NEUTRON IRRADIATION IMPACT ON ITER GRADE INSULATING MATERIAL

SEJAL SHAH[1,2], M. BANDYOPADHYAY[1,2], A. CHAKRABORTY[1]
[1]ITER-INDIA, Institute for plasma research, Gandhinagar, India
[2]Homi Bhabha National Institute, Mumbai, India
Email: sshah@iter-india.org

SUNIL KUMAR[3], S. VALA[3], R. KUMAR[3], M. ABHANGI[3], S. PRASAD[4]
[3]Institute for plasma research, Gandhinagar, India
[4]FCIPT, Institute for Plasma Research, Gandhinagar, India

Abstract

Ceramics are widely being used for insulation in high voltage systems of nuclear reactors. They are subjected to different type of radiations in the reactors hence the impact on radiations on the insulating material and thereby its performance in long operating period need to be ensured. Present study focus on the impact of irradiation on Al_2O_3 (alumina) ceramic, the one being used for high voltage isolation in neutral beam injectors of ITER. Production proof samples of required sizes of high purity alumina are prepared. Samples are irradiated by thermal and high energy neutrons using two different neutron sources. Analytical simulation is performed for two neutron spectra prior to irradiation, to estimate probable transmutation and damage. In-situ and ex-situ characterizations are performed to study irradiation impact to ensure its structural and electrical compatibility. SEM of low energy neutron irradiated sample showed defect cluster formation on ceramic surface. It is observed that surface morphology is getting affected mainly due to low energy neutrons whereas electrical and structural properties are being affected by high energy neutrons. Proposed study will help in selection of precise grade of insulating material for nuclear reactor applications, based on the expected radiation dose.

1. INTRODUCTION

Insulation is an essential part of any high voltage system, which can be provided by either gaseous, liquid or solid insulators based on the application and operational environment. In case of nuclear reactors, most of the systems are non-replaceable during operating period. Hence, precise selection of the insulator is the key aspect of successful operation of the machine. Alumina is one of the functional materials which is widely being used in fusion reactors.

In case of International thermonuclear experimental reactor (ITER) [1], neutrons are inevitable outcome. One of the possible fusion reaction is:

$$D + T \rightarrow He\ (3.5\ MeV) + n\ (14.1\ \text{MeV})$$

High energy neutrons, generated through this reaction, interact with the reactor material. In presence of radiation shields, expected neutrons at component location may not be mono-energetic. Hence assessment is required to estimate the neutron energy spectrum at the area of interest using neutron transport code.

Compatibility of insulator for vacuum, high voltage and radiative environment need to be ensured for the operation of high voltage system. The study is initiated using Kyocera A 479 grade, high density ~$3.8\text{x}10^3$ kg/m^3 alumina which is widely being used in high voltage bushings [2-4] of neutral beam injector systems [5, 6] of ITER and classified as protection important class (PIC). Vacuum, structural and high voltage compatibility of this insulator has already been established during prototype experiments [7, 8]. However, its operational performance in presence of radiative environment need to be established. Further, Kovar©-Ceramic brazed configuration is commonly used for ceramic to metal transition as a vacuum sealing boundary [3]. Silver is used to perform this brazing by active titanium brazing process. Upon neutron irradiation, Ag gets transmuted into Cd. It is recommended that the materials to be used in vacuum must show low rate of outgassing unlike Cd due to its evaporation properties [9]. To estimate the amount of generated Cd, preliminary assessment of nuclear activation and transmutation is carried out using FISPACT [10]. For the estimated requirement of Ag during brazing for the ceramic ring of DNB, transmuted Cd is ~$1\text{x}10^{-7}$ grams which can be considered as negligible amount. This exercise helped in finalizing the acceptable amount of Ag during brazing. Further, total damage is calculated for given neutron energy spectrum analytically and experimental assessment is initiated to study the impact of irradiation on material properties.

Step by step approach is taken to understand the impact of damage created due to neutron irradiation on the material properties. The irradiation experiment is initiated by creating very low damage using 14.1 MeV neutron source. Estimation of the damage in term of displacement per atom (DPA) is carried out by online available SPECTER code. Considering limitations of the source flux and operation time, irradiation is planned for 2 hours. In-situ and ex-situ characterizations of the pristine and irradiated samples are performed. Based on the outcome, the sample are further irradiated for long time using thermal neutrons to create higher damage than the earlier case. Pristine and irradiated samples are characterized for structural properties and surface morphology. However, it is difficult to create ITER equivalent damage using lab scale neutron source hence further study is extended using ion beam irradiation and will be reported separately.

2. NEUTRON IRRADIATION

For the neutron irradiation, samples are prepared from production proof material with the required size followed by ultrasonic cleaning and necessary characterizations prior to irradiation. They are irradiated using two available neutron sources at Institute for Plasma Research (IPR) viz (i) 14.1 MeV mono-energetic neutrons & (ii) Am-Be source (max. flux @ ~0.5 MeV). The irradiation parameters, characterizations and their results are presented in subsequent section.

2.1. 14.1 MeV irradiation

Monoenergetic neutrons generated through D-T reactions. 200 keV deuterium beam is targeted on Tritium to generate 14.1 MeV neutrons by fusion reaction as explained in introduction. The neutron flux at sample location is $2x10^6$ n/cm^2.s. The samples are irradiated for 120 minutes and calculated damage is ~$3x10^{-15}$ DPA. Despite of very low damage interesting trend is observed during in-situ electrical measurement.

2.1.1. In-situ electrical measurement

In-situ Insulation Resistance (IR) is measured with time up to 120 minutes for applied voltage 2.5 kV DC. IR is observed to improve with time, as a typical characteristic of any excellent insulator. However, IR is abruptly changing at the time of switching on/off the neutron source. Rapid reduction in IR is attributed to radiation induced conductivity [11] in the insulator as shown in Fig. 1.

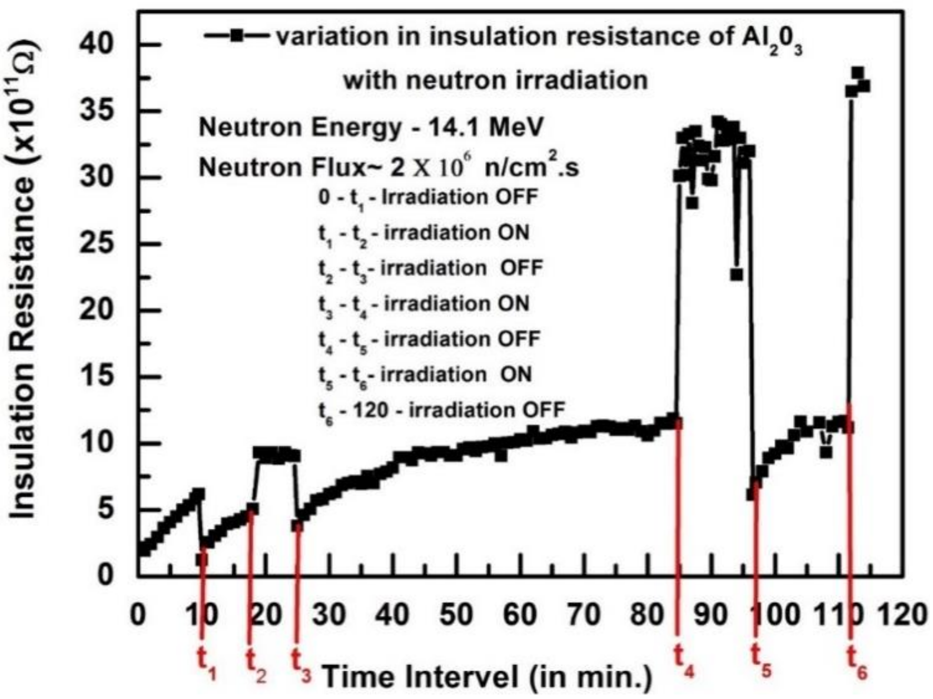

Figure 1 In-situ IR measurement

Despite of the limitation of precise current measurement through megger, the trend of reduction in leakage current with time shows good correlation with Curie-von Schveindler law $I(t) = I_0 x t^{-n}$, where n is the decay constant, calculated to be 0.3 by fitting the curve and it matched with the requirement of 0<n<1 according to the law. IR is observed to recover after irradiation which indicates that the electrical functionality of the material does not get altered for the damage up to ~$3x10^{-15}$ DPA. The impact of irradiation on the structure is studied by X-ray diffraction analysis.

2.1.2. X-ray diffraction

XRD peak intensity of irradiated sample is observed to increase post irradiation with 14.1 MeV neutrons. Detailed analysis of the peaks is as shown in Table-1.

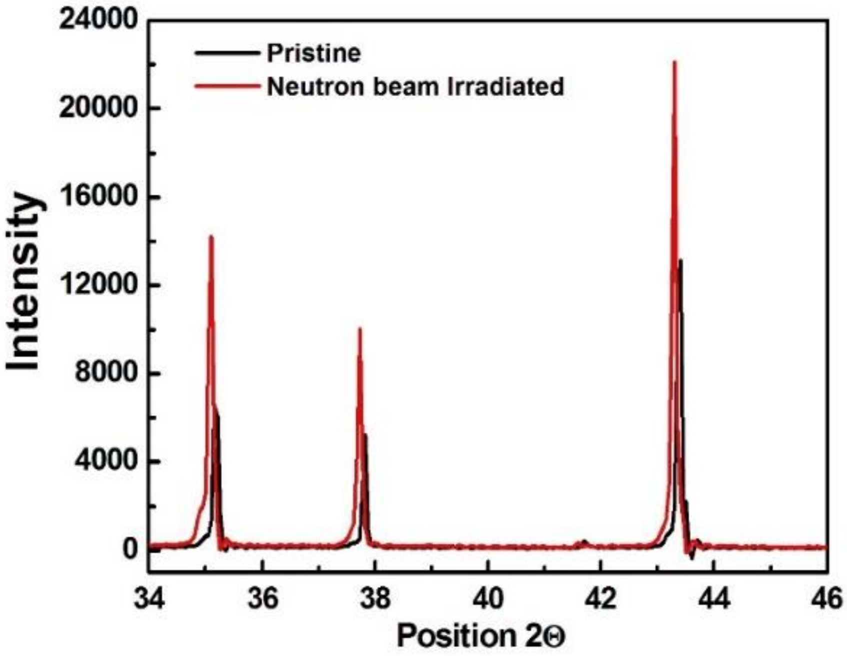

Figure 2 XRD of 14.1 MeV irradiated neutrons

Fig. 2 shows that after 14 MeV neutron irradiation, XRD peaks position shifted to lower angle. Peak shift can be caused by strain or by changes in chemical composition [12]. In our case change in chemical composition possibility is ruled out as transmutation of Al and O is negligible and any elementary particles are not detected in EDX. Neutron irradiation create defects which may develop strain. As XRD peak shifted to lower angle, the developed strain is compressive in nature [13].

TABLE 1. XRD PEAK ANALYSIS

Position		Width		Intensity	
Pristine	Irradiated	Pristine	Irradiated	Pristine	Irradiated
35.18	35.09	0.051	0.076	980	1951
37.80	37.72	0.040	0.079	572	1240
43.38	43.29	0.032	0.063	2097	2723

From the Table-1, it is noted that the width (FWHM) is increases after irradiation which is clear evidence of defect creation post neutron irradiation. The Intensity of peak increases after irradiation which signifies the improvement in crystalline plane after irradiation. Further assessment by changing neutron dose is ongoing.

2.1.3 Scanning Electron Microscopy (SEM)

SEM is performed for 14.1 MeV irradiated samples shows, there is no significant change in surface morphology post irradiation. Fig. 3 reveals that high energy neutrons mainly interact with the bulk property of the material while surface morphology remain unaltered post irradiation.

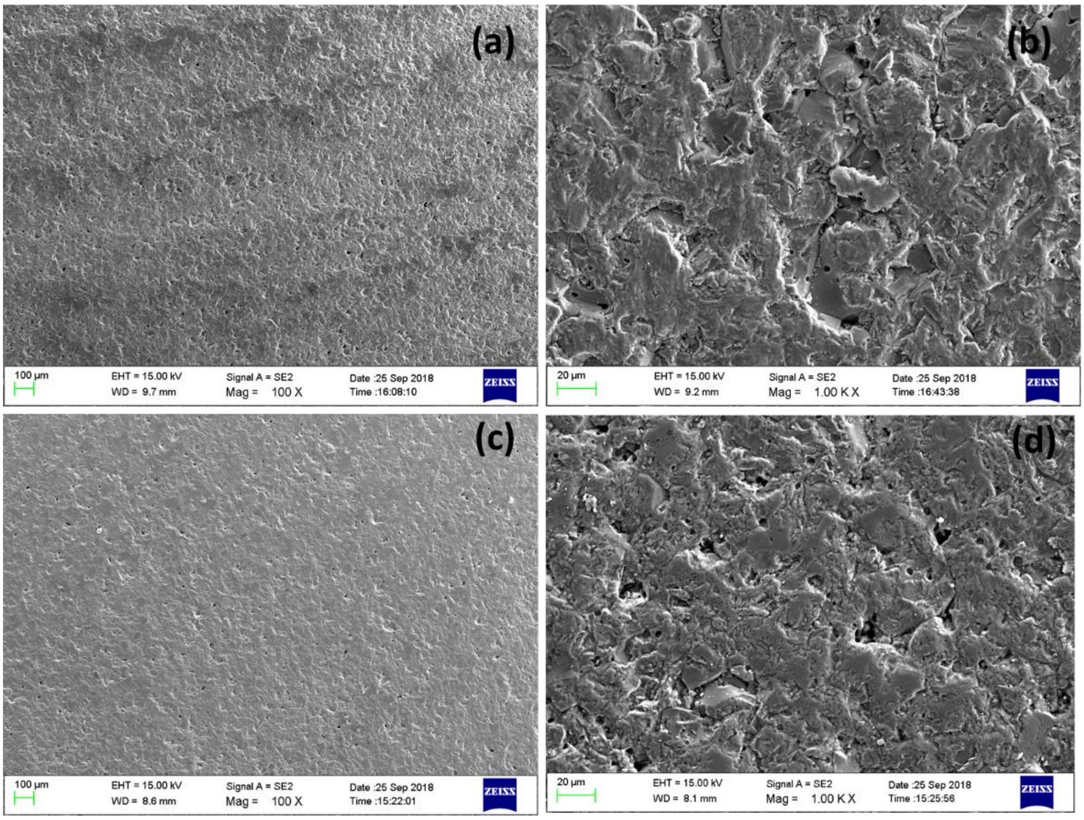

Figure 3 SEM of (a,b)Pristine and (c,d) irradiated Al_2O_3

No significant change in surface morphology reveals, no major surface interaction of high energy neutrons whereas structural and electrical properties are observed to change post irradiation. However, the insulation resistance recovered after stopping the irradiation which implies that the proposed material can be used for the DPA up to $3x10^{-15}$. To increase the damage further, another source is used for irradiation.

2.2. Thermal neutron irradiation

With Am-be source, the neutron spectrum obtained at the sample location with the maximum flux for the energy 0.5 MeV. Total flux is calculated at sample location ~ $2.8x10^4$ n/cm^2.s. Due to low flux, the irradiation time is kept longer up to 96 days and estimated damage is ~$3x10^{-9}$ DPA. Despite of low damage, the impact on surface property is prominent.

2.2.1. *Scanning electron microscopy (SEM)*

The samples are ultrasonically cleaned. Because the samples are non-conducting, gold coating is performed on the surface prior to SEM characterization. Images are captured at different location after scanning entire surface of the sample. Agglomeration is observed post irradiation. Similar results are observed during our earlier studies [8] when the sample was irradiated up to 45 days using the same source. Cluster formation or agglomeration is observed only for irradiated samples, which indicates the irradiation induced surface defects. Neutron/Ion beam induced surface defect has been reported earlier for different materials [14-16]. Fig. 4 (a, b) and Fig. 4 (c, d) shows morphology of pristine and irradiated sample respectively. Due to long irradiation time, in-situ characterization was not possible however, post irradiation its insulation resistance is observed to recover.

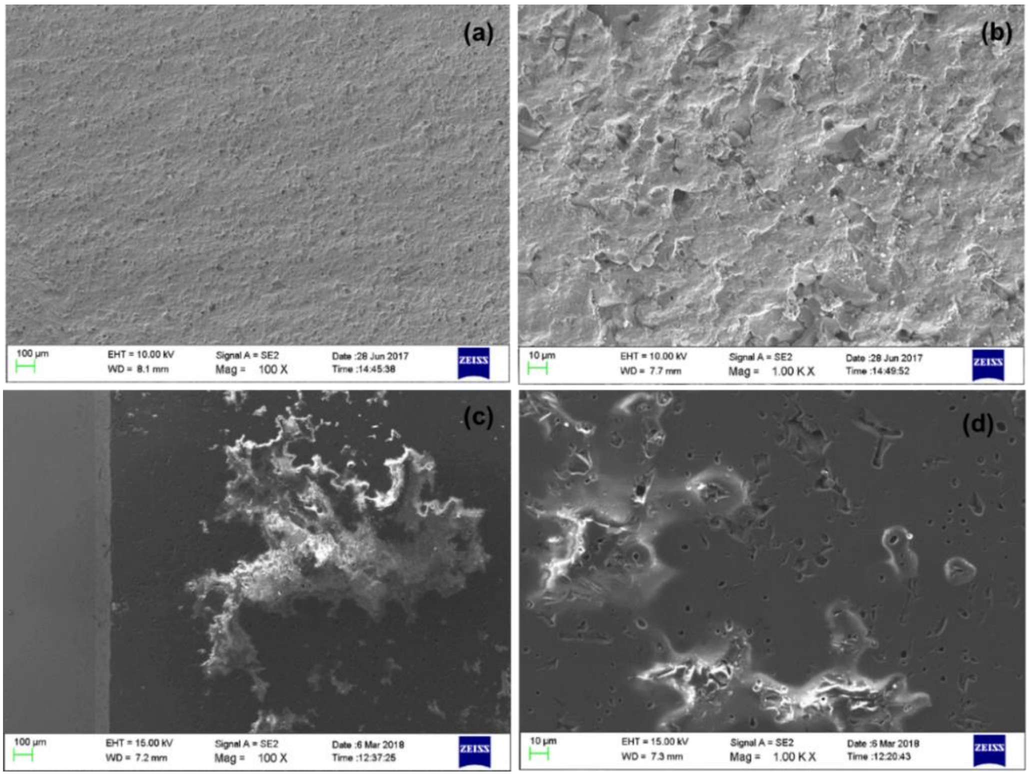

Figure 4SEM of (a,b)Pristine and (c,d) irradiated Al_2O_3

To ensure that there is no surface contamination or stray particle, Energy Dispersive X-ray analysis (EDX) is performed to study elemental composition at the damaged region as shown in Fig. 5.

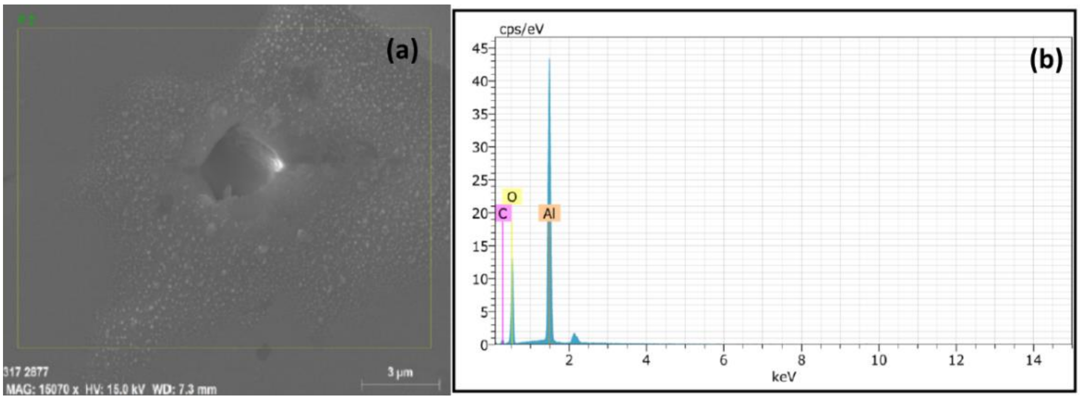

Figure 5 EDX spectrum of irradiated Al_2O_3

As conducting layer of gold is coated on alumina only to perform SEM characterization, gold peak is eliminated in EDX analysis. Major peak obtained by EDX are Al and O implies the agglomeration or cluster formation of the same material post irradiation. Structural changes are further studied using Photoluminescence (PL).

2.2.2. *Photoluminance*

Fig.6 shows photoluminescence (PL) spectra of pristine and neutron irradiated Al_2O_3 samples. There are doublet peak name R_1 and R_2. This doublet structure is due to excitation and de-excitation of Cr^{3+} which is present in Al_2O_3 at Al^{3+} lattice sites [17]. The change in intensity of R_1 and R_2 give the information about crystalline nature of Al_2O_3, defects introduction and stress/strain development after neutron irradiation [18].

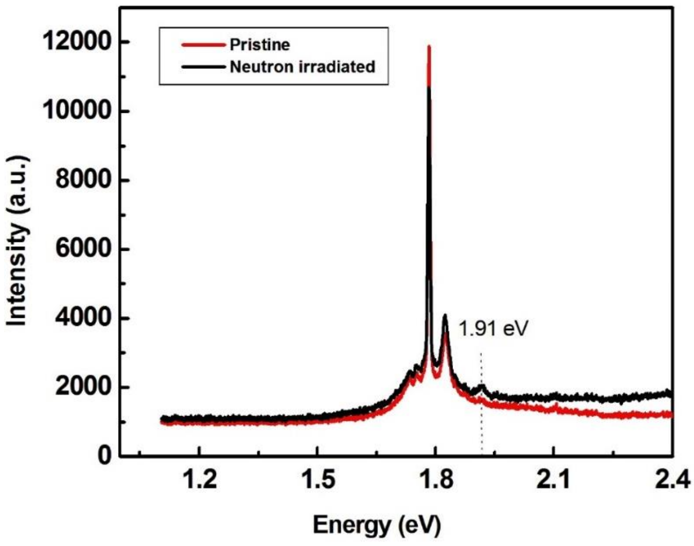

Figure 6 PL of pristine and irradiated Al_2O_3

It can be seen that the intensity of most pronounces R1 peak decreases after neutron irradiation. Intensity decrease is attributed to either surface roughness change or defects production after irradiation [19, 20]. Smoothing of Al_2O_3 increases surface reflectivity which may lead to increased PL intensity. As AFM images shows there is no significant change in surface roughness, there is possibility of defect production after neutron irradiation. Peak at position 1.918 eV also gets pronounced which indicate the stress development in Al_2O_3 after neutron irradiation which will be further validated by XRD. Also the impact of this surface damage and change in structural properties on electrical insulation of Al_2O_3 is being studied.

3. DISCUSSION

Ceramics are widely being used in various high voltage systems due to their excellent vacuum, electrical and environmental compatibility. However, different grade of ceramic exhibits different behavior depending on the operating conditions. Also, in nuclear reactors, along with neutrons several high dose radiation is expected which may alter the material properties and its operational performance. It is, therefore, becomes important to choose proper grade of insulating material for specific application. There are not much experimental data available to study the impact on radiation on the material properties. This paper mainly describes the impact of low and high energy neutrons on the material properties of ITER grade ceramic material.

Neutron irradiations experiments are performed in lab to create defect in the material. Different level of defects are created using two neutron sources available at Institute for Plasma Research, Gandhinagar, India. The defect is calculated analytically for the expected flux of neutrons, its energy spectrum and irradiation time. In-situ electrical measurement set up is prepared and insulation resistance (IR) of the ceramic is measured. Instantaneous change in IR is observed for high energy neutrons which is due to radiation induced conductivity. Also crystallinity observed to increase post irradiation which might be due to thermal spike at lattice sites. However, detailed material-neutron interaction based on the energy of the incoming neutrons is being carried out analytically. No noticeable change in surface morphology observed for 14.1 MeV neutrons. On the contrary, significant surface defect is observed due to thermal neutron interaction. It is observed that high energy neutron impacted the bulk properties of the material whereas thermal neutron mainly impact the surface properties of the material.

Studies have been performed for the defect of the order of $3x10^{-15}$ DPA and $3x10^{-9}$ DPA. Despite of low damage, properties of the material is observed to get altered. This study will provide a database for material selection in the radiative environment where low damage is foreseen. Step by step study is ongoing to evaluate the operational performance of insulator for higher DPA up to the damage level of ITER. Due to operating limitation of lab scale neutron sources, it is difficult to create the required damage. Nevertheless, it is known that the use of ion beams to mimic the irradiation damage produced in a reactor is a vital tool for the understanding the changes in material properties in radiative environment. Hence further study has been initiated using low and high energy ion beam and shall be reported separately. This study will help in realization of safe radiation limit for the material to maintain its operational performance and thereby selection of proper grade of the material for reactor based applications.

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