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Electrical in situ and post-irradiation properties of ceramics relevant to fusion irradiation conditions

Tatsuo Shikama [a], S.J. Zinkle [b,*]

[a] Tohoku University, Katahira, Sendai 980-8577, Japan

[b] Oak Ridge National Laboratory, Metals and Ceramics Division, P.O. Box 2008, Oak Ridge, TN 37831-6138, USA

Abstract

Electrical properties of ceramic candidate materials for the next-generation nuclear fusion devices under relevant irradiation conditions are reviewed. A main focal point is placed on the degradation behavior of the electrical insulating ability during and after irradiation. Several important radiation induced effects play important roles: radiation induced conductivity, thermally stimulated electrical conductivity, radiation induced electrical charge separation, and radiation induced electromotive force. These phenomena will interact with each other under fusion relevant irradiation conditions. The design of electrical components for the next-generation fusion devices should take into account these complicated interactions among the radiation induced phenomena.

1. Introduction

Functional ceramic materials are employed for many purposes such as electrical insulators and optical windows in plasma diagnostic components in nuclear fusion devices [1]. While major interests in radiation effects are placed on structural materials for reactor systems, functional ceramic materials will provide important theaters for displaying impacts of radiation effects in the next-generation nuclear fusion devices, such as the international thermonuclear experimental reactor (ITER). In particular, their properties will be affected by smaller irradiation doses than those of metallic structural materials in general and some of their properties will be affected from the beginning of the operation by phenomena called dynamic irradiation effects [2]. Here, the next-generation fusion devices are assumed to have a nuclear burning plasma composed of deuterium and tritium, and to generate a substantial intensity of neutron and gamma-ray fluxes.

Extensive international studies have been carried out concerning radiation effects in functional ceramic materials in the course of the engineering design activity of the ITER (ITER-EDA), which was successfully completed in July 2001 [3]. Several important aspects of radiation effects in functional ceramic materials have been surveyed and evaluated experimentally. General conclusions were that most of the functional ceramic materials, which are candidates for applications in the ITER, could withstand degradation of their properties caused by radiation effects in the course of their services in the ITER. Several international collaborative experiments were set up to study the behavior of functional ceramics during irradiation. Typical examples were studies on degradation phenomena of electrical insulating ability, called radiation induced electrical conductivity (RIC) and radiation induced electrical degradation (RIED) [4–6], and on radiation effects on optical transmission properties in fused silica core optical fibers [7]. Also, the phenomenon called radiation induced electromotive force (RIEMF) was studied by international collaborative groups, as it might affect the performance of important diagnostic components such as magnetic probes and bolometers [3,8].

Electrical insulators play crucial roles in electrical devices deployed near the burning plasma in the ITER.

* Corresponding author. Tel.: +1-865 5767220; fax: +1-865 2413650.
E-mail address: zinklesj@ornl.gov (S.J. Zinkle).

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Several important experimental enterprises were initiated to study radiation effects in the electrical properties of ceramic insulators in four major players in the ITER-EDA, namely the European Union (EU), Japan (JA), Russian Federation (RF), and the United States of America (US). Controversial results were reported, concerning the phenomena of the RIED and intense discussions were exchanged among the international research groups. Up to now, several excellent review papers were published on this topic [9–13] and these activities were summarized in the IEA (International Energy Association) workshop held in Cincinnati in 1998 [14]. To sum up the discussions, some of the ceramics, such as highly pure alumina (Al_2O_3) could keep their electrical-insulating ability through their expected service periods in the next-generation fusion devices represented by the ITER [5,6].

Some mechanisms were proposed for explaining the observed degradation of the electrical insulating ability, namely the increase of the electrical conductivity in the course of irradiation. The most plausible one will be that energy deposited in electronic systems of irradiated materials, electronic excitation effects of radiation, will be converted into structural defects, occasionally resulting in formation of new phases such as gamma-alumina in alpha-alumina reported by Morono and Hodgson [15]. The proposed mechanism has a strong correlation with the phenomenon called radiolysis well known in dielectric insulators of alkaline salts, with assistance of an outer electrical field [4]. However, no single mechanism proposed up to now could explain the complicated and controversial experimental results comprehensively.

Post-irradiation measurements of the electrical conductivity were carried out on several grades of alumina irradiated in the high flux isotope reactor (HFIR) of Oak Ridge National Laboratory. Results were analyzed extensively with experimental results obtained in situ [5,6] during the HFIR irradiation. This is the first successful measurement of the electrical conductivity of ceramic insulators whose electrical properties was already measured in situ in a fission reactor under conditions relevant to those expected in the next-generation fusion devices such as the ITER. Results revealed complicated aspects of degradation of the electrical insulating ability of the alumina [16]. There, several fundamental phenomena are thought to play important roles, such as RIC, RIEMF, thermally stimulated electrical conductivity (TSC), and radiation induced electrical charge separation (RICS). The complicated behavior of the electrical conductivity of irradiated materials will imply important technological issues to be handled for the development of the next-generation fusion devices.

Several excellent review papers [2,9–13] have been already published, which describe general aspects of radiation effects in ceramic insulators for applications in the next-generation fusion devices. The present paper will concentrate to review the recent understandings of electrical properties of ceramic insulators, mainly alumina which is the primary candidate ceramic insulator for ITER.

2. Experimental results on the electrical conductivity of alumina during and after irradiation in HFIR

Fig. 1 shows the electrical conductivity of a highly pure single-crystal alumina (Crystal Systems Hemex UV grade sapphire) under irradiation in the HFIR. The irradiation was carried out at about 730 K with an averaged fast ($E > 0.1$ MeV) neutron flux of 4.9×10^{18} n/m^{2}s and a gamma-ray dose rate of 10–12 kGy/s. The irradiation extended to three HFIR irradiation cycles, corresponding to 75.78 effective full power days [17]. The dc voltage of about 100 kV/m was applied to the material through the irradiation. Measurements of the electrical conductivity were carried out in situ during the reactor operation, with procedures recommended by the IEA research group for the radiation effects in ceramic insulators chaired by Zinkle, with a so-called guard-ring configuration [18]. The scheme of the measuring circuits was depicted in Fig. 2. An electrical current from a guarded center electrode to the ground potential was measured to evaluate the bulk electrical conductivity. Details of the measuring procedures and obtained results were already reported elsewhere [5,6,17–21].

General results could be summarized as follows:

1. Nine kinds of commercially available alumina, single crystals and polycrystals, highly pure of 99.9% up to moderate grade of 97%, kept their electrical conductivity below 10^{-6} S/m, up to a displacement dose of 2.8 dpa (displacement per atom) and an electronic ex-

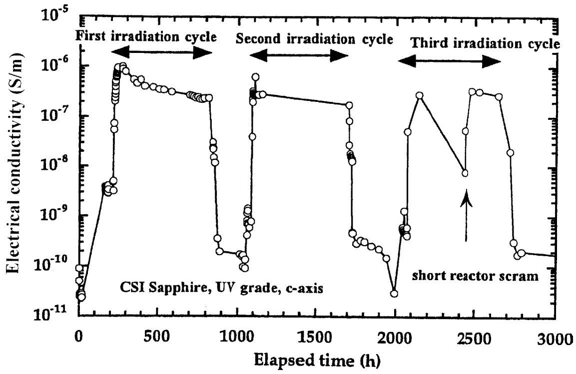

Fig. 1. Electrical conductivity of highly pure alumina single-crystal, crystal systems Hemex, under HFIR irradiation (in situ measurement).

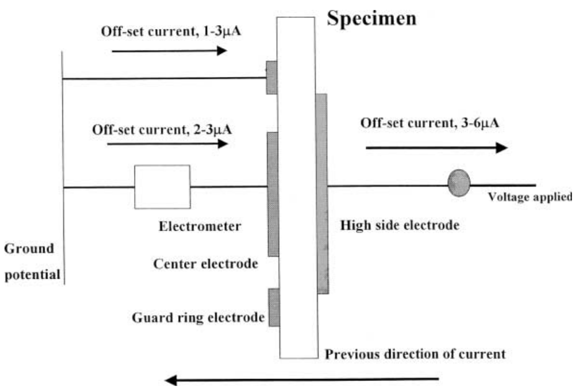

Fig. 2. Scheme of measuring circuit and direction of off-set current in relation with previous direction of current due to applied outer voltage.

citation dose up to 8×10^{10} Gy/s, except for one specimen, a chromium doped sapphire. However, it should be pointed out that the measurements of the electrical conductivity were interrupted at early stages of irradiation, on some of the specimens. The electrical conductivity of 10^{-6} S/m is the upper limit for designing diagnostic components in the ITER [3].
2. A moderate increase of the electrical conductivity was observed at the beginning of the irradiation up to about 0.1 dpa on many specimens [5].
3. A chromium doped alumina single crystal, ruby, showed a relatively large increase of the electrical conductivity. It showed a large surface conductivity, suggesting that the measured bulk electrical conductivity was affected by a surface leakage current [5].
4. A large off-set current (electrical current measured even with no applied voltage), was observed in many specimens. Direction and magnitude of the off-set current are schematically shown in Fig. 2 and its magnitude is plotted as a function of the irradiation time in Fig. 3.

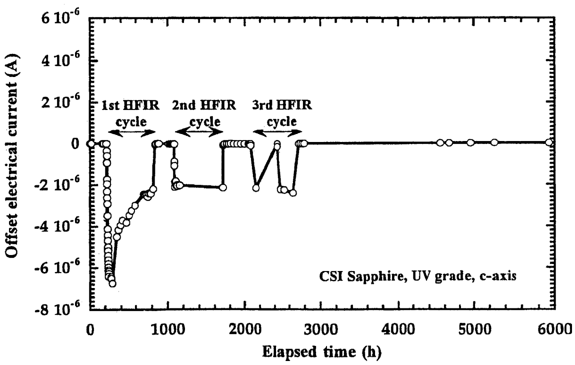

Fig. 3. Off-set current observed under HFIR irradiation as a function of irradiation time (in situ measurement).

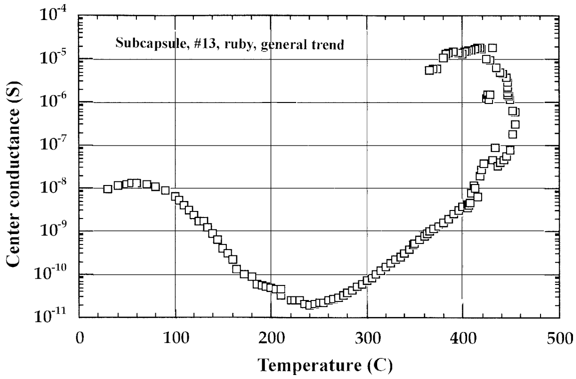

Fig. 4. Electrical conductance of chromium doped single crystal alumina, ruby, after HFIR irradiation, as a function of measuring temperature (post-irradiation measurement).

Fig. 4 shows the temperature dependence of the electrical conductivity of the irradiated ruby. Detailed procedures of the post-irradiation measurements of the electrical conductivity of irradiated alumina are given elsewhere [16]. Due to the subcapsule holding the specimen had an induced radioactivity from the HFIR irradiation (an electronic excitation dose rate at the specimen of about 0.01 Gy/s), a residual electrical conductivity of about 10^{-10}–10^{-12} S/m, occurred in the specimens due to the radiation induced conductivity (RIC). A measuring limit of the electrical conductivity ranged from 10^{-11} S/m at 300 K to 10^{-8} S/m at 670 K in the post-irradiation tests performed in a remote-handling hot cell.

General features of the measured electrical conductivity of the irradiated specimens, especially which were irradiated in the HFIR with a dc voltage, could be summarized as follows:

1. The electrical conductivity (σ_e) increased with increasing temperature (T) smoothly up to about 500–730 K, except for values measured at temperatures below 500 K, as shown in Fig. 4. The temperature dependence, $d\sigma_e/dT$, above 500 K, was about the same as that reported on the unirradiated high-purity alumina. However, absolute values of the measured electrical conductivity were far larger than those of unirradiated specimens (due in part to surface contamination of the irradiated specimens). The measured electrical conductivity is summarized in Fig. 5 along with that measured on the unirradiated alumina [22].
2. The electrical conductivity showed a large increase above certain temperatures, usually above 750 K, and it exceeded 10^{-4} S/m. After the specimen had the maximum electrical conductivity and the current was larger than 1 mA/cm^2, the specimen showed electrical breakdown.

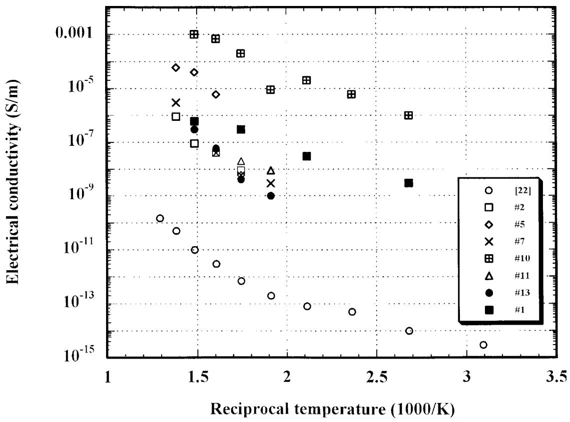

Fig. 5. Temperature dependence of electrical conductivity of HFIR irradiated alumina (post-irradiation measurement).

3. The electrical conductivity showed an abrupt increase when the heating rate was occasionally increased as shown in Fig. 6. This increase and the larger electrical conductivity observed below 400 K (shown in Fig. 4) appear to be caused by the phenomenon called thermally stimulated electrical conductivity (TSC) [23–25]. The existence of TSC means that there would be a large number of trapped electrical charges.
4. The irradiated specimens behaved like an electrical battery. They provided an electrical current of a few μA, after a dc voltage was applied for more than several minutes. The decaying behavior of the discharge current had a time constant longer than 100 s.

Results of in situ and post-irradiation measurements showed a complicated behavior for the electrical conductivity of alumina. One important feature is that there would be a large number of trapped charges (large number of charge-trap sites) in irradiated specimens. With voltage application, the specimen would have a localized charge separation, which will compensate the outer electric field locally in some places but will also amplify the outer electric field in some places. This amplified electric field may contribute to radiolysis, converting electronic defects into structural defects and also to stabilization of unstable structural defects. Also, trapped electrical charges will be liberated when there is a temperature perturbation, which will cause a large localized electrical current. Then, this large current could result in localized electrical breakdown. It was estimated that the critical current would be in the range of 1 mA/cm^2 as described above.

A threshold damage level should be needed to have a large number of charge trap-sites. This will explain the behavior of RIED reported by Hodgson [4]. Also, application of an outer electric field will assist electrical charges to be trapped and it will assist the trapped charges to be liberated. These would be the role of the applied electric field in RIED [4]. In the case of electron irradiation, where RIED was frequently observed [4], some of irradiating electrons will be trapped within a specimen. Thicker specimens will be needed to realize a large number of localized charge-traps, which may explain why RIED was not observed in ion-beam irradiation experiments. The perturbation of irradiation temperatures will be the most important trigger to initiate the electrical breakdown, which was consistent with the results of the HFIR experiment [15].

Liberation of trapped electrical charges is a phenomenon well known as TSC [23–25]. Usually, electrical charges will be trapped in pre-existing trap sites in the crystals in the TSC with assistance of electronic excitation irradiation. Then, the spectra of released charges are rather simple and well-forecast as a function of temperature. However, the displacement damage will introduce a variety of trap sites whose trapping energies will vary in a wide range. So, the conductivity induced by the TSC would be far more complicated in electrical insulators irradiated in the conditions relevant to next-generation fusion devices.

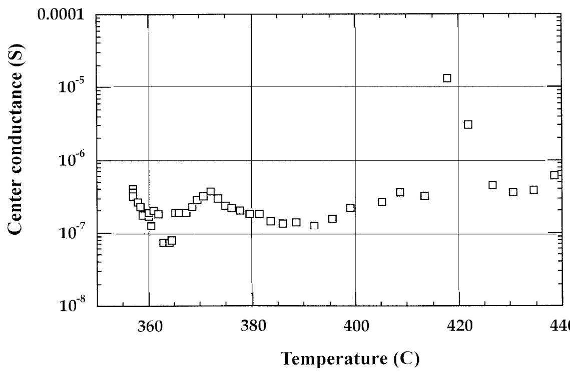

Fig. 6. Increase of electrical conductance in the course of heat-up of specimens (post-irradiation measurement).

3. Radiation induced electromotive force

Electrical voltage and current are generated between two metallic parts, that are separated by an electrical insulator under irradiation. This is called RIEMF. The phenomenon has been recognized from the beginning of the development of nuclear fission systems since 1950, especially in mineral-insulated cables (MI-cables) [26,27]. The cause of RIEMF was implied to be an electrical charge transportation through the insulator between two metallic parts. The primary cause of the electrical charge transportation has generally been considered to be the high-energy electrons generated by the beta-decay of neutron-activated components.

However, the magnitude of the induced current in MI-cables could not be explained by the density of beta-emitters generated in metallic parts as well as those generated in the insulator layer. Furthermore, a polarity change of RIEMF was occasionally observed in MI-cables, which could not be explained by the beta-emitter mechanism. A detailed analysis of the observed RIEMF implies that low energy electrons activated by the irradiation processes, such as photoelectrons, are essential in RIEMF. In that case, interface structures between the metallic part and the insulator should play important roles to control movements of the low energy electrons [28].

The influence of RIEMF on the performance of a magnetic coil, which should play an essential role in controlling the plasma at the beginning of operation, has been extensively studied in the ITER-EDA [29]. At present, the results strongly support that RIEMF itself would not cause a serious influence on the performance of the magnetic coil relevant to ITER. However, the present design of ITER forces the size of the magnetic coils to be as small as possible. Then, the allowable long-term voltage drift should be less than 1 μV/s. In that case, any asymmetric contribution of RIEMF to the coil leads might cause drift voltages of this magnitude. Comprehensive irradiation tests of the coil relevant to the ITER design will be carried out in the JMTR under international collaboration.

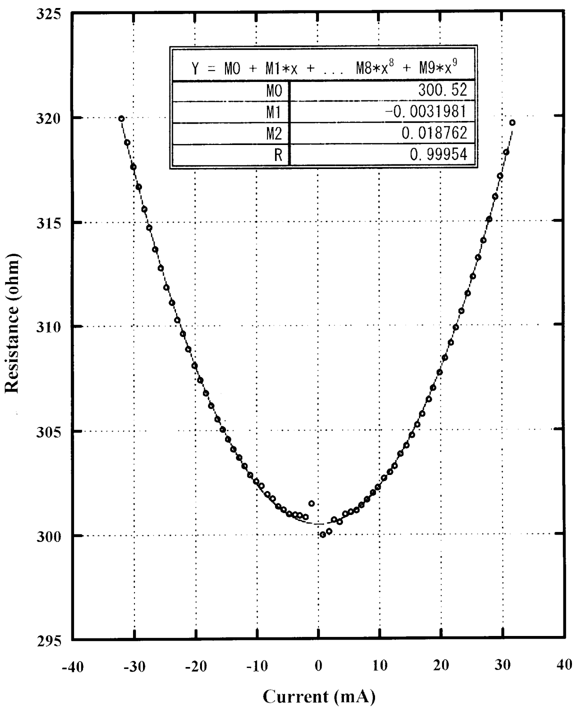

Fig. 7. Electrical resistance of gold meander composing ITER-relevant bolometer as a function of electrical current, under irradiation in JMTR (in situ measurement).

Another example of the effects of RIEMF on diagnostic components of a bolometer is shown in Fig. 7 [30–32]. The electrical resistance of gold meander will be a monitor for a small change of temperature when the bolometer receives heat from the plasma. The relationship between the received power and the electrical resistance clearly follows in Fig. 7 a parabolic law under JMTR irradiation. However, small scatters of the data could be identified near zero power, which was caused by RIEMF generated in the gold meander. The analysis revealed that the data scatter would not be a problem for the bolometer diagnostics in ITER.

4. Behavior of electrical insulators in fusion devices

Fig. 8 depicts the schematic behavior of the electrical conductivity of a highly pure alumina in fusion devices. Permanent degradation of the electrical insulating ability, called RIED, will proceed gradually and the intrinsic electrical conductivity would exceed the critical value of 10^{-6} S/m only after the insulator received a few dpa. This will correspond to the dose which electrical insulators in in-vessel components would receive in a few years. The electrical insulating ability will be determined by the radiation induced electrical conductivity, which is roughly proportional to the electronic excitation dose rate. The value of RIC would be in the range of about 10^{-8}–10^{-6} S/m at the electronic excitation dose rate of 10 kGy/s, which will be about 10 times larger than that expected at the first wall of ITER. Thus, in general, the candidate alumina would satisfy the design criteria for the electrical insulator in ITER.

However, the electrical insulator would show a transient increase of the electrical conductivity, when the temperature is temporarily increased, due to the phenomenon of the TSC. Depending on the rate of the temperature change, the TSC-induced electrical current can be much larger than that of RIC. When the electrical current induced by TSC could exceed the critical value (about 1 mA/cm^2 for highly pure alumina), the alumina would then experience electrical breakdown. This would cause malfunctions of the electrical components.

The irradiated electrical insulator will show a capacitive behavior, which will affect the performance of the electrical insulators. When a voltage is applied to insulators, a far larger current will be initially introduced across the insulators than the steady-state current. A current will flow across the insulator for a while after the voltage is cut-off. These currents may cause malfunctions of some electrical components.

RIEMF will cause another problem. Along with the off-set current described above, it will induce electrical noise in diagnostic components. Also, RIEMF may

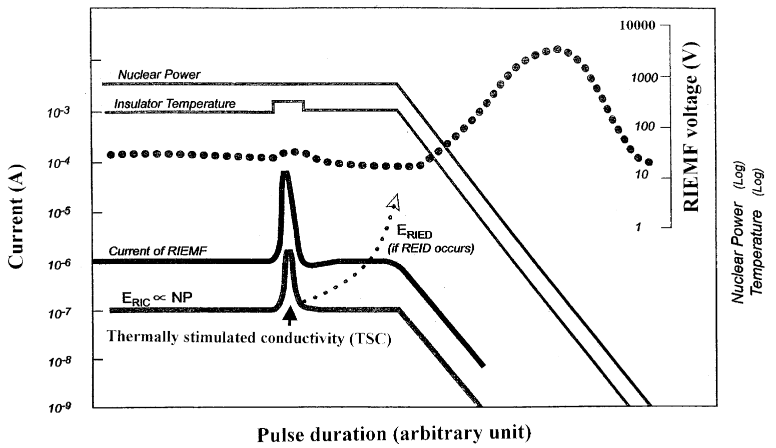

Fig. 8. Scheme of behavior of electrical insulators under nuclear fusion relevant irradiation (summarized by T. Shikama and C. Walker in the ITER-EDA [3]). Electrical current through 1 cm^2 and 1 mm thick electrical insulator with 100 V applied.

generate substantial voltage, even up to a few kV, as RIEMF is generally a current-driven phenomenon that would be imposed on circuits with high internal impedance. When some electrical components are electrically floated (such as those included for redundancy, which would be electrically disconnected during stand-by), a large voltage induced by RIEMF may cause a electrical breakdown.

Finally, Hodgson reported that RIED would result in the degradation of mechanical properties. Occasionally, micro-cracks were found in specimens exposed to irradiation with voltage applications. One interpretation was that the specimen preparation procedures introduced these micro-cracking and enhanced diffusion of electrically conductive materials along these crackings would cause the RIED-like behavior. However, another interpretation would be that RIED caused the formation of these micro-cracks, through radiolysis-enhanced accumulation of displacement damage and resultant formation of secondary phases, as proposed by Hodgson. Also, the phenomenon of radiation-enhanced sub-critical crack growth (SCCG) reported by Pells et al. [3] may degrade the mechanical properties under irradiation.

Recently, diagnostic components were disassembled after they were irradiated in a fission reactor, JMTR. It was found that some of ceramic plates became brittle and were easily broken in the processes of disassembling. The total displacement damage was about a few tenths of dpa, and the degradation of the mechanical properties was not usually expected in ceramic materials such as alumina and aluminum nitride. The ceramic plates, which showed mechanical degradation, were irradiated with an applied mechanical stress and electrical voltage. The applied stress and voltage may cause enhancements of degradation of the mechanical properties by irradiation. Synergistic effects of mechanical stress, electrical field and other extensive environmental effects should be extensively studied for applications of ceramic materials in actual components for the next-generation fusion devices, to minimize the risk of unexpected failures during their service.

5. Summary

Recent understandings of radiation effects in ceramic materials are reviewed, mainly focused on their electrical properties and dynamic irradiation effects. Individual elementary processes of radiation effects on ceramic materials have been well surveyed and quantitatively evaluated through the international collaborative efforts of ITER-EDA. However, synergistic effects of multi-processes of radiation effects may induce new physical phenomena and may cause unexpected degradation. Further investigations of synergistic effects, under irradiation conditions relevant to the next-generation fusion devices, are needed.

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