ELSEVIER

Journal of Nuclear Materials 258–263 (1998) 1867–1872

journal of nuclear materials

Electrical properties of ceramics during reactor irradiation

T. Shikama [a], S.J. Zinkle [b,*], K. Shiiyama [c], L.L. Snead [b], E.H. Farnum [d]

[a] Oarai Branch, Institute for Materials Research, Tohoku University, Ibarakiken 311-13, Japan
[b] Oak Ridge National Laboratory, Metals and Ceramics Division, P.O. Box 2008, Oak Ridge, TN 37831-6376, USA
[c] Department of Nuclear Engineering, Kyushu University 36, Fukuoka 812, Japan
[d] Los Alamos National Laboratory, P.O. Box 1663, Los Alamos, NM 87545, USA

Abstract

Twelve different types of polycrystal and single crystal Al_2O_3 (alumina and sapphire) specimens of varying grades of purity were irradiated for three reactor cycles in a removable beryllium position in a High Flux Isotopes Reactor (HFIR) at Oak Ridge National Laboratory at a temperature of 720–760 K up to a maximum dose of 3 dpa while a dc electric field of 200 V/mm was applied. The recently completed Temperature Regulated In Situ Test (TRIST) facility in the HFIR was used to perform in situ measurements of electrical conductivity. In addition, three Al_2O_3 specimens were simultaneously irradiated without a continuously applied dc electric field. In situ electrical conductivity measurements were performed on the specimens before, during and following each irradiation cycle. Behavior of electrical conduction in Al_2O_3 was studied, with special emphasis on detection of any long-term increase of the electrical conductivity.

1. Introduction

More and more sophisticated irradiation studies are being demanded, as research and development of fusion reactor materials are advancing. There, studies of irradiation effects in high flux fission reactors are appreciated well [1,2]. A typical example is in situ experiments for studying dynamic irradiation effects in high flux fission reactors [2,3]. Functional properties of ceramic materials will be affected dynamically by irradiation, which can be studied only by in situ type measurements. A Japan/USA collaboration on fusion reactor materials known as the JUPITER (Japan/USA Project on Irradiation TEst of fusion Research) project was established in 1995 after an extensive preparation for a few years. It was initiated to mobilize research activities of both parties, namely those in organizations under Japanese Ministry of Education and those in organizations under US Department of Energy. The project will utilize expensive instrumented-irradiation facilities in high flux fission reactors, cooperatively and efficiently.

Dynamic irradiation effects on electrical properties of ceramic insulators have been attracting strong concerns, in conjunction with use of ceramic insulators in important instruments near the burning plasma. The JUPITER project approved an in situ study of electrical conductivity of ceramic insulators as one of its major research topics. The experiment used the recently completed Temperature Regulated In Situ Test (TRIST) controlled irradiation facility in the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory. And the experiment was denoted as HFIR–TRIST–ER (Electrical Resistivity). Reliable in situ experiments in high flux reactors have many technical difficulties to overcome. Extensive efforts for improving then-existing techniques have been done collaboratively [4–6]. Preliminary irradiation tests were done in the High Flux Beam Reactor (HFBR) in Brookhaven National Laboratory [4] and in the Japan Materials Testing Reactor (JMTR) in Japan Atomic Energy Research Institute [5].

The TRIST–ER irradiation study was carried out in the HFIR from March, to June in 1996. The study yielded quantitative data concerning so-called radiation induced conductivity (RIC) of several different aluminas (Al_2O_3) and sapphires (single crystal Al_2O_3). The study also looked for evidence of a long term (permanent)

* Corresponding author. Tel.: +1 423 576 7220; fax: +1 423 574 0641; e-mail: zinklesj@ornl.gov.

0022-3115/98/$19.00
PII: S0022-3115(98)00300-6

increase of electrical conductivity under irradiation, which is so-called radiation induced electrical degradation (RIED) originally reported by Hodgson [7] under electron irradiation.

Detailed analysis of the obtained data did not reveal catastrophic increase of electrical conductivity up to 3 displacement per atom (dpa) in single crystal alumina specimens, which were reported to be most susceptible to the RIED under electron irradiation [8]. Details of the irradiation techniques [6,9] and irradiation history [10] for the TRIST–ER experiment were already reported and the primary results either have or will be reported elsewhere [9,11,12]. The present paper gives general overview of the Japan/USA collaborative joint venture of the TRIST–ER1 irradiation experiment.

2. Experimental procedures

Twelve different Al_2O_3 specimens of varying grades of purity shown in Table 1 were irradiated for 3 reactor cycles in a HFIR removable beryllium position at a temperature of 720–760 K up to a maximum dose of 3 dpa while a dc electric field of 200 V/mm was applied. In addition, three Al_2O_3 specimens were simultaneously irradiated without a continuously applied dc electric field. The irradiation temperature and the electric field were chosen to be in the range where previous irradiation experiments on Al_2O_3 have reported pronounced RIED following low-dose ($\ll$0.1 dpa) irradiation. In situ electrical conductivity measurements were performed on the specimens before, during and following each irradiation cycle. Polycrystal and single crystal aluminas are leading candidates as an electrical insulator in a fusion reactor, as they have the most extensive database and well-established industrial experiences. Their electrical conduction is thought to be understood relatively well, namely, electron-conduction dominant at concerned temperatures [13]. Accumulation of results of in situ measurements, however, are inferring that dynamic electrical conductivity of Al_2O_3 would depend on some of material properties which might not be controlled well in conventional manufacturing processes, such as impurities and grain size. Thus, it is important to study in situ electrical conductivity of variety of Al_2O_3 in the same irradiation conditions. Fig. 1 shows the structure of subcapsule used in the present study [6].

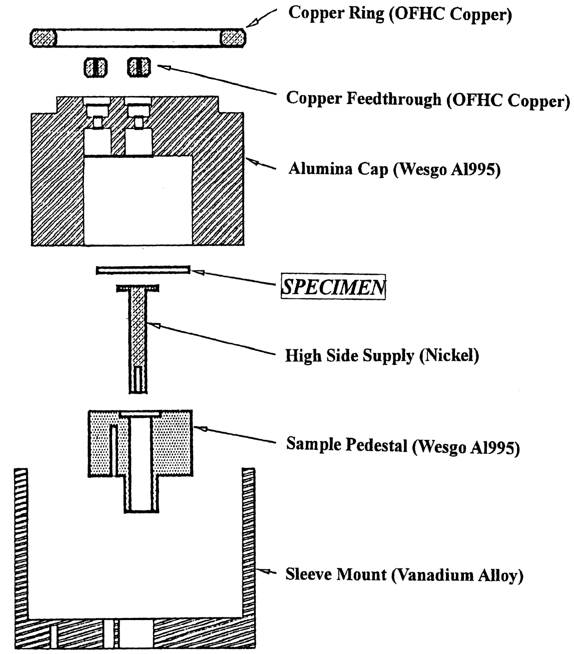

Fig. 1. Cross sectional view of subcapsule for in situ electrical measurement.

Table 1
Specimen list for the HFIR TRIST–ER1 in situ electrical conductivity capsule

HFIR position	Material	Appl. Voltage	Vendor and grade
1	Al_2O_3, single crystal	150 V	Crystal systems (Hemex UV grade) *a*-axis
2	Al_2O_3, single crystal	150 V	Crystal systems (Hemex UV grade) *c*-axis
3	Al_2O_3, single crystal	150 V	Crystal systems (Hemex regular) *c*-axis
4	Al_2O_3, single crystal	150 V	Crystal systems (Hemex regular) *a*-axis
5	Al_2O_3, polycrystalline	150 V	Vitox (99.9% purity, Morgan Matroc, Anderman Div.)
6	Al_2O_3, polycrystalline	150 V	Kyocera A-480 (99.9% purity)
7	Al_2O_3, polycrystalline	150 V	Wesgo AL300 (97.0% purity)
8	Al_2O_3, polycrystalline	150 V	Kyocera A-479 (99.0% purity)
9	Al_2O_3, polycrystalline	150 V	Coors AD998 (99.8% purity)
10	Al_2O_3, polycrystalline	150 V	Wesgo AL995 (99.5% purity)
11	Al_2O_3, polycrystalline	0 V	Wesgo AL995 (99.5% purity)
12	Al_2O_3, single crystal	0 V	Crystal systems (Hemex regular) *c*-axis
13	Al_2O_3+Cr, single crystal	150 V	Union carbide (UV grade), 60° from *c*-axis
14	Al_2O_3, single crystal	150 V	Kyocera SA 100 ($1\ \bar{1}\ 0\ 2$ orientation)
15	Al_2O_3, single crystal	0 V	Kyocera SA 100 ($1\ \bar{1}\ 0\ 2$ orientation)

The subcapsule was filled with helium to provide an inert environment for the irradiation. Although a good-vacuum environment is preferable for electrical measurements, it is impossible to maintain a suitable low pressure in subcapsules in the HFIR reactor due to material outgassing effects and the long distance from the reactor core to experimentally-accessible regions where vacuum pumps could be located. Also, effective heat-removal from irradiated specimens is essential for the temperature control, under a high rate-gamma-heating irradiation. The present subcapsule was designed to have good heat-removal paths from the specimen to the outer wall of the irradiation capsule which was effectively cooled by flowing water [6,10]. The helium environment would help to minimize unexpected hot spots caused by localized gamma-heating and by unnoticed thermal barriers. The low voltage side electrode and its electrical lead, which are the measuring side of electrical current for bulk conduction, utilized a standard guarded configuration consisting of a center electrode, guard ring electrode and a triaxial mineral insulated cable.

The subcapsule was designed to minimize unexpected electrical paths between high and low voltage side electrodes. However, realization of the ideal guarded-configuration is nearly impossible in a fission reactor irradiation. A heavily ionized helium gas environment will make unexpected electrical paths. Energetic photo-electrons generated by intense high energy gamma-rays will transport and redistribute electrical charges among materials composing of the subcapsule and a capsule accommodating the subcapsules. To analyze effects of such unexpected electrical leakage, extensive measuring program was developed using National Instrument Labview III program. Electrical currents among high and low voltage side electrodes and a guard ring electrode were measured as a function of applied voltage in the course of irradiation.

3. Experimental results and discussions

The following is a summary of results obtained in the HFIR–TRIST–ER experiment.

The phenomenon of RIED was not confirmed up to 3 dpa in the helium environment in the present irradiation conditions as shown in Fig. 2. However, most of the measurements on the 15 specimens were prematurely terminated before the end of irradiation and measurements for the full 3 dpa irradiation dose were only obtained on 3 specimens. According to bench-top tests, the premature interruption of the measurements is thought to be due to a conventional electrical-breakdown of a glass seal of high side Mineral Insulated (MI) cables [9,11]. Analyses of electrical currents did not reveal any evidence for initiation of catastrophic increase of the

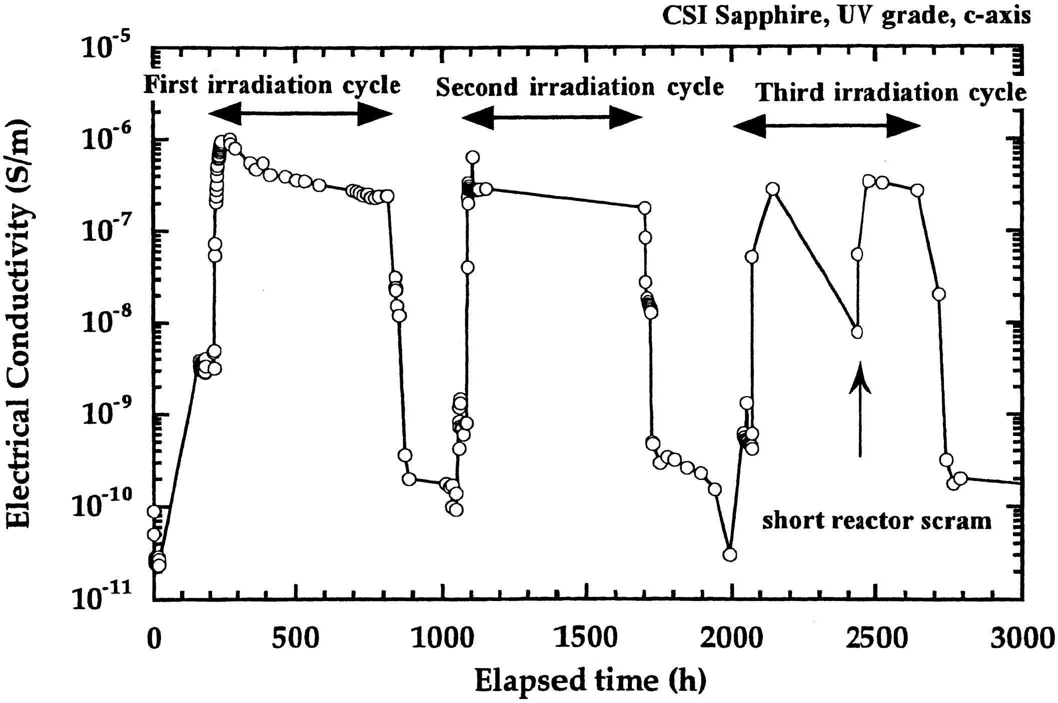

Fig. 2. Electrical conductivity of crystal system sapphire as a function of time.

electrical conductivity prior to the premature interruption of measurements. Some specimens showed increase of electrical conductivity in the initial stage of irradiation up to about 0.1 dpa [12], but it was generally very slight (less than a factor of 5 increase compared to the initial RIC value).

As described in a separate paper [12], ruby (chromium-doped sapphire) showed a substantial (about three orders of magnitude) increase of electrical conductivity during irradiation up to a dose of about 0.1 dpa. The conductivity of all specimens either remained constant or gradually decreased with increasing dose above about 0.1 dpa [9]. The voltage/current (V–I) relation was measured through the irradiation and the non-ohmic behavior of electrical conduction of aluminas and sapphires was confirmed up to 3 dpa as shown in Fig. 3 [14]. This non-ohmic behavior demonstrates that continuous-current-measurements with a fixed voltage (conventional measurements) would not be appropriate to evaluate change of electrical conductivity under irradiation. The newly developed measuring program made it possible to evaluate the V–I behavior through the irradiation. In the present analyses, the electrical conductivity was evaluated by differentiating a current-voltage curve (dI/dV) at negative applied voltages [5]. This procedure minimized effects of leakage current and other parasitic currents that might be independent of an applied voltage.

The non-ohmic behavior was observed in a previous JMTR experiment [5], where a prototype subcapsule was irradiated under the JUPITER program. There, the non-ohmic behavior was concluded to be caused by a leakage current through ionized gas (the gas effect). In the JMTR prototype subcapsule, there existed an electrical path through a gas environment from the high side to the low side. In the TRIST–ER experiment, efforts were exerted to minimize a possibility of current leakage from the high voltage side to the low voltage side. The observed non-ohmic behavior does not appear to be simply attributable to a gas effect in the HFIR–TRIST–ER subcapsules. The cause of the non-ohmic behavior is under analysis and effects such as the difference in work functions between the metal electrode and the alumina specimen appear to be partly responsible for the non-ohmic behavior [14].

A fundamental irradiation study under the Japan/USA collaboration was proposed for studying the non-ohmic behavior and also the physical mechanisms responsible for the observed large offset currents in the HFIR–TRIST–ER experiment (Fig. 3). A large electrical current was observed even at 0 V, when the electrical current was measured as a function of applied voltage from +100 V to −100 V with a voltage step of 20 V. This current at 0 V, denoted as an offset current, sometimes exceeded a few tens of micro-amperes at the beginning

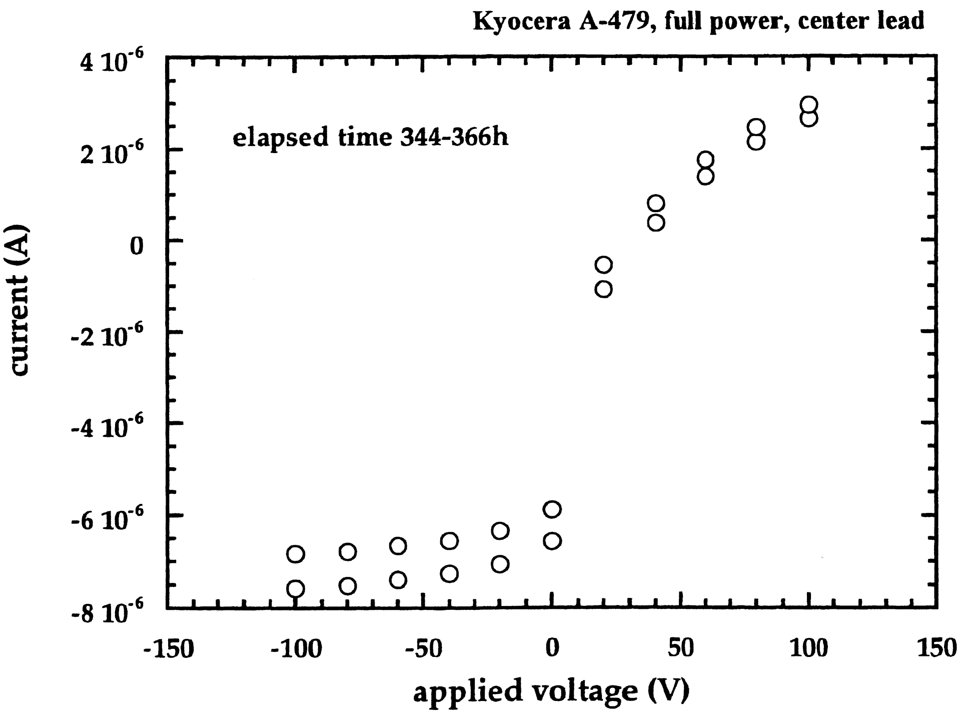

Fig. 3. Non-ohmic behavior of alumina of Kyocera A479. Measured current as a function of applied voltage.

of irradiation as shown in Fig. 4. A steady state offset current is in the range of several micron ampere and was observed for all of the TRIST–ER subcapsules. The offset current usually went from ground to the low side of a specimen and from the high side of a specimen to ground. Occasionally, a reverse offset current was observed. As described above, a new experiment was proposed to study cause of the offset current in the JMTR and will be carried out in 1998. This offset current phenomenon may have an important impact on the accuracy of magnetic probes (used in conjunction with current integrators) which are used to detect the position of the plasma. The plasma position must be accurately known in order to make adjustments in the tokamak magnetic field which allows long-term operation of the plasma. Also, there is some possibility that the offset current might reveal microstructural modification of alumina under irradiation. A charge separation might be the cause of the observed offset current. There is another possibility that the offset current is caused by a phenomenon so-called radiation induced electromotive force (RIEMF) [15].

The observed RIC was approximately linearly dependent on ionizing dose rate in the range of 10^2–10^4 Gy/s, though it showed some sublinear behavior above 10^3 Gy/s at the beginning of irradiation. The magnitude of the RIC depended on the grade of Al_2O_3 in the following way up to 10^3 Gy/s.

Normal grade polycrystal $\ll$ High grade polycrystal $\ll$ doped sapphire(Ruby) $\ll$ normal grade sapphire $\ll$ UV-grade sapphire.

At the ionizing dose rate of 10^4 Gy/s, the dependence of the RIC on the grade of Al_2O_3 became very weak. All of the examined alumina specimens showed an RIC value of about 10^{-6} S/m at 10^4 Gy/s.

4. Conclusions

Admitting that there were technical problems, which were especially represented by the premature interruption of the measurements on 12 specimens, RIED was not observed phenomenologically in the TRIST–ER experiment. The maximum dose rates of electronic excitation and atomic displacement were about 10^4 Gy/s and about 5×10^{-7} dpa/s, respectively. The maximum total doses were about 6×10^{10} Gy and 3 dpa, respectively. Above these doses, mechanical properties of alumina would degrade substantially [2] and their usable limit of total dose would be in the range of a few dpa based on degradation of thermomechanical properties.

The TRIST–ER supplied the engineering database of polycrystal and single crystal aluminas that are the primary candidate for the electrical insulators in a fusion reactor. Also, the TRIST–ER supplied ample data for studying fundamental aspects of electrical conduction

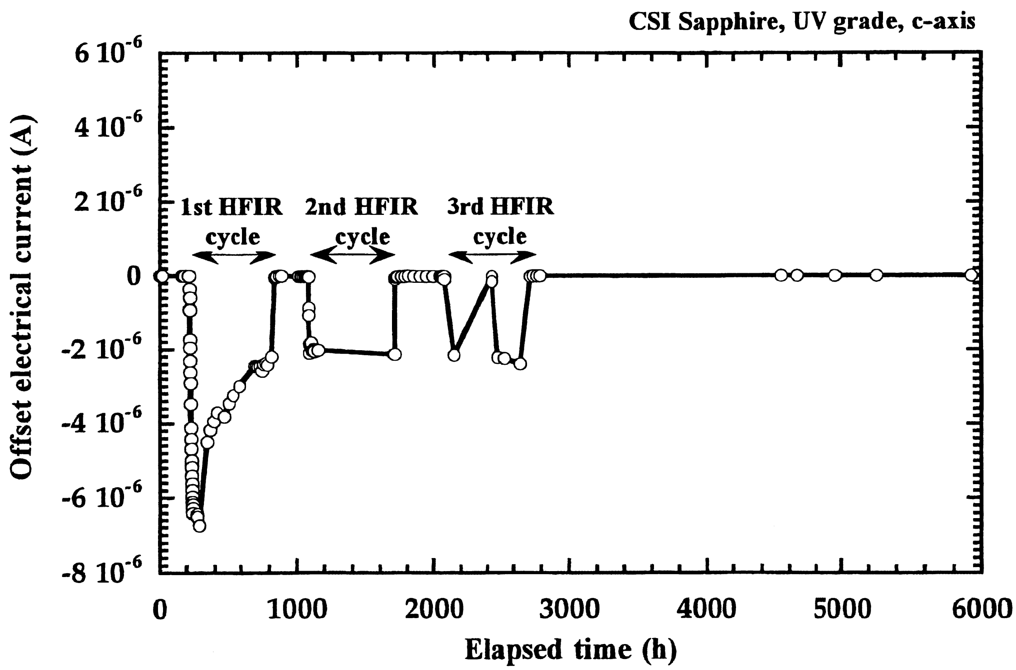

Fig. 4. Observed offset current as a function of time.

through different grades of aluminas under a "fusion relevant" irradiation condition. Here, the fusion relevant irradiation environment means irradiation environments that have an appropriate ratio of an electronic excitation rate and an atomic displacement rate.

The TRIST–ER has provided convincing data that RIED would not be a problem for fusion reactor development, as long as effects of irradiation atmosphere are not contributing to the irradiation behavior such as preferential removal of oxygen from surface of alumina, "surface RIED", etc. It is concluded that the TRIST–ER completed its primary mission, namely studying electrical conduction behavior of ceramic insulators under a fusion relevant irradiation environment and clarifying whether or not RIED would be a problem. To confirm the above mentioned conclusion; that RIED did not occur in a helium environment up to 3 dpa, post irradiation examination of irradiated specimens will be needed. In particular, the temperature dependence of the electrical conductivity of irradiated specimens should be evaluated in a post irradiation examination.

There still exist several scientific and technical concerns about the electrical conduction behavior of ceramic insulators under irradiation. The TRIST–ER experiment revealed interesting aspects of electrical conduction of polycrystal and single crystal aluminas under irradiation, such as non-ohmic behavior and a large offset current. The large offset current would have a possibility to become a serious problem in a plasma diagnostics using alumina insulators. Also, there is some possibility that the offset current might reveal microstructural modification of alumina under irradiation.

Acknowledgements

Many participants from the USA and Japan contributed to successful execution of the TRIST–ER experiment. Authors wish to express their special thanks to Dr. F.W. Wiffen of USDOE, and Prof. K. Abe of Tohoku University, who are representatives of the JUPITER project. They also wish to give their hearty appreciation of invaluable contributions to Mr. W.S. Eatherly, D.W. Heatherly, M.T. Hurst, D.G. Raby, R.G. Sitterson, R.L. Wallace and Drs. A.L. Qualls, K.R. Thoms, E.E. Bloom in Oak Ridge National Laboratory, and to Mr. K. Scaborough in Los Alamos National Laboratory, and to Mr. M. Narui in Tohoku University, and Mr. T. Sagawa in JAERI, and to Prof. A. Kohyama in Kyoto University and C. Namba in NIFS. Authors regret sincerely that they can not mention all of collaborators who gave the authors precious help and guidance. This study has been carried out under the Japan/USA Program of Irradiation TEst for fusion Research (JUPITER) project. Japanese Ministry of Education supported the project through National Institute for Fusion Science (NIFS). Research was sponsored in part by the Office of Fusion Energy Sciences, US Department of Energy under contract DE-AC05-96OR22464 with Lockheed Martin Energy Research Corporation.

References

[1] S.J. Zinkle, Nucl. Instr. and Meth. B 91 (1994) 234.
[2] T. Shikama G.P. Pells, J. Nucl. Mater. 212–215 (1994) 80.
[3] T. Shikama, Mater. Trans. JIM 36 (1995) 997.
[4] L.L. Snead, D.P. White, S.J. Zinkle, J. Nucl. Mater. 226 (1995) 58.
[5] E.H. Farnum, T. Shikama, M. Narui, T. Sagawa, K. Scaborough, J. Nucl. Mater. 228 (1996) 117.
[6] W.S. Eatherly et al., in: Fusion Materials Semiannual Progress Report for the period ending 31 December, DOE/ER-0313/19, 1995, p. 241.
[7] E.R. Hodgson, J. Nucl. Mater. 191–194 (1992) 552.
[8] E.R. Hodgson, J. Nucl. Mater. 212–215 (1994) 1123.
[9] S.J. Zinkle et al., in: Fusion Materials Semiannual Progress Report for the period ending 30 June, DOE/ER-031320 1996, p. 257.
[10] A.L. Qualls et al., in: Fusion Materials Semiannual Progress Report for the period ending 30 June, DOE/ER-0313/20, 1995, p. 267.
[11] S.J. Zinkle, T. Shikama, presented at Joint workshop of IEA and JUPITER on radiation effects in ceramic insulators, held in Cincinnati, May 5–8, 1997; also S.J. Zinkle et al., in: Fusion Materials Semiannual Progress Report for the period ending June 30, DOE/ER-0313/22, 1997, p. 188.
[12] T. Shikama, S.J. Zinkle, these Proceedings.
[13] F. Agullo-Lopez, C.R.A. Catlow, P.D. Townsend, in Point Defects in Materials, Academic Press, London, 1988, p. 235.
[14] K. Shiiyama, et al., these Proceedings.
[15] T. Shikama, M. Narui, T. Sagawa, Nucl. Instr. and Meth. B 122 (1997) 650.