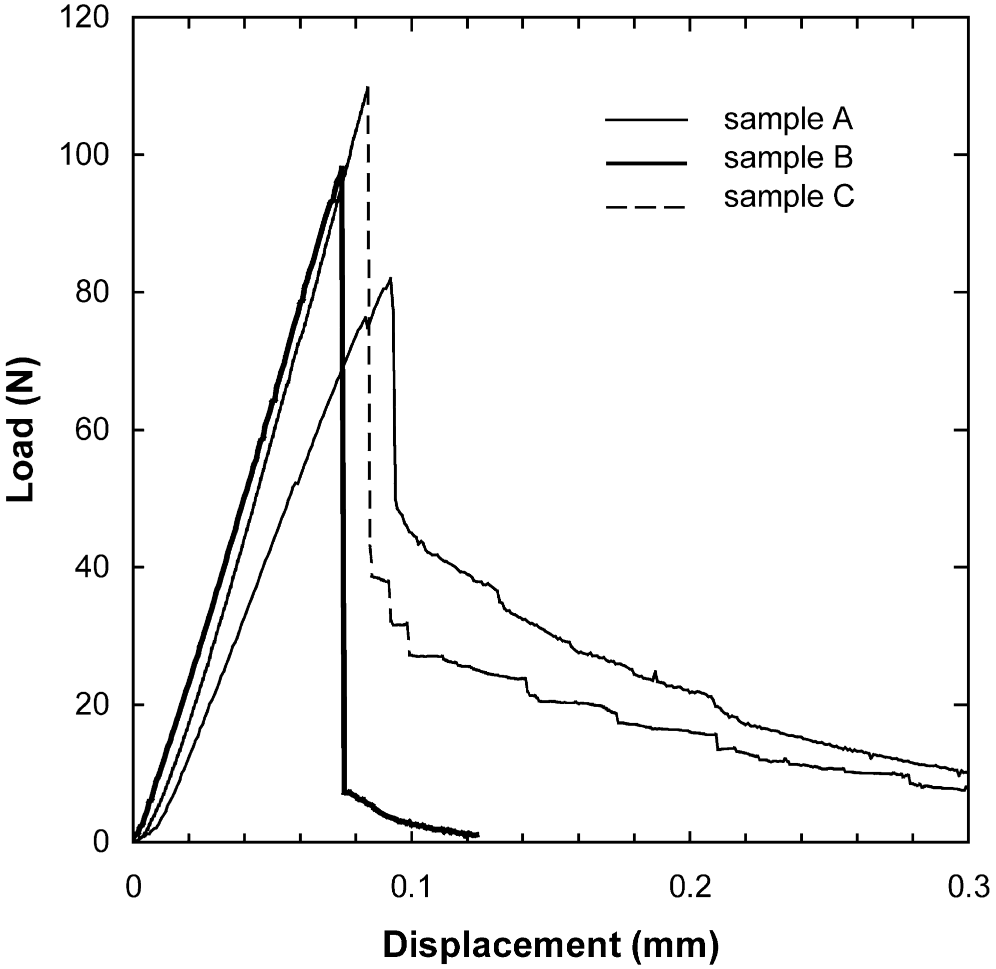

Fig. 5. Load–displacement curves of composites sintered at 1225 °C with different size of matrix powder.

B samples were immersed into nano-sized alumina powder suspension with a concentration of 30 wt.% and submicron-sized powder suspension with the same concentration, respectively. The same infiltration and dry process was repeated 3–4 times as described above. Then sample A with nano-sized powder matrix and sample B with submicron-sized powder matrix were prepared. While sample C was first immersed into the nano-sized alumina suspension with concentration of 10 wt.% for five rounds, and then it was immersed into the submicron-sized alumina suspension with concentration of 30 wt.% for another 3 cycles. The infiltration and dry process was the same as that of sample A and B.

After infiltration, eight infiltrated cloths were laminated. Then it was heat-treated at 300 °C for 24 h with 20 kPa pressure, and finally was hot-pressed at 1225–1325 °C under 40 MPa in Ar atmosphere for 1 h (Hi-multi 5000, Fuji Dempa Kogyo Co. Japan). Final composites with fiber volume fraction of about 40% were cut into 3.5 mm×35 mm×~3.5 mm bars along fiber direction of 0/90° for the measurement of bulk density and flexural strength. Bulk density of the hotpressed composites was measured by the Archimedes method using water. Caution was paid to restrict over removing of water from surface of porous specimens. Flexural strength was measured at room temperature by three-point flexural method using a universal testing machine (Type-1185, Instron Co Ltd., USA). Crosshead speed was 0.05 mm/min and the span was 30 mm. Maximum strength value was an average of five tested specimens. Fracture surface of composites were observed by scanning electron microscopy (SEM). The phases produced in the composites were analyzed by X-ray diffractometry (XRD).

3. Results and discussion

Bulk density, porosity and flexure strength of the composites with different matrix powder size were summarized in Table 1. Bulk density and flexural strength of the composite generally increased with the increase of sintering temperature regardless of the raw powder size of matrix. However, the composites containing the

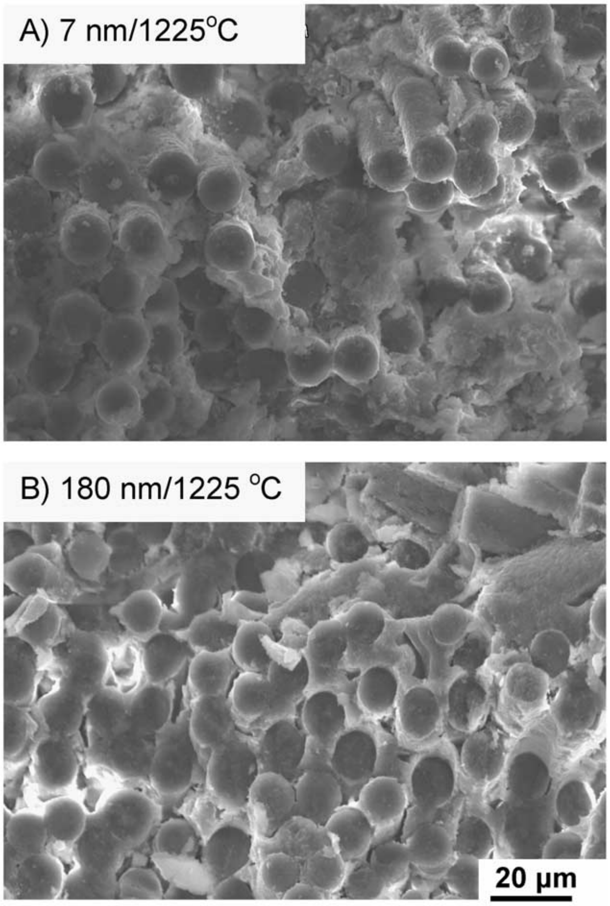

Fig. 6. SEM photographs of fracture surface of composites sintered at 1225 °C with (A) 7 nm and (B) 180 nm powder as matrix.

submicron-sized α-alumina powder as matrix (sample B and C) have much higher bulk density and flexure strength than those of the composites only using the nano-sized alumina powder as matrix (sample A). It seems that the nano-sized γ-alumina powder can not let the matrix of composite as dense as the submicron powder matrix. Those results correspond to that of R. Dong et al.,[12] who reported that the strength of composites using a nano-sized powder as matrix showed only half strength of the composites using a submicron-sized powder as matrix. Furthermore, in this study, the composites using the combination of nano-sized and submicron-sized powder as matrix (sample C) showed a little higher density as well as flexure strength in all examined sintering temperatures than that of the composite only using the submicron-sized powder as matrix (sample B). Porosity of the sample C was not obviously lower than that of sample B, even in some of the sintering temperature, it was a little higher than that of sample B, indicating that the nano-sized powder might play a role in the densification process of composites. G. Li et al.[6] found that an increase of nano-sized alumina powder content resulted in decrease of density of porous alumina ceramics, suggesting that raw powder size influenced on the densification process, and then affected on the mechanical properties of the ceramics.

Fig. 2 shows fracture surface of the composites using the nano-sized powder (sample A) and submicron-sized powder (sample B) as matrix sintered at different temperatures. For both sample A and sample B, grains in the matrix of composites sintered at 1325 °C (Fig. 2(C) and (D)) were obviously larger than that of the composite sintered at 1225 °C [Fig. 2(A) and (B)]. Cavities in the matrix of sample A are obviously more than that of the sample B regardless of the sintering temperature. Furthermore, the mean grain size of matrix after sintering using the nano-sized raw powder is larger than that of the matrix using the submicron-sized raw powder, suggesting that the powder size might greatly affect the densification process. Thus, strength as well as density of composites would be affected. About sample C (not shown in Fig. 2), the matrix near fibers, which was formed from the nano-sized powder, shows much looser packing than that of the matrix far from fibers, which was formed from the submicron-sized powder.

X-ray diffraction spectra of the composites sintered at different temperatures using the nano-sized powder as matrix are shown in Fig. 3. It is known that γ-alumina could transform into α-alumina above ∼1060 °C. In these spectra, only α-alumina and La-monazite presented and no other phases presented for all composites, indicating all γ-alumina in the matrix has transformed into α-alumina after sintering.

Load–displacement curves of the composites using the nano-sized powder and sintered at different temperatures are shown in Fig. 4. The composite sintered at 1225 °C showed inelastic fracture and relatively good pseudo-ductility was obtained. Wood-like fracture surface of the composite could be observed, as shown in Fig. 5(A), indicating that fibers were debonded from matrix during fracture of the composite. With increasing sintering temperature to 1275 °C, the composite still had good pseudo-ductility. At 1325 °C, ductility of the composite disappeared and it became completely brittle. Meanwhile, alumina grains both in fiber and matrix became obviously coarse [Fig. 2(E) and (F)], and it is supposed to be an important reason for the reduction of ductility of the composite, even though the fiber/matrix direct bonding is resisted by monazite coating on the fiber surface. Degradation of fiber strength should be another reason for brittle fracture.

Fracture behavior of the composites with different matrix powder size was compared in Fig. 5. These composites were hot-pressed at 1225 °C under 40 MPa for 1 h. The composite using the nano-sized powder as matrix showed the highest pseudo-ductility among the three kinds of composites. On the contrary, the composite containing only the submicron-sized powder showed a little pseudo-ductility. The composite using both the nano-sized and submicron-sized powder as matrix (sample C) showed medium ductility in the three kinds of composites, however, the fracture energy is almost the same with composite using only the nano-sized powder as matrix. It was observed that looser matrix around fibers of the nano-sized powder infiltrated matrix [Fig. 6(A)] contrasted to denser matrix around fibers in the submicron-sized powder infiltrated matrix [Fig. 6(B)], suggesting that density of the matrix plays an important role in the fiber debonding during fracture, since the fibers in the composite need to overcome higher resistance force during pulling-out when the composite is consisted of denser matrix, even all the fibers were coated by monazite. Furthermore, the loosely packed matrix formed by nano-sized powder can branch cracks and absorb fracture energy, which was supposed as another important reason for fibers debonding. It thus confirmed again that the ductility of the composites not only depends on the fiber, and fiber/matrix interface, but also depends on the matrix.

4. Conclusions

Mechanical properties of the alumina fiber-reinforced alumina composites were influenced by the matrix powder size. Nano-sized powder as matrix raw material could improve pseudo-ductility of the composites, while its strength as well as bulk density would be decreased. The composites using the submicron-sized powder as matrix showed the opposite tendency. Using both nano-sized and submicron-sized powder as matrix showed highest strength as well as enough ductility. Increasing sintering temperature could increase maximum strength and density of the composites regardless of the powder size, but ductility of the composites was reduced. For the continuous fiber reinforced ceramics matrix composites, the matrix properties, combined with fiber and fiber/matrix interface, plays an important role in mechanical properties of composites.

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