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Neutron-irradiation effect on the mechanical properties of alumina fiber

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Abstract

This paper describes the neutron irradiation effects on the deterioration of alumina fiber (made by Mitsui Mining Material, Almax), a typical electrical insulation material. The material was irradiated at the research reactor at Kyoto University Research Reactor Institute with a maximum fluence of 5.6×10^{23} n/m^2 (energy: $E > 0.1$ MeV). Tensile strength and tensile modulus of the specimen scarcely changed. Observation with a scanning electron microscope (SEM) and a transmission electron microscope (TEM) did not indicate any changes in crystal or pore structure. However, the Weibull coefficient of tensile strength decreased as the irradiation dose increased. This suggests an increase in the defect size distribution. © 1998 Elsevier Science B.V.

1. Introduction

Alumina ceramics have common characteristics such as high heat resistance, high electrical insulation, excellent wear resistance and weakness in embrittlement. Alumina is usually very difficult to form into a fibrous state. However, remarkable advances in fabrication technology in recent years have enabled us to form it in a fibrous state, which has a potential to be used in many areas by partially overcoming its weak points and by retaining its high heat — as well as radiation — resistance. Because of its excellent properties in addition to relatively low price, alumina has become one of the most popular materials in the fineceramics area. Several papers [1–6] have reported that, in spite of its excellent radiation resistance, irradiation in alumina may produce swelling mainly due to induced point defects, changing the physical properties, degrading its electrical resistance. These papers, however, deal with alumina plate or block and no reports have been published on alumina fiber. Papers describing radiation effects on ceramics fibers have been confined to silicon carbide (SIC) and carbon (graphite) fibers [7,8]. As described above, high purity alumina in a long fiber state has been difficult to fabricate, but recently some products have been manufactured on a commercial basis. Products containing more than 80% alumina are divided into three categories: high purity alumina (Al_2O_3), alumina–silica (Al_2O_3–SiO_2), and alumina–silica–boria (Al_2O_3–SiO_2–B_2O_3). High purity alumina fiber includes Almax made by Mitsui Mining Material Co., Nextel 610 made by 3 M Co, and Saphicon made by Saphicon Inc. Table 1 shows the properties of the three fibers [9]. The tensile moduli of these high purity alumina fibers range from 330 to 373 GPa and their breaking elongations are from 0.5 to 0.6%.

Saphicon is a single crystal and thick monofilament fiber whose diameter is 125 μm (±25 μm). Its thick diameter makes it inappropriate for manufacturing clothes. Nextel 610 is a fiber recently developed by 3 M Co. Its diameter is about 10 μm. SiO_2 and Fe_2O_3 are successfully

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Table 1
Dimensions of various high purity alumina fibers [9]

Brand name	Almax	Nextel 610	Saphicon
Manufacturing company	Mitsui Mining Material Co.	3 M Co	Saphicon Inc.
Chemical composition (wt%):			
Al_2O_3	99.5	99	100
SiO_2		0.2–0.3	
Fe_2O_3		0.4–0.7	
Fiber diameter (μm)	10	10–13	125
Filament number (No./yarn)	1000	390	monofilament
Crystal phase	α, polycrystal	α, polycrystal	α, monocrystal
Density (kg/m^3)	3.6×10^3	3.75×10^3	3.97×10^3
Tensile strength (GPa)	1.8	1.9	2.1–3.4
Tensile elastic modulus (GPa)	330	373	352

added to limit the crystal grain of alpha-Al_2O_3 to 500 nm or less. Almax is a continuous fiber with an alumina purity of 99.5% and was developed by Mitsui Mining Material Co. It has a crystal grain size of about 0.5 μm and is famous for its convex-concave fiber surface [9]. Crystal grain growth tends to give the fiber a brittleness which makes it difficult to handle. A similar product called Fiber FP, previously made by DuPont, was thick with a diameter of 20 μm making it difficult to use in fabricating clothes. On the other hand, the fiber strength of Almax and its thin 10 μm diameter, make it very suitable for products such as cloth, tape, sleeves, and board.

In the present study, Almax made by Mitsui Mining Material Co. is selected to investigate the neutron irradiation effect on its texture and strength.

2. Experimental method

2.1. Neutron irradiation of specimen

Alumina fiber used for the experiment is a polycrystalline alpha-alumina fiber (Almax) with the alumina purity of 99.5 wt% or more, with the fiber diameter of 10 μm, the crystal diameter of 0.5 μm and the porosity of about 8%. These are three categories of fiber: a non-size, an epoxy resin size, and a polyvinyl alcohol size. This paper reports on the nonsize. A yarn is composed of 1000 filaments which are specimens. Ten yarns cut to about 0.1 m were contained in a quartz tube filled with 1/3 atm helium and the quartz tube was contained in an aluminum tube with 2/3 atm helium. Four aluminum tubes were then irradiated in the core of the Kyoto University Reactor (KUR) at the Kyoto University Research Reactor Institute. KUR is basically operated from Tuesday through Friday every week at a thermal power of 5 MW for about 75 h continuously. The temperature of the coolant water is kept below 50°C during operation. The neutron flux of the irradiation point of the reactor is as follows; thermal neutron flux $\phi_{th} = 4.7 \times 10^{17}$ n/m^2 s, epithermal neutron flux $\phi_{epi} = 1.7 \times 10^{16}$ n/m^2 s and fast neutron flux $\phi_f = 1.7 \times 10^{17}$ n/m^2 s ($E > 0.1$ MeV).

The four aluminum tubes which contained specimens were irradiated in the reactor for two weeks, one month, three months and six months, resulting in four different fast neutron (>0.1 MeV) fluences of 0.45, 1.0, 3.1, 5.6×10^{23} n/m^2, respectively. The temperature of the specimens during irradiation was not measured, but was estimated to be about 90°C. Because the shape memory alloys were applicable to the temperature measurement [10], and by means of this technique, Kodaka et al. measured the temperature of aluminum specimens under the same condition and reported that it was below 88 ± 2°C [11]. Gamma dose rate of the specimens was estimated 3×10^2 Gy/s and it raised the temperature by about 40°C.

As is shown in Table 2, the fractional concentration of displacements (C_d) of each irradiated specimen was calculated using the following equation [12]:

$$C_d \cong t\phi \frac{\overline{\sigma}_s}{A}\left(\frac{E_f}{E_d}\right) \Bigg/ \log\left(\frac{4E_f}{AE_d}\right), \tag{1}$$

where E_d is the threshold energy of displacement, E_f is

Table 2
Fractional concentration of displacements (C_d) at each level of neutron irradiation (ϕ)

ϕ (n/m^2)	0.45×10^{23}	1.0×10^{23}	3.1×10^{23}	5.6×10^{23}
C_d of aluminum (dpa)	4.51×10^{-3}	9.80×10^{-3}	3.04×10^{-2}	5.49×10^{-2}
C_d of oxygen (dpa)	5.08×10^{-3}	1.35×10^{-2}	4.19×10^{-2}	7.56×10^{-2}

Table 3
Tensile strength (σ), Weibull coefficient of tensile strength (m) and tensile modulus (E) at each level of neutron irradiation (ϕ)

ϕ (n/m^2)	0.00×10^{23}	0.45×10^{23}	1.0×10^{23}	3.1×10^{23}	5.6×10^{23}
σ (GPa)	1.54 ± 0.15	1.60 ± 0.13	1.54 ± 0.17	1.52 ± 0.17	1.50 ± 0.19
m	11.2	13.3	9.2	9.6	8.4
E (GPa)	291 ± 26	292 ± 21	279 ± 5	285 ± 17	263 ± 15

the cut-off energy of fast neutrons, $\bar{\sigma}_s$ is the mean elastic scattering cross-section, A is the atomic mass (amu), ϕ is the flux of fast neutrons and t is the irradiation time.

The E_d values of aluminum and oxygen in alumina are 18 and 75 eV at 300 K [13].

2.2. Tensile test and observation by scanning and transmission electron microscopes

A filament was fixed on a pasteboard using an epoxy resin adherent to create a specimen with a span length of 25 mm. About 35 specimens were prepared from which 31 to 35 specimens were used for tests, excluding specimens which lacked the proper shape. Tensile tests were performed at a stroke speed of 1 mm/min using a tensile test machine (Autograph: AG-500A made by Shimadzu). Maximum breaking load and corresponding elongation ratio were measured. The elongation ratio was about 0.5–1% and maximum breaking load was $1.4–1.5 \times 10^{-2}$ kg. Under these conditions, there was no influences of the stiffness of the machine on the measurement. The tensile strength was calculated from the maximum breaking load and the diameter of each filament was measured by an optical microscope prior to the test. A tensile modulus was obtained by dividing the tensile strength by the elongation ratio.

Fractured surfaces of the fiber were observed using a scanning electron microscope (SEM: ABT-150F made by Topcon). An observation specimen was prepared from the fractured specimen in the tensile test using vaporized gold.

To observe defects in alumina grain fiber, a transmission electron microscope (TEM: H-9000 made by Hitachi) was used. To prepare TEM specimens, several alumina fibers were placed on a single hold mesh (supporting plate of 3 mm in diameter) with a bore of 0.5 mm in diameter and ion-polished (7 kV Ar$^+$, 100 mA) for 3 h.

3. Results and discussion

3.1. Tensile test

Table 3 shows the relationship between neutron irradiation (ϕ), tensile strength (σ), tensile modulus (E), and the Weibull coefficient (m). Figs. 1 and 2 indicate changes in tensile strength and in tensile modulus due to irradiation, respectively. Fig. 3 shows changes in the Weibull coefficient of the tensile strength due to irradiation. As shown in these figures, tensile strength and tensile modulus scarcely decreased under neutron irradiation up to $\phi = 5.6 \times 10^{23}$ n/m^2. On the other hand, as shown in Fig. 4, the Weibull coefficient decreased as the fluence increased, thus proving its distribution spread. This suggests an increase in defects that cannot be observed by an electron microscope.

3.2. Observation by scanning electron microscope

Fig. 5(a) is a SEM photograph of a fracture surface of the specimen before neutron irradiation. The figure shows an alumina fiber fractured at the grain boundary that is surrounded by crystalline facets, indicating a weak contrast due to a semispherical dent. Also, it shows a transgranular fracture of alumina where pores less than 0.1 μm in diameter are observed. These cavities are significantly smaller than alumina grain, which is not expected to affect the tensile strength.

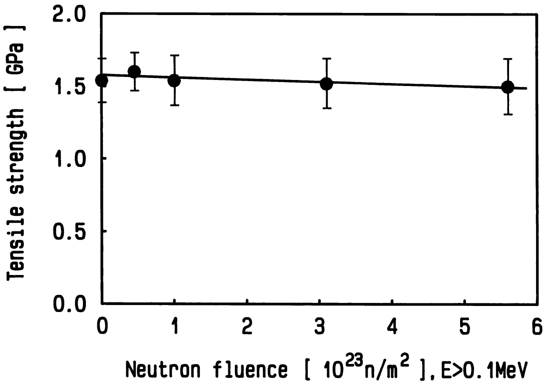

Fig. 1. Change in tensile strength due to neutron irradiation.

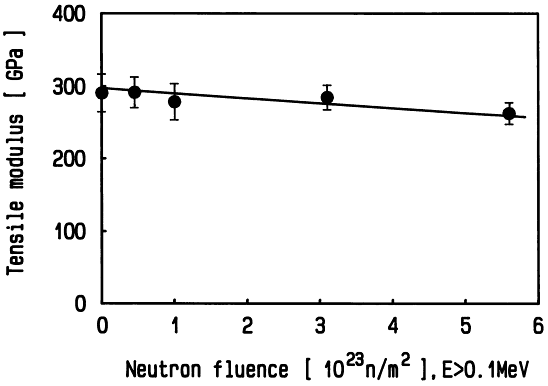

Fig. 2. Change in tensile modulus due to neutron irradiation.

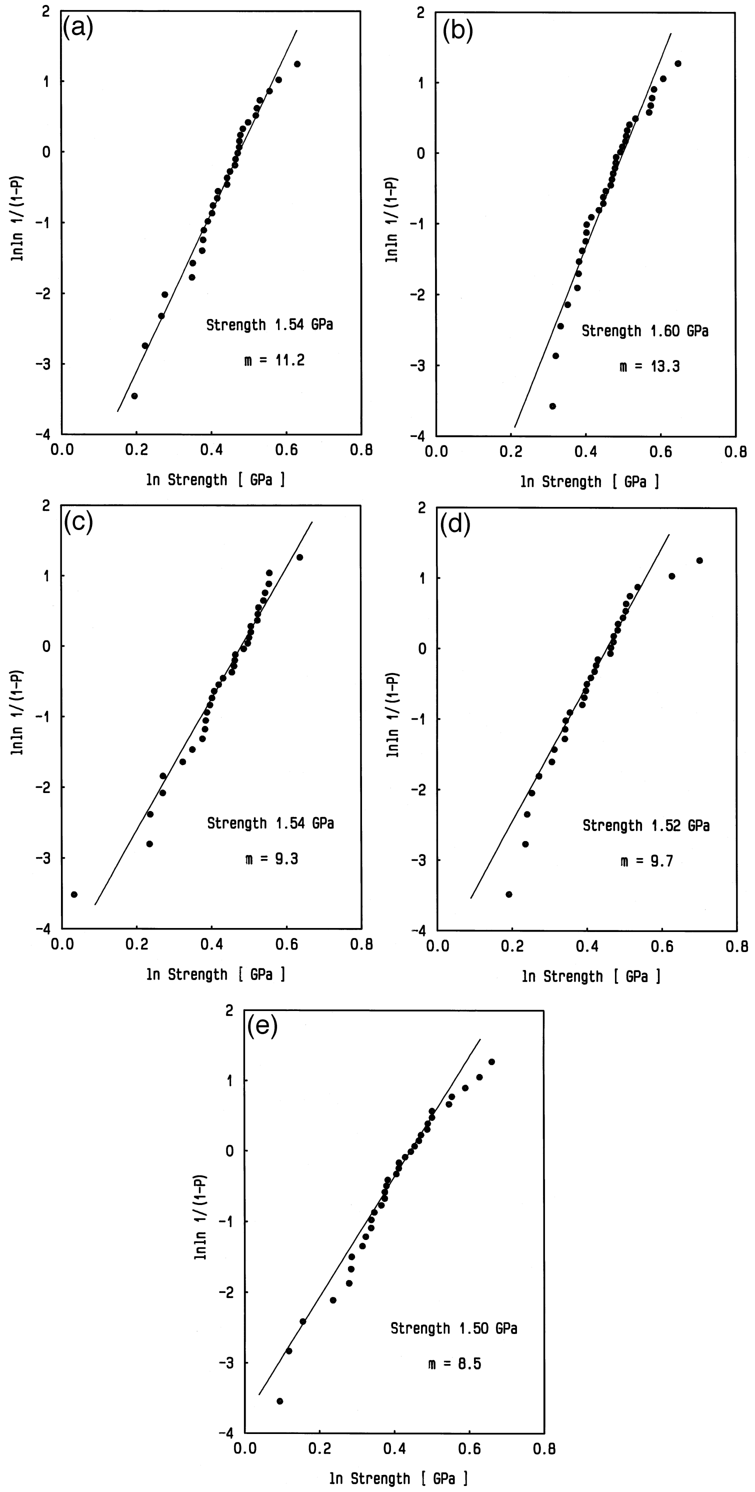

Fig. 3. Weibull coefficient of tensile strength at each level of neutron irradiation, (a) before irradiation; (b) $\Phi = 0.45 \times 10^{23}$ n/m^2; (c) $\Phi = 1.0 \times 10^{23}$ n/m^2; (d) $\Phi = 3.1 \times 10^{23}$ n/m^2; (e) $\Phi = 5.6 \times 10^{23}$ n/m^2.

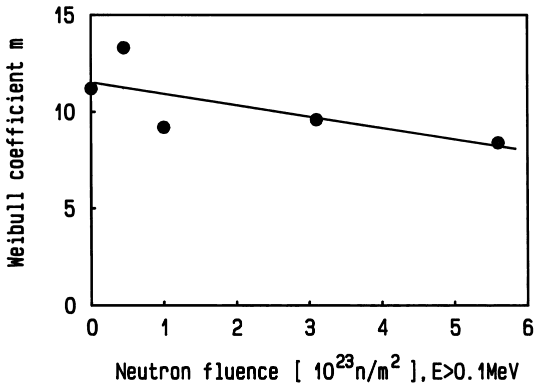

Fig. 4. Neutron irradiation and Weibull coefficient.

Fig. 5(b) is a photograph of a fracture surface after neutron irradiation of 5.6×10^{23} n/m^2. This figure shows nearly an equal number of alumina grains for the grain boundary fracture and transgranular fracture. Unclear contrast in the photograph appears due to the lack of gold evaporation for observation. The pores are smaller than those seen in Fig. 5(a) due to differences between specimens, and larger pores are observed even after neutron irradiation. This suggests that neutron irradiation has no affect at all on the SEM microstructure.

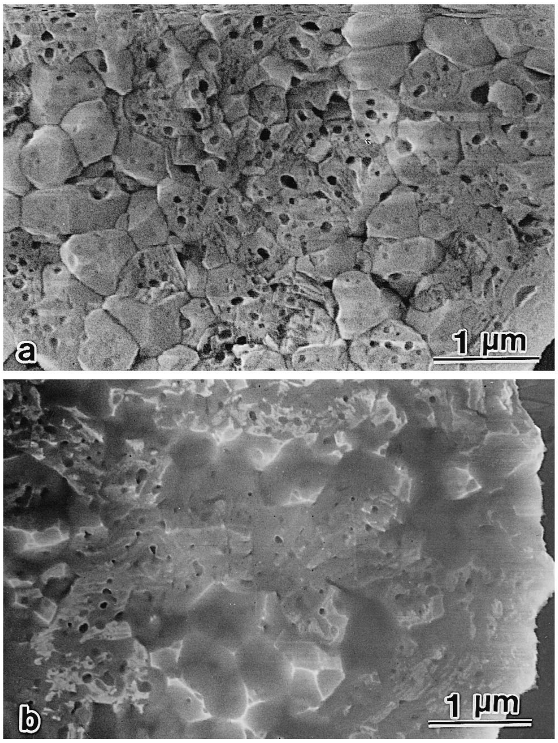

Fig. 5. Scanning electron microscope photograph of alumina fiber cross-section before (a) and after (b) irradiation.

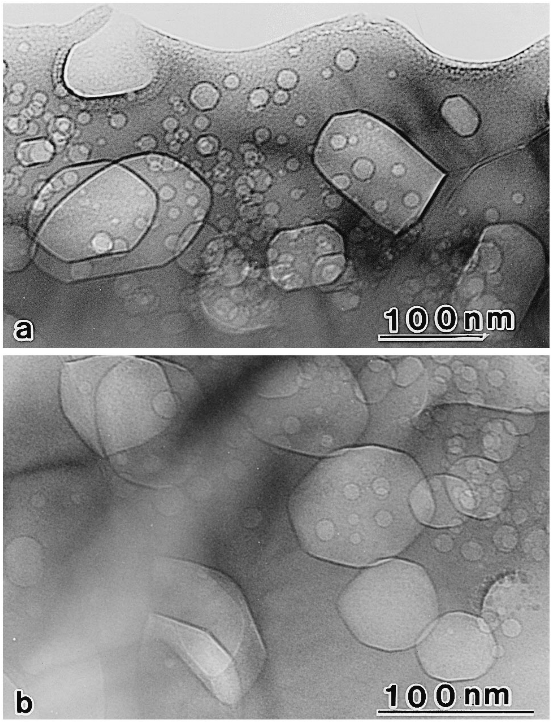

Fig. 6. Transmission electron microscope photograph of alumina fiber cross-section before (a) and after (b) irradiation.

3.3. *Observation by a transmission electron microscope*

Fig. 6(a) is a TEM photograph of an alumina fiber ion-polished before neutron irradiation. The photograph shows one alumina grain in which several large pores of 50 to 100 nm in diameter and many small pores of 10 nm or less are observed. It has been observed that smaller pores sometimes overlapped the large pores as they are located at different depths. A pore seen in the left of the figure is an example of a dent which has moved to the end of the specimen due to ion-polishing. Such cavities were also observed in Nextel 610 specimens made by 3 M Co.

Fig. 6(b) is a TEM photograph after neutron irradiation of 5.6×10^{23} n/m^2. The size and shape of the pores do not change before or after irradiation, and no defects due to irradiation are observed.

4. Conclusion

The alpha-alumina fiber Almax showed the following behavior with a neutron fluence of 5.6×10^{23} n/m^2 ($E >$ 0.1 MeV) or less:

(1) Strength and tensile modulus scarcely decreased as the irradiation proceeded.

(2) An increase in pores and swelling were not observed by SEM and TEM observations.

(3) The Weibull distribution spread with an increase in irradiation dosage suggesting an increase in defects which were not observed by SEM and TEM observations.

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