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RADIATION RESISTANCE INVESTIGATION OF QUARTZ GLASS KU-1

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Results of investigation of the quartz window radiation resistance to fusion reactor conditions are described. The tests of a few selected materials have shown that the best for a visible spectrum region is quartz KU-1 with a large hydroxyl (OH) content. In comparison with other tested types of quartz it has a lowest loss of the transparency, the lowest radioluminescence(RL) intensity and in comparison with other materials (sapphire, cerium glass and spinel) preserved its properties in the widest region of the optical spectrum after neutron, electron and gamma irradiation.

1. Introduction

Despite of previous numerous investigations [1] a problem of the transparent material choice for fusion reactor conditions needs in additional study. Reasons for this are: (1). The expected neutron fluence for windows in diagnostic channels may reach of 10^{18}–10^{19} n/cm^2 at ionization dose of 10–100 MGy(Si) (below all dose units are given for Si) (2). Fusion reactor radiation conditions (neutron and gamma fluxes and the ratio between these fluxes) will differ from that of other sources (3). The choice must be done from materials of different characteristics.

Therefore the next main problems had to be solved: (i) The choice of the best material from those which traditionally are used in thermonuclear devices. (ii) The study of transparency dependence of the chosen material (or induced optical density) on fluxes, doses and type of irradiation. (iii) Radioluminescence (RL) of material measurement and comparison its intensity with the plasma visible radiation. (iv). Measurements the transparency and (RL) in dependence on the temperature, and search a possibility of thermal annealing for restoring material characteristics.

Below the results of investigations of some indicated problems which were accomplished in European, Japanese and Russian laboratories are described.

2. The choice of the window material

Materials selected for preliminary tests were three types of the quartz glass with different content of hydroxyl – KUVI-S (~10^{-4} % OH), KUVI-M (~0.01% OH) and KU-1 (~0.1% OH); sapphire, aluminum-magnesium spinel $MgAl_2O_4$ and cerium glass S-96. Samples of all these materials had diameter of 2 cm and thickness 0.8 cm.

Several radiation sources were used for tests [2]: ^{60}Co gamma source (dose rate $dD_\gamma/dt \leq 12$ Gy(Si)/s and dose $D_\gamma \leq 1$ MGy), pulse electron accelerators LU-40 in Kharkov (pulse duration τ=2.9 μs, repetition frequency f=12.5-150 Hz, electron energy E_e=10 MeV, average dD_γ/dt = 0.15-1.0 kGy/s, $D_\gamma \leq 8$ MGy) and LU-50 in Sarov (τ=10 ns, f ≤ 2.4 kHz, E_e=50 MeV, average $dD_\gamma/dt \leq 30$ Gy/s, $D_\gamma \leq 700$ kGy); neutron generator SNEG-13, (neutron flux on samples $dF_n/dt \approx 3\text{x}10^{11}$ n/cm^{2}s at $dD_\gamma/dt \approx 0.15$ Gy/s); nuclear pool-type reactor (for $E_n > 100$ keV, $dF_n/dt \approx 2\text{x}10^{12}$n/cm^{2}s $D_\gamma \leq 2.5$ MGy). In all cases the temperature during and after irradiation was not higher than 30-50^{o}C and the relative transparency was measured after irradiation at room temperature.

The main results of these tests are as follows: (1). Transparency of all tested materials is very sensitive to all kinds of the radiation in a near ultraviolet spectral region; (2). Spinel and cerium glass totally lost transparency at the wavelength region λ < 400 nm and were excluded from further tests, also their transparencies at λ > 400 nm are almost not sensitive to irradiation; (3). Sapphire is very sensitive in all visible region (Fig. 1) and may be used only in an infrared spectral region [3]; (4). The difference in the spectral transparency for three types of quartz glasses is rather weak and all they were left for additional tests.

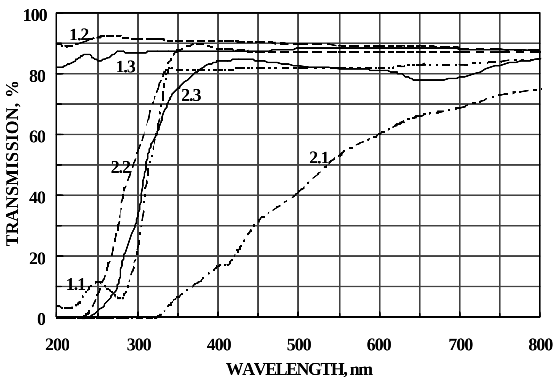

Fig.1. Optical transmission spectra of sapphire (curves 1.1 and 2.1), quartz glass KU-1 (1.2 and 2.2) and glass KUVI-S (1.3 and 2.3) before (1.1, 1.2 and 1.3) and after (2.1, 2.2 and 2.3) irradiation to nuclear reactor neutron fluence of 10^{18} n/cm^2 and g-dose of 45 MGy [2].

3. Quartz glass optical density measurements

Irradiation of KU-1 samples in the nuclear reactor up to neutron fluence of $6\text{x}10^{19}$ n/cm^2 (total ionization dose of ~3 GGy) and with electrons (up to 10 MGy) have shown that the optical density at λ>350 nm practically does not changed with exception of a week absorption band at $\lambda \approx 620$ nm [4]. From the irradiation start the optical density at λ < 350 nm increases gradually after the dose of ~10 kGy and then, beginning from ~1MGy, either saturates or even starts to decrease (Fig.2). Unfortunately time variation was investigated only under γ-irradiation and only at λ≅215 nm [5,6], so far as all irradiation effects at this wavelength are most pronounced, and at λ>400 nm are almost absent [7]. After the irradiation stops the optical density in the short wavelength region abruptly decreases and for λ < 350 nm and then shows less values than measured *in situ*.

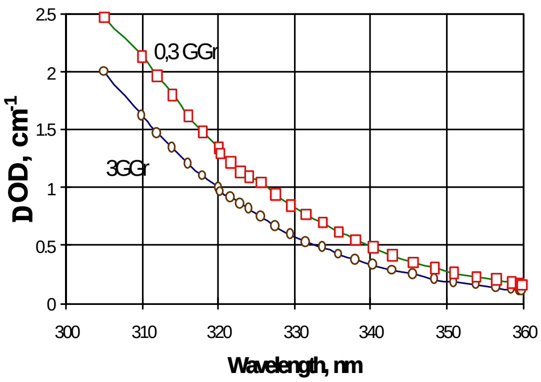

Fig. 2. Radiation induced optical) spectra in the short wavelength region after irradiation to different total ionization doses [4].

Such results were obtained for the optical transparency dependence on the neutron flux and gamma dose rate (Fig.3): the dose rate practically has no effects in the wavelength region of λ > 400 nm, but at shorter λ the larger flux leads to the lower transparency at equal fluences.

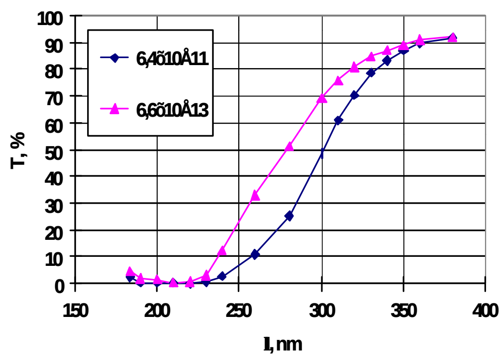

Fig.3 Optical spectra in the UV region for two samples KU-1 with thickness of 2 mm [8]. The values of the neutron flux under irradiation are given in frame in units of [n/cm^2 c].

Temperature influence on the optical density during irradiation was studied mainly for λ≅215 nm [5,6]. At λ ≥ 350 nm its influence is very week [6]. The higher is the KU-1 sample temperature the faster increases the optical density during irradiation and the lower its value is in a saturation region at equal gamma dose and dose rate. This effect indicates the possibility to restore the quartz transparency by heating. The direct check has shown that the transparency of irradiated to neutron fluence of (1-6)x10^{19} n/cm^2 KU-1 samples really was restored after gradual heating up to 700-800^{o}C [8].

4. Radioluminescence of quartz glass KU-1.

It is necessary to be sure that the luminescence of the chosen window material under neutron and gamma irradiation will not prevent any optical measurements. Therefore after the candidate materials where defined, the study of their luminescence under neutron, electron and gamma radiation was started. The main difficulties of such experiments consist in requirement of measurements *in situ* and rather low luminescence intensity. For these measurements the radiation source should permit the access to the tested samples without introducing addition radiation sources. This request excludes the use of stationary nuclear reactors. For low luminescence intensity measurement it is necessary to have either very sensitive registration system or high power radiation source. Actually in experiments were used ^{60}Co gamma source and neutron generator with sensitive detection system [9, 10], electron beam from Van de Graaff accelerator with dose rate of 700 Gy/s [11] and pulse nuclear reactor with an average dose rate of 155 kGy/s [12].

To compare luminescence of different materials the of sapphire, anhydroguide quartz (analog of KUVI-S) and KU-1 were tested under electron beam [11]. Anhydroguide quartz emission has strongly pronounced maximum at λ≈450 nm (this maximum was observed also under X-ray irradiation [5]), which is doubled after increasing the absorbed dose from 5 to 20 MGy, weak maximum at λ≈650 nm and maximum in the short wavelength region. KU-1 glass emission has maximum around 450 nm and

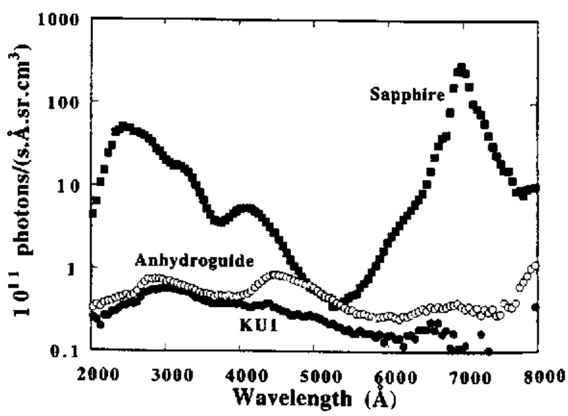

Fig.4. RL spectra for two types of SiO_2 and Union Carbide UV grade sapphire after 2 h irradiation with electrons at 15^{o}C, 700 Gy/s, and. ~10^{-10} dpa/s [11]

weakly pronounced maximum at 650 nm. Common for both glasses is increased emission at λ≤300 nm. Results of absolute measurements of the RL intensity for tested materials are shown in Fig. 4.

Measurements at different temperature *T* have shown that KU-1 light emission does not depend on *T*. Emission of anhydroguide quartz at ~450 nm decreased with *T* and disappears at ~350^{o}C. The spectrum both of them, corrected for the self-absorption, looks like the Cherenkov emission (λ^{-2}).

Two identical samples of anhydroguide SiO_2 were irradiated with D-T neutrons and ^{60}Co gammas. Luminescence results are shown in Fig.5 [9]. The difference in these spectra consists not only in emission intensity but also in existence in the lower spectrum of peak at ~630 nm that is absent in other spectrum. Data adduced in Table 1 [9,10] allow to compare luminescence efficiencies for different materials under irradiation with D-T neutrons and ^{60}Co gamma quanta. These data show that from the point of view of minimal RL intensity in a visible spectral region the best materials are silica glasses and the best of these - the quartz glass KU-1.

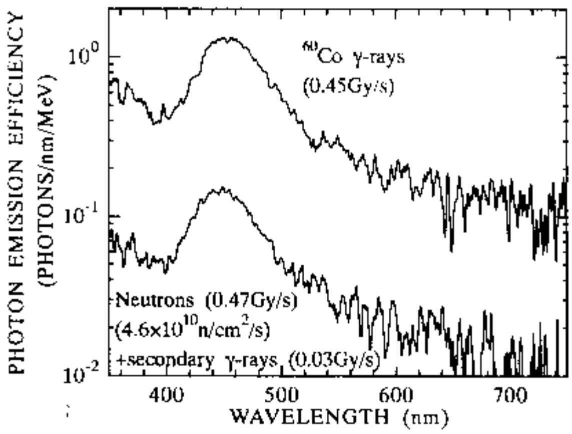

Fig.5. Spectra of photons emitted from high-purity SiO_2 in DT-neutron and ^{60}Co gamma ray irradiation experiments at room temperature [9,10]

RL measurements using pulse nuclear reactor have some advantages as compared with other sources: ratio of neutron and gamma fluxes is near to expected for the fusion reactor and pulse character of irradiation allows to avoid of possible influence of the absorbed dose and also to measure in one pulse luminescence dependence on irradiation flux. Luminescence spectrum for this case is shown in Fig.6. Measurements of the temperature and preliminary irradiation influence have shown that temperature increasing from 18 to 100 ^{o}C does not changed luminescence intensity and after preliminary irradiation up to 6x10^{18} n/cm^2 emission intensity at $\lambda\approx$620-670 nm became ~2.5. times larger, but after further increasing neutron fluence up to 6x10^{19} n/cm^2 intensity decreased by ~20%. Other part of the spectrum remains unchanged. Results of KU-1 RL measurement at different radiation sources are given in Table 2.

Comparison of estimated values of plasma continuum emission and windows RL for expected ITER conditions has shown [12, 13] that both quartz types luminescence intensity will not prevent from plasma diagnostics but sapphire emission intensity is near the brink of permissible.

5. Discussion

One can see from Fig.1 that mostly radiation induced absorption is in the UV-region of spectra, and includes absorption bands at 215 and 260 nm. The 215 nm band is observed in all silica glasses and is caused by E' centers, that is formed by trapping a radiolitic hole in Si-Si bond at the site of oxygen vacancy. The 260-nm band is more intense for OH- free silica glass (KUVI-S). It has been attributed to oxygen-related defects, such as non-bridging oxygen hole centers (NBOHC). Comparison of influence of reactor's neutron and electron irradiation [2,5] on optical absorption leads to conclusion, that silica glass with high concentration of OH group (wet glass) is more radiation resistant to compare with dry glass.

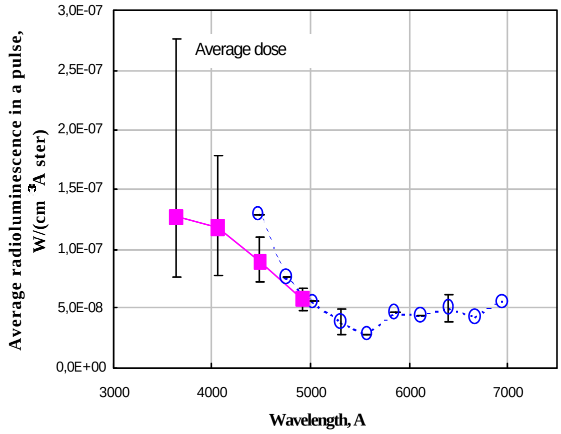

Fig.6. RL spectrum of KU-1 glass under irradiation in the pulse nuclear reactor. Different signs refers to different measurement cycles [12].

From the spectra of RL under electron excitation one can distinguish bands in SiO_2 at 290, 450 and 650 nm [5]. The nature of indicated bands were discussed in numerous review papers. Most probable the 650 nm band is related to NBOHC, the 450 nm is related to the center of the family of the so-called oxygen-deficiency centers (ODC's) - specifically to di-coordinated silicon -ODC(II). The band at 290 nm also related to ODC(II) - oxygen di-vacancy. It was found that relative intensities of bands very sensitive to the source of irradiation [10,11]. Under electron excitation at dD/dt = 700 Gy/s the intensity of different bands are comparable. In DT-neutron experiments dD_γ/dt=0.03 Gy/s the intensity of the 450 nm band is the lowest despite the simultaneous irradiation with neutrons at the dF_n/dt= 4.6x10^{10} n/cm^{2}s. Under irradiation only in ^{60}Co gamma ray source at higher excitation dD_γ/dt=0.45 Gy/s the intensity of this band increases by the order of magnitude. Thus, most of this luminescence comes from the defects (either intrinsic or newly produced) in electron excitation. From Table 1 one can see that in DT experiments the number emitted photons per unit of absorbed energy is the lowest for SiO_2 to compare with other potentially used materials for optical windows.

Table 1. *Number of emitted photons per unit absorbed energy in visible range (350-650 nm)*

Sample	Peak wavelength (nm)	D-T neutrons ++secondary γ-rays (photons/MeV)	^{60}Co γ-rays (photons/MeV)
Anhydroguide SiO_2	450	17±6	170±60
KU-1 quartz glass	650	14±5	-
Ge doped silica	390	83±30	410±140
Sapphire	410	2500±1000	27000±11000

Table 2. *RL in the spectral region of 350-650 nm of KU-1 irradiated from different radiation sources.*

Radiation Source	Operating condition	Ionizing dose rate, (Gy/s)	Displacement dose rate, (dpa/s)	Photons number at 350-650 nm per 1MeV of total absorbed energy	Ratio of radiated energy in a region of 350-650 nm to absorbed energy
Electron accelerator	stationary	700	$\sim 10^{-10}$	80*)	$\sim 2x10^{-4}$
Neutron generator	stationary	0.5	$\sim 10^{-10}$	14*)	$\sim 4x10^{-5}$
Neutron generator	pulse, FWHM 60µs	in one pulse at an average155,000	$\sim 4x10^{-5}$	4*)	$\sim 1x10^{-5}$

*) measurement accuracy 30-50 %

Comparison of the dose rate dependencies of optical properties of the KU-1 glass irradiated with pulse electron beam and continuous neutron bombardment indicates that formation of the stable optical centers involves the transient processes, that are dependent on the type of excitation and density of energy deposition.

The pulsed dose rate at the electron irradiation comes to instant energy deposition of 2.2 MGy/s [5] in material that leads to creation the high density of defects. Post irradiation evolution and interaction between these transient defects facilitate creation of the state optical centers. On the other hand, the continuous irradiation even at the highest neutron dose rate of $6.6x10^{13}$ n/cm^2 s produces transient defects with large spatial separation excluding their mutual interaction and nonlinear effect of stable center formation.

6. Conclusion

These investigations were undertaken with the aim to find and test a suitable material for ITER diagnostic systems. Obtained data show that in ITER radiation conditions all types of quartz glass may be used in the spectral region of 350-2000 nm. Glass KU-1 can stand at the higher neutron fluence and has a lower RL intensity than anhydroguide or high purity glass.

At the same time there are some questions which are left without answers. (i) Expected in a fusion reactor the irradiation temporal regime can not be reproduced in test process. (ii) Almost all data on optical absorption were attained after irradiation. Optical density in the short wavelength region during and after irradiation may be different. (iii) The ratio between neutron and gamma radiation in a fusion reactor will distinguish from this one used in experiments. But it is very unlikely that all these circumstances can change the main result of present studies – the quartz glass KU-1 is the most suitable material for diagnostic windows.

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