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Journal of Nuclear Materials 318 (2003) 307–312

journal of nuclear materials

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2.5 MeV electron irradiation effect of alumina ceramics

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Abstract

In order to choose an alumina ceramic material for use as a vacuum beam duct in a rapid cycling synchrotron, several kinds of alumina ceramics, having different microstructures, were examined under radiation fields of 2.5 MeV electrons. Since a long ceramic duct can only be manufactured by glazing duct segments, the mechanical strength and deterioration not only of the ceramics but also in the glazing joint were measured after irradiation. These ceramics have a sufficiently high flexural strength of more than 300 MPa before electron beam irradiation, and the experimental results showed no deterioration of the flexural strength after 1000 MGy electron beam irradiation. Also no noticeable changes could be seen in the measured tensile strength of Ti-ceramic brazed samples after 1000 MGy electron beam irradiation.

1. Introduction

Japan Atomic Energy Research Institute (JAERI) and High Energy Accelerator Research Organization (KEK) have been working on a joint project for a high intensity proton accelerator. The accelerator complex consists of a 400 MeV linac, a 3 GeV rapid cycle synchrotron, and a 50 GeV synchrotron ring [1].

The 3 GeV synchrotron ring accelerates a 400 MeV proton beam from a proton linac up to 3 GeV and then supplies it to the 50 GeV synchrotron ring. The 3 GeV synchrotron ring also supplies a neutron production target with a 3 GeV proton beam in order to produce a high intensity neutron beam. For this purpose, the 3 GeV synchrotron ring aims to generate a high power beam of 1 MW by setting 25 Hz as the repetition rate of acceleration and 8.3×10^{13} as the proton number per acceleration. In the rapid cycling magnetic field of the 3 GeV synchrotron, an eddy current effect in a metal duct produces an unacceptable perturbation of the magnetic field and unnecessarily large ohmic losses.

Since a specific alumina ceramics material has a low outgassing rate and is durable under heat treatment, it is widely used in accelerator vacuum systems as electrical insulator, such as rf windows and high-voltage seals [2]. Further, a beam duct made of alumina ceramics is often used in a rapid cycling magnetic field in order to avoid generation of eddy current [3]. The alumina ceramics duct to be installed in the ring should meet the following requirements [4]: (1) 3.5 m long duct (elliptical inner cross-section of 187 × 240 mm), bending by 15 degrees, within the dipole magnets (210 mm of gap distance), (2) 1.2 m long duct (264 mm in inner diameter) within the quadrupole magnets (290 mm of bore diameter), (3) mechanical strength not only in the base ceramics material, but also in brazed joint to a titanium flange, (4) low outgassing rate for ultra high vacuum, (5) equipped with rf shield so as to realize a low impedance, and (6) coated for suppressing secondary electron emission in order to avoid becoming charged or gather an electron cloud.

In an accelerated high current proton beam, a few protons will not remain in appropriate trajectories, but will hit a halo-collection device or a duct wall. These will generate secondary radiation such as neutrons as well as beta and gamma rays over a wide range of the energy spectrum. The cumulative energy dose due to radiation is approximately estimated to be on the order of 10^8 Gy for 30 years of operation [5]. The vacuum system should

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doi:10.1016/S0022-3115(03)00018-7

be constructed using components with high reliability and a long lifetime so as to minimize maintenance. Upon choosing alumina ceramics materials, the deterioration concerning electrical and mechanical properties must be taken into account when the alumina duct is installed in an accelerator with a higher level of radiation.

In this work, the radiation effects due to 2.5 MeV electron irradiation on the electrical/electronic and mechanical properties were measured for several kinds of alumina ceramics of different purity and grain structure.

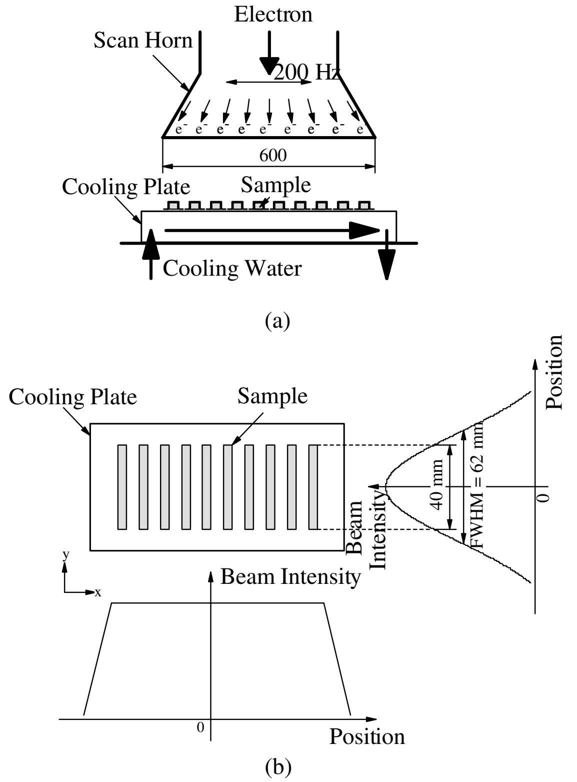
2. Experimental apparatus

2.1. Electron irradiation system

The electron beam irradiation facility, which has been operated at Takasaki Radiation Chemistry Research Establishment of JAERI, was used for this irradiation experiment. An electron beam is supplied to sample over a range of 600 mm by a repetition frequency of 200 Hz. The schematic view of the electron irradiation experiment and spatial distribution of electron beam intensity is shown in Fig. 1. The electron beam passes through a thin window of a scanning horn. The samples wrapped one by one with thin aluminum foil are fixed on the cooling plate with high heat conductivity paste to improve cooling efficiency. The beam intensity distribution is almost uniform across the 600 mm along the beam scanning direction (x) and has a Gaussian distribution along the y-axis. The full width half maximum of the electron beam intensity for the y-direction is 62 mm and is centered on the samples.

2.2. Examined sample

In order to choose an alumina ceramics material for use as a vacuum beam duct, radiation effects on the mechanical properties were measured for thirteen kinds of alumina ceramics having difference microstructures and purity. The characteristics of examined alumina ceramics are summarized in Table 1. Flexural strength of alumina ceramics and tensile strength of Ti-ceramics brazed sample were measured. Schematic views of



(a)

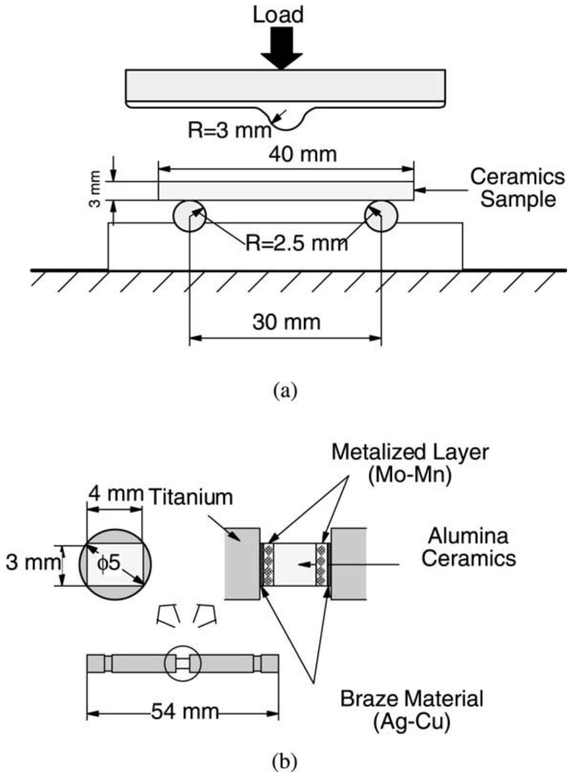
(b)

Fig. 1. (a) Schematic view of electron irradiation experiment and (b) electron beam distribution.

Table 1
Characteristics of examined alumina ceramics

	Purity (%)	Specific gravity	Grain size (μm)	ε @ 3.4 GHz	$\tan\delta(\times10^{-5})$ @ 3.4 GHz
A473	92	3.6	4–8	9.15	22
HA92	92	3.6	3–4	9.10	21
KP95	95	3.7	3–4	9.25	20
H538	95	3.7	5–8	9.21	25
H555	95	3.6	5–7	9.23	25
H525	96	3.6	4–5	9.24	18
UHA99	99	3.9	2–4	9.81	9
KP990	99.5	3.9	2–4	9.66	8
A479SS	99.5	3.8	4–8	9.73	<4
SSA-S	99.6	3.92	8–10	10.21	<4
HA997	99.7	3.95	10–16	9.95	4
H580	99.8	3.9	5–16	9.71	15
KP999	99.9	3.9	10–12	9.7[a]	30[a]

[a] @ 10 GHz.



(a)

(b)

Fig. 2. Schematic view of the mechanical strength measurement. (a) 3-point bend test and the shape of sample. (b) The shape of Ti-ceramics brazed sample to measure the tensile strength.

samples and mechanical testing techniques are shown in Fig. 2. A ceramic bar of 3 (thickness) × 4 (width) × 40 (length) mm in dimension was used as a test sample for flexural strength measurement. A metalized ceramics brazed with titanium rod was used as test sample for tensile strength measurement. The brazed area was about 10 mm^2.

3. Results and discussion

Energetic electrons are able to produce displacements through the Coulomb potential by direct interaction with the nuclei of the solid [6]. In order to calculate the number of atoms displaced by electron bombardment, we estimated the scattering cross-section of the nucleus for incident electrons. Since the threshold energies to produce displacement of aluminum (Al) and oxygen (O) were reported on 18 and 76 eV, respectively [7], we estimated at 52 eV for an average threshold energy (E_d) of alumina (Al_2O_3). In the case of $E_\mathrm{d} = 52$ eV, the displacement cross-section (σ_d) becomes 6.1×10^{-24} cm^2 (6.1 barn). As the incident electron energy is such that each collision transferring energy in excess of E_d produces only one displacement, the rate of atomic displacement is $\mathrm{d}n/\mathrm{d}t = \sigma_\mathrm{d}\phi_\mathrm{e}$ where ϕ_e is the incident electron flex density per unit area. Also since an electron beam of 3 mA peak current was irradiated to the alumina ceramics continuously on the area 60 cm × 6.2 cm, the displacement per atom per second (dpa/s) was estimated at 3×10^{-10} dpa/s. Since the electron energy is in excess of the minimum electron energy to produce displacements, the range of electrons in alumina ceramics is estimated at 3 mm using the empirical Katz–Penfold relations for 2.5 MeV electrons [8]. Therefore, a fraction of 1.7×10^{-8} atoms are displaced at a range of 3 mm from the surface of alumina ceramics due to the 1 MGy electron irradiation.

3.1. Flexural strength

The flexural strength of unirradiated alumina ceramics is usually 200–400 MPa, depending on the microstructure, such as the grain size, void distribution and sintering additive materials. Under a high radiation field, not only electron excitation (or further ionization) but also vacancy creation, take place. The latter would directly cause the production of dislocations, which grow to become voids (swelling) in alumina. The former is considered to enhance the mobility of dislocations.

A ceramics bar of 3 (thickness; t) × 4 (width; w) × 40 (length) mm in dimensions was used as a test sample, and was subject to the 2.5 MeV electron beam; the absorption dose rate was 16.92 kGy/s. Using the 3-point bend test, the flexural strength was measured; the distance L between two supports was 30 mm, From the loading force value (P) at break, one can derive the tensile strength by $\sigma = (3/2) \times (PLw^{-1}t^{-2})$ as a flexural strength. Changes in the strength, as the absorption dose is increased up to 1000 MGy, were observed for thirteen kinds of alumina ceramics for practical use (Table 1).

The absorption dose dependence of the flexural strength of alumina ceramics is shown in Fig. 3. The horizontal axis is absorption dose or dpa and the vertical is the flexural strength of the electron irradiated alumina ceramics. The examined ceramics are classified in three groups according to the alumina contained. The ceramics having a large content of sintering additives (92% alumina contained ceramics) are classified in group-1, the ceramics of about 95% alumina content are classified in group-2, and the ceramics having a high purity (more than 99% alumina contained ceramics) are classified in group-3, respectively. As shown in Fig. 3, the strength is not so greatly changed, as far as the absorption dose given here is concerned. However it seems that the reinforcement of the flexural strength is shown for almost all examined ceramics around 700 MGy absorption dose. The changes in the flexural strength observed here cannot always be explained by dislocation growth, as in case of a single crystal. The reinforcement shown in

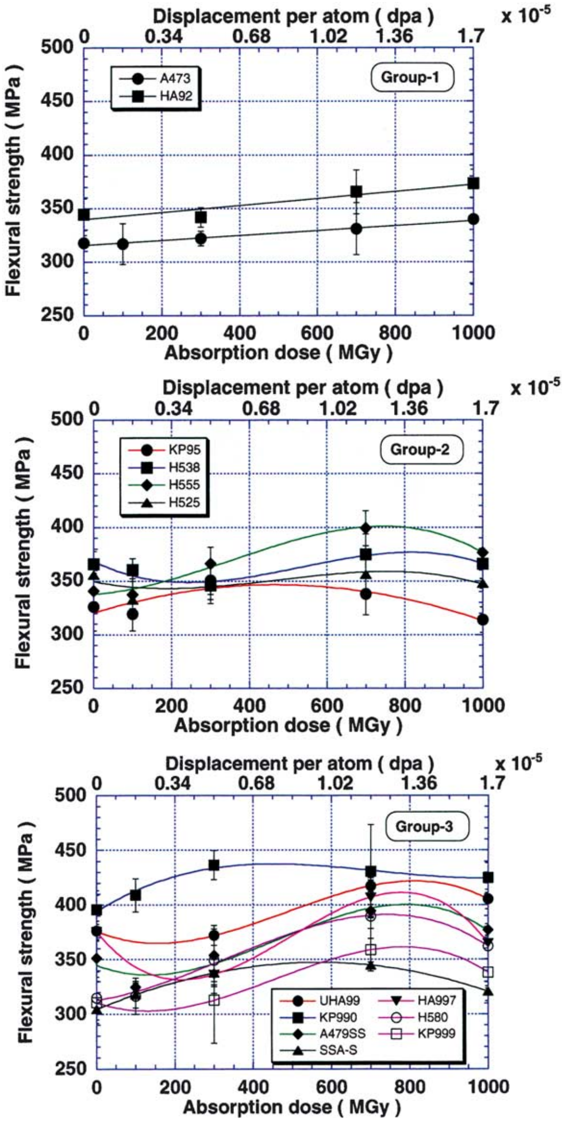


Fig. 3. Flexural strength of electron irradiated alumina ceramics measured by 3-point bend test. 2.5 MeV electron beam are irradiated to the ceramics for a maximum dose of 1000 MGy. The dose of 1000 MGy is correspond to 1.7×10^{-5} dpa. Group-1: 92% alumina contained ceramics, group-2: about 95% alumina contained ceramics, group-3: more than 99% alumina contained ceramics.

group-3, especially SSA-S, H580, and KP-990, is possibly due to electron transfer among trapping levels in grains and boundaries. These ceramics, having been molded by a spray dry method, and then sintered almost without additives, may have defects and thus have residual stresses in crystalline interfaces. The energy dose by electron irradiation seems to have reduced the stresses by redistributing the trapped electrons.

3.2. Tensile strength

The duct segment of alumina ceramics is to be jointed so as to form a 3.5 m long beam duct, as described before. A brazing method is planned to apply not only to the segment joint, but also to a joint between a titanium flange and a duct. In order to know the radiation effect on the interface, a tensile strength measurement of Ti-ceramics brazed samples (brazed area of 10 mm^2) was performed before and after the irradiation of a 2.5 MeV electron beam.

Fig. 4 shows the tensile strength of electron irradiated Ti-ceramic brazed samples. The horizontal axis is absorption dose or dpa and the vertical is the flexural strength of the electron irradiated alumina ceramics. The examined ceramics are classified in three groups according to their content of alumina. The ceramics having a large content of sintering additives (92% alumina contained ceramics) are classified in group-1, the ceramics with about 95% alumina are classified in group-2, and those having a high purity (more than 99% alumina contained ceramics) are classified in group-3, respectively. No noticeable change could be seen in the measured tensile strength after irradiation with a cumulative adsorption dose of 1000 MGy. The values of the tensile strength for brazed joints scatter around 50–80 MPa, which is lower than that of bulk alumina ceramics. Fig. 5 shows the picture of the ruptured side of Ti-ceramics brazed sample after tensile strength measurement. As shown in Fig. 5, a crack upon breaking, observed for both unirradiated and irradiated samples developed in neither metallized layer (Mo–Mn component) nor brazed material (Ag–Cu), but occurred in alumina ceramics adjacent to the joint. It is considered that the metallizing process followed by brazing introduces a residual stress, and that the stress, dominated by the mechanical strength, is not affected by radiation with a dose of 1000 MGy.

3.3. Another approach

Alumina ceramics used as a high-voltage insulator in vacuum very often becomes yellowish. As for an rf window, multipactoring electrons, having an incident energy of several keV, are considered to change the electronic states in the ceramics due to bremsstrahlung, causing coloring even inside the alumina bulk (3 mm thick); the effective range of the incident electron is less than 1 μm.

Fig. 6 shows the optical-absorption (or transmittance) coefficients measured for some alumina ceramics before and after X-ray irradiation; the samples were set on a beam collector of the klystron being operated with a beam voltage of 300 kV. The desorption coefficient of a ceramics (HA-997, 99.7% of purity), having been specially sintered in order to crystallize sintering addi-

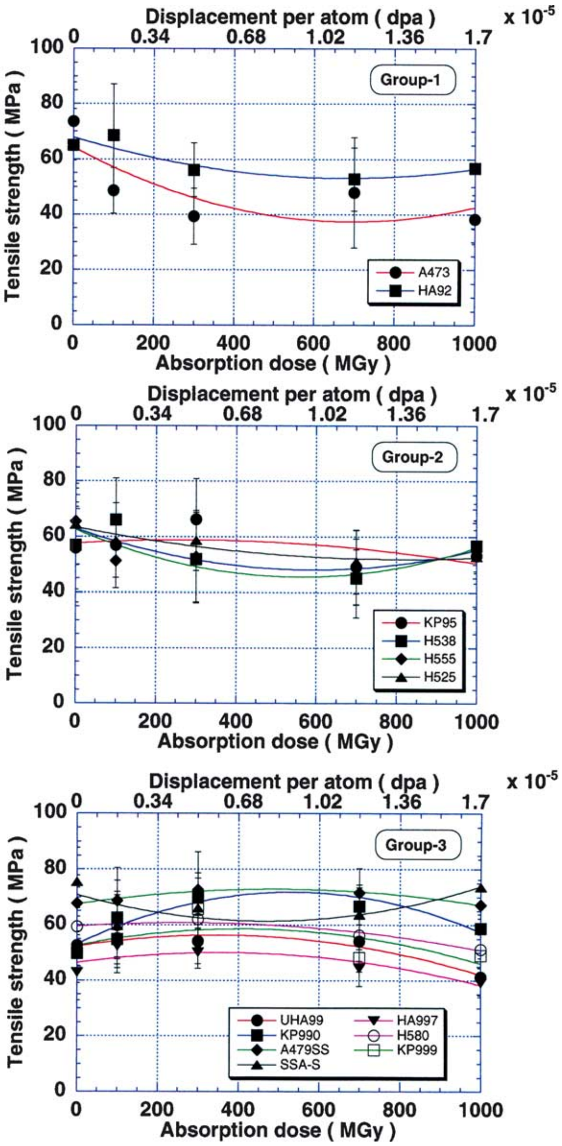
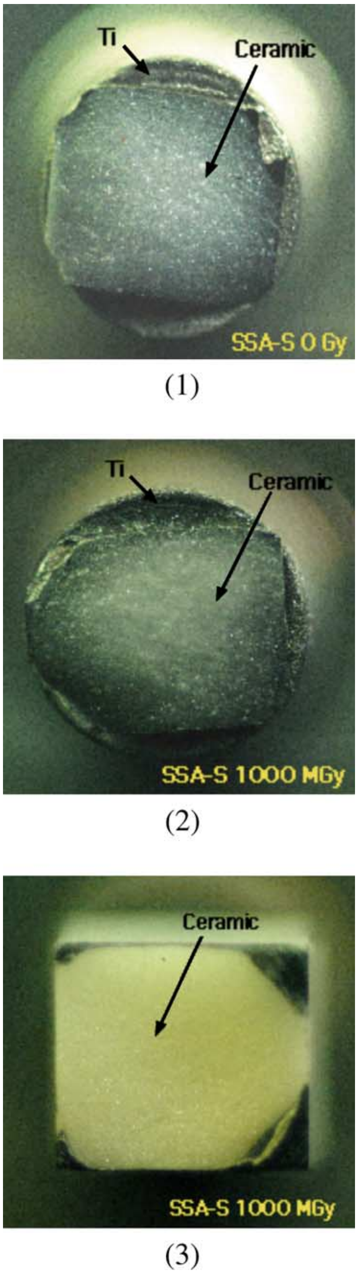


Fig. 4. Tensile strength of electron irradiated alumina ceramics which have been connected with titanium. 2.5 MeV electron beam are irradiated to the ceramics for a maximum dose of 1000 MGy. The dose of 1000 MGy is correspond to 1.7×10^{-5} dpa. Group-1: 92% alumina contained ceramics, group-2: about 95% alumina contained ceramics, group-3: more than 99% alumina contained ceramics.

tives of SiO_2 and CaO, is changed the least compared to another ceramics (KP-990, 99.0% of purity), after irradiation. From cathodoluminescence (CL) measurements, observing the F^+-and F-centers of oxygen vacancies in alumina ceramics, the X-ray irradiation does not create a new defect, which contributes to alumina luminescence.



(1)

(2)

(3)

Fig. 5. Picture of the rupture side of Ti-ceramic brazed sample after tensile test. The ceramic sample is SSA-S: (1) without irradiated sample, (2) after 1000 MGy electron irradiated sample: side-a, (3) after 1000 MGy electron irradiated sample: opposite side of side-a.

Coloring in alumina ceramics is, therefore, mainly caused by a change in the electronic states (excitation) in sintering additives of amorphous structure. The annealing process (1500 °C, in air) after the irradiation recovers the optical properties, indicating relaxation of the excited electrons by thermal activation [9].

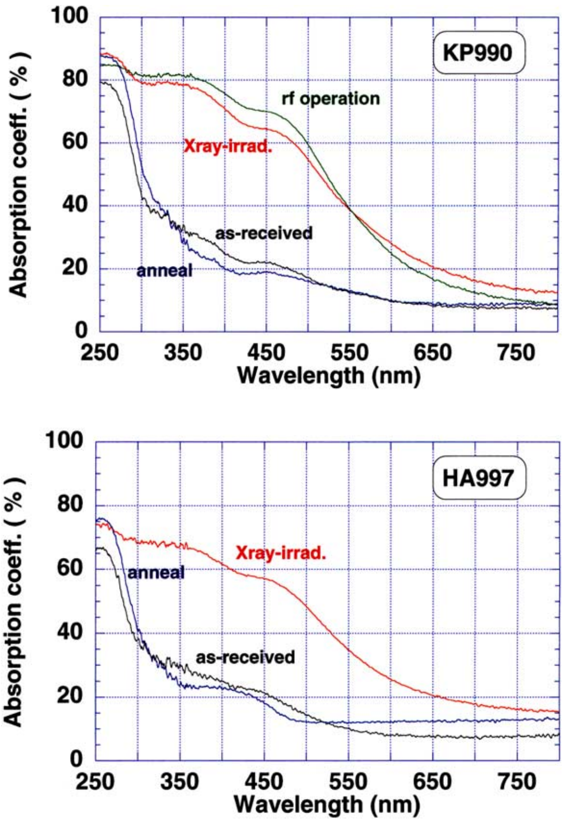


Fig. 6. Optical absorption coefficient observed for alumina ceramics irradiated by X-rays. Annealing was performed at 1500 °C in air. Effects of multipactoring during rf operation is indicated in the KP990 ceramics.

4. Conclusion

Radiation effects on the electrical/electronic and mechanical properties were measured for several kinds of alumina ceramics. From the results, some of the properties, after irradiation, are found to be affected not only by purity, but also by microstructures of the ceramics. Changes of flexural strength after electron irradiation depend on the sintering process.

Basically, irradiation excites vacancy trapped electrons as well as valence band electrons in alumina ceramics; the latter also deform the crystal lattice and produce vacancies. The excited electrons may redistribute in pre-excitation and radiation induced trap sites. The above-described results, therefore, indicate that the trap sites are concentrated adjacent to the crystalline interface (grain boundary), the property of which strongly depends on the sintering process.

As far as the irradiation dose given here is concerned, detrimental degradation by irradiation has not been observed for the examined alumina ceramics. However, since the irradiation has been performed in the atmosphere so far, an investigation is necessary with irradiation in vacuum to find any effects by oxygen and hydrogen during irradiation in order to choose an alumina ceramics for use as a vacuum beam duct in a high current proton synchrotron accelerator.

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