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REACTOR RADIATION DOSIMETRY USING FUSED QUARTZ

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The characteristics of quartz glass for separate dosimetry of γ radiation from a VVR-SM reactor in the presence and absence of neutron fluxes are investigated. Comparing the absorption and photoluminescence spectra of samples irradiated with γ rays from a stopped reactor and mixed with neutron and γ radiation from an operating reactor shows that in both cases oxygen defects are produced and the dose dependences are linear. The dosimetric bands are stable with respect to light and temperatures up to 400°C. The stationary γ-ray flux from the reactor, after the reactor is stopped, is calibrated according to a known source of γ-ray source ^{60}Co and is ~15 Gy/sec.

Radiation-induced formation of defects in oxides is a serious problem for disposal of radioactive wastes by methods of vitrification and cementation. The combined effect of neutron and γ radiation on oxides has been investigated in detail, and it has been believed that neutrons with energy >100 keV are capable of displacing atoms and the γ component gives rise only to ionization heating of materials and electron transfer between atoms. However, even back in 1973, it was shown [1] that average Compton-electron energy $E_{av} = 5E_g = 40$–45 eV is sufficient to produce one electron-hole pair in a SiO_2 lattice with gap width $E_g = 8$–9 eV. Then the concentration of optical centers produced by γ radiation is related to dose by the empirical relation $N(\text{cm}^{-3}) \approx 10^{10} D_\gamma$ (Gy). It has been proved experimentally that ^{60}Co γ rays with $E \sim 1.25$ MeV can shift light anions (O, F) in crystals and glasses by an inelastic mechanism.

In a certain interval, the concentration of radiation-induced optical centers in an oxygen sublattice is proportional to the absorbed energy. Consequently, the intensity of the corresponding optical absorption band or photoluminescence can be used as a dosimetric parameter if the optical sensors are stable with respect to the effect of light and temperature (up to 400°C) and also do not overlap in spectra with other centers and do not convert into other centers. For such requirements, the material must be either free of impurities down to 10^{-4}% or contain only one optically active impurity, the change of whose valence can be used to determine the absorbed dose of ionizing radiation. In addition, it is desirable that high-temperature annealing of irradiated samples would restore the initial optical spectra and the material can be fused repeatedly. Such a material turned out to be germanium-doped SiO_2 glass, which is applicable as a solid dosimeter for determining the absorbed dose of neutron and γ radiation [2].

Investigations [3] have shown that most quartz glasses acquire color under the action of ^{60}Co γ radiation. This can be used in dosimetry. The intensity of the optical absorption at wavelength $\lambda = 550$ nm increases almost linearly with increasing absorbed dose of γ radiation up to dose 10^5 Gy, which attests to the possibility of using KI quartz glass for γ-radiation dosimetry. However, KGS and KSIII glasses acquire almost no color and remain transparent. Consequently, the band at 215 nm may be better as the working band because it is related with the formation of oxygen vacancies (structural defects designated in the literature as E_1'-centers) and has a wider dose range [4–7]. The absorbed dose of the γ component of reactor radiation can be determined by the increase of the concentration of oxygen vacancies and the flux of fast neutrons – according to the increase

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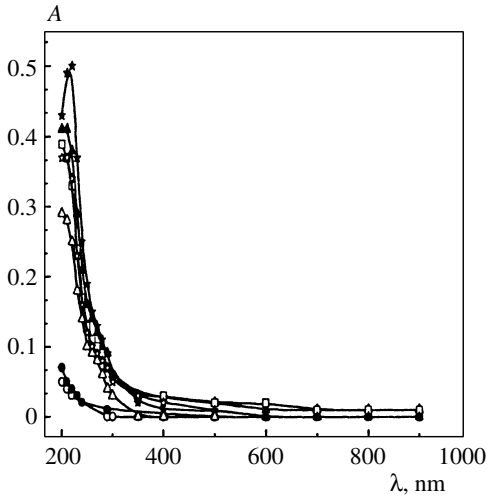

Fig. 1. Absorption spectra of pure SiO_2, irradiated for 3 (●), 30 (▵), 54 (▲), 64 (□), 90 (✲), and 120 h (✶) in water of a stopped reactor; ○) unirradiated reference substance.

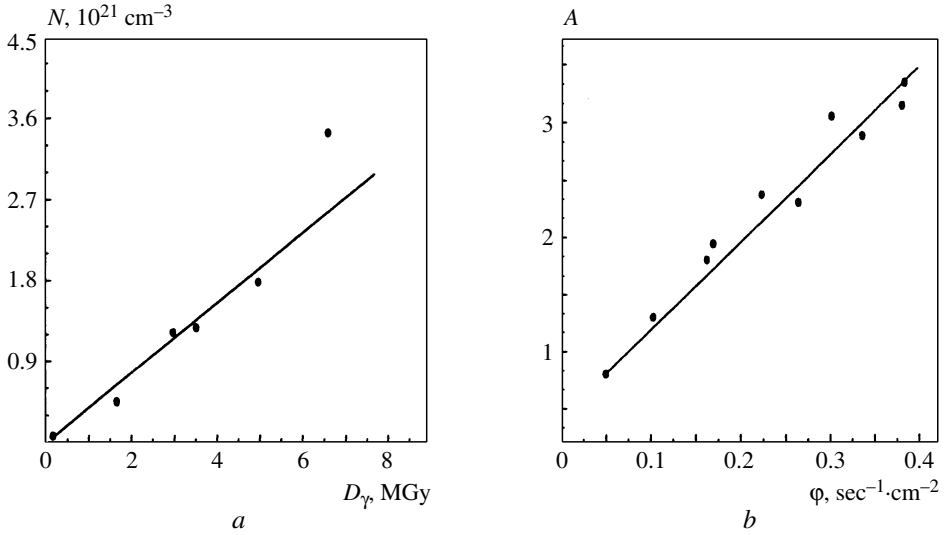

Fig. 2. Concentration of E_1'-centers in SiO_2 samples versus the γ-ray dose from a stopped reactor (*a*) and optical absorption of pure glass at wavelength λ = 215 nm versus the neutron flux density in the channels in the core of a working reactor with short-time irradiation (*b*).

in the content of silicon vacancies in ultrapure quartz glasses. The separate dosimetry of reactor-radiation components is a topical problem in connection with the switching the research VVR-SM reactor to low-enrichment fuel.

The purpose of the present work is to investigate the dosimetric characteristics of pure fused quartz to determine the intensity of γ radiation in the presence of fluxes of fast neutrons with energy above 0.1 MeV in the core and after the reactor is stopped.

Experimental Part. Dosimetric investigations of ultrapure quartz glass were performed using spectrometric optical absorption and photoluminescence. The SiO_2 absorption spectra were measured in the range 200–900 nm with a

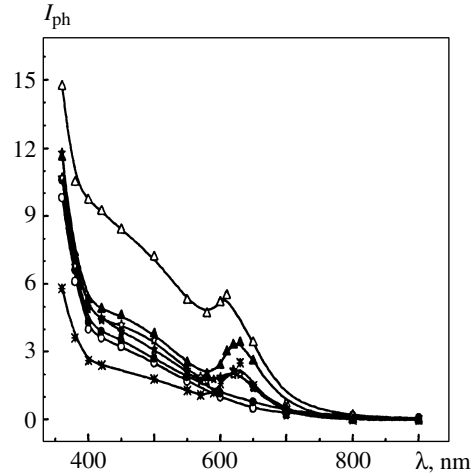

Fig. 3. Photoluminescence of pure SiO_2 under laser excitation at wavelength 337 nm, irradiated for 3 (●), 30 (△), 54 (▲), 64 (✲), 90 (✶), and 120 h (✳) in water of a stopped reactor; ○) unirradiated reference sample.

Specord M-40 spectrophotometer (Germany), the photoluminescence spectra were measured with laser excitation at wavelength 337 nm (LGI-21 nitrogen laser, Russia) in the range 300–900 nm using a SPM-2 monochromater (Germany). The samples consisted of polished wafers of pure fused quartz, containing uncontrollable impurities (less than 10^{-4} mass%). The possibility of the formation of known pairs of defects in an oxygen sublattice was investigated: E_1'-centers (absorption band 215 nm), associated with the formation of an oxygen vacancy, and displaced oxygen atoms, called nonbridge oxygen (photoluminescence band at 620–640 nm excited at wavelength 270 nm) under the action of γ radiation with energy 0.2–3 MeV from a stopped reactor.

Ten vertical channels in the VVR-SM core were chosen. Some of these channels were adjacent to fuel assemblies with different degrees of fuel burnup, and the remaining channels were near the beryllium reflector. A nickel threshold detector, which is insensitive to γ radiation, was used to determine the flux of neutrons with energy >1 MeV. To obtain a map of the flux distribution, ten samples of SiO_2 glass were loaded simultaneously into ten channels and irradiated for 2 h together with samples of nickel with the same mass. Next, irradiation for 3, 30, 54, 64, 90, and 120 h was conducted in vertical channels in the core of a stopped reactor, which were placed next to the fuel assemblies. A period of time where almost a constant current 10–20 nA was established in an ionization chamber, corresponding to the average γ flux intensity, was chosen.

To compare the measurements and determine the absorbed dose of γ radiation, ^{60}Co was also irradiated with a known dose rate of 7.65 Gy/sec. It was assumed that the concentration of induced centers in the oxygen sublattice with the same irradiation is proportional to the dose rate of the γ radiation from the reactor.

Results and Discussion. The absorption spectra of pure SiO_2 were measured before and after irradiation. Figure 1 shows that the absorption spectrum of an irradiated sample differs little from the reference sample. UV absorption is observed at 215 nm, which is associated with an electron captured by triply coordinated silicon near a bridge-oxygen vacancy. The γ irradiation produces the vacancy. As the duration of irradiation is increased to 30 h, the intensity of the absorption band increases sharply and continues to grow under further γ irradiation for 64, 90, and 120 h.

The comparison sample of ^{60}Co was irradiated for 54 h. The dose $1.4 \cdot 10^6$ Gy was accumulated; it corresponds to E_1'-center concentration $1.4 \cdot 10^{20}$ cm^{-3}. A simple proportion was used to estimate the dose rate of γ radiation from the stopped reactor as ~15 Gy/sec (correspondingly, the current of the ionization chamber was 10 nA). A monotonic dose dependence of the optical absorption band at 215 nm was obtained in the experimental range of irradiation times (Fig. 2). Comparison shows that the dose dependence of E_1'-centers is linear and makes it possible to establish a correlation between the fast-neutrons and γ radiation fluxes.

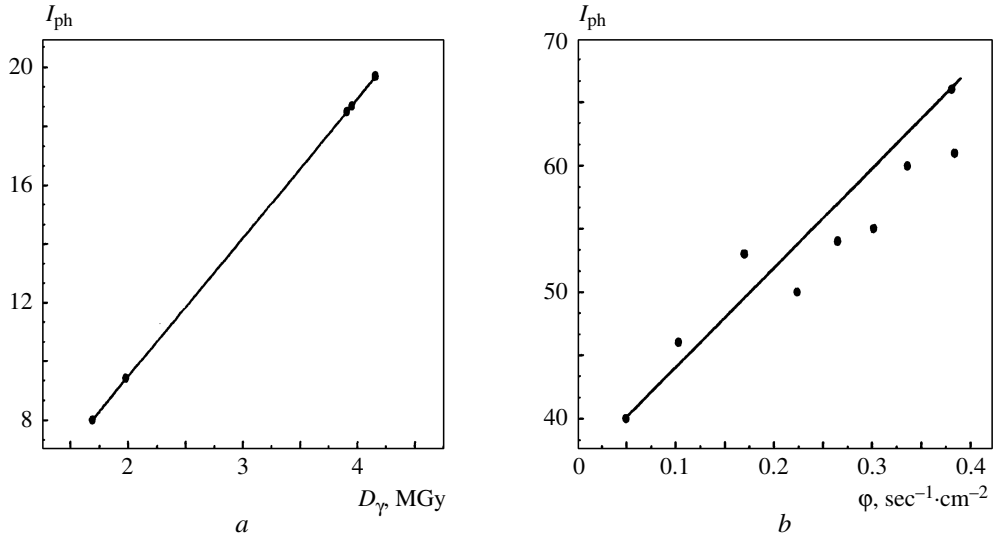

Fig. 4. Intensity of the photoluminescence band at 640 nm under laser excitation at 270 nm of pure SiO_2 glass versus the γ dose from a stopped reactor (*a*) and neutron flux density in the channels in the core of an operating reactor (*b*).

The photoluminescence spectra of samples of unirradiated and irradiated SiO_2 after the reactor is stopped was measured with laser excitation of a hole center of nonbridge oxygen (λ = 337 nm). It is known that unirradiated pure glass SiO_2 does not luminesce under UV excitation. For irradiation for 3 h in water, after the reactor is stopped, the photoluminescence spectrum of the unirradiated reference material is identical to that of the irradiated material; this dose is too low to produce optically detectable quantities of defects (Fig. 3). Irradiation with γ rays for 30 h produces photoluminescence bands at 400 and 500 nm, associated with oxygen-deficient centers, and the band 630 nm, which is due to nonbridge oxygen [5]. Further irradiation (54 h) changes the form of the photoluminescence spectrum, which approaches that of the reference spectrum; the peak becomes sharper and then becomes the same as the reference peak. Under irradiation for 64, 90, and 120 h, the photoluminescence spectrum is almost identical to the reference spectrum because the intensity of the 400–500 nm band decreases and the 630 nm band remains unchanged. After a high dose of irradiation, the photoluminescence at wavelength at 400–500 nm is even weaker than that in the reference material. Thus, for γ radiation in a reactor nonbridge oxygen forms as a result of a silicon vacancy as a pair defect for a E_1'-center associated with an oxygen vacancy.

Since laser radiation at 337 nm does not coincide with maximum excitation of photoluminescence of nonbridge oxygen at 270 nm, measurements were performed using a xenon lamp. The results of the photoluminescence of nonbridge oxygen and the dose dependences are shown in Fig. 4. Comparing the dose dependences of the growth of E_1'-centers (see Fig. 2) and nonbridge oxygen (see Fig. 4) shows that they coincide and are linear in the dose range 1–7 MGy; for this reason they can serve as dosimetric parameters for determining the γ-ray fluxes from a reactor. The dosimetric bands are stable with respect to light and temperatures above 400°C.

In summary, pure quartz glass can be used for separate dosimetry of γ-ray fluxes in the presence of fast neutrons.

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