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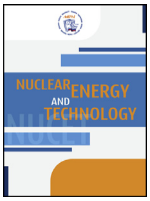

The temperature monitoring during the reactor core material irradiation by analyzing the structure of graphite-like boron nitride

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Abstract

Possibility has been shown for determining the temperature of in-core irradiation using a single structural analysis of irradiated boron-nitride ceramics without a time-consuming research stage, including post-irradiation annealing. This is explained by the fact that steady structural states, depending on temperature and not depending on exposure dose, may develop in nanocrystalline graphite-like ceramic boron nitride in high-dose environments. A temperature dependence of the position of the X-ray line (002) has been obtained for the radiation-induced steady structure of ceramics. This dependence can be used for determination of temperature in a fast reactor in a range of 690 to 1870 K.

It has been found using X-ray structural analysis that reaction-sintered boron-nitride ceramics in the initial and irradiated states comprises two structural components. The first one corresponds to defect-free hexagonal and rhombohedral structures. The X-ray line displacements relate to the second component the nanocrystallites of which contain a great deal of microdeformations stabilized by clusters of vacancy discs.

Keywords: In-pile irradiation; Temperature monitors; Structural studies.

Introduction

Often, temperature of equipment components cannot be determined directly in the course of the development and operation of new advanced nuclear facilities when exposure doses exceed 10^{22} neutron/cm^2 and temperature rises to above 700 K. The explanation is that the properties of temperature detectors change greatly in conditions of radiation, and readings thereof turn out to be inaccurate. A method was proposed in [1] to determine the temperature in the reactor exposure conditions by analyzing the radiation-induced changes in the material structure. The method is based on the fact that materials exposed to radiation tend to accumulate only damage which is stable in conditions of irradiation and are annealed at temperatures exceeding the test or operating temperature. It is possible to determine the irradiation temperature by the start of the change in a structurally sensitive physical parameter of the monitor material with an increase in the post-irradiation annealing temperature.

Special requirements are placed on the monitor material. It is required to possess a broad temperature range in which reliably fixed structural changes take place under the action of radiation exposure. It was also proposed in [1] that high-temperature dielectric oxide, nitride and carbide materials should be used, in which the annealing temperature for the radiation-induced structural damage may reach the melting point (above 2000 K). It was found out that the temperature in a fast reactor can be determined in principle through a series of sequential annealing sessions and a structural analysis of the irradiated BN materials.

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It has been shown as part of the study that steady structural states, depending on temperature and not depending on exposure dose, may develop in nanocrystalline graphite-like ceramic boron nitride in high-dose environments. This makes it possible to determine the temperature of in-core irradiation through a single analysis of the irradiated BN ceramics structure without a time-consuming research stage, including post-irradiation annealing.

Method justification

Irradiation by fast neutrons or ions leads to cascades of atom-atom collisions formed in materials. Depending on the energy of the primary knocked-on atom (PKA), and the material temperature and type, the cascade represents either a group of small regions of Frenkel pairs or a compact region in which all of the PKA energy is spent to ionize the environment and displace atoms. The efficiency of the formation of dense high-energy cascades in the course of fast-neutron irradiation is $\eta \sim 3-4\%$ and, as shown by calculations in [2] and by estimates based on the measurement of the cascade glow intensity [3], the cascade region has an average size of $d \sim 6-7$ nm and contains 10^4 to 10^5 atoms. With times of up to 10^{-11} s, all energy of the dense cascade (up to 100 keV, over 1 eV/atom) is concentrated in it, after which pressures of up to 10^{11} Pa ($\sim E_{\text{cascade}}/d^3$) occur on the region boundary, and most of the energy in the compact continuous medium is entrained by deformation waves [3]. This results in a plastic deformation of the material's regions much greater of the cascade size. Structural damage is so caused not only by structural relaxation inside the cascade regions but also by plastic deformation outside the cascades. A dose growth leads to a continuous increase in the material damageability as the result of plastic deformation in conditions of an excessive content of radiation-induced point defects up to the material failure in the form of swelling.

There is no elastic long-range action in ceramics. Structural changes take place only within some of the crystallite grains. If the dimensions of ceramic grains are comparable to that of the cascades, damageability depends only on the processes inside the cascade regions. One can determine the saturation dose at which dense cascades span the whole of the volume, and above which no structural changes take place any longer with no long-range action being present:

$$D \approx 1/(d^3 \eta \sigma N), \tag{1}$$

where η is the efficiency of the formation of high-energy cascades; σ is the cross-section of the interaction between the neutron and the material atoms; and N is the concentration of atoms. For many materials, the saturation dose exceeds 10^{20} neutron/cm^2.

Such estimates are valid in the event that the nanodimensional grain structure remains stable in conditions of high-dose irradiation. Materials with an equilibrium stable nanostructure, in which the crystallite grain dimensions are always nanometric, include BN-based materials. Due to an intense anisotropy of the atomic bonds in the basal plane and between close-packed layers (the bond energies differ by a factor of 100 [4,5]), the size of the graphite-like BN crystallite grains (up to μm along the basal plane) is limited thermodynamically. When this size is exceeded, there is a greater probability of the formation of defects which break down the crystallite. Boron-nitride materials are obtained by hot compaction, reaction sintering [6] and pyrolytic synthesis [7]. These materials consist of differently sized crystallite grains with dimensions of 50 to 1000 nm along the basal plane and up to several nanometers across [6,8].

Apart from structural damage processes, fast-neutron irradiation of nanostructured ceramics involves phenomena leading to the structure recovery. With nanometric dimensions of grains containing up to 10^6 atoms, the temperature across the grain may rise up to the melting temperature in conditions of the so-called thermal peak of the atomic displacement cascade. The explanation is that the heat transfer is complicated on the boundaries between grains. It can be shown that, in BN grains with dimensions of 10 to 100 nm along the basal plane and with a thickness of 1 to 10 nm, the temperature across the volume at the cascade thermal peak stage may reach the melting point of 3000 K. Above the saturation dose (1), the radiation-induced degradation is compensated by the structure recovery in thermal peaks. This results in a steady nanostructure of ceramics.

Ceramics obtained by reaction sintering through the annealing in a nitrogen atmosphere of a compacted mixture of boron powders and turbostratic boron nitride (ORPE Tekhnologiya [6]) was used to determine the correlation dependence between the developing steady structure of graphite-like BN and the irradiation temperature. The ceramics consisted of crystallite grains with the basal-plane dimensions of 100 to 1000 nm. Samples were irradiated in the BR-10 fast reactor (IPPE) with an above-threshold dose of $D = 1.5–10^{21}$ neutron/cm^2 ($E > 0.1$ MeV) in an argon environment at a temperature of 690 K. The irradiation was followed by a series of annealing sessions (at a temperature of up to 1870 K) in a vacuum of $\sim 10^{-3}$ mm Hg. X-ray spectra of the boron nitride samples were obtained using the radiation of Cu•K$_\alpha$ on a DRON-2 diffractometer in a continuous recording mode. Structural changes were recorded by the position of the base line (002) in X-ray diffractograms.

The position of the X-ray maximum (002) versus the annealing temperature is shown in Fig. 1. In-pile irradiation at 690 K led to a minor displacement (by 0.6 deg.) of the line maximum (002) into the low-angle region. The higher was the temperature, the smaller was the displacement. The dependence consists of two linear segments in the intervals of 690 to 1100 K and 1300 to 1870 K, the second segment having three times as small slope as the first one. It can be seen that there is an explicit correlation between the temperature and the position of the X-ray maximum (002) across the temperature interval. Major X-ray displacements are explained by peculiarities of the graphite-like BN crystallite structure defect composition which stabilize the nanostructured state of ceramics.