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Long term degradation of electrical insulation of Al_2O_3 under high flux fission reactor irradiation

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Abstract

Long term increase of electrical conductivity under irradiation was observed in some Al_2O_3 (polycrystal and single crystal alumina) specimens in in situ electrical measurements in the High Flux Isotope Reactor in the framework of the Japan/USA collaboration called JUPITER project. Especially, chromium doped single crystal alumina showed substantial increase of electrical conductivity at moderate doses of ~0.1 dpa, which had similarity to the phenomenon called radiation induced electrical degradation (RIED). Irradiation to doses above 0.1 dpa did not cause a further degradation of electrical conductivity, but instead resulted in an improvement in insulating ability. Behavior of the observed increase of electrical conductivity is described in conjunction with the reported phenomena in RIED and some mechanisms responsible for the observed increase are speculated from the results obtained.

1. Introduction

Degradation of electrical insulating ability of ceramic insulators is one of the major concerns for functional fusion reactor materials, ever since a phenomenon called radiation induced electrical degradation (RIED) was reported by Hodgson [1]. Although some of previous results showed similar but moderate increase of electrical conductivity in fission reactors irradiation [2,3], recently accumulated results on Al_2O_3 (polycrystal and single crystal alumina) strongly support that there would not be a catastrophic increase of electrical conductivity of Al_2O_3 in a fission reactor irradiation [4–6]. Recent results also indicate that the electrical conductivity under irradiation, radiation induced conductivity (RIC), may decrease in the course of atomic displacement associated irradiation [7]. While accepting that RIED would take place under charged particle irradiation, where eccentric phenomena such as spatial electrical charge-up may cause microstructural evolution resulting in permanent increase of electrical conductivity [8], fission reactor irradiation is thought to be more relevant to fusion irradiation environments among presently available irradiation sources [9].

In situ measurements of electrical conductivity of Al_2O_3 were carried out in the Temperature Regulated In Situ Test facility (TRIST) in High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory under the collaboration program JUPITER (Japan/US Project on Irradiation TEsts of fusion Research) [6]. The experiment was denoted as HFIR-TRIST-ER (Electrical Resistivity measurements) and the HFIR-TRIST-ER showed that there would not be a catastrophic increase of electrical conductivity of Al_2O_3 in a helium environment in a high flux fission reactor. Details of the experiment and its major results were described elsewhere [6,10,11].

However, the HFIR-TRIST-ER did show that moderate but distinct increase in electrical conductivity took place for some of Al_2O_3 specimens. The present paper will describe the observed increase of electrical conductivity as well as the observed peculiar behavior of electrical conduction of Al_2O_3, which might be related with the observed increase of electrical conductivity.

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2. Experimental procedures

A detailed description of experimental procedures and a specimen list may be found elsewhere [6,10]. Twelve different grade of Al_2O_3 specimens, including a 0.05% chromium doped ruby specimen, were irradiated for three irradiation cycles of the HFIR at 720–760 K up to a maximum dose of 3 dpa (displacement per atom) and 6×10^{10}Gy, for the atomic displacement and the electronic excitation, respectively in a helium environment. A three cycle irradiation, to a fast neutron ($E > 0.1$ MeV) flux and fluence of about 4.5×10^{18} n/m^2 s, and 3×10^{25} n/m^2, respectively, was carried out in HFIR. A dc electric field of 200 kV/m was applied to the specimens, while no electric field was applied to 3 specimens. Bulk electrical conductivity was measured by the established guard-ring-configuration method, while leakage electrical currents, which would affect estimation of the bulk electrical conductivity, were monitored through the irradiation by the developed measuring system [5].

3. Results and Discussions

Fig. 1 shows electrical conductivity of a ruby (chromium doped single crystal alumina) manufactured by Union Carbide as a function of time for a whole experimental period including periods of reactor intermission. The electrical conductivity only under irradiation is plotted as a function of dpa logarithmically in Fig. 2, emphasizing its initial increase. It is clearly shown that the electrical conductivity increased for an irradiation dose up to about 0.1 dpa [8]. The increase of electrical conductivity was substantial from about 2×10^{-7} S/m at the beginning to about 1×10^{-4} S/m at about 0.1 dpa, more than 500 times increase. At the same time, the electrical conductivity during reactor intermission (hereafter denoted as base electrical conductivity) increased substantially from less than 10^{-9} S/m to more than 10^{-6} S/m. The electronic excitation rate was less than 1 Gy/s and the temperature was about 300 K during reactor intermission. These observations were similar to those reported as the RIED by Hodgson [1].

However, the increase of electrical conductivity did not proceed to electrical breakdown. After it attained its maximum value at about 0.1 dpa, the electrical conductivity decreased continuously to the end of irradiation. Also, the base electrical conductivity decreased substantially as shown in Fig. 1. By the end of the 3 cycles irradiation, the ruby attained its initial electrical conductivity of about 2×10^{-6} S/m under irradiation. Similar results were obtained on some of polycrystal and single crystal alumina, though magnitude of the increase was much small. The initial increase of electrical conductivity of a normal grade single crystal alumina manufactured by Crystal System was shown in Fig. 3.

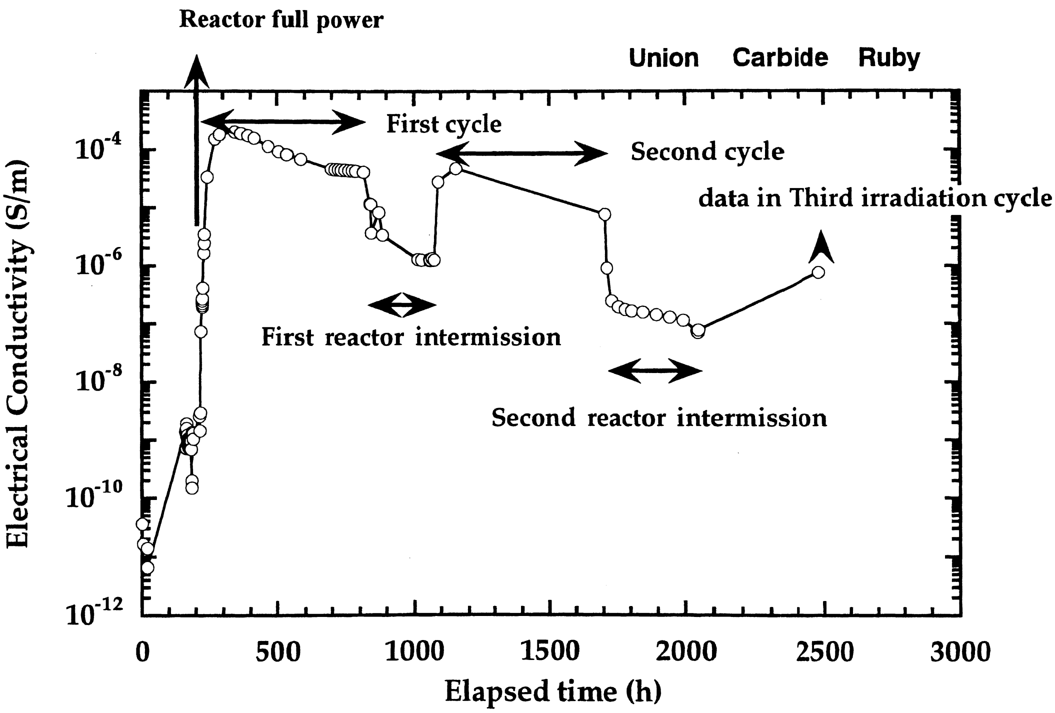

Fig. 1. Electrical conductivity of ruby as a function of time.

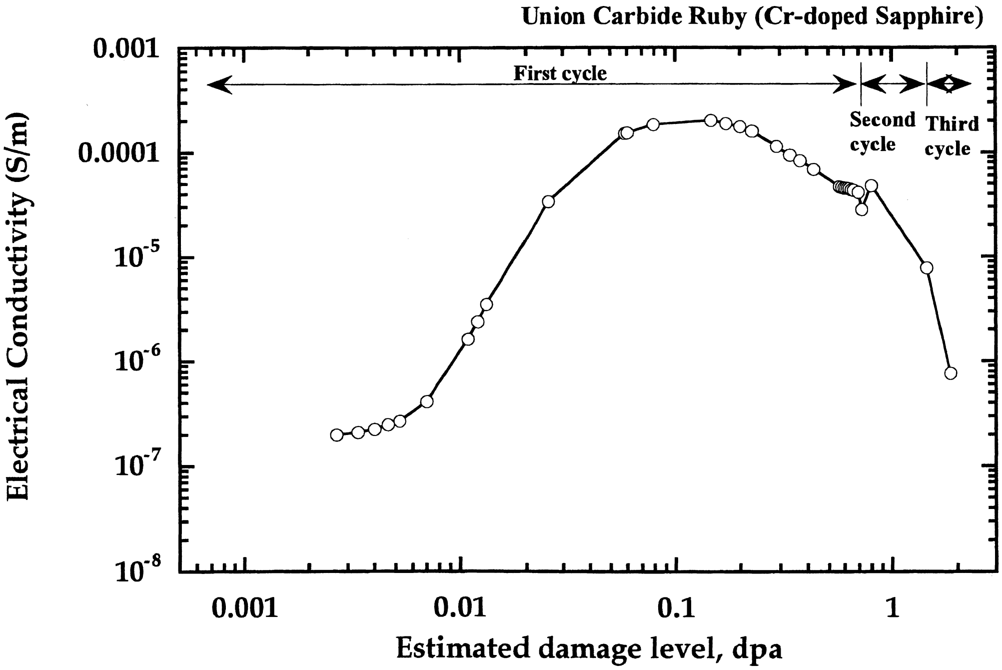

Fig. 2. Logarithmic plot of electrical conductivity of ruby as a logarithmic function of dpa. Initial increase of electrical conductivity is emphasized.

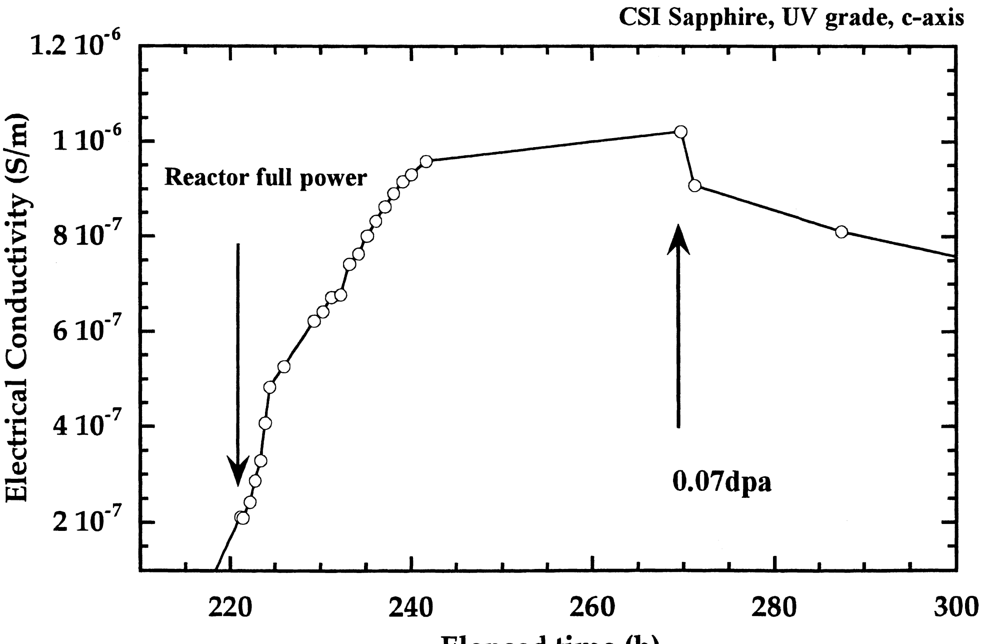

Fig. 3. Initial increase of electrical conductivity of UV-grade sapphire. Maximum is attained at about 0.07 dpa, namely at about 270 h elapsed time.

There, the electrical conductivity attained its maximum also at about 0.07 dpa and then decreased continuously toward the end of irradiation up to 3 dpa. Detailed analyses showed that surface leakage current from the high voltage side electrode to the guard ring electrode was high in the ruby at the initial state of irradiation and that the estimation of a bulk electrical conductivity there might be affected by the leakage current. However, except for this case in the ruby, the measured leakage currents among three electrodes of high voltage side, low voltage side and guard ring were below criteria quantitatively described by Kesternich et al. [12]. The subcapsule was designed to minimize possible electrical leakage paths from a high voltage side electrode to a low voltage side electrode, which is not easy in a reactor irradiation. For example, an electrical path through an ionized gas is screened. Also, precaution was made to avoid microcrackings in the specimens in the procedures of the subcapsule preparation [13]. Thus, the observed apparent increase of electrical conductivity may be due to an increase of intrinsic bulk conductivity although specimen cracking cannot be ruled out.

Moderate increase of the electrical conductivity up to a few tenth dpa was reported previously under irradiation in fission reactors, but their experimental techniques were not well qualified to clearly demonstrate the RIED effect. The present results show confidently that the bulk electrical conductivity increases in the course of irradiation relevant to fusion irradiation conditions, namely having an appropriate ratio of the electronic excitation rate to the atomic displacement rate. However, observed increase of electrical conductivity did not proceed to the catastrophic increase and the resultant electrical breakdown as reported in the phenomenon of RIED. This will imply that the increase of electrical conductivity observed in the present experiment would be different from the phenomenon of RIED or that some mechanisms retard the increase of electrical conductivity in the fission reactor irradiation in a helium environment.

One interpretation of the present results is as follows. As reported before [6,8,10], the value of RIC in the course of the first startup of HFIR did not depend linearly on the reactor power, namely on the electronic excitation rate as schematically shown in Fig. 4, refer-

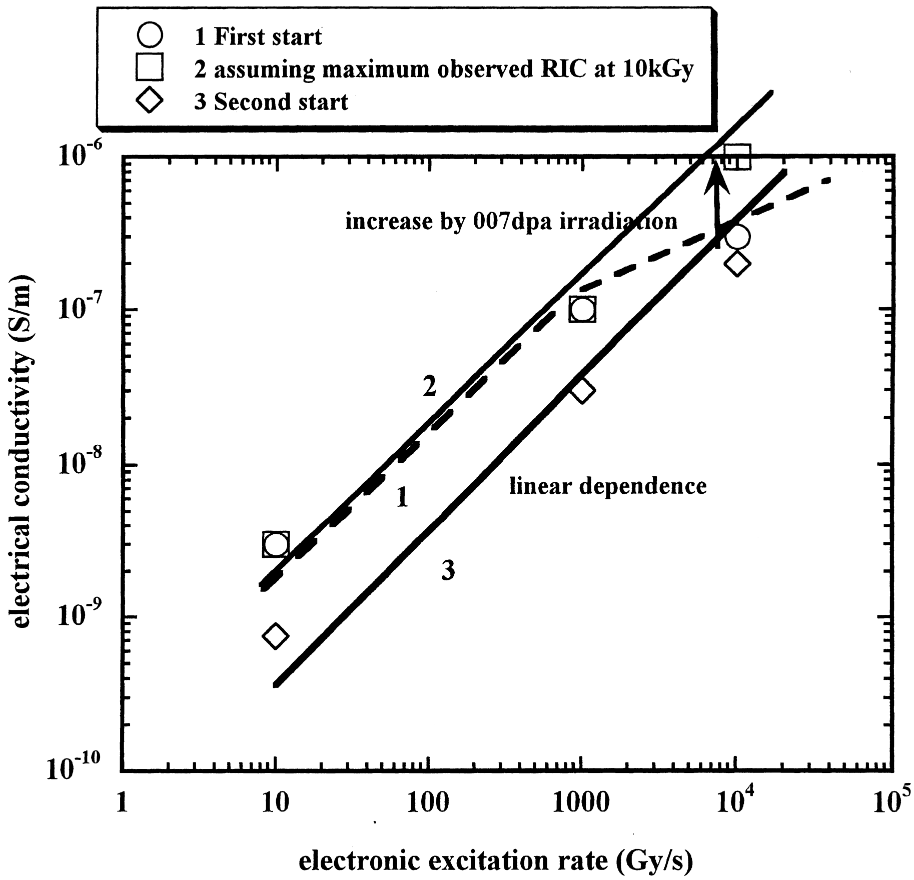

Fig. 4. Schematic display of RIC dependence on electronic excitation rate.

ring data of single crystal alumina. RIC showed saturating behavior at the electronic excitation dose rate larger than 10^3Gy/s. However, RIC showed near-linear dependence on the electronic excitation at the second startup, namely after the specimens were irradiated with an atomic displacement dose of 1 dpa and an electronic excitation dose of 2×10^{10}Gy. The results imply that the irradiation modified rate determinant processes of the phenomenon of RIC. The following speculation may explain the results. Occupation of the shallow donor levels saturated as the electronic excitation level exceeds 10^3Gy/s in the first startup, thus the value of RIC caused by the thermal excitation of electrons from the shallow donor levels, would show a saturation behavior. Then, the atomic displacement increased the number of the shallow donor levels and increase the saturation level and result in the increase of RIC, as observed as the gradual increase of electrical conductivity as shown in Figs. 2 and 3. RIC would then have better linear dependence on the electronic excitation rate, which would be the case in the electronic conductivity at 10^4Gy/s after 0.07–0.1 dpa irradiation plotted in Fig. 4. The decrease of the electrical conductivity above 0.07–0.1 dpa may be due to a decrease of the mobility of conduction electrons suggested by Pells [7] or due to a decrease of effective life time of conduction electrons, which would result from an increase of acceptor levels. In this region, the value of RIC should keep linear dependence on the electronic excitation rate as in the case shown in Fig. 3.

A large offset current was observed in the present study as reported elsewhere [11]. Substantial current was measured from the low voltage side electrode to ground and from ground to the high voltage side electrode, even when electric voltage was not applied to the specimen. Magnitude of the current was initially in the range of 5–10 μA. Then, the current increased and attained its maximum of more than a few tens of micro amperes at about 0.07–0.1 dpa as shown in Fig. 5, about the same dpa where the electrical conductivity attained its maximum as shown in Fig. 3. The behavior of the offset current, being concurrent with that of the electrical conductivity, may support the speculation that some electronic modification took place during the irradiation up to a few tenths of a dpa.

The ruby is speculated to have lower concentration of shallow donor levels than normal sapphires, which resulted in the lower RIC, observed in the beginning of irradiation. However, the concentration of donor levels will increase due to the displacement irradiation even during the reactor startup. Then, RIC of the ruby should reach the similar value of the sapphire at higher reactor power, as observed in the present experiment. However, this model does not explain the large increase of electrical conductivity of the ruby in the course of irradiation. Also, the offset current should maintain its maximum value through the irradiation or continue to increase, if it had some correlation with the concentration of shallow donor levels.

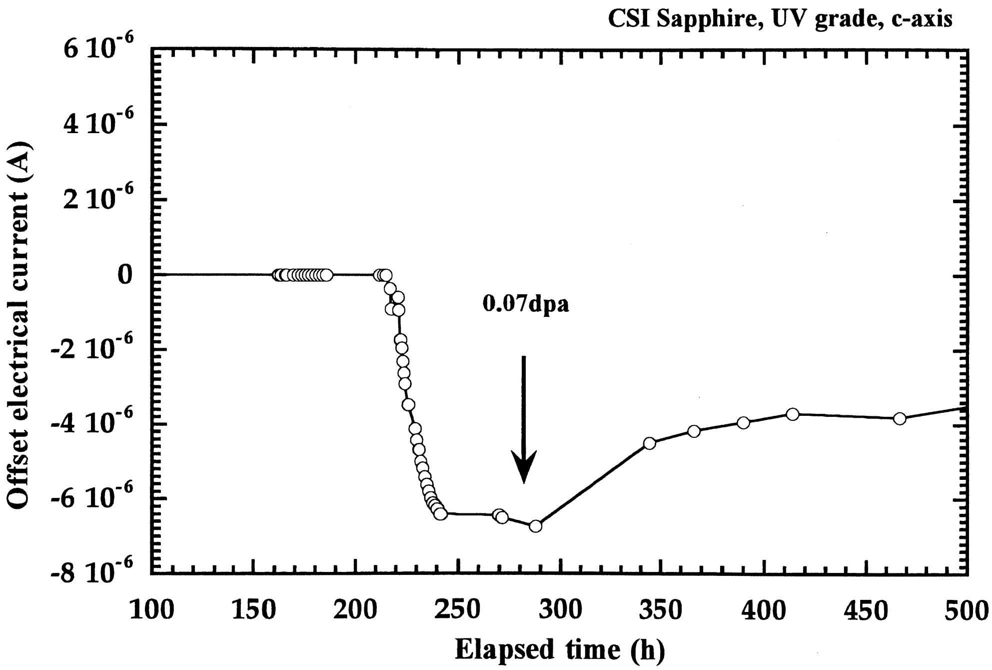

Fig. 5. Behavior of offset electrical current in the initial irradiation stage as a function of irradiation time. Offset electrical current attains its maximum at about the same dpa, where the electrical conductivity attains its maximum shown in Fig. 3.

Another explanation is that an increase of electrical conductivity similar to the phenomenon of RIED is taking place in the course of the reactor irradiation. Here, the impurity of chromium would play an important role in the increase of electrical conductivity. The offset current might reflect microstructural modification which resulted from the RIED-like phenomenon, such as localized electrical charge-up caused by long-range ion migration. Electrical impurities such as chromium in the ruby would assist such microstructural modification and would result in the larger increase of electrical conductivity as described above. However, some phenomena may retard its procession as the irradiation proceeded. The spatial charge-up itself mentioned above might make an barrier to retard the catastrophic procession of the electrical-charge-migration.

However, detailed analyses of behavior of electrical currents as a function of applied voltage have not revealed occurrence of distinct charge-up or any other abnormal behavior up to now. One result which might support the RIED-like degradation is that the electrical measurements were prematurely terminated on many specimens as described elsewhere [11]. The high side electrode was grounded by some cause. The most probable cause of the termination is the normal breakdown at the glass seal at the terminal of a mineral insulated cable at the high voltage side [6,10,11]. However, some correlation between the timing of the premature intermission of the measurements and the grade of alumina could be seen. When we compare the results on Crystal System normal grade and UV-grade sapphires, the specimens for which the electric field was applied along *a*-axis showed earlier timing of termination of measurements. In the case of polycrystal alumina, specimens having higher purity had earlier timing of termination when the polycrystal aluminas made by the same manufacturing company are compared. These observations may support the existence of RIED-like phenomenon in the present irradiation or may simply be a coincidence.

Further more detailed analyses of the present results are needed to clarify the cause of the observed increase of electrical conductivity.

4. Conclusion

Some long term increase of electrical conductivity of Al_2O_3 was observed in the course of HFIR irradiation for displacement dose up to a few tenth dpa. A ruby, chromium doped single crystal alumina showed substantial increase of electrical conductivity, which is similar to the phenomenon reported as RIED. Complicated behavior of electrical conduction in the course of irradiation implied complexity of behavior of the irradiation-affected electrical conduction in the alumina. The substantial increase of electrical conductivity in the ruby suggests that the impurity plays an important role. Although accumulated experimental results indicate that RIED would not be a problem for the ITER development, further detailed studies are needed to understand the complicated behavior of electrical conduction in alumina under irradiation.

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References

[1] E.R. Hodgson, J. Nucl. Mater. 191–194 (1992) 552.
[2] V.A. Neverov, Yu.L. Revyakin, to be published in Plasma Devices and Operation.
[3] T. Shikama et al., J. Nucl. Mater. 191–194 (1992) 544.
[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] S.J. Zinkle et al., Fusion Materials Semiannual Progress Report for the Period ending June 30, DOE/ER-0313/20, 1995, p. 257.
[7] G.P. Pells et al., Harwell report AERE-13222, 1988.
[8] S.J. Zinkle, Proceedings of the 9th IEA workshop on radiation effects in ceramic insulators, ORNL/M-6068, 1997.
[9] T. Shikama, G.P. Pells, J. Nucl. Mater. 212–215 (1994) 80.
[10] S.J. Zinkle et al., Fusion Materials Semiannual Progress Report for the Period ending June 30, DOE/ER-0313/22, 1997, p. 188.
[11] T. Shikama et al., these Proceedings.
[12] W. Kesternich, F. Scheuermann, S.J. Zinkle, J. Nucl. Mater. 206 (1993) 68.
[13] W.S. Eatherly et al., Fusion Materials Semiannual Progress Report for the Period ending December 31, DOE/ER-03131/19, 1995, p. 241.