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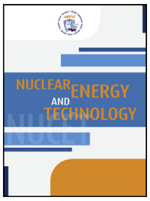

The high-dose and high-temperature monitors of reactor irradiation based on insulators

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Abstract

Radiation-induced changes of the structure and properties have been investigated for oxide and nitride materials, and the use of high-temperature insulators as temperature/dose monitors for in-reactor irradiation of materials test assemblies has been validated.

It has been experimentally shown that the use of Al_2O_3 single crystals and BN ceramics provides means of monitoring the temperature of irradiation from 370 to 1900 K. The temperature is derived from measurements of the optical absorption or X-ray diffraction line shifts after post-radiation annealing of the monitors. We discuss the applicability of (a) the optical absorption and F-center luminescence spectroscopies of irradiated Al_2O_3 single crystals for gamma dose evaluation and (b) the isotopic analysis of irradiated BN ceramics for neutron dose evaluation.

Keywords: Radiation material science; Reactor irradiation; Insulators; Structure characterization; Optical measurements; Temperature monitor; Dose monitor.

Introduction

Clarification of the conditions of high-temperature and high-dose nuclear reactor irradiation of materials is an important and topical problem for the experimental radiation materials science. In-situ monitoring is of extreme importance for nuclear power reactors, for which the local irradiation conditions (temperature, doses) are usually calculated (for a given reactor power, neutron and gamma-ray spectra, heat flux, etc.) or evaluated with the help of various passive monitors placed in the locations of interest. Post-radiation examination of the monitors, their structure, properties and isotopic composition, is capable of providing necessary data on the irradiation conditions (doses, and temperatures).

Radiation-induced changes of structure (various radiation-induced defects) and physical properties of materials are usually dose- and temperature dependent. During long-term continuous irradiation, the defects accumulated in the materials are solely those survived at the maximum irradiation temperature, and other defects are annealed at lower temperatures. Accordingly, the irradiation temperature can be derived from a dependence $A(T)$ of a structure-sensitive property A of the irradiated material on the post-irradiation annealing temperature T (Fig. 1). This method is not reliable if the irradiation temperature varies significantly in the course of irradiation. In this case, the dependence $A(T)$ must be associated with the temperature regime at the end of irradiation.

Nevertheless, at steady-state temperature conditions this method can be useful, especially in combination with other methods of temperature measurement, for example, with the use of fuse monitors (fuse temperature monitors), or computational methods.

In this paper we substantiate the use of high-temperature insulators as monitors for a wide range of irradiation temperatures

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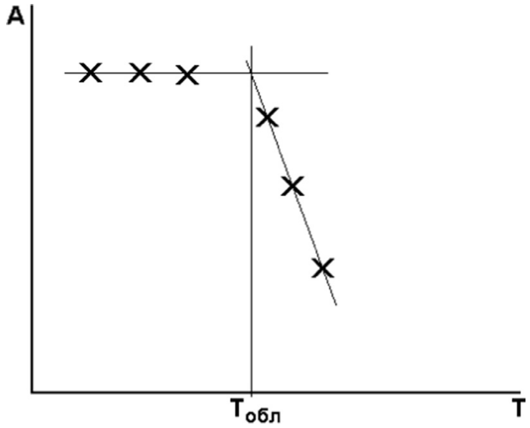

Fig. 1. Determination of the irradiation temperature with the help of series of post-radiation annealing and measurements of a material property *A*.

and doses. To substantiate the use we consider radiation-induced structural changes and related post-irradiation physical properties of insulators.

Radiation monitors based on oxides and nitrides

To observe the irradiation temperature at the plot of dependence $A(T)$, it is necessary to measure the property A of a monitor material in a wide temperature range. High-temperature insulators, for example, oxides or nitrides (Al_2O_3, MgO, $MgAl_2O_4$, BN, Si_3N_4, SiC) with melting temperatures from 2000 (Si_3N_4) to 3000 K (BN), provide the temperature range for annealing of radiation defects that is much wider than the metals can provide, and the upper annealing temperature limit can reach the melting temperature of the insulator. The latter is due to the fact that irradiation of oxide or nitride insulators not only generates various lattice defects, as it happens in irradiated metals, but also results in the formation of local nonstoichiometry and, moreover, the precipitation of new phases [1]. It is worth noting that a much larger number of structure-sensitive properties $A(T)$ can be analyzed for the insulator monitors, including dielectric permittivity, loss tangent, luminescence, UV–vis–IR optical absorption, electrical resistance, etc. To study the phase composition and stoichiometry of irradiated insulators, both X-ray and lattice-vibration spectroscopy can be used [2,3].

As an example, optical measurements conducted for sapphire (α-Al_2O_3 single crystal) can be mentioned. Irradiation generates oxygen-deficient regions and optically active oxygen vacancies (F-centers) in sapphire [4]. Typical spectra of radiation-induced optical absorption of sapphire are shown in Fig. 2. Effect of thermal annealing on optical absorption of the sapphire irradiated in a BR-10 reactor (SSC RF – IPPE) has been studied. Reactor irradiation to the fluence of $1.5 \cdot 10^{21}$ neutrons/cm^2 gave rise to the background absorption in the wavelength range from 215 to 560 nm, and bands of optical absorption at 413, 256, 227 and 205 nm were observed. During thermal annealing, the optical absorption changed at temperatures ranging from 350 to 900 K. The temperature of the annealing onset for the 413 nm band corresponded to the temperature of post-irradiation storage.

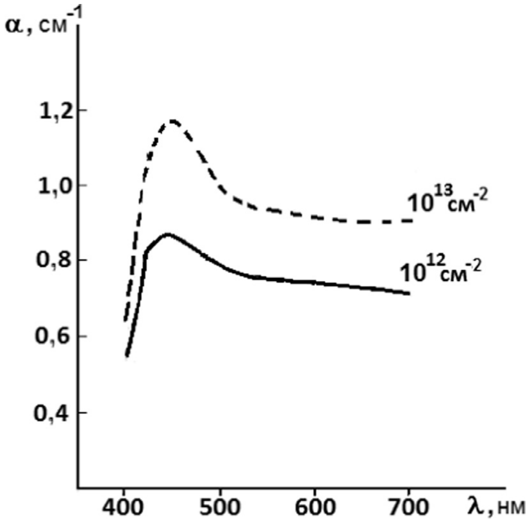

Fig. 2. Optical absorption spectra of sapphire irradiated by 210 MeV Kr^+ ions (300 K).

During irradiation, radiation-induced oxygen vacancies perpetually change their valence states from F^+- to F-centers (positively charged and neutral oxygen vacancies, respectively) and vice versa. These centers give rise to optical absorption bands at 205–256 nm, as well as the photoluminescence bands at 413 nm (F centers) and 328 nm (F^+-centers) [4]. As is also, optical measurements allow one to evaluate the absorbed gamma dose via optical absorption and luminescence intensity. Changes of optical absorption of gamma-irradiated sapphire in the range of the fundamental absorption edge (200–300 nm) were observed at annealing temperatures exceeding of 370 K.

Temperature monitors based on BN

Various materials sorts of graphite-like boron nitride (BN) are known:

- ceramics fabricated by the hot pressing;
- ceramics fabricated by the reaction sintering of a pressed mixture of boron powder and turbostratic boron nitride [7] via annealing in nitrogen atmosphere;
- pyrolytic layers fabricated by the method of chemical vapor deposition, using BF_3 (BCl_3) and NH_3 [8] as reagents.

Ceramic boron nitride is an isotropic material with a crystal grain size in the range of 100–1000 nm. Pyrolytic boron nitride is a significantly textured nanocrystalline material with a specific structural hierarchy [9]. Aggregates of sizes up to 2000 nm consist of grains of size approximately 100 nm, the latter consists of weakly misaligned tiny crystals of 30–60 nm in size (Fig. 3).

A specific feature of the crystal structure of the graphite-like BN is the presence of isomorphic phases with different interlayer distances. Their structures comprise both the irregular order of basic planes of the hexagonal phase AA’AA’ and the stacking faults of AA’BAA’ type that are in fact fragments of the rhombohedral phase [5]. Different phases can join coherently within