Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 362 (2007) 222–226
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Radiation-optical characteristics of quartz glass and sapphire

A.Kh. Islamov, E.M. Ibragimova *, I. Nuritdinov

Institute of Nuclear Physics AS RUz, pos, Ulugbek, 100214 Tashkent, Uzbekistan

Abstract

The systems registering plasma radiation comprise optical elements, which may distort a diagnostic signal if they are not radiation resistant. The work was aimed at experimental study of optical absorption and γ-induced luminescence of KU-1 quartz glass and sapphire crystals in the range of 200–900 nm, which were irradiated in the ^{60}Co γ-ray source of 6.7 Gy s^{-1} up to the dose of 10^8 Gy and in the water cooled nuclear reactor of WWR-SM type with neutron fluences of 10^{17}–10^{20} cm^{-2}, in order to imitate plasma effect. The transparency window (minimal optical losses) and the intrinsic luminescence bands were determined in the studied materials. The advantages of KU-1 type glass over sapphire as optical elements at 300–350 K have been demonstrated.

PACS: 29.40.Mc; 61.80.Fe; 70

1. Introduction

Optical radiation within 170–2000 nm in various accelerators and international thermonuclear experimental reactor (ITER) condition is used for plasma diagnostics. So, optical elements of a plasma registration system should transmit a light signal with minimal loss and distortion, i.e. be radiation resistant [1].

Irradiation with mixed fluxes of nuclear particles and gamma-rays can imitate plasma effect. Although optical elements of plasma diagnostic systems do not undergo such a high irradiation flux as in the nuclear reactor, the obtained data are important for evaluation their radiation-optical resistance and correct placing in the protected diagnostic channels. This work was aimed at studying the influence of mixed neutron and gamma-ray fluxes on the optical characteristics of pure quartz glass and sapphire crystals to determine their applicability as optical elements for plasma diagnostics.

2. Experiment

Samples of pure quartz glass of KU-1 type (contains 8.5×10^{-2} wt% OH) and pure sapphire crystals (basing neutron activation analysis, the impurity contents were determined as Cr – 4×10^{-5} wt%, Mo – 5.5×10^{-7} wt%, Au – 3.4×10^{-8} wt%, no Fe was found) were obtained from State Optical Institute (St. Petersburg, Russia) and having a high optical stability against radiation, were irradiated in the Tashkent water cooled reactor of WWR-SM type of 10 MW at the temperature of 350 K with integral

* Corresponding author. Fax: +998 71 1503080.
E-mail addresses: islamov-s@mail.ru (A.Kh. Islamov), ibragimova@inp.uz (E.M. Ibragimova).

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.01.047

fluxes of neutrons within 10^{17}–10^{20} cm^{-2} accompanied by gamma-radiation of a wide energy spectrum. The flux density for neutrons of energy above 2.4 MeV, as determined with Ni monitor, came to 3×10^{13} cm^{-2} s^{-1}. For estimating the contribution of gamma-radiation of the reactor into the optical characteristics change, the samples were also irradiated in ^{60}Co gamma-source of the energies 1.17 and 1.32 MeV at 300 K in the dose range $D \sim 10^3$–10^8 Gy at the dose rate of 6.7 Gy s^{-1}.

Spectra of optical absorption (OA) and photoluminescence (PL) prior and after irradiations were taken at 300 K with fluorescence spectrophotometer MPF-2A (Hitachi, Japan) and 'Specord-M40' (Carl Zeiss Jena, Germany) in the wavelength region of 200–900 nm. Gamma-luminescence (GL) spectra were registered at spectrometer SPM-2 (Carl Zeiss Jena, Germany) with PMT-100 in the same spectral region at 300 K and the ^{60}Co intensity of 4.4 Gy s^{-1}.

3. Results and discussion

3.1. KU-1 glass

No band occurs in the OA and GL of non-irradiated samples. After ^{60}Co gamma-irradiation to $D = 10^4$ Gy there appears OA band at 215 nm (Fig. 1), ascertained to radiation induced E′-centers (≡Si•) [2,3], which grows up to the dose 5×10^6 Gy and saturates. At higher gamma-doses $D \geqslant 10^6$ Gy and mixed (n, γ)-irradiation to 10^{17} cm^{-2} the long wave smooth descending of this OA band is due to overlapping of a few bands (Fig. 2). The OA band at 245–250 nm and PL at 280 nm are known [4] to relate with oxygen vacancy defect (≡Si − Si≡). And the OA band at 270–280 nm is ascribed to E′(Ge)-centers at Ge impurity [5,6] or non-bridging oxygen hall center (NBOHC), the latter being also characterized by OA band at 620–630 nm and PL at 660 nm [2,7]. To clear up the nature of the OA band at 270–280 nm, we studied PL spectra of non-irradiated samples and gamma-irradiated to $D = 10^7$ Gy at excitation in the interval of 200–350 nm. The PL bands at 282 and 465 nm are excited in 250 nm, and the band 650 nm is excited in 280 nm, as in [2,7,8], i.e. they have nothing to do with Ge impurity.

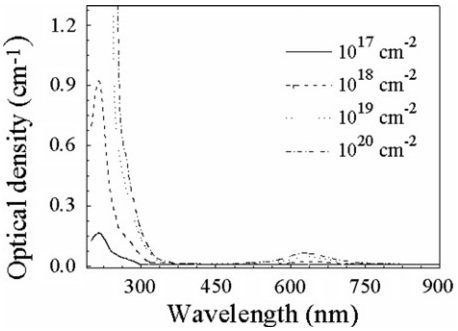

Fig. 2. Absorption spectra of KU-1 glass after (n, γ)-irradiation in the reactor to the fluences indicated.

There appears only one GL band at 460 nm after gamma-irradiation to $D \geqslant 10^7$ Gy, while after the reactor irradiation within 10^{17}–10^{20} cm^{-2} (Fig. 3) there is also a weak band at 650 nm ascertained to NBOHC centers. The both bands grow with the fluence up to 10^{19} cm^{-2} and then saturate. Since the intensity of GL at 460 nm after the gamma-dose of 10^8 Gy equals to that after neutron + gamma irradiation to $\Phi = 10^{17}$ cm^{-2} (Fig. 3), one can conclude about prevailing gamma-contribution into excitation of this band.

The results obtained for KU-1 glass confirm the suggestion made in [8], that E′-centers are generated

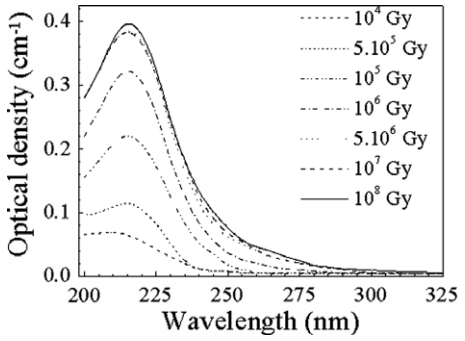

Fig. 1. Absorption spectra of KU-1 glass after ^{60}Co γ-irradiation to the doses indicated.

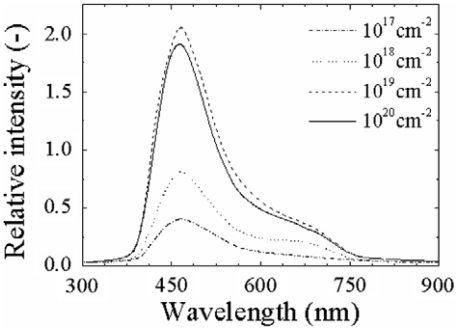

Fig. 3. Gamma-luminescence spectra of KU-1 glass after (n, γ)-irradiation in the reactor to the fluences indicated.

in hydrogen containing glasses under ^{60}Co-gamma-irradiation to a dose $>10^4$ Gy, and at higher doses $\geqslant 5\times10^6$ Gy there also appear NBOHC as the result of rupture of hydrogen bonds in ≡Si–H and ≡Si–OH, respectively. Perhaps hydrogen transits into molecular bound state in the glass network. Oxygen single vacancies (≡Si − Si≡) are generated at doses $\geqslant 5\times10^6$ Gy by means of migration of E′-centers and their interaction by the reaction:

$$\equiv Si^{\cdot} + {}^{\cdot}Si\equiv + \gamma \rightarrow \equiv Si - Si\equiv \quad (1)$$

GL band at 460 nm observed in KU-1 after irradiations to gamma $\rightarrow 5\times10^7$ Gy and neutron-gamma fluence of 10^{17}–10^{20} cm^{-2} (Fig. 3) can also be induced under 2 MeV-electron beam [9], neutrons of DT-generator [10] and pulse nuclear reactor [11]. Authors of [9] ascribe the luminescence band at 450 nm to chlorine impurity at low hydrogen content, and in [10] it is explained by the self-trapped excitons (STE) at oxygen vacancies. But, there is PL band at 390 nm in the chlorine doped glasses, which is due to the centers (≡Si − Cl Cl − Si≡) [12], which does not occur in Fig. 3 and [9]. On the other hand, the radioactive decay of STE was observed at 500–530 nm in pure SiO_2 glass by [13,14]. Therefore the GL band at 460 nm can be ascribed to e–h-recombination at single oxygen vacancies, the concentration of which grows with the neutron fluency. The following saturation of this band may be due to transformation of the single vacancies into divacancies and other complexes. The mechanism for gamma-luminescence at 460 nm is the following. Effect of ^{60}Co gamma-radiation on the neutron + gamma-induced centers (≡Si − Si≡) results in their ionization and transformation into (≡Si$^{\cdot}$ − $^{+}$Si≡) localizing a non-paired charge. The following radiative recombination of electron on this center gives rise to the GL band at 460 nm by the reaction:

$$\begin{aligned}\equiv Si - \equiv Si + \gamma &\rightarrow \equiv Si^{\cdot} - {}^{+}Si\equiv - e\\ &\rightarrow [(\equiv Si^{\cdot} - {}^{+}\equiv Si) + e]^{*}\\ &\rightarrow \equiv Si - \equiv Si + h\nu\ (460\ nm) \quad (2)\end{aligned}$$

3.2. Sapphire

The authors of [15] observed OA bands at 205 and 308 nm and also a non-elementary band at 403 nm after γ-irradiation of synthetic sapphire to the dose of 5×10^3 Gy, all bands growing up to 1.5×10^5 Gy, which were ascribed to lattice defects. No band was observed in OA spectrum for both non-irradiated sapphire crystals and those after ^{60}Co-gamma-irradiation even to the dose of 10^8 Gy. The GL spectrum comprises 300–330 nm (lattice defect) and 695 nm (Cr^{3+}), the intensities of the both decrease with the dose to 10^8 Gy. Besides, since the dose 10^7 Gy there appears GL band at 420 nm (Fig. 4) related with F- (means farben = color) centers [16], which increases with the gamma-dose. In our opinion, the gamma-irradiation results in decreasing of the amount of the existing F^{2+} (charged farben center) and Cr^{3+}-centers by means of their transformation into F^{+} and Cr^{4+} centers, respectively. And GL at 420 nm is due to electron trapping at F^{+}-center by reaction:

$$F^{+} + e^{-} \rightarrow F^{*} \rightarrow F + h\nu\ (420\ nm)$$

Thus the gamma-irradiation up to 10^8 Gy does not create additional structure defects in sapphire, but just transforms the defects and impurities generated under the crystal growing as in the case of [15].

Unlike ^{60}Co gamma-radiation, the reactor (n, γ) irradiation induces a number of OA bands at 205, 230, 257, 300, 355 and 450 nm (Fig. 5), which grow with the fluency up to 10^{20} cm^{-2}. According to [17,18], the band at 205 nm is due to F-center, and 230 and 255 nm are to F^{+}-center (oxygen vacancy trapping one and two electrons, respectively). Besides effective generation of oxygen vacancy F- and F^{+}-centers under neutron fluency growth, there are also formed interstitial ions (Al_i^{+}) responsible for the OA band at 305 nm and PL at 510 nm [19]. The observed OA bands at 355 and 450 nm are related with formation of F-aggregate centers, consisting of two oxygen vacancies with different charges [20,21].

Under ^{60}Co gamma-excitation of the sapphire crystals pre-irradiated in the reactor, the intensity

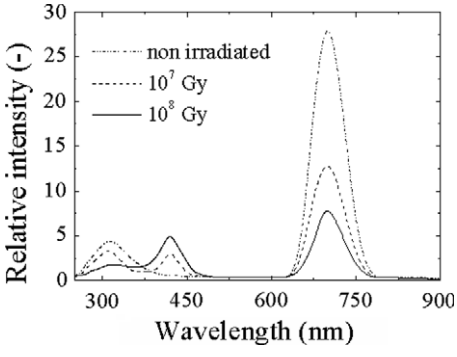

Fig. 4. Gamma-luminescence spectra of sapphire crystals prior and after ^{60}Co γ-irradiation to the doses indicated.

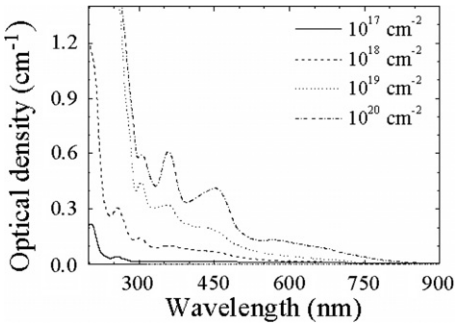

Fig. 5. Absorption spectra of sapphire crystals after (n,γ)-irradiation in the reactor to the fluences indicated.

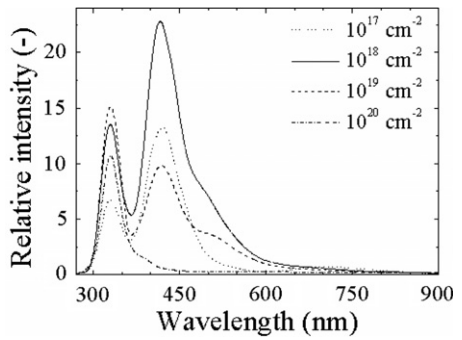

Fig. 6. Gamma-luminescence spectra of sapphire crystals after (n,γ)-irradiation in the reactor to the fluences indicated.

of GL bands at 330 nm (F^+-center) and 420 nm (F-center) increases up to the fluency of 10^{18} cm^{-2} (Fig. 6), then at higher fluences the band at 330 nm descends slower than that at 420 nm. The decrease of these GL band intensities may be due to its partial reabsorption by the color centers induced in this spectral range (Fig. 5), which related with F-aggregate centers. Thus, at mixed n-γ-irradiation of sapphire single oxygen vacancies are generated effectively, that promotes production of interstitial cation defects Al_i^+ and at higher fluences F-aggregate centers.

4. Conclusions

The obtained data on the radiation induced changes in the optical absorption and γ-luminescence of the studied materials allow one to make a choice and determine the proper places for optical windows and lenses in the radiation environment.

Sapphire can be used as highly transparent windows in the γ-radiation fields up to doses of 10^8 Gy, as no optical absorption band is induced. While in KU-1 glass UV color centers appear in the range of 200–300 nm, and the optical losses in the range of 340–900 nm are negligible. However, when registering plasma radiation, one should take into account the irradiation induced luminescence bands of sapphire at 330, 420 and 695 nm in accordance with an absorbed dose, especially the dominating band at 420 nm, the intensity of which at the dose of 10^8 Gy is ~20 times more than that of 460 nm in the glass. Therefore, a window made from KU-1 glass contributes less distortion in a diagnostic signal, than sapphire.

In the case of mixed neutron-gamma fields in the fluence range of 10^{17}–10^{20} cm^{-2} the KU-1 glass preserves two transparency gaps within 350–550 and 750–900 nm, but a diagnostic signal will be distorted by the GL band at 460 nm. Sapphire has a wide transparency gap within 300–900 nm at the fluence of 10^{17} cm^{-2}, which narrows to 600–900 nm at 10^{18} cm^{-2}, and the maximal distortion of a signal will be at bands 420 and 330 nm in the fluence range of 10^{17}–10^{19} cm^{-2}.

Acknowledgements

The authors are grateful to Dr Amosov A.V. for kind providing samples. The work is supported by grants from CST Uzbekistan F2.1.2, F2.1.17 and NATO CBP.EAP.CLG.981765.

References

[1] K. Vukolov, B. Levin, Fus. Eng. Design 66-68 (2003) 861.
[2] D.L. Griscom, in: Proceedings of the XXXII Frequency Control Symposium, Atlantic City, 1979, p. 98.
[3] A.V. Amosov, Phys. Chem. Glass. 6 (1980) 218.
[4] A.V. Amosov, G.T. Petrovski, DAN SSSR 268 (1983) 66.
[5] J.H. Mackey, J. Chem. Phys. 39 (1963) 74.
[6] A.Kh. Islamov, I. Nuritdinov, E.M. Ibragimova, A.V. Amosov, W. Cooke, in: IEEE Nuclear Science Symposium and Medical Imaging Conference, Costa-Rica, 2005, p. 14.
[7] L.N. Skuja, A.R. Silin, Phys. Status Solidi 56 (1979) k11.
[8] G.B. Blinkova, Sh.A. Vakhidov, A.Kh. Islamov, I. Nuritdinov, Kh.A. Khaydarova, Phys. Chem. Glass. 14 (1988) 494.
[9] A. Morono, E.R. Hodgson, J. Nucl. Mater. 224 (1995) 216, also 249 (1997) 128.
[10] F. Sato, T. Iida, Y. Oyama, F. Maekawa, Y. Ikeda, J. Nucl. Mater. 258–263 (1998) 1897.
[11] A. Gorshkov, D. Orlinski, V. Sannikov, K. Vukolov, S. Goncharov, Yu. Sadovnikov, A. Kirillov, J. Nucl. Mater. 273 (1999) 271.
[12] A.V. Abramov, N.S. Karpychev, V.O. Sokolov, Phys. Chem. Glass. 16 (1990) 769.
[13] A.N. Trukhin, A.E. Plaudis, Solid State Phys. 21 (1979) 1109.

[14] D.W. Cooke, B.L. Bennett, E.N. Farum, D.E. Thomas, A.M. Porits, J. Nucl. Mater. 255 (1998) 180.
[15] L. Fuks, C. Degueldre, J. Nucl. Mater. 280 (2000) 360.
[16] K.H. Lee, J.H. Crawford, Phys. Rev. B19 (1979) 3217.
[17] K.H. Lee, J.H. Crawford, Phys. Rev. 15 (1977) 4065.
[18] T.J. Turner, J.H. Crawford, Phys. Rev. B13 (1976) 1735.
[19] B.D. Evans, M. Stapelbrock, Phys. Rev. B18 (1978) 7098.
[20] P.F. Gulchuk, L.A. Litvinov, P.V. Peterenko, E.A. Chernina, J. Appl. Spectr. 63 (1985) 132.
[21] I.Kh. Abdukadirova, J. Appl. Spectr. 70 (2003) 788.