Remittances Review
June, 2023
Volume: 8, No: 4, pp. 1206-1212
ISSN: 2059-6588 (Print) | ISSN 2059-6596 (Online)
remittancesreview.com

Received: 11 November 2022 Accepted: 28 March, 2023
DOI: https://doi.org/10.33182/rr.v8i4.84

Change of properties of medicine optical materials under gamma-irradiation

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Abstract

The effect of irradiation with 60Co gamma-quanta in the dose range of 104–106 R on the optical absorption of SiO2 glasses containing BaO or PbO. After such irradiation the glass acquires nice golden or brown colors, absorbs actively the UV-radiation, but transmits yellow-orange light, does not have residual radioactivity and gets sterilized. The obtained results allow us to offer the SiO2 glasses containing barium and led oxides for personal protection of eyes from UV-radiation in medicine.

Keywords: *SiO2 glasses containing oxides of barium or lead, absorption spectra, color centers.*

Introduction

In the field of nuclear energy, space technology, medical tomography, radiation therapy, non-destructive testing, the operation of optical and optoelectronic devices is often carried out under conditions of increased radiation load. A change in the optical properties of oxide glasses under the action of ionizing radiation can significantly affect the operating parameters of equipment in which many elements, such as light guides, lenses, etc., are made of glass. It is known that defects can form in glasses under irradiation [1, 2]. It is known that high-energy ionizing radiation creates not only point structural defects, but also dimensional defects, such as dislocations and cascades of atomic displacements [3]. Theoretical calculations of displacement cascades and particle tracks in silicon or quartz crystals have shown that their sizes reach 100 nm, and then, as a result of relaxation, their sizes and shapes change. We also showed that, as a result of gamma irradiation, color centers are formed in crystal glass with 24% PbO, the optical absorption of which indicates the accumulation of charge carriers at the glass-lead-containing phase interface [4].

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For example, using X-ray diffraction analysis, we previously detected BaO_2 nanocrystallites ~28 nm in size and ~15 nm α-SiO_2 nanocrystals, as well as amorphous BaO particles 0.78 nm in unirradiated SiO_2 glass samples with Ba content of 26.8 + 0.2%. Under ^{60}Co gamma irradiation (~1.25 MeV) with dose of 10^8 R, as a result of phase transitions, tridymite nanocrystallites from cristobalite and $BaSiO_3$ nanocrystallites are formed due to the dissolution of BaO_2 nanoparticles [5]. Also, we [6] studied the microhardness and photoluminescence (PL) spectra upon laser excitation at 337 nm of industrial samples of SiO_2 glasses (UV windows, substrates with a $BaSiO_3$ film coating) irradiated with ^{60}Co gamma rays and a mixed flux of neutrons and gamma quanta of the reactor. Nanocrystalline phases were found in the initial samples. It is shown that phase transformations of SiO_2 cristobalite - tridymite, BaO-BaO_2, decomposition of $BaSiO_3$ and $BaCO_3$ occur during reactor irradiation, which causes a decrease in microhardness and quenching of PL.

Silicate glass is widely used as a chemically inert material for vials and ampoules containing liquid medicines. When sterilizing in γ-sources and especially in electron accelerators, it is important to know how the parameters of ionizing radiation change when passing through glass 1–2 mm thick in order to assess the quality of drug sterilization [7].

It is known that after X- or γ-irradiation, glasses of some brands darken in the violet region of the spectrum, while others are colored much weaker. Windows or tubes in UV sterilizers should not be painted, so as not to absorb UV radiation. The most suitable glass for this purpose is glass welded from highly pure SiO_2 ($<10^{-4}$ % impurities), which is not stained even by gamma quanta. At the same time, to protect the eyes of medical workers and patients, on the contrary, glass with strong absorption in the UV region is needed, where violet light filters are usually used, which are difficult to manufacture.

The purpose of this work is to study the possibility of gamma irradiation to improve the protective properties of optical materials used in medical technology.

Object and methods of research

Objects. Colorless clean glass (GOI St. Petersburg, Russia) was used for the study. The samples had the form of polished discs 16 mm in diameter and 1 to 2 mm thick. As comparison standard, we took extra-pure SiO_2 glass with impurities less than 0.001% (grades KU or KSV), which does not stain upon irradiation, which is made in the form of optically polished elliptical laser windows (5×3 mm^2) with thickness of 1 mm. We also studied industrial crystal glass - SiO_2 containing PbO (according to the glass passport, at least 24%), the samples were cut in the form of plates with a diameter of 10–15 mm and a thickness of 2-4 mm. The elemental composition of impurities in wt.% was determined by the X-ray radiometric method using the Am-241 radioisotope source, which excites X-ray radiation from elements with an atomic mass greater than 40, and confirmed by 24% PbO.

Gamma irradiation. The samples were irradiated with γ-quanta (1.17 and 1.32 MeV) of the ^{60}Co

isotope at pool-type facility at the Institute of Nuclear Physics of the Academy of Sciences of the Republic of Uzbekistan in air at power of 520 R/s in the dose range of 10^5–10^9 R at 320 K.

Experimental technique. Optical absorption spectra were measured on Specord M-40 "Carl-Zeiss" spectrometer (Germany) and on an SF-56 instrument (LOMO) at 300 K in the wavelength range 190–1100 nm.

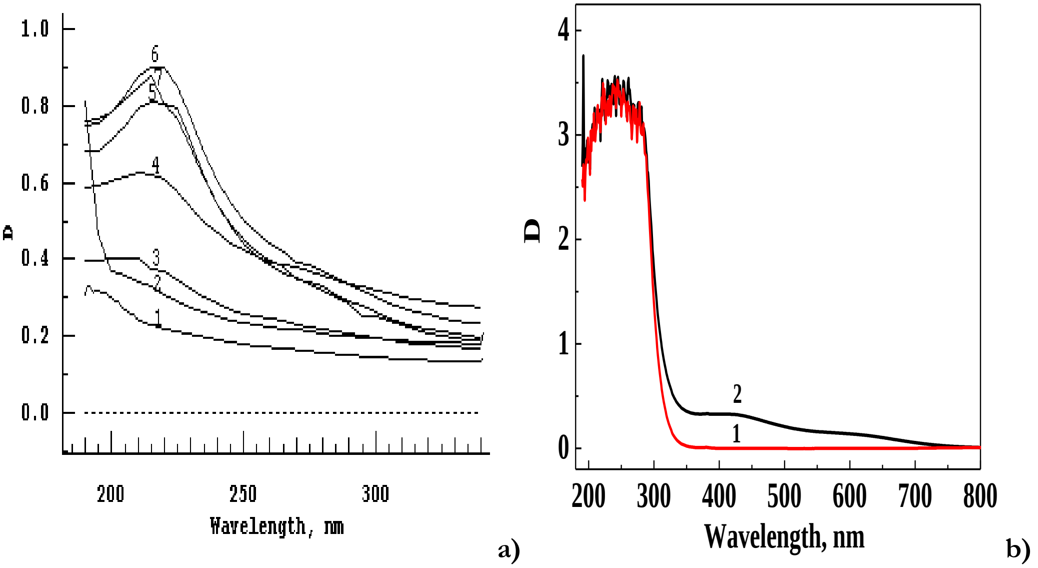

Fig.1. a) Sample spectra of SiO_2 optical density before (1) and after ^{60}Co gamma irradiation at ~520 R/s with doses: $2{\cdot}10^6$ R (2),), $8{\cdot}10^6$ R (3), $3.4{\cdot}10^7$ R (4), $3.1{\cdot}10^8$ R (5), $1{\cdot}10^9$ R (6), $2.7{\cdot}10^9$ R (7); b) 1 – non-irradiated, 2 – stained glass after irradiation.

Fig. 1. shows the optical density spectra of SiO_2 before (1) and after gamma irradiation. In unirradiated samples, weak (D=0.3) optical absorption band with maximum at 190 nm (Fig. 1, curve 1) is visible, which is attributed to E_s- centers on the surface [8]. Known bands at 215 and 260 nm associated with the electron E'_1 - and hole O^0_1 centers of the nonbridging oxygen atoms (NBO) [8-12]. Particular attention should be paid to curves 3 and 7 in Fig.1, taken immediately after irradiation, showing the beginning of splitting of the broad band at 215 nm.

The concentration of centers induced in extra pure glass at $2.7{\cdot}10^9$ R to saturation is $N_E = 1{,}54{\cdot}10^{17}$ cm^{-3}. The spectra in Fig. 1 are consistent with those of KV glass upon irradiation with electrons $2.4{\cdot}10^{16}$ cm^{-2} (10 MeV) [8,13], although the energy of gamma radiation is lower (1.25 MeV) and the dose $2.43{\cdot}10^{19}$ cm^{-2} is higher.

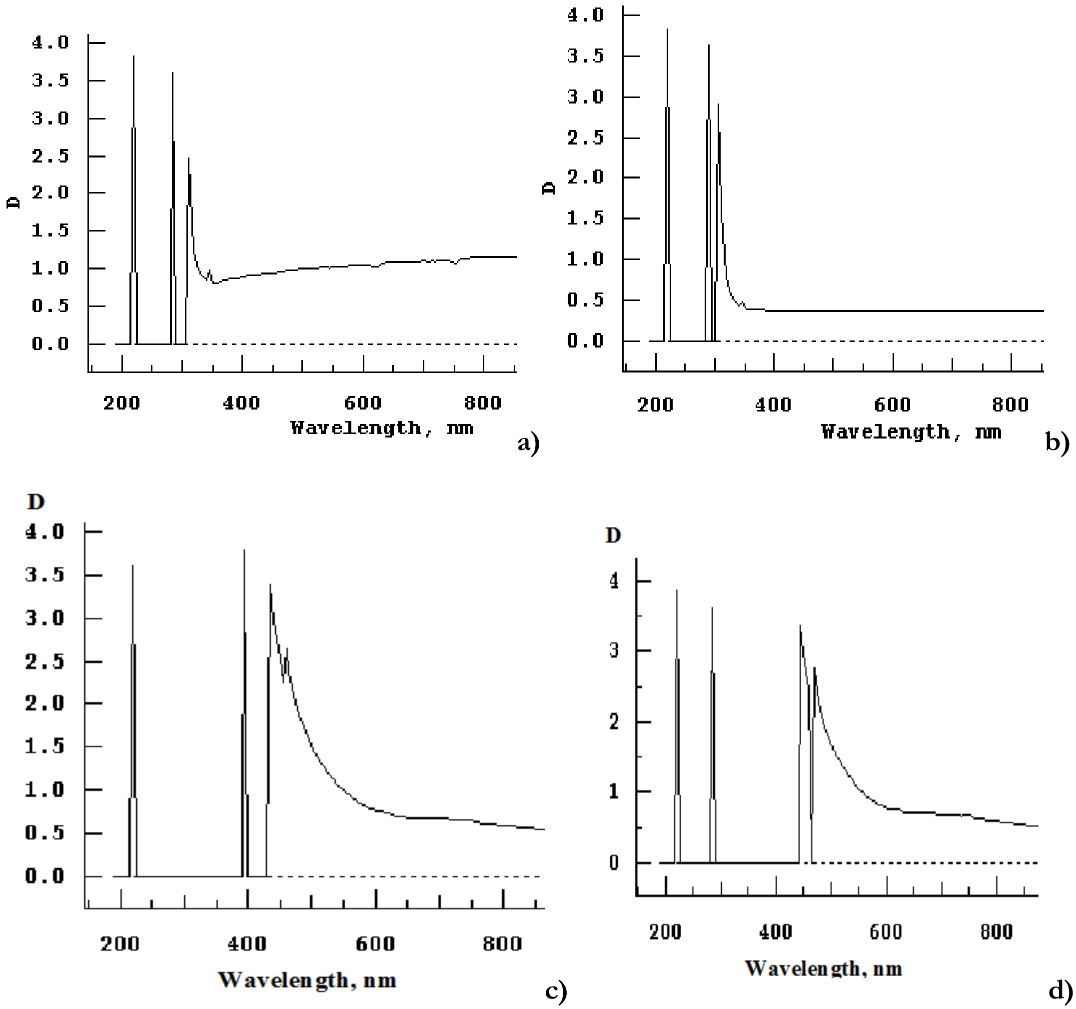

Fig. 2. Optical absorption spectra of SiO_2-PbO: a) before irradiation and after irradiation in the channel of the ^{60}Co gamma-installation at power of 477 R/s with doses: b) $8.5 \cdot 10^4$ R, c) $1.7 \cdot 10^5$ R, d) $1.2 \cdot 10^6$ R

In the fig. 2. shows the optical density spectra a) of non-irradiated colorless sample and after gamma irradiation with dose of: b) $8.5 \cdot 10^4$ R, c) $1.7 \cdot 10^5$ R, d) $1.2 \cdot 10^6$ R. Before irradiation, narrow intense resonances are observed, which are characteristic of quantum dots, since their optical density exceeds 3, which is the limiting value for optical transitions with charge transfer. The resonance at 220 nm is due to oxygen-deficient electronic centers in SiO_2 nanocrystals [5], 285 nm can be attributed to lead, since it is close to the 283.3 nm emission line of lead atoms when excited by an electric arc. The formation of 0-dimensional metal nanoparticles in a glass matrix occurs by the mechanism of phase segregation [14, 15].

The asymmetric resonance profile at 310 nm characterizes the longitudinal plasmon of a 1D-particle and the interaction between metal particles. Scattering of light in the visible region, which depends weakly on the wavelength, is associated with the presence of opaque molecular centers of the Pb-Si eutectic mixture. After irradiation, the population of the 310 nm resonance increased, and significant bleaching occurred in the visible region, which was associated with the dissolution of the Pb–Si eutectic mixture. The result is yellow color.

Next, the samples were irradiated in the dose range from 10^5 R to $1.2\cdot10^6$ R, while the color changed from yellow to brown as the absorbed dose increased, and the optical absorption spectra were measured before and after each irradiation session. Under gamma irradiation in the dose range of 10^4–10^7 R, charges are transferred between them, which manifests itself in a change in the ratio of optical densities of UV resonances associated with localized dipole plasmons. In addition, there is redshift of the absorption band associated with longitudinal surface plasmonic polaritons and elongation of lead nanoparticles.

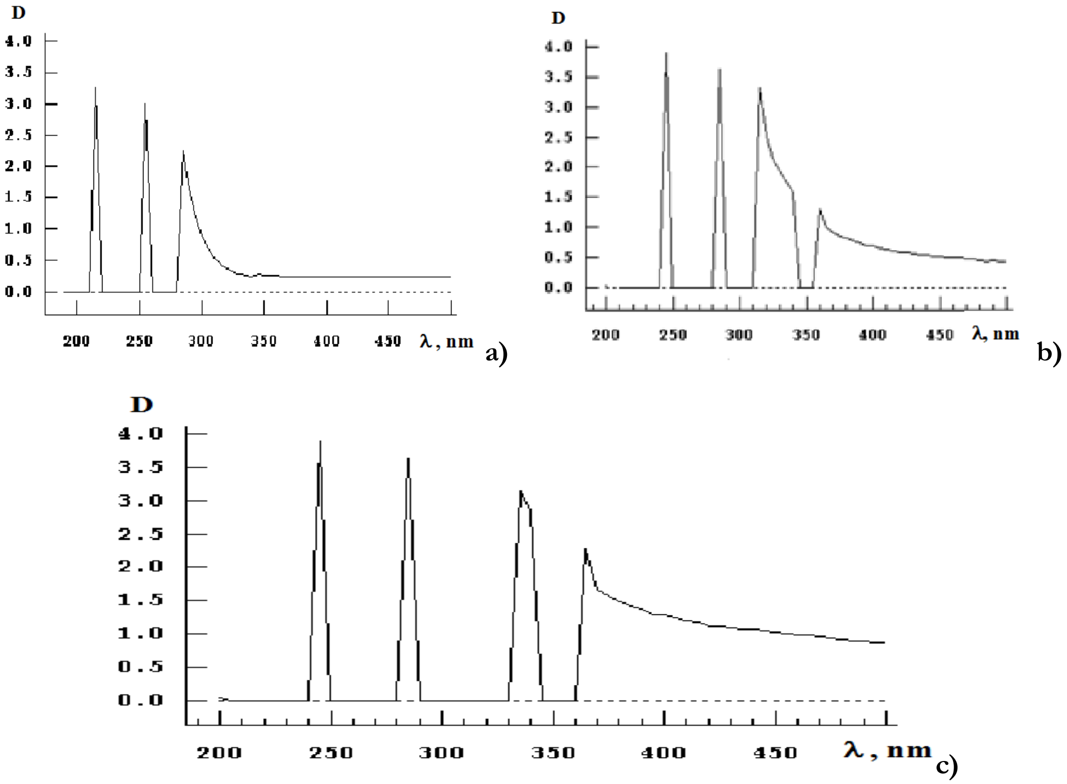

Fig. 3. Optical density spectra of barium glass before (a) and after irradiation at power of 210 R/s with doses of $1.5\cdot10^6$ R (b) and after gamma irradiation at a power of 600 R/s with doses of $4.3\cdot10^6$ R (s).

On fig. 3 (a-c), show the optical density spectra before and after gamma irradiation of barium glass. It should be noted right away that only a weak absorption band <200 nm (F center) is present in extra pure glass. Irradiation creates an oxygen-deficient electronic E'-center with a band of 215 nm [2], which grows to D=0.9 at 10^9 R. It can be seen from fig. 3 (a), that there are narrow intense absorption peaks in the initial SiO_2-BaO_2 samples at λ=215 and 255 nm, as well as an asymmetric band at 285 nm, separated by intervals with D=0. Such linear "molecular" spectrum and transparency windows in the UV region are characteristic of coherent plasmon oscillation in nanoparticles with sizes $L<<\lambda$, when only the dipole term affects the extinction cross section (i.e., the internal size effect of nanoparticle polarization) [16, 17]. After irradiation at 210 R/s with a dose of $1.5 \cdot 10^6$ R (Fig. 3b), the 215 nm line disappeared (together with nano-quartz), and two new ones appeared: 245 nm (Si-Si), associated with nano-tridymite and 285 nm with the $BaSiO_3$ nanophase. A noticeable increase in scattering and absorption, as well as splitting in the region of 320 and 360 nm, are due to the formation of color centers (brown) with a high concentration of localized charges. The 310 nm band is assigned to Q_{2+} hole centers near the alkaline earth metal [2]. In this case, the Ba content is about 27 wt.%; therefore, the band is very intense, wide, and splits, which is typical not for point centers, but for an ensemble of bound nanoparticles [16, 17].

After dose of $4.3 \cdot 10^6$ R (Fig.3 c)) narrow bands 245 nm (tridymite 40 nm in size) and 285 nm ($BaSiO_3$), 340 nm and a wide scattering band 370 nm are visible. The absorption spectra in Fig. 3 in the region of wavelengths longer than 350 nm are in good agreement with the spectra of ion-synthesized cobalt nanoparticles in an amorphous silicon dioxide matrix [18]. The authors attribute a very wide absorption band in the region of 400-500 nm to the absorption of free electrons in Co nanoparticles (that is, surface plasmons [16]. The differences in the UV part of the spectrum are due to the fact that Co nanoparticles are located only in the near-surface implanted layer, while Ba-containing nanoparticles throughout the volume of glass.

Conclusions

We have studied the effect of irradiation with ^{60}Co gamma rays in the dose range of 10^4–10^6R on the optical properties of SiO_2 glasses containing BaO or PbO. After irradiation, such glass acquires a beautiful golden or brown color, actively absorbs radiation in the UV region, but transmits yellow-orange light, has no residual radioactivity, and becomes sterile. The results obtained make it possible to offer SiO_2 glass containing barium or lead oxides for personal eye protection from UV radiation in medicine. The research was carried out with financial support from the Scientific Researches Program to President Degree 4526 of 21.11.2019.

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