Journal of Alloys and Compounds 527 (2012) 5–9

Contents lists available at SciVerse ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

DSC, ESR and optical absorption studies of Cu^{2+} ion doped in boro cadmium tellurite glasses

P. Gayathri Pavani*, M. Prasad, V. Chandra Mouli

Glassy Material Research Laboratory, Department of Physics, Osmania University, Hyderabad 500 007, India

ARTICLE INFO

Article history:
Received 1 October 2011
Received in revised form 17 February 2012
Accepted 24 February 2012
Available online xxx

Keywords:
Amorphous materials
Differential scanning calorimetry (DSC)
Glass transitions
Electron resonance

ABSTRACT

Physical, optical absorption, ESR and DSC studies on 50 B_2O_3–(50 − x) CdO–$x$$TeO_2$ glasses containing Cu^{2+} spin probe have been carried out. Density measurement is carried out by Archimedes principle. Variation in glass transition temperature is discussed in terms of physical parameters. ESR results show that $g_\parallel > g_\perp$ indicating that the Cu^{2+} ions is in tetragonal distorted octahedral site and its ground state is $d_{x_2-y_2}$. There are considerable changes in $g_\parallel$, $g_\perp$ and $A_\parallel$ values with increasing the concentration of CdO in BCT glass systems. The optical absorption spectra results show that the absorption peak of Cu^{2+} is a function of composition. The observed optical absorption peak of Cu^{2+} has been found to be minimum at 776 nm for x = 30 mol.% of CdO content. The variations in the physical, optical and bonding parameters clearly indicate the structural changes in the present glass system with varied CdO content.

1. Introduction

Structural studies in glasses are of important owing to the inter-relation between the atomic arrangement and properties. A great deal of work has been carried out in the preparation and analysis of different kinds of glasses which was based on oxides, bismuthates, borates and tellurites. To comprehend structural details, a common strategy is to change the glass structure either by adding network modifiers or by interrogating the material with some external stimuli such as high heat or pressure. Tellurium oxide is an interesting case where the basic building block of structure experiences considerable changes either by modifiers or by increasing temperature, though being of three-dimensional geometry. According to the earlier works, it has been found that addition of TeO_2, into the glass matrices enhance glass quality with an improvement in transparency, refractive index, durability toward moisture and enhancement in the IR transmission in the present work.

Tellurite glasses system is an important amorphous system that possess interesting glass-forming ability, glass structure, no hygroscopic properties and low melting point [1–4] large thermal expansion [5] high refractive indices, high dielectric constants and good IR transmissions, thus they have been considered as promising materials for use in optical fibers and non linear optical devices [6–14]. Tellurite glasses are also important for their high electrical properties [15–19]. These peculiar characteristics of tellurium based glasses attracted researchers toward preparation of combination of glass formers like B_2O_3, V_2O_5, etc. for their desired optical properties. B_2O_3 is one of the best and well known glass former. Addition of small amount of TeO_2 into borate glass network enhances the glass quality with an improvement in transparency, refractive index. The modifier oxides ZnO and CdO are expected to shorten the time taken for solidification of glasses during the quenching process. Both ZnO and CdO are thermally stable, sublime and appreciably covalent in character. In view of these qualities, these modifier oxides are interesting oxides and make the glasses more stable against devitrification and resistant to moisture.

Transition metal ions are being greatly used in the present days to probe the glass structure since their outer d-electron orbital function has a broad radial distribution and due to their high sensitive response changes in the surrounding actions. Among various transition metal ions, the copper ion, a para-magnetic metal ion, when dissolved in glass matrices in very small quantities make the glasses colored and has a strong influence over the optical properties of the glasses.

In the present study, preparation and characterization of 50 B_2O_3–(50 − x) CdO–$x$$TeO_2$ glasses (where x = 0–50 mol.%) has been done by using DSC, ESR and optical absorption studies.

2. Experimental details

2.1. Glass preparation

Glasses were prepared by the conventional melt quenching method. The starting materials, TeO_2 (99.9% pure, Sigma–Aldrich), H_3BO_3, CdO were taken in appropriate proportions and 1 mol.% of CuO was added to each composition as a spin probe. Appropriate amounts of weighed chemicals to constitute a 5 g batch were grounded thoroughly in a motor to produce homogeneous mixture. The batches were then melted in a platinum crucible in a high temperature furnace at 850–1050 °C for

---

* Corresponding author. Tel.: +91 40 27682242; fax: +91 40 27099020.
E-mail address: gayathri.potturi@gmail.com (P. Gayathri Pavani).

0925-8388/$ – see front matter © 2012 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2012.02.142

Table 1
Composition of boro cadmium tellurite glass system.

Sample	C_0	C_1	C_2	C_3	C_4	C_5
B_2O_3 mol.%	50	50	50	50	50	50
CdO mol.%	0	10	20	30	40	50
TeO_2 mol.%	50	40	30	20	10	0

about half an hour until a bubble free liquid was formed, stirred frequently to ensure homogeneity, and quenched on stainless steel plate which is kept at 200 °C and subsequently annealed at 350 °C/2 h to relieve mechanical strains and cracking of the glass samples. The composition of the glass samples employed in present study are shown in Table 1.

2.2. Glass characterization

The amorphous nature of the glass samples was confirmed by X-ray diffraction measurements using Philips X-ray diffractometer PW(1140) at room temperature using copper target (K_α = 1.5406 Å). The density of the samples was determined by standard Archimedes principle with xylene as an immersion liquid on a single pan electric balance to the nearest 0.001 mg. The densities were calculated by using the formula

$$\rho = \frac{a}{a-b}\rho_x(\mathrm{g/cm^3})$$

where a is the weight of the glass sample in air and b is the weight of the sample when immersed in xylene of density (ρ_x) 0.865 g/cm^3. The molar volume (V_m) of each sample glass sample was calculated using the formula

$$V_m = \frac{M}{\rho}(\mathrm{cm^3/mol})$$

where M is the molecular weight of the sample. Molar volume of the oxygen (volume of glass in which one mole of oxygen is contained) is calculated using the equation

$$V_0 = () \times \left(\frac{1}{\sum x_i \times n_i}\right)$$

x_i is mole fraction of each component of oxide I and n_i is the number of oxygen atoms in each oxide.

The glass transition temperature (T_g) was measured using a temperature modulated differential scanning calorimeter (TA instruments DSC). For this purpose, the powdered glass sample was heated in an aluminum pan at standard heating rate of 10 °C/min in the temperature region 30–500 °C using nitrogen as pure gas.

2.3. ESR and optical absorption studies

ESR spectra of all the glass samples were recorded for all the glass samples at the X-band using JEOL-JM FE 3X spectrometer with 100 kHz field modulation at room temperature. DPPH was used as a standard gmarker. The optical absorption spectrum of the present glass samples was recorded at room temperature using a double beam spectrophotometer (Schimadzu) in the wave length region 300–800 nm taking air as reference medium. The peak pick facility provided in the spectrometer was used to measure the peak position.

3. Results and discussion

3.1. Characterization of glasses

The X-ray diffraction pattern of an amorphous material is distinctly different from that of crystalline material and consists of few broad diffuse haloes rather than sharp rings. The XRD patterns of all the samples showed broad humps typical of amorphous materials as shown in Fig. 1.

The density is a powerful tool to explore the structural compactness of the glasses and is affected by structural softening/compactness, changes in geometrical configurations, coordination numbers, crosslink densities and dimensions of interstitial spaces of glass system. Fig. 2 shows the compositional dependence of density and molar volume. In zinc tellurite glass system with an increase of ZnO content density is found to increases [20] and in boro tellurite glasses the density increases with an increase of TeO_2 [21]. However the density of the following system tends to decrease on addition of CdO at the expense of TeO_2. Such result was observed in boro zinc tellurite glass system [22]. This behavior is generally observed when content of modifier oxide is increased in borate glasses. The addition of modifier oxide in the glass system changes the density of the structural units by changing the density of the glass system. Because density is an additive property it can be proposed that each oxide in glass would have its contribution in density. Density at x = 50 mol.% of CdO is 4.288 g/cm^3 (Table 2) and its value increases as content of CdO into glass network decreases (Fig. 2). The decrease in density on increasing the modifier content CdO is due to the substitution of higher molecular weight substance TeO_2 (atomic mass 159.599 g/cm^3) by lower molecular weight substance CdO (atomic mass 128.4 g/cm^3) there by net molecular weight decreases resulting in weak connectivity in glass network [23]. Oxygen packing density (O) is calculated using the relation

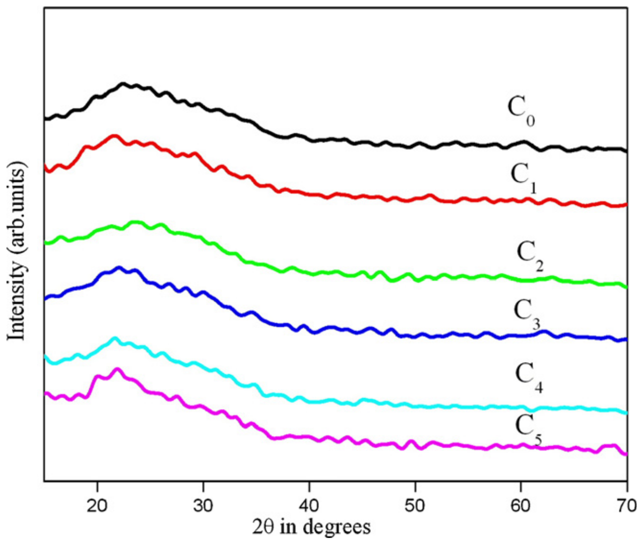

Fig. 1. XRD Spectra of boro cadmium tellurite glass system.

$$\text{Oxygen packing density}(O) = \frac{\rho \times o}{M}$$

O is number of oxygen atoms per formula units and these values are tabulated in Table 3.

It is clear from Fig. 2 that with decrease of CdO, both density and molar volume decreases. In general, the density and molar volume are inversely related. But in the present glasses the behavior is different. However, this anomalous behavior was earlier reported in many glass systems, TeO_2–NbO–Bi_2O_3 [24], Na_2O–B_2O_3–TeO_2

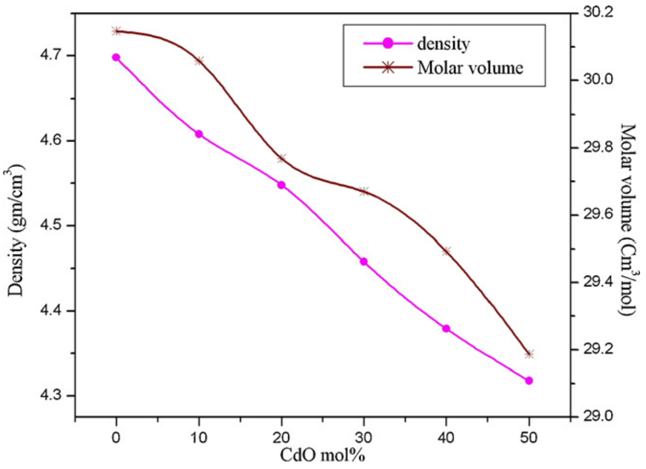

Fig. 2. Variation of density and molar volume with CdO mol.%.

Table 2
Physical parameters of boro cadmium tellurite glass system.

Parameter	$x = 0$	$x = 10$	$x = 20$	$x = 30$	$x = 40$	$x = 50$
Density, ρ (gm/cm^3)	4.698	4.608	4.548	4.458	4.379	4.318
Molar volume, V_m (cm^3/mol)	30.147	30.059	29.769	29.67	29.493	29.187
Oxygen packing density, OPD (mol/l)	82.928	79.844	77.261	74.148	71.203	68.523
Number of bonds per unit volume, n_b (10^{21} cm^{-3})	82.204	82.777	83.929	83.996	85.492	86.191
Cross link density, n_c	1.333	1.333	1.333	1.333	1.333	1.333
Average stretching force constant, F (N/m)	398.813	438.22	477.627	517.034	556.441	595.848
Glass transition temperature, T_g (°C)	317	323	345	410	445	478

Table 3
Spin Hamiltonian parameters of Cu^{2+} ion in boro cadmium tellurite glass system.

Sample	C_0	C_1	C_2	C_3	C_4	C_5
$g_\parallel$	2.208	2.228	2.208	2.194	2.181	2.188
$g_\perp$	2.069	2.076	2.076	2.077	2.079	2.079
$A_\parallel$ (10^{-4}) cm^{-1}	115.6	106.32	114.96	133.26	129.49	140.01

[25], Bi_2O_3–Li_2O–B_2O_3 [26]. In the present study molar volume is directly proportional to glass density and molecular weight. The increase in molar volume from 24.458 to 25.644 cm^3 mol with decrease of CdO indicates an increase in free volume. The larger values of radii and bond length of TeO_2 compared to CdO gave rise to an increase of molar volume.

Variation of oxygen packing density and oxygen molar volume with the composition of CdO is shown in Fig. 3 and the values are given in Table 2. From Table 2 it is clear that oxygen packing density (OPD) value decrease with increase of CdO content, as the substitution of TeO_2 by CdO decreases the number of oxygen atoms in the ratio 1/2.

Differential scanning calorimetry (DSC) is used to characterize the glasses and to determine glass transition temperature (T_g), which is useful in suggesting structural changes that takes place by the compositional changes. This is because T_g is very sensitive to any change of the coordination number of the network-forming atoms and also to the formation of non-bridging oxygen. Typical DSC plot of boro cadmium tellurite glass system (C_3 sample) is shown in Fig. 4. The inset of Fig. 4 shows variation of glass transition temperature with CdO mol.%. The transformation to a glass does not take place at on, strictly defined temperature, but within a temperature range, representing the transformation region. The width of the later depends on the properties of the material studied (low temperature edge) and on the thermal history of the sample (high temperature edge). The single endothermic glass transition peak indicates the homogeneity of the glass. T_g represents the 'strength' or 'rigidity' of the glass structure. Density of boro cadmium tellurite glasses decreased on increasing CdO content. The glass molar volume also decreased. On increasing the CdO content at the expense of heavier TeO_2, the influence of molar mass on density is minimized by the opposite effect of decrease in volume. The glass transition temperature increases with CdO content which indicates that network connectivity increase. This result is consistent with molar volume data, which shows that the network becomes more compact on increasing CdO concentration.

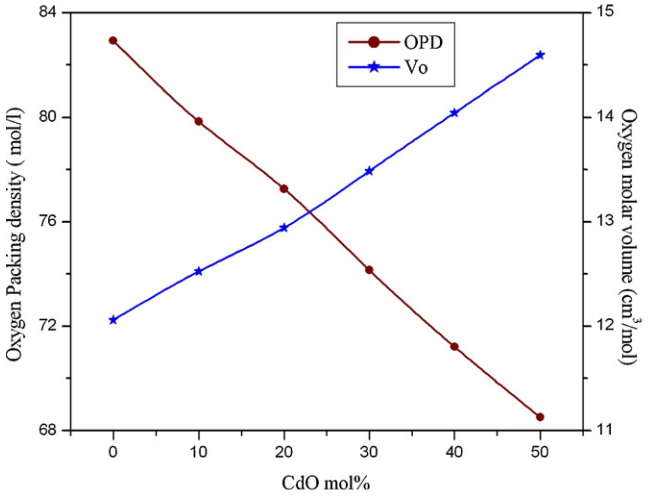

Fig. 3. Variation of oxygen packing density and oxygen molar volume with CdO mol.%.

The glass transition temperature (T_g) depends on a number of parameters. It is more informative for analyzing the glass network to correlate data between thermal and structural properties, between (T_g) and average cross-link density (n_c), number of bonds per unit volume (n_b) and the average stretching force constant (F). The average cross-link density (n_c) of the glass is calculated using the relationship [27–29].

$$\bar{n}_c = \frac{\sum_i (x_i) + (n_c)_i + (N_c)_i}{\sum_i x_i + (N_c)_i}$$

where x_i is mole fraction; n_c is number of cross-links per cations and is equal to the number of bonds minus two; N_c is number of cations per glass formula unit. The increase of CdO content at the expense

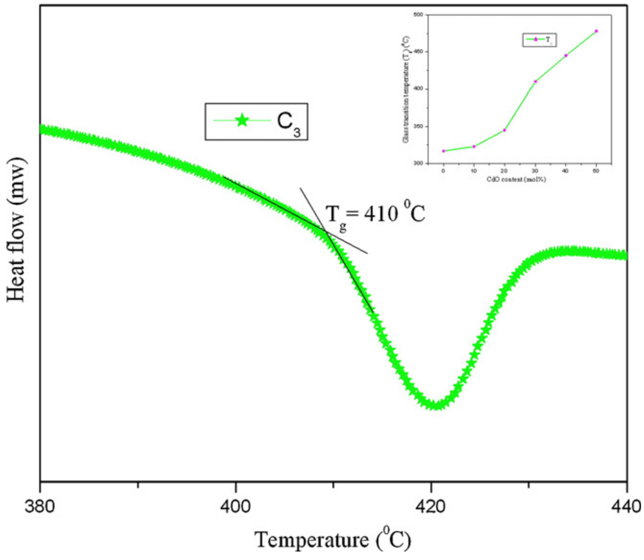

Fig. 4. DSC thermogram and inset representing glass transition temperature with CdO mol.%.

of TeO_2 from $x = 0–10$ mol.% maintains a constant average cross-link density 1.333 as shown in Table 2. This is because the coordination number of CdO and TeO_2 is same. The number of bonds per unit volume (n_b) of the glass is calculated using the relationship [27–29].

$$n_b = \frac{N_a}{V_m}\sum_i (x \times n_f)_i$$

where N_a is Avogadro's number; V_m is molar volume; n_f is coordination number of the cation; and x is mole fraction. The coordination numbers of borate, cadmium and Te are 3, 4 and 4, respectively .The value of n_b for pure TeO_2 is $7.74 \times 10^{28}\,m^{-3}$ [30] The number of bonds per unit volume of the present glass system is found to decrease from 8.619×10^{28} to $8.220 \times 10^{28}\,m^{-3}$ with increasing TeO_2 content as shown in Table 2. Average stretching force constant (F) of the glass is calculated using the relationship [27–29].

$$F = \frac{\sum_i (x \times n_f \times f)_i}{\sum_i (x \times n_f)_i}$$

where n_f is the coordination number of cation; x_i is mole fraction and f_i is given by

$$\bar{f}_i = \frac{17}{r^3}$$

where $\bar{f}_i$ is the stretching force constant of oxide and i denotes the component oxide, and r is the bond length between cation and anion. The average stretching force constant of the bond Te—O is 216 N/m [30]. The average stretching force constant (F) of the present glass system is found to decrease from 215.10 to 213.21 N/m with increasing TeO_2 content (Table 2). Therefore the decrease in T_g of the present glass system can be ascribed due to the decrease in number of bonds per unit volume (n_b) and average stretching force constant (F). The observed decrease in T_g of the present glass system with increase of TeO_2 content in the glass matrix, is shown in inset of Fig. 4. The observed variation in oxygen packing density (OPD) and oxygen molar volume (V_o) values can also be correlated to the decrease in T_g. The glass transition temperature (T_g) is a measure of the strength of a material [31]. Hence, from DSC results, it is concluded that the strength of the present glass system decreases with CdO content.

3.2. Electron spin resonance of the glass system

The electron spin resonance (ESR) is a very powerful technique for investigating paramagnetic centers in oxide glasses containing transition metal oxides and useful for identifying the local environment of a paramagnetic impurity and mapping the crystal field. The ESR spectra of BCT glasses doped with Cu^{2+} ions are shown in Fig. 6. The spectrum closely resembles that of the Cu^{2+} ion in most oxide glasses [32–34]. From the spectrum analysis, the spin-Hamiltonian parameters are calculated using the equation due to [35] Dance et al. and are presented in Table 3. The ESR spectra were analyzed using spin Hamiltonian:$H = g_\parallel \beta H_Z S_Z + g \perp \beta (H_X S_X + H_Y S_Y) + A_\parallel S_Z I_Z + A_\parallel (S_X I_X + S_Y I_Y)$

Here Z is the symmetry axis of individual copper ions, β is the bohr magneton, $A_\parallel$ and $A_\perp$ are parallel and perpendicular components of the hyperfine coupling tensor 'A', $g_\parallel$ and $g_\perp$ are parallel and perpendicular components of 'g' tensor. H_X, H_Y and H_Z are the components of magnetic field. S_X, S_Y, S_Z are the components of spin operators of the electron and I_X, I_Y and I_Z are the spin operators of nucleus. The values of $A_\parallel$ are calculated using the following equation due to Kivelson and Lee [36].$H_\parallel\left(-\frac{3}{2}\right) - H_\parallel\left(+\frac{3}{2}\right) = 3A$

From Table 3 it is clear that $g_\parallel > g_\perp > g_e = 2.0023$ relation, evidencing the Cu^{2+} ions presence in the predominant axially distorted octahedral symmetric site [37]. The g_e is spectroscopic splitting factor corresponding to the free electron. Lack of equidistance between the hfs peaks denotes pronounced ligand and fluctuations in the Cu^{2+} ion is $d_{x_2-y_2}$ orbitals. $g_\parallel$ and $g_\perp$ and $A_\parallel$ values change with the composition CdO in BCT glasses. $g_\parallel$ is maximum for BTC_1, minimum for BTC_3 and $g_\perp$ reaches to maximum for BTC_4 minimum for BTC_1 indicating continuous structural changes in the glass system. Thus from Fig. 5 and Table 3 it is clear that ESR spectra of the boro cadmium tellurite glasses are strongly concentration dependent.

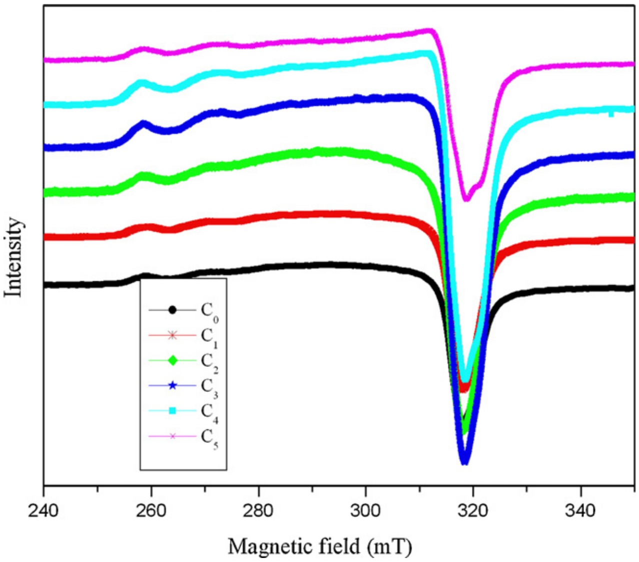

Fig. 5. ESR spectra of boro cadmium tellurite glass system.

From Table 3 it is observed that $g_\parallel$ decreases and reach a minimum in BTC_1 to BTC_3 at about 30% CdO and increase abruptly and reach a maximum at about 40 mol.% CdO. This indicates distortion around Cu^{2+} ion is changing with increase in content of CdO and also indicates a continuous structural change in the glass system.

The variation in Spin-Hamiltonian parameters with increase in concentration of CdO up to 30% in BCT series is due to the structural changes in glass system. This behavior agrees with IR spectra data, which reveal that partial conversion of BO_3 units into BO_4 units as CdO increases.

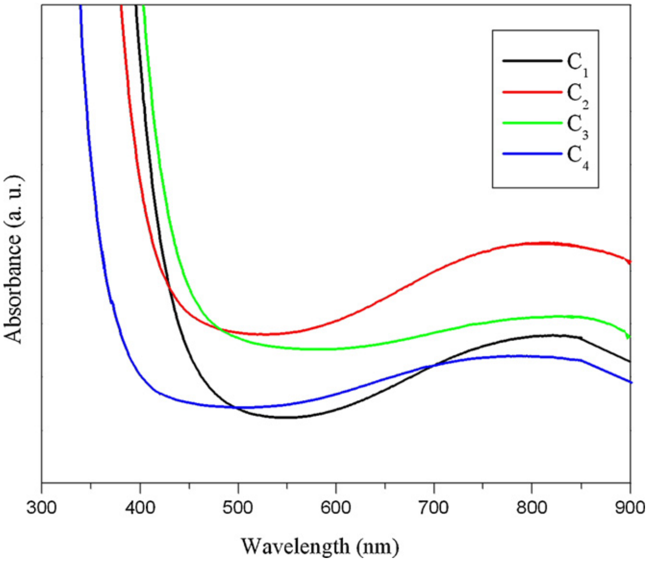

Fig. 6. Optical absorption spectra of boro cadmium tellurite glass system.

Table 4
Absorption peak of Cu^{2+}, bonding parameters and % of bonding symmetry of Cu^{2+} doped in boro cadmium tellurite glass system.

Sample	C_0	C_1	C_2	C_3	C_4	C_5
Cu^{2+} peak (nm)	832	821	812	776	785	772
α^2	0.599	0.596	0.6	0.637	0.614	0.651
β^2	0.985	0.99	0.983	0.926	0.96	0.906
β_1^2	0.623	0.695	0.637	0.584	0.558	0.557
T_σ (%)	75.387	61.017	72.549	83.227	88.389	88.442
T_π (%)	87.419	88.146	87.138	79.149	84.049	76.062

3.3. Optical absorption spectra

Optical absorption spectra for all glass compositions exhibit a single broad peak in the range 772–831 nm in the near infrared region, which can be attributed to presence of Cu^{2+} in glasses [38,39] is shown in Fig. 6. Absorption can be assigned to spin allowed transition from $^2E_g(D) \rightarrow {}^2T_{2g}(D)$ energy levels in distorted octahedral symmetry of Cu^{2+} ions in the glasses. The absorption peak of Cu^{2+} in BCT glasses at different concentrations of CdO is given in Table 4.

The absorption peak of Cu^{2+} in BCT glasses is shifted to lower wavelength with increase in concentration of CdO up to 30% and then peak is shifted toward higher wavelength with further increase in concentration of CdO from 30 to 40%. The variation in peak position with CdO concentration increases fluctuation in ligand field around Cu^{2+} probe ion which is related to change in polarizability of oxygen ions surrounding the Cu^{2+} and its dependence on field strength of network formers [39–41].

The maximum absorption peak in optical absorption in the present system can be explained on the basis of structural change in the glasses with variation of composition of CdO. With increasing CdO content, structural changes take place due to partial conversion of BO_3 units into BO_4 units, TeO_4 units to TeO_3 via TeO_{3+1}. The bonding parameters are calculated using ESR and optical data by the following equations [35]$g_\parallel = 2.0023\left[\frac{1-4\lambda\alpha^2\beta_1^2}{\Delta E_{xy}}\right] g_\perp = 2.0023\left[\frac{1-4\lambda\alpha^2\beta_1^2}{\Delta E_{xz,yz}}\right]$ where λ, spin orbit coupling parameter is equal to 828 cm^{-1} for Cu and $\beta^2 \approx 1$ for octahedral environment. ΔE_{xy} and $\Delta E_{xz,yz}$ are the heights of d_{xy} and $d_{xz,yz}$ and molecular orbital levels above the ground state $d_{x_2-y_2}$, respectively, and these values are estimated from optical absorption spectra [42]. In optical absorption spectra, the position of observed absorption maximum of Cu^{2+} indicates the value of ΔE_{xy} from Table 3, it is observed that the bonding parameters are changing with the percent of CdO. The bonding coefficients α^2, β^2 and β_1^2 characterize respectively, the in-plane σ bonding, in-plane π bonding and out -of-plane π bonding of the copper (II) complex, their values lie between 0.5 and 1.0, the limits of pure covalent and pure ionic bonding. From Table 3 it is clear that, the present system is ionic in nature.

4. Conclusions

Transparent and stable boro cadmium tellurite glass system was obtained by melt quenching technique. The density of the glass system is found to increase with decrease of CdO as higher molecular weight is substituted by lower molecular weight. The larger radii and bond length of TeO_2 compared with CdO caused a decrease in oxygen packing density and molar volume with increase of CdO. The values of network bonds and stretching force constant decreases with decrease in CdO content which is in agreement with decrease in T_g. ESR studies indicate that the structural changes takes place as the concentration of CdO varies. It is observed that $g_\parallel > g_e$. The variation in $g_\parallel$ and $A_\parallel$ values may be due to the change in the environment of Cu^{2+} ion which may be attributed to the structural changes in the glass structure. From the optical absorption it is observed that absorption peak is minimum at $x = 30$ mol.% of CdO. The bonding parameters calculated from both ESR and optical absorption are found to change with CdO concentration concluding that the system is concentration dependent.

Acknowledgment

The author is thankful to DRDO for the financial support and Hyderabad Central University for providing ESR facility.

References

[1] M.A. Sidkey, A. Abd El-Moneim, L. Abd El- Latif, Mater. Chem. Phys. 61 (1999) 103.
[2] M. Dawy, Mater. Chem. Phys. 77 (2002) 48.
[3] J.E. Stanworth, Nature 169 (1952) 581.
[4] J.E. Stanworth, J. Soc. Glass Technol. 38 (1954) 425.
[5] K. Tanaka, T. Yoko, H. Yamada, J. Kamiyama, J. Non-Cryst. Solids 103 (1988) 250.
[6] R. El Mallawany, J. Mater. Res. 7 (1992) 250.
[7] J. Heo, D. Lam, G. Sigel, E. Mendoza, D. Hensley, J. Am. Ceram. Soc. 75 (1992) 277.
[8] A. Abdel Kader, M. Elkholy, J. Appl. Phys. 73 (1993) 71.
[9] J.S. Wang, E.M. Vogel, E. Snitzer, Opt. Mater. 3 (1994) 187.
[10] P. Balaya, C.S. Sunandana, J. Non-Cryst. Solids 162 (1993) 253.
[11] D. Tatar, M.L. Ovecolu, G. Ozen, B. Erim, J. Eur. Ceram. Soc. 29 (2009) 329.
[12] G. Karaduman, M. Celikbilek, A.E. Ersundu, N. Solak, D. Tatar, S. Aydin, Mater. Ceram. 63 (1) (2011) 40.
[13] G. Upender, S. Ramesh, M. Prasad, V.G. Sathe, V.C. Mouli, J. Alloys Compd. 504 (2) (2010) 468.
[14] P. Gayathri Pavani, S. Suresh, V. Chandra Mouli, Opt. Mater. 34 (1) (2011) 215.
[15] A. Ghosh, J. Phys.: Condens. Matter 1 (1989) 7819.
[16] A. Pan, A. Ghosh, Phys. Rev. B 60 (1999) 3224.
[17] D. Dutta, A. Ghosh, Phys. Rev. B 72 (2005) 024201.
[18] A. Ghosh, A. Pan, Phys. Rev. Lett. 84 (2000) 2188.
[19] A. Pan, A. Ghosh, Phys. Rev. B 59 (1999) 899.
[20] J.M. Stevels, Proc. 11th Int. Congr. Pure Appl.Chem. 5 (1953) 519.
[21] M.K. Halimah, W.M. Daud, H.A.A. Sidek, A.T. Zainal, H. Zainul, J. Hassan, Am. J. Appl. Sci. (2005) 63 (special issue).
[22] P. Gayathri Pavani, K. Sadhana, V. Chandra Mouli, Physica B 406 (2011) 1242.
[23] B.V.R. Chowdari, P. Pramoda kumari, Solid State Ionics 113–115 (1998) 665.
[24] Y. Wang, S. Dai, F. Chen, T. Xu, Q. Nie, Mater. Chem. Phys. 113 (2009) 407.
[25] Y. Sadeek, L. Abd El Latif, Physica B 348 (2004) 475.
[26] Y.B. Sadeek, E.R. Shaaban, E.S. Moustafa, H.M. Moustafa, Physica B 403 (2008) 2399–2407.
[27] B. Bridge, A.A. Higazy, Phys. Chem. Glasses 27 (1) (1986) 1.
[28] B. Bridge, Phys. Chem. Glasses 28 (1987) 70.
[29] E. Lambson, G. Saunders, B. Bridge, R.A. El-Mallawany, J. Non-Cryst. Solids 69 (1984) 117.
[30] R.A. El-Mallawany, Mater. Chem. Phys. 63 (2000) 109.
[31] S.R. Elliott, Physics of Amorphous Materials, Longman Scientific and Technical, London, 1990.
[32] L.D. Bogomolova, A.V. Jackhin, J. Non-Cryst solids 63 (1984) 223.
[33] S. Suresh, Swapna, V. Chandramouli, Ferroelectrics 41 (2003) 954.
[34] R.P. Srikanth Chakradher, B. Yasoda, J. Lakshman Rao, N.O. Gopal, J. Non-Cryst. Solids 352 (2006) 3864.
[35] J.M. Dance, J. Dranudery, H. Baudry, M. Moneraye, Solid State Commun. 39 (1981) 199.
[36] D. Kivelson, S.K. Lee, J. Chem. Phys. 47 (1986) 11.
[37] S. Suresh, J. Chinna babu, V. Chandramouli, Phys. Chem. Glasses 46 (2005) 27.
[38] S. Sakka, K. Kamira, K. Matika, Yamanato, J. Non-Cryst. Solids 63 (1984) 223.
[39] P.W. Franc, S.F. Carter, J.M. Parker, Phys. Chem. Glasses 46 (2005) 27.
[40] M.S. Selim, E. Metwalli, Mater. Chem. Phys. 78 (2002) 94.
[41] T. Sekiya, N. Mochida, A. Ohtsuka, M. Tonokowa, J. Ceram. Soc. Jpn. 96 (1988) 973.
[42] J. Krosh-Moe, Phys. Chem. Glasses 6 (1965) 46.