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Physical and optical properties of Na_2O doped BaO containing boro-Tellurite glasses for battery applications

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Abstract:
The structural, physical and optical properties of glasses of composition (50-x)TeO_2-40B_2O_3-10BaO-xNa_2O (x=0, 2.5, 7.5 and10 mol%) and the role of Na_2O have been studied in this work. The amorphous phase of the glasses was confirmed through an X-ray diffraction study. The optical band gap energy (E_{opt}) was decreased with the addition of Na_2O, whereas, the refractive index values were increased. Urbach energy (ΔE) of the glasses varied from 0.18 to 0.26 eV and and refractive index of the glass system also increased. Density (ρ) of the studied glasses was found to increase from 3.46 g/cm^3 to 3.87 g/cm^3 with Na_2O content. Glass transition temperature (T_g) was increased with Na_2O content.

Keywords: *Density, cut-off wavelength, Urbach energy, indirect band gap*

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I. Introduction

Glassy materials are the most promising materials to fulfill current breakthrough technological needs due to their particular structure and optical characteristics and the ability to make variety of compositions very easily. Tellurite-based glasses (TeO_2-based glasses) are considered as promising materials for their promising characteristics like transparency in a wide (~ wavelength range 400 nm to 6 μm), stable with thermal, low melting point, chemical durability, and high linear refractive index. It is important to point out that TeO_2 forms glass by only adding any suitable glass [**1-3**]. Mixing of boron oxide (B_2O_3) with (TeO_2) improves the forming ability of the glass system and enhances the thermal stability and optical characteristics of the obtained glasses. The resultant boro-tellurite (B_2O_3-TeO_2) glasses can be used in electronic and optical applications [**4-5**]. The structure, UV–Vis–NIR spectroscopy, thermal, and non-linear optical characteristics of tellurite glasses modified by different oxides such as ZnO, BaO, SrCl2, LiF, Bi_2O_3, and others have been reported by several researchers [**6-9**].

There are clear advantages of using tellurium oxide (TeO_2)-based glasses for a wide range of applications due to their excellent third-order optical nonlinearity, a high index of refraction, large infrared transparency, low melting temperature, and low glass transition temperature in comparison to regular glasses. Moreover, because of the unshared TeO_4 electron pair , these glasses have high dielectric constant and electrical conductivity **[10-16]**. The TeO_2 glass features have been developed to make them ideal for different applications, such as in photonic devices, sensor systems, optical fibre technology, laser technology and CD memory devices [**17-20**]. It has been established that TeO_2 can act as a conditional glass former that requires the addition of modifier oxides such as alkali, alkali earth, and transition metal oxides (TMOs), or other glass modifiers for glass formation **[21-22]**. TeO_4 trigonal bipyramids (tbp's) and TeO_3 trigonal pyramids (tp's) with a lone pair at the equatorial position form the basic structure of pure TeO2. Furthermore, the network structure is depolymerised and TeO_4 tbp's are converted into TeO_3 tp's through the TeO_{3+1} polyhedra when modifier oxides are added into TeO_2 based-glasses. The structural and optical properties of the glass can be affected by modifications to the TeO_2 network structure.

Borate glasses have several significant applications over other glass formers like silicate, bismuthate, phosphate, tellurite and draw the researcher's interest owing to their chemical stability, strong thermal stability, mechanical characteristics, and strong solubility of alkaline earth metals along with different transition metal oxides **[23-27]**. Borate based glasses contain two combinations of boron groups called trigonal BO_3 and

tetragonal BO_4. BO_3 groups play a dominant role in pure borate glasses, whereas BO_4 groups are glass modifying groups with different oxidation states formed with non-bridging oxygen (NBOs) in the glass matrix **[28-30]**.

Large numbers of glass research laboratories are increasing day-by-day for the creation of optically transparent radiation shielding (OTRS) materials. Optically transparent materials for the use of optical communication, modern optical devices, and radiation shielding materials are used as shielding where the protection is required from radiation. The basic and essential features of OTRS materials are high density and refractive index for radiation shielding and optical related applications respectively. Borate glasses have become best substitute for concrete shielding because of their exceptional properties such as high thermal stability, toughness, chemical compatibility, and so on, and boro-tellurite glasses have further enhancement in physical properties such as transparency and refractive index. Density and transparency can be improved by incorporation of heavy metal oxides (HMO) to the boro-tellurite glasses. Most of the HMOs including BaO, PbO, and so on act as modifier or former role in the glass network based on their percentage in the composition. Among various HMOs, BaO and PbO have more compatibility with the borate glasses and incorporation of these elements to the borate glasses improves the radiation absorption.

The aim of the present work is to study physical and optical properties of the selected glass system.

II. Experimental

2.1 Glass preparation

Glass compositions of (50-x)TeO_2-40B_2O_3-10BaO-xNa_2O (x=0, 2.5, 7.5 and10 mol%) were synthesized by conventional melt quench technique. AR grade chemicals like H_3BO_3, TeO_2 and Na_2CO_3 were taken in powder form in the stoichiometric ratios and mixed in a platinum crucible. The crucible that containing the starting materials was kept in the electrical carbide rod furnace which is maintained at 1000^0C for half an hour. The molten mixtures was thoroughly stirred to get homogeneity and poured on the metal mold at 200^0C and quenched with a metal disc to get transparent glass samples of the desired shape. The obtained glass samples were annealed at 300^0C for 12 hours in the temperature controlled furnace to subtract thermal strains. The prepared glasses were kept inside the paraffin oil to avoid hygroscopic nature. Various experimental studies were carried out on the prepared glass samples.

2.2 Characterization techniques

X-ray diffraction studies:

X-ray diffraction patterns of the glass system were recorded on diffractometer (Philips PW 1140) at the room temperature with k_α radiation (1.54 A^o) to confirm the amorphous nature of the glass samples. The XRD spectrographs were recorded with Bragg's angle (2θ) from 10^0 to 80^0 with count 0.2/sec.

Physical parameters (Density related)

The density (ρ) of the glasses was measured on VIBRA HT analytical balance by using Archimede's principle with xylene (ρ=0.86 g/cm^3) as the immersion liquid according to the following relation.

$$\text{Density}(\rho) = \frac{a}{a-b} x 0.86 \quad (1)$$

Where a- weight of glass in air, b - weight of glass in xylene

The molar volume (V_m) and the Oxygen packing density (OPD) can be calculated from the following relations

$$\text{Molar volume}(V_m) = \frac{M}{\rho} \quad (2)$$

$$\text{Oxygen packing density}(OPD) = \frac{1000\rho C}{M} \quad (3)$$

Where M-Molecular weight, ρ-density and C- No. of oxygen atoms per unit formula

2.3 UV-Visible spectroscopy

UV-Vis. absorption spectra of the glass system were recorded at room temperature in the wavelength range 200 nm-1000 nm by using UV-Visible spectrometer (Shimadzu 8400S model) with a spectral resolution of 1nm. Cut-off wavelength (λ_c) has been estimated by the tangent which is drawn along the cut-off edge onto X-axis in the spectra. The absorption coefficient (α) is a function of the thickness and absorbance of the sample. The direct and indirect optical band gap energies (E_{opt}) of the glasses were obtained from the UV-absorption edge using Davis and Mott relation.

$$\alpha h\upsilon = B(h\upsilon - E_{opt})^n \quad (4)$$

Where α -is the optical absorption coefficient at a frequency (ν), E_{opt}- Optical band gap energy, h- Planck's constant, n=2, $\frac{1}{2}$ for indirect, direct band gap respectively.

The refractive index (n) values of the present glass system were calculated using the equation Vesselin Dimitrov and Sumio Sakka. The molar refraction(R_M) and electronic polarizability (α_e) of the glass system were calculated using the relations given by Komatshu and Duffy.

III. Results and discussion

3.1 Physical properties of TBBN series

XRD

Fig.1 shows the XRD patterns of glasses (50-x) TeO_2 -40 B_2O_3 -10BaO- xNa_2O where x = 0, 2.5, 5, 7.5 and 10 mole% of various glass formers and network modifiers. In these figures no sharp peak is observed that means the prepared glasses are amorphous in nature. Hence, after X ray diffraction studies the glasses will be given to the further characterization.

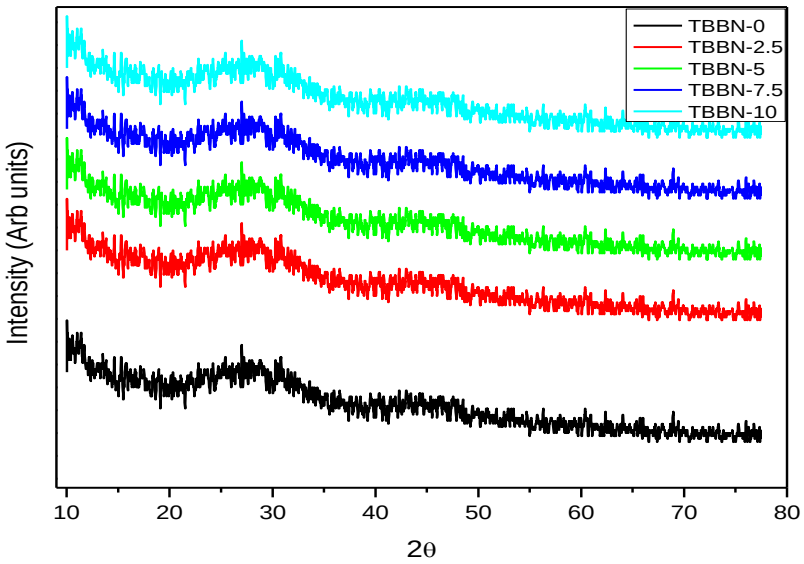

Fig.1 XRD spectrograms of TBBN glass system

Table 1. Compositions of (50-x) TeO_2 -40 B2O3-10BaO- xNa_2O glass system in mole% and glass transition temperature Tg (^{0}C)

Glass Code	TeO_2	B_2O_3	BaO	Na_2O	T_g(^{0}C)
TBBN-0	50	40	10	0	340
TBBN-2.5	47.5	40	10	2.5	310
TBBN-5.0	45	40	10	5	358
TBBN-7.5	42.5	40	10	7.5	360
TBBN-10	40	40	10	10	372

Density

Chemical composition of the present glass system is (50-x) TeO_2 -40B_2O_3 -10 BaO-xNa_2O where x = 0, 2.5, 5, 7.5 and 10. In the present system, B_2O_3 and BaO mole percentages were fixed and TeO_2 percent decreases whereas Na_2O mole percentage increased. It is found that the density of the TBBN series of glasses increased from 3.46 g/cm^3 to 3.87g/cm^3 linearly and molar volume has shown opposite behaviour as shown in **Fig.2**. The density is increased due to heavier TeO_2 occupies the lighter Na_2O in the glass network [**31-32**].

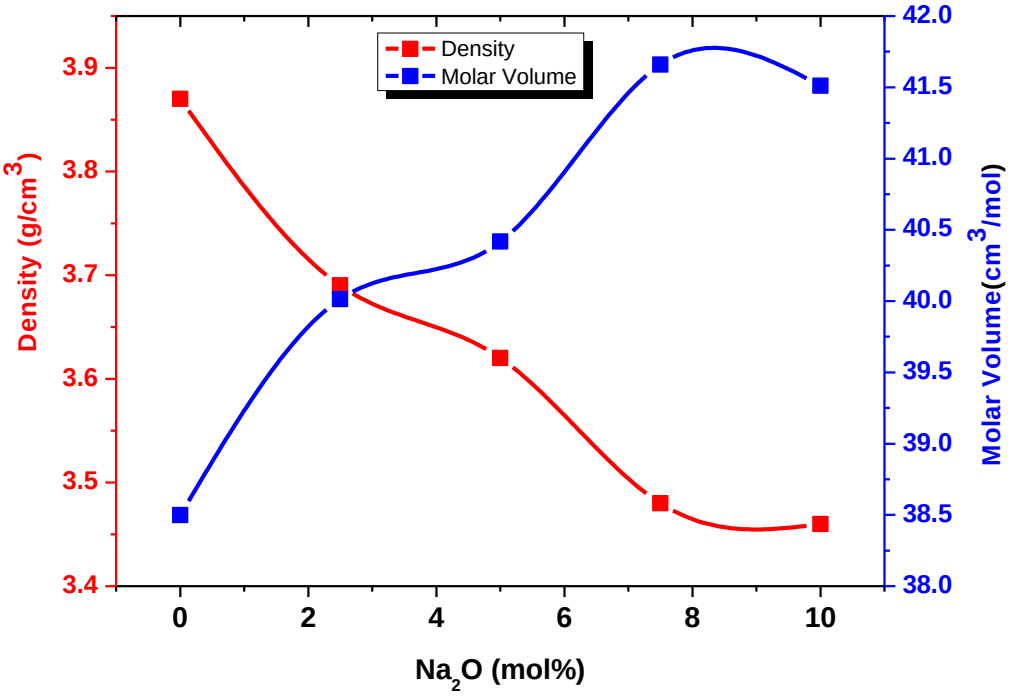

Fig.2. Variation of density and molar volume with Na_2O

The changes may be due to breaking of network and bonds with addition of TeO_2. The density also influences the other physical parameters such as molar volume, oxygen packing density (OPD) etc.

Molar volume of **TBBN** series decreases with the increase of Na_2O mole percentage from 0 to 10 cc/mole in a linear manner. Both density and molar volume shows opposite behaviour. Oxygen packing density increasing, because more number of oxygen atoms are added to the glass network as the increase of Na_2O mole percentage. Variation of OPD with Na_2Omole percentage is shown in **Fig. 3** All the physical parameters of present glass system are given in **Table 2.**

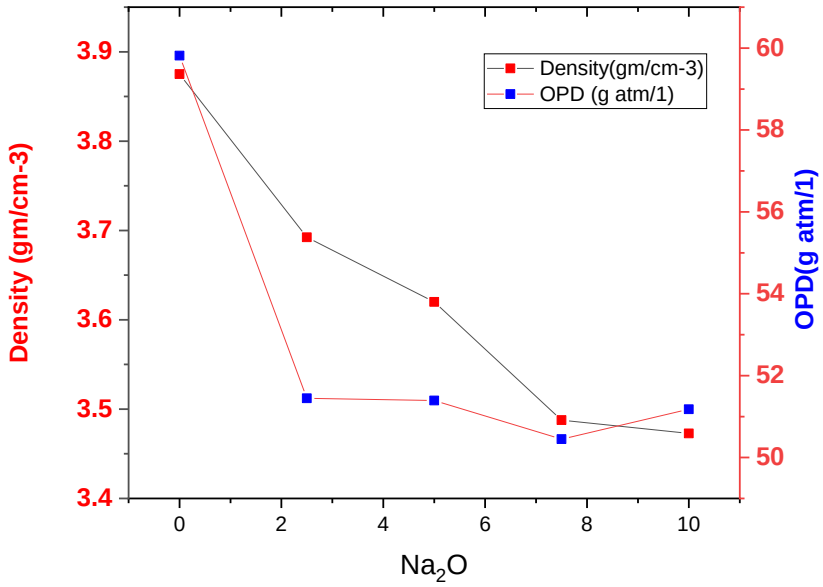

Fig.3 Variation of density and OPD with Na_2O

TABLE.2 Physical and optical parameters of TBBN series

Properties	TBBN 0	TBBN2.5	TBBN5.0	TBBN7.5	TBBN-10
Density (g/cm^3)	3.87	3.69	3.62	3.48	3.46
Molecular weight (g/mol)	148.99	147.65	146.31	144.97	143.63
Molar volume (cm^{-3}/mol)	38..49	40.01	40.41	41.65	41.51
OPD (g atm/1)	59.81	51.44	51.39	50.44	51.17
Cut-off wavelength (nm)	338	421	396	370	342
Indirect band gap energy (eV)	3.72	2.74	2.80	2.97	3.56
Direct band gap energy (eV)	3.74	3.02	3.18	3.28	3.60
Urbach energy(eV)	0.183	0.185	0.389	0.256	0.240
Molar refraction(cm^{-3}/mol)	21.90	25.20	25.29	27.73	27.67
Refractive index	2.22	2.47	2.45	2.40	2.26
Electronic polarizability(*10^{-24} cm^{-3})	8.67	9.98	10.02	10.14	9.50

3.2 Optical properties of TBBN series

(50-x)TeO_2 -40B_2O_3 -10 BaO-x$Na_2O$with different mole fractions obtained are shown in **Fig.4** No sharp absorption edge is observed, which is the characteristic nature of a glass material. It is observed that fundamental absorption edge shifting towards higher wavelengths as the concentration of Na_2O increases. Formation of non-bridging oxygens (NBOs) results in the lower rigidity of the glass system [**33-37**].

In the absence of Na_2O, cut off wavelength is highest which 338nm. In this case only two network modifiers play the role with glass former TeO_2. By adding Na_2O to the glass system, cut off wavelength varies between 338-421nm. Indirect band gap of the present system obtained from the **Fig.5**. Indirect optical band gap energies decreases from 3.72eV to 2.74 eV due to non-bridging oxygens. Optical band gap energy is lowest (3.02eV) for the TBBN2.5 sample.

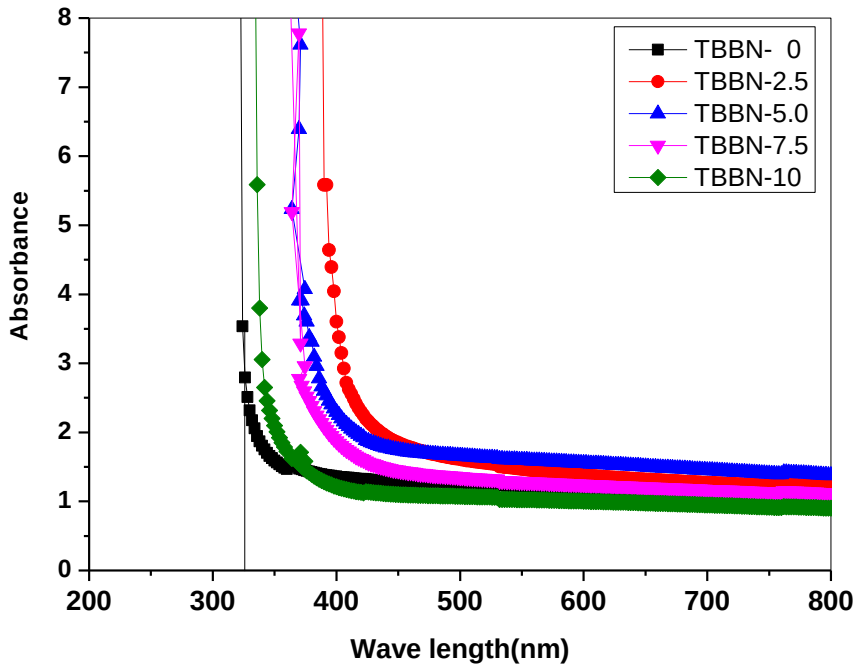

Fig.4 Optical absorption spectra of TBBN glass composition

Without TeO_2, refractive index is high for TBBN7.5 sample. The increase in refractive index may be due to the non-bridging oxygen (NBOs) bonds in the glass network. Electronic polarizability of the prepared TBBN system of glasses is decreased from 10.14 to 8.67 in the order of 10^{-24} cm^3 non-linearly. All the optical properties of TBBN series are given in **Table 2**.

Direct band gap of the present glass system are calculated from the **Fig. 6**. It is found that direct optical band gap without Na_2O is 3.74eV. By adding Na_2Oit is suddenly decreased up to 3.02eV again increased to 3.60eV. Because of non-bridging oxygens, direct band gap is changed with the effect of Na_2O. Refractive index of these glasses is increased from 2.28 to 2.35 with addition of Na_2O.

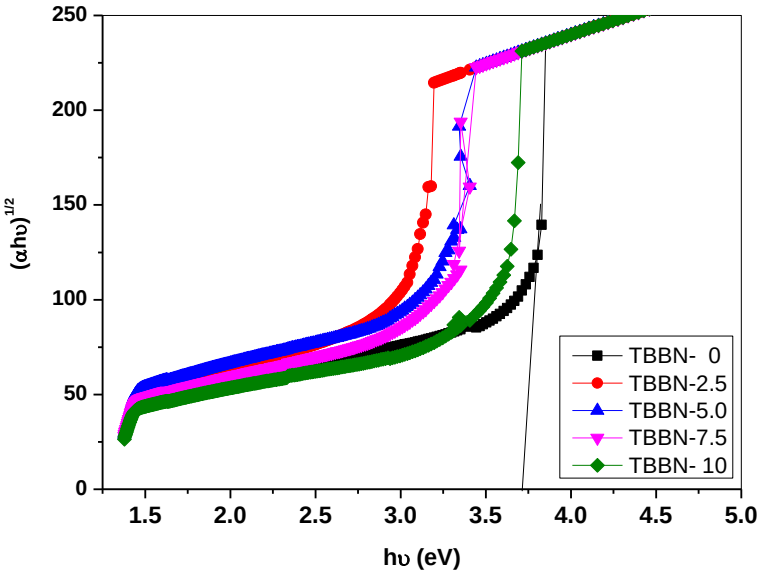

Fig.5.Tauc plots of TBBN glass composition for indirect band gap.

Urbach Energy

Urbach plots of TBBN series obtained are shown in **Fig.7**. Urbach energy and optical band gap values of the TBBN series are presented in the **Table 2.** Urbach energy is lowest for TBBN-0 sample attributed to the structural rearrangement of the three modifiers. Urbach energy values are found to increase and lie between 0.18 and 0.23 with the increase of Na_2Omole percentage. When an alkali oxide (Na_2O) added to the pure boron oxide, the basic structural unit of borate BO_4 converts into BO_3 [38]. These units are responsible for the Urbach energy or increase in the connectivity of the glass network. Urbach energy is a measurement of disorder in the glasses and hence increase in Urbach energies confirms an increase of disorderness in the glass [**39-40**]. The optical band gap energy values of TBBN series are having a good agreement with the earlier reports [**41**].

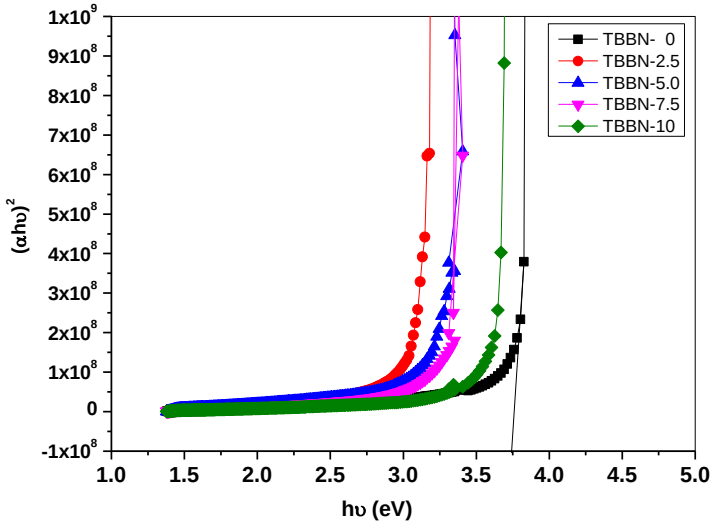

Fig.6 Tauc plots of TBBN glass composition for direct band gap.

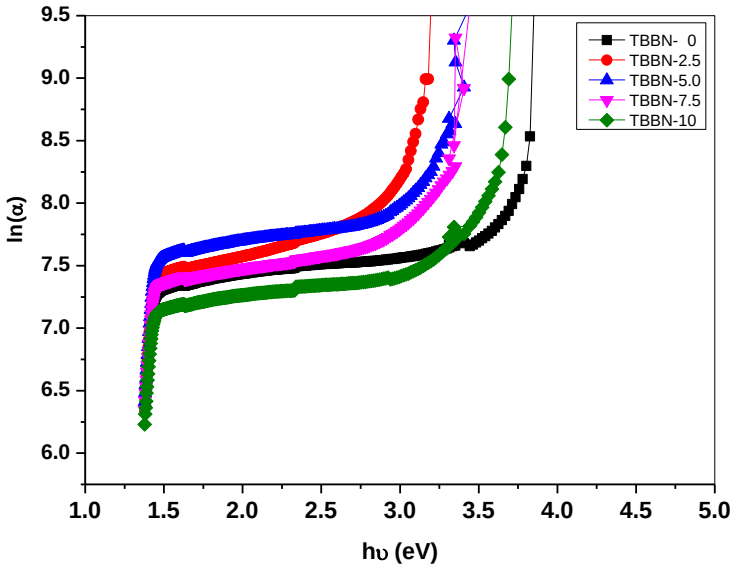

Fig.7 Urbach plots of TBBN glass composition

IV. Conclusions

The glass compositions (50-x)TeO_2-40B_2O_3 -10 BaO-xNa_2O where x = 0, 2.5, 5, 7.5 and 10 mol% were prepared through melt quench technique. XRD spectra revealed the amorphous nature without showing any crystalline peaks. With the addition of Na_2O the density increased linearly. Shifting of cut-off wavelength towards higher frequencies and optical band gap energies towards lower frequencies were significant evidences of the creation of NBOs. The refractive index of the glass system was observed to be high for all the glass samples which suggest that the present glasses are most useful in the fabrication of non-linear optical devices.

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