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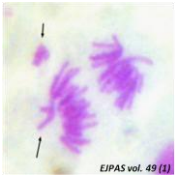

Fast Neutron Irradiation Effect on Some Optical Properties of Lead Borate Glass Doped with Samarium Oxide

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A B S T R A C T

Neutron irradiation creates a variety of defects in glass that results in changes in the optical properties, thus it is important to measure how much optical parameters are altered by this process. Optical densities have been measured experimentally before and after fast neutron irradiation for Al_2O_3-PbO-B_2O_3 glasses doped with Sm_2O_3. These values have been used to calculate optical parameters of such glass such as absorption coefficient (α), direct and indirect optical energy gap (E_{ogd} and E_{ogind}), Urbach's energy (E_u) and Fermi energy (E_f). It was found that E_f and E_u increase while E_{ogd} and E_{ogind} decrease with increasing neutron irradiation dose. The changes in these optical parameters may be attributed to structural changes and redistribution of the electrons in the glass samples due to collision of energetic neutrons, and indicates an enhancement of the conduction and valence electron levels to the forbidden energy gap.

Introduction

More recently, interest in radiation effects in glass, generally, has been renewed as glass has been used in different technological applications. Rare-earth doped glasses have been paid much attention because of their high potential use for optical applications such as fibers, amplifiers, lasers and sensors[1].

The effect of irradiation on glass is believed to depend on the type and energy of irradiation, glass composition and sample parameters[2].

Neutrons being uncharged interact with matter quite differently than the charged particles or the electromagnetic radiation. As neutrons pass through matter, they lose energy by a series of collisions, in scattering or capture events. After a neutron has lost a significant portion of its kinetic energy through scattering events it may be absorbed by a target nucleus in a capture event. The result of this event is that the new atom has its mass number increased by one, and as such will undergo one of many possible nuclear events. The result is often the emission of ionizing radiations, which result in the production of defects in the material via electron and hole productions[3].

The electrons are initially excited from the valence band if the incident energy is greater than the band gap. The excess energy is converted to kinetic energy and as these electrons travel through the material, they will either recombine the positively charged holes, become trapped to form color centers or produce a secondary electron cascade by knock-on collision with other bound electrons. Additional bound electrons are ionized by the secondary electrons through Coulomb interactions, the secondaries losing approximately 20 eV for each ionization. Finally, when the electrons energy becomes too low to ionize other electrons they will either be trapped or recombine with holes that give rise to optical absorption bands. These bands are associated with radiation-induced intrinsic and extrinsic defects. Vacancies and self-interstitials cause intrinsic defects. These defects can be introduced by ionization or atomic displacement mechanisms or via the activation of the preexisting defects[4]. Extrinsic defects related with impurities such as alkali, alkaline earth and transition metals in the glass which increase radiation- induced defects[5]. Materials which have such defects absorb photons at particular wavelengths resulting in a modified absorption spectrum. The difference between the absorption spectra before and after irradiation is an important characteristic and is called the induced absorption spectrum.

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The purpose of our work is to study the optical absorption spectrum to get information about the changes in energy gap and some optical parameters of this glass due to neutrons irradiation.

Experimental procedures

Preparation of glass samples

Glasses having composition $(B_2O_3)_{50}$ $(PbO)_{45}$ $(Al_2O_3)_5$ $(Sm_2O_3)_x$,where x = 0, 1.7 were prepared using the normal melt quench technique from AR grade chemicals of B_2O_3, PbO, Al_2O_3 and Sm_2O_3 . The samples were mixed in porcelain crucibles and then heated at 950°C for two hours under normal atmospheric conditions. The melt was stirred from time to time to promote complete mixing and finally poured into preheated moulds made of stainless steel of radius 1.3 cm. All samples were properly annealed at 350°C in a muffle furnace to eliminate mechanical and thermal stresses. The samples were ground and highly polished using alpha alumina polishing suspension to obtain optical flatness. The thickness of each sample was measured by a micrometer.

The amorphous nature of these glasses was examined by x-ray diffraction analysis at room temperature. The x-ray diffraction spectra of the samples showed the diffused bands characteristic of the x-ray diffraction pattern of amorphous materials; the spectra did not show any sharp peaks and confirms that the glass samples are amorphous in nature.

UV-visible absorption measurements

The Optical properties of highly polished samples were measured at room temperature before and after each neutron irradiation dose using a UV/VIS spectrophotometer covering the wavelength range 190–1100 nm type JASCO, corp., V–500, Japan.

Fast neutron irradiation

Glass samples were irradiated with neutron from an Am–Be neutron source of 5 Ci activity, which has a neutron yield of $1.1\text{x}10^7$ n s^{-1}. Three irradiation doses of $3.229\ \text{x}10^{10}$, $5.045\ \text{x}10^{10}$ and $1.7\text{x}10^{11}$ n cm^{-2} were used.

Results

As shown in Fig.1, optical absorption spectra can be divided into three particular regions. The first is weak absorption tail, which originates from defects and impurities, the second in the middle, is the exponential edge region which is strongly related to the structural randomness of the system and the last one is the high absorption region that determines the optical energy gap.

Glass Fermi energy

The imaginary refractive index, *k*, can be represented by Fermi-Dirac distribution function[6]:

$$k = \frac{1}{1+\exp\left(\frac{E_f - E}{K_B T}\right)} \quad , \qquad (1)$$

where E is the energy of the incident photon, T is the temp. in Kelvin where K_B is Boltzmann constant and E_f is the Fermi energy which is defined as the highest occupied energy level when electron configuration is in its ground state. Equation (1) can be written as

$$K_B T \ln\left(\frac{1}{k} - 1\right) = E_f - E \qquad (2)$$

Linear fitting of the last equation is used to determine the glass Fermi energy. The recorded data reveal that E_f increases with increasing neutron irradiation dose, as shown in tables 1, 2. It has been noted also that E_f increases with doping of samarium oxide as shown in Fig. 2.

Optical band energies

The absorption of an optical medium can be quantified in terms of optical density (O.D.). This is sometimes called *absorbance*, *A*, and is defined as:

$$A = -\log_{10}(I / I_o), \qquad (3)$$

and according to Beer's law:

$$I = I_o e^{-\alpha x}, \qquad (4)$$

the absorption coefficient, α, can be obtained from

$$\alpha = 2.303\,(A / x), \qquad (5)$$

where I and I_o are optical intensities at thickness x and $x = 0$ respectively. The optical absorption coefficient changes rapidly for photon energies comparable to that of the band gap, E_g.[7]. According to Mott and Davis[6], the absorption of light by amorphous solid depends on the energy E of the incident photon and on the optical gap of the material.

It is found that[8] this behavior may be represented by an equation of the form:

$$\alpha E = \text{B}\,(E - E_g)^{\text{r}} \qquad (6)$$

where B is a constant and r is an index which assumes the values 1/2, 3/2, 2 and 3 depending on the nature of the electronic transition responsible for the absorption. In the present case r is taken equal to 1/2, for allowed direct transition and equal to 2, for allowed indirect transition[1]. The direct optical band gap energy, E_{ogd}, can be obtained by plotting $(\alpha E)^2$ versus the photon energy E. The indirect optical band gap energy, E_{ogind}, can be obtained by plotting $(\alpha E)^{1/2}$ versus the photon energy E extrapolating the linear portion of the curve to intersect the energy axis. As shown in tables 1, 2 we notice a small decrease in E_{ogd} and E_{ogind} with increasing neutron irradiation dose.

Urbach energy:

In the exponential region of the absorption versus the photon energy curve, the absorption coefficient, α, is given by the relation:

$$\alpha = \text{C}\exp(E / \Delta E), \qquad (7)$$

where C is a constant and ΔE characterizes the slope of the exponential edge (Urbach's energy, E_u) region and it is the width of the band tails of the localized states. The existence of the long tails of the localized states is attributed to the amorphous nature of the material. Since the glass we are using in the present study is an amorphous material, that is why ΔE (or E_u) is relevant in this particular study. To calculate the width, ΔE, of the energy tail, a model proposed by Urbach (1953) can be used[9]. From (7):

$$\ln(\alpha) = \ln(C) + (E / \Delta E),$$

or $$\ln(\alpha) = \ln(C) + (E / E_u), \qquad (8)$$

Thus, from eq. (8), E_u can be obtained from the linear plot of ln(α) as a function of photon energy. The values of E_u listed in tables 1, 2 reveal that E_u increases with increasing neutron fluence.

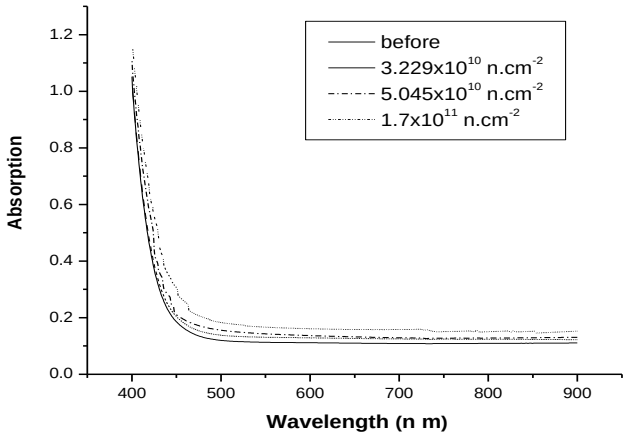

Fig. 1: UV–VIS spectra of free Sm glass before and after neutron irradiation.

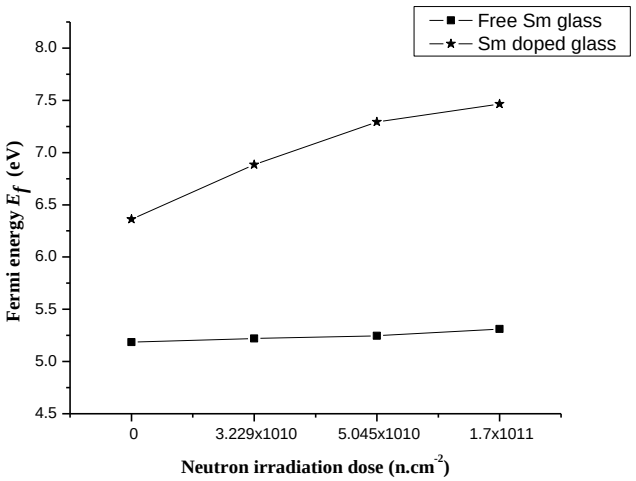

Fig. 2: Fermi energy at different neutron irradiation doses for free Sm glass and Sm doped glass.

Discussion

As shown before, successive neutron irradiation doses cause changes in all optical parameters. These changes attributed to compositional changes in the glass matrix result from the collision of energetic neutrons with atoms dissipate their energy through the solid. These collisions cause displacement of atoms from their normal positions[10]. The neutron irradiation may cause densified structural change in glass or compact state which may include that some bond angles become smaller, change in density and refractive index and induced color center that responsible for induced absorption bands. However, this decrease in the optical band gap may be due to the fact that in neutron irradiation, the electrons leave their normal positions and they move through the glass network[9], this results in redistribution of the electrons in the glass samples. Urbach's energy increasing indicates an enhancement of the conduction and valence level electrons to the forbidden energy gap and may cause an increase in the conductivity of the glass due to neutron irradiation. From table 2 we notice that E_{ogd} and E_{ogind} decrease with Sm_2O_3 doping, while other parameters, E_f and E_u, increase with Sm_2O_3 doping. Upender *et al.*[11] showed that addition of Sm_2O_3 appears to suppress the effect of PbO whereby a progressive shift of BO_4 and BO_3 bands toward higher wavenumbers is noted. Usually, a shift of absorption bands to higher wavenumbers occurs as a result of an increase in the degree of polymerization of structural network units of glass system. But here in case of adding rare earth element, Sm, the absorption bands move to lower wavenumbers[12] for which these changes in the listed parameters may be attributed to.

Conclusion

From the above study it is clear that optical parameters of the glass samples have been changed due to neutron irradiation. It was found that the values of E_f and E_u increase while E_{ogd} and E_{ogind} decrease with increasing neutron irradiation dose. These changes may be attributed to structural changes and redistribution of the electrons in the glass samples due to collision of energetic neutrons. In addition to this, Sm_2O_3 doping affected those optical parameters.

Table 1: Optical parameters at different neutron irradiation doses for Sm free glass.

Dose (n.cm^{-2})	E_f (eV)	E_{ogd}(eV)	E_{ogind}(eV)	E_u(eV)
0	5.186	3.293	2.963	0.182
3.229x10^{10}	5.219	3.225	2.769	0.201
5.045x10^{10}	5.245	3.219	2.765	0.215
1.7x10^{11}	5.312	3.153	2.754	0.247

Table 2. Optical parameters at different neutron irradiation doses for Sm doped glass.

Dose (n.cm^{-2})	E_f (eV)	E_{ogd}(eV)	E_{ogind}(eV)	E_u(eV)
0	6.362	3.193	2.671	0.331
3.229x10^{10}	6.883	3.091	2.543	0.358
5.045x10^{10}	7.294	3.093	2.573	0.451
1.7x10^{11}	7.465	3.066	2.510	0.413

References

1) Nogami M., Hagiwara T., Kawamura G., Ghaith E., Hayakawa T. (2007). Journal of Luminescence 124: 291–296.

2) Rajaram, M., Friebele E. J. (1989). J. Non-Cryst. Solids 1081: 1.

3) Sandhu, A.K., Singh S., Pandey O. P. (2009). Materials Chemistry and Physics 115: 783–788.

4) Agrawal, A., Tomazawa M. (1996). J. Non-Cryst. Solids 209: 166.

5) Friebele, E.J., Gingerich M. E., Long K.J. (1982). Appl. Opt. 21: 547.

6) Mott, N.F., Davis E.A. (1971). Electronic Processes in Noncrystalline Materials, Clarendon, Oxford.

7) Elliott, S.R., in: R.W. Cahn, P. Haasen, E.J. Kramer (Eds.), Materials Science and Technology: Glasses and Amorphous Materials, vol. 9, 1991, p. 420, Published jointly by VCH, Weinheim Germany, and New York, USA.

8) Hasegawa, S., Yazaki S., Shimizuol T. (1978). Solid State Commun. 26: 4070.

9) Singh S., Sandhu A. K., Prasher S., Pandey O. P. (2007). Radiation Measurements 42: 1328 – 1331.

10) Primak W., Fuchs L. H., Day R. (1995). J. Am. Ceram. Soc. 38: 134.

11) Upender G., Vardhani C. P., Kamalaker V., Mouli V. C. (2010). SRX Physics, 1-6-doi: 10.3814/2010/927056.

12) Ni Yaru, Chunhua L., Yan Z., Qitu Z., Zhongzi X. (2007). Journal of rare earths, Vol. 25, Suppl., Jun., p. 94