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## Research Article

### SPECTROSCOPIC PROPERTIES OF LEAD BORATE GLASSES DOPED WITH COPPER IONS

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#### ABSTRACT

40PbO-(60-x)B<sub>2</sub>O<sub>3</sub>: (x)CuO glasses with the values of x ranging from 0.1-0.4 mol % in steps of 0.1 were prepared. A number of Spectroscopic studies viz. x- ray diffraction, optical absorption, IR Spectra and ESR Spectra were carried out. The Optical absorption spectra suggests that Cu<sup>2+</sup> ions occupy octahedral positions in larger concentrations in the titled glass network. The EPR spectra of the glasses suggests that With successive replacement of B<sub>2</sub>O<sub>3</sub> with CuO, the values of g tensors are observed to decrease. The IR spectral studies point out an increase in the degree of disorder of the glass network. As the concentration of CuO is increased in the glass matrix, the intensity of the vibrational band due to BO<sub>3</sub> groups is observed to increase while the band due to BO<sub>4</sub> structural units is observed to decrease. Overall the analysis of the results of various spectroscopic studies of CuO doped PbO-B<sub>2</sub>O<sub>3</sub> glasses, there is a gradual increase in the degree of depolymerization of the glass network.

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#### INTRODUCTION

Copper oxide based glasses, like PbO-B<sub>2</sub>O<sub>3</sub> glasses, have attracted extensive investigation in recent years, in view of their potential applications in non-linear optical devices in a number of solid state ionic devices. Attempts were made to improve the physical properties of PbO based glasses by the addition of alkali oxides (like Li<sub>2</sub>O), alkali halides (like NaCl, NaI) and alkali sulphides (like Na<sub>2</sub>S, K<sub>2</sub>S) [1-3]. Alkali fluorides are expected to make the glasses more moisture resistant when compared with the alkali oxides and iodides. Copper ions have strong bearing on electrical, optical and magnetic properties of glasses. A large number of interesting studies are available on the environment of copper ion in various inorganic glass systems [4-8]. These ions subsist in different surroundings (ionic, covalent) in glass matrices. The content of copper in different environments exist in the glass depends on the quantitative properties of modifiers and glass formers, size of the ions in the glass structure, their field strength, mobility of the modifier cat ion etc. Hence, the connection between the position of the copper ion and the physical properties of the glass is highly interesting. Cu<sup>2+</sup> ions are well-known paramagnetic ions and it is also quite likely for

copper ions to have link with borate groups. Though considerable number of studies is available on some CuO containing glasses most of them are restricted to spectroscopic studies [9-12]. Most of the studies available on lead borate glasses doped with copper ions are on the understanding of their structural aspects [3, 13, 14] and some electrical properties [15, 16]. In this work we propose to investigate the spectroscopic properties of glasses which may help to throw some light on the structural aspects of the glasses [17,18]. Thus the clear objective of this paper is to have a comprehensive understanding over the influence of change of environment of copper ions (by a progressive replacement of B<sub>2</sub>O<sub>3</sub> by CuO) in lead borate glasses by investigating their spectroscopic properties.

#### MATERIALS AND METHODS

Within the glass forming region of PbO-B<sub>2</sub>O<sub>3</sub> glass system, the following particular compositions with successive increase in the concentration of PbO (with simultaneous decrease in the concentration of B<sub>2</sub>O<sub>3</sub>) and with variable concentrations of CuO (0.1-0.4 mol %) are chosen for the present study. The details are as follows:

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- C<sub>0</sub>: 40PbO-60 B<sub>2</sub>O<sub>3</sub>
- C<sub>1</sub>: 40PbO-59.9B<sub>2</sub>O<sub>3</sub>: 0.1CuO
- C<sub>2</sub>: 40PbO-59.8 B<sub>2</sub>O<sub>3</sub>: 0.2CuO
- C<sub>3</sub>: 40PbO-59.7 B<sub>2</sub>O<sub>3</sub>: 0.3CuO
- C<sub>4</sub>: 40PbO-59.6B<sub>2</sub>O<sub>3</sub>: 0.4CuO

The glasses used for the present study are prepared by the melting and quenching techniques[19-21]. The starting materials used for the preparation of the present glasses were Analytical grade reagents (99.9 % pure) of PbO, H<sub>3</sub>BO<sub>3</sub>, CuO. The compounds of required compositions were thoroughly mixed in an agate mortar and melted in a platinum crucible. The furnace used was a PID temperature controlled furnace. The glasses were melted at about 950 -1000 °C for an 1 h till a bubble free liquid was formed. The resultant melt was poured on a rectangular brass mould (containing smooth polished inner surface) held at room temperature. The samples were subsequently annealed at 300 °C in another furnace. The glasses were then ground and optically polished. The approximate final dimensions of the glasses used for studying the electrical and optical properties are 1 cm x 1 cm x 0.2 cm.

The density (d) of the glasses was determined by the standard principle of Archimedes’ using o-xylene (99.99 % pure) as the buoyant liquid. A direct reading balance (capacity 100 g, readability 0.1 mg) was used for weighing. The bulk glass was suspended on a very thin copper strand was set in the immersion liquid container and weighed in the liquid and air. Using density and average molecular weight other physical parameters such as Copper ion concentration N<sub>i</sub>, the inter atomic distance r<sub>i</sub> of these glasses are evaluated. The crystalline phases in the glass ceramic samples were checked by X-ray diffraction spectra recorded on SEIFERT Diffractometer Model SO-DEBYE FLUX 202 fitted with copper target and nickel filter operated at 40 kV, 30 mA. The optical absorption spectra of the present glasses were recorded using a Shimadzu-311 UV-VIS-NIR Spectrophotometer in the wavelength range 200-1200 nm. The electron Paramagnetic resonance spectra of PbO-B<sub>2</sub>O<sub>3</sub> glasses doped with copper metal ions were recorded using commercial X-Band microwave spectrometer in the frequency range 8.8 to 9.6 GHz manufactured by JOEL, Model No. FA series 200. Infrared transmission spectra were recorded on a Bruker IFS 66 V – IR spectrophotometer with a resolution of 0.1 cm<sup>-1</sup> in the range 400-2000 cm<sup>-1</sup> using potassium bromide pellets (300 mg) containing pulverized sample (1.5 mg).

## RESULTS

### Characterization and Physical Parameters

#### X-ray diffraction Spectra

As glass or amorphous materials do not have long range order, a diffraction pattern containing sharp peaks is not expected as in crystalline materials.

The X-ray diffraction pattern for PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses recorded in the range 10° ≤ 2θ ≤ 80° is shown in fig1. The absence of sharp peaks in the pattern indicates the amorphous nature of the samples.

From measured values of density (d) and calculated average molecular weight (M̄), various physical parameters such as

copper ion concentration (N<sub>i</sub>), mean copper ion separation (R<sub>i</sub>), are evaluated for understanding the physical properties of these glasses using the standard formulae and are presented in Table 1.

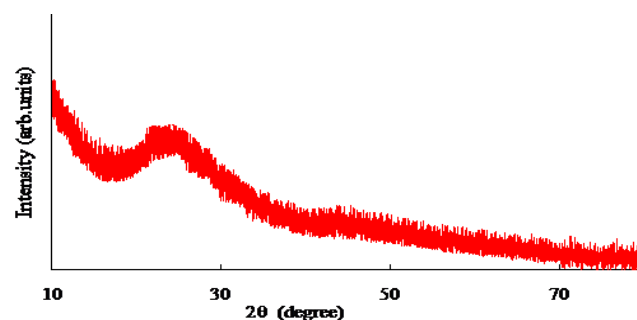


Fig 1 XRD Pattern of PbO – B<sub>2</sub>O<sub>3</sub> glass doped with CuO.

Table 1 Summary of data on various physical parameters of PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses.

Glass	d (g/cm <sup>3</sup> )	N <sub>i</sub> (10 <sup>21</sup> /cm <sup>3</sup> )	r <sub>i</sub> (Å)
C <sub>0</sub>	5.27	--	--
C <sub>1</sub>	5.28	0.18	8.20
C <sub>2</sub>	5.29	0.35	6.53
C <sub>3</sub>	5.30	0.54	5.69
C <sub>4</sub>	5.31	0.71	5.18

#### Infrared transmission spectra

Fig.2 represents the FT-IR spectra of PbO-B<sub>2</sub>O<sub>3</sub>:CuO glasses were recorded in the wave number region 400-1600 cm<sup>-1</sup>. The spectra exhibited bands due to B-O vibrations in BO<sub>3</sub> and BO<sub>4</sub> structural units at about 1370 and 1030 cm<sup>-1</sup>, respectively. The band due to vibrations of B-O-B linkages is also observed at 705 cm<sup>-1</sup>. The peak at 470 cm<sup>-1</sup> related to symmetric bending vibration of Pb-O in PbO<sub>4</sub> tetragonal pyramid (PbO covalent bond) is also located in the spectra. The summary of the peak positions of various vibrational bands observed the IR spectra is presented in Table 2.

Table 2

Glass	BO <sub>3</sub> units	BO <sub>4</sub> units	B-O-B linkages	PbO <sub>4</sub> units
C <sub>0</sub>	1338	1005	705	470
C <sub>1</sub>	1335	1010	707	473
C <sub>2</sub>	1332	1013	710	475
C <sub>3</sub>	1329	1020	712	478
C <sub>4</sub>	1330	1017	715	480

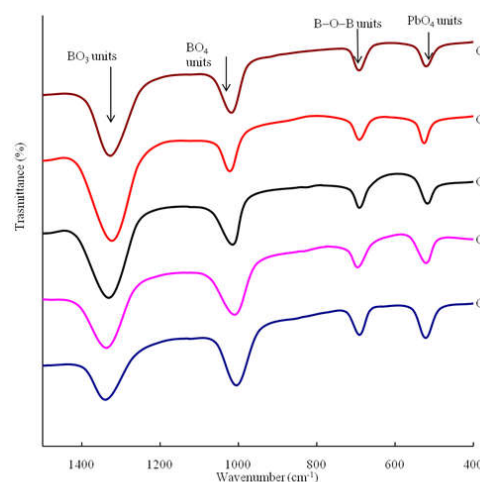


Fig. 2 IR spectra of PbO-B<sub>2</sub>O<sub>3</sub> glasses doped with CuO

Further, it is also observed that there is an increase in the intensity of the band due to BO<sub>3</sub> structural units and the decrease in the intensity of the band due to BO<sub>4</sub> units is decreased with the concentration of CuO in the glass matrix. Summary of data on the positions (cm<sup>-1</sup>) of the bands in IR spectra of PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses

**Optical absorption studies**

Fig.3 represents the optical absorption spectra of PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses recorded at room temperature in the wavelength region 300-1000 nm. The spectrum of CuO free glass has not exhibited any absorption bands. The spectra of CuO containing glasses have exhibited a broad absorption band at about 765 nm corresponding to <sup>2</sup>B<sub>1g</sub>→<sup>2</sup>B<sub>2g</sub> transition of Cu<sup>2+</sup> ions [22, 23]; the successive replacement of B<sub>2</sub>O<sub>3</sub> by CuO has caused a shift of peak position towards slightly higher wavelength. The details of the optical absorption data of the present glasses are given in Table 3.

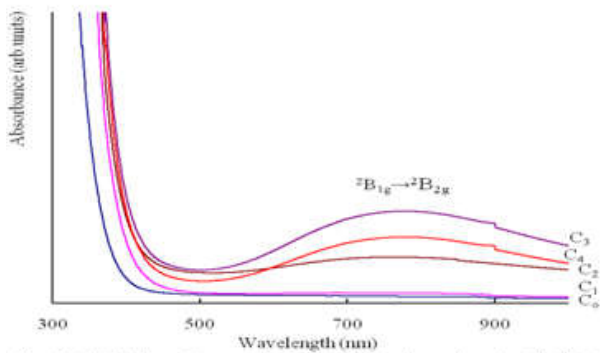


Fig. 3 Optical absorption spectra of PbO-B<sub>2</sub>O<sub>3</sub> glasses doped with CuO

**Table 3** Data on optical absorption and EPR spectra of PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses.

Glass	<sup>2</sup> B <sub>1g</sub> → <sup>2</sup> B <sub>2g</sub> Band position (nm)	g <sub>  </sub>	g <sub>⊥</sub>	α <sup>2</sup>	β <sub>1</sub> <sup>2</sup>
C <sub>0</sub>	—	—	—	—	—
C <sub>1</sub>	765	2.350	2.085	0.66	0.79
C <sub>2</sub>	766	2.431	2.076	0.68	0.84
C <sub>3</sub>	768	2.427	2.077	0.73	0.88
C <sub>4</sub>	767	2.447	2.078	0.78	0.98

**Electron paramagnetic resonance (EPR) studies**

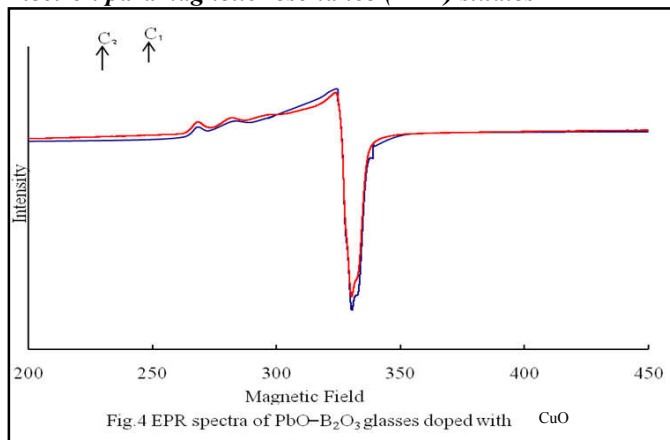


Fig. 4 EPR spectra of PbO-B<sub>2</sub>O<sub>3</sub> glasses doped with CuO

EPR spectra of PbO-B<sub>2</sub>O<sub>3</sub> glasses containing different concentrations of CuO recorded at room temperature. The spectra obtained for two of the glasses are shown in the Fig.4. The structure and the intensity of the signal are observed to be

strongly dependent on the concentration of CuO. The spectra of the glass C<sub>1</sub> exhibited an asymmetric signal with a hyperfine structure partially resolved with a value of g<sub>⊥</sub> = 2.085 and a shallow quadruplet at g<sub>||</sub> = 2.35. As the concentration of CuO is increased in the glass matrix the signal broadened progressively and the hyperfine structure smeared and exhibited a broad absorption at g<sub>⊥</sub> = 2.085. The hyperfine parallel components of the spectra have also exhibited a similar behaviour. The pertinent data corresponding to the EPR spectra of PbO-B<sub>2</sub>O<sub>3</sub>: CuO glasses are presented in the Table 3.

**DISCUSSION**

B<sub>2</sub>O<sub>3</sub> is a strong glass network former and participates in the glass network with BO<sub>3</sub> and BO<sub>4</sub> structural units. Pb and CuO act as modifiers. When the concentration of CuO is increased the intensity of band due to BO<sub>3</sub> structural units is observed to increase with increase of CuO content. The observed increase in the intensity of the band due to BO<sub>3</sub> structural units at the expense of band due to BO<sub>4</sub> units with the increase in the concentration of CuO indicates an increase in the degree of disorder in the glass network.

The free ion term Cu<sup>2+</sup> (d<sup>9</sup>) is <sup>2</sup>D. In octahedral crystal field it splits into <sup>2</sup>E<sub>g</sub> and <sup>2</sup>T<sub>2g</sub> with <sup>2</sup>E<sub>g</sub> being the lower level. <sup>2</sup>E<sub>g</sub> generally splits due to Jahn-Teller effect. Therefore Cu<sup>2+</sup> is rarely found in regular octahedral site. Accordingly, in the present investigation, Cu<sup>2+</sup> is taken to be octahedrally coordinated by six oxygen atoms and octahedron is tetragonally distorted. Hence in the tetragonally distorted octahedral environment, the <sup>2</sup>E<sub>g</sub> level splits into <sup>2</sup>A<sub>1</sub> and <sup>2</sup>B<sub>1</sub>, and <sup>2</sup>T<sub>2g</sub> level into <sup>2</sup>E and <sup>2</sup>B<sub>2</sub>, the ground state being <sup>2</sup>B<sub>1</sub>. With this model the EPR spectra are explained on the basis of the conventional spin-Hamiltonian [24]

$$\mathcal{R} = \beta_e H_g S + SAI \tag{1}$$

here β<sub>e</sub> is Bohr magneton, S=1/2 for Cu<sup>2+</sup> ions and I=3/2 for copper nucleus, <sup>63</sup>Cu / <sup>65</sup>Cu. The first term represents electronic Zeeman interaction and the second term represents the interaction of unpaired electron spin with the nuclear spin.

The observed g<sub>||</sub> and g<sub>⊥</sub> values (g<sub>||</sub> =2.35 and g<sub>⊥</sub> = 2.085 ) in EPR spectra of PbO-B<sub>2</sub>O<sub>3</sub>:CuO glasses are characteristic of Cu<sup>2+</sup> ions coordinated by six ligands which form an octahedron elongated along the z- axis [25]. Because g<sub>||</sub> > g<sub>⊥</sub> > 2.0023, the ground state of the paramagnetic electron is the d<sub>x<sup>2</sup>-y<sup>2</sup></sub> orbital (<sup>2</sup>B<sub>1g</sub> state). The EPR and optical absorption spectra data can be correlated to understand the environment of Cu<sup>2+</sup> ions PbO-B<sub>2</sub>O<sub>3</sub> glass network as follows [22]:

$$g_{||} = 2.0023 \left[ 1 - \frac{4\lambda\alpha^2\beta_1^2}{E(^2B_{2g})} \right] \tag{2}$$

$$g_{\perp} = 2.0023 \left[ 1 - \frac{\lambda\alpha^2\beta^2}{E(^2E_g)} \right], \tag{3}$$

$$\alpha^2 = E_1/\lambda [1 - g_{\perp}/g_e]$$

$$\beta_1^2 = E_2/4\lambda\alpha^2 [1 - g_{||}/g_e]$$

where λ is the spin-orbit coupling coefficient and the bonding coefficients α<sup>2</sup>, β<sub>1</sub><sup>2</sup> and β<sup>2</sup> (=1.00) characterize respectively,

the in-plane  $\sigma$  bonding, in-plane  $\pi$  bonding and out-of-plane  $\pi$  bonding of the Cu(II) complexes. Their values lie between 0.5 and 1, the limits of pure covalent and pure ionic bonding [23]. The value of  $\beta^2$  may be expected to lie sufficiently close to unity as to be indistinguishable from unity in the bonding coefficients calculations [25]. Since only one absorption band corresponding to the transition  ${}^2B_{1g} \rightarrow {}^2B_{2g}$  has been observed, the position of the second band has been estimated using the equation [23]:

$$E({}^2B_{1g} \rightarrow {}^2E_g) = \frac{2k_{\perp}^2 \lambda}{2.0023 - g_{\perp}} \quad (4)$$

where  $k_{\perp}$  is the orbital reduction factor ( $k_{\perp}^2 = 0.77$ ) and  $\lambda$  the spin-orbit coupling constant ( $= -828 \text{ cm}^{-1}$ ) [26].

The values of  $g_{\parallel}$  and  $g_{\perp}$  are obtained for the present glasses are comparable to those obtained for a variety of other glass systems (Table 4)

**Table 4** Comparison of  $g$  values of  $\text{Cu}^{2+}$  ions in different glass systems.

System	$g_{\parallel}$	$g_{\perp}$	Ref.
SrO-B <sub>2</sub> O <sub>3</sub>	2.308	2.044	[27]
ZnO-B <sub>2</sub> O <sub>3</sub>	2.321	2.039	[28]
PbO-B <sub>2</sub> O <sub>3</sub>	2.323	2.042	[28]
CdSO <sub>4</sub> -B <sub>2</sub> O <sub>3</sub>	2.422	2.084	[29]
Li <sub>2</sub> SO <sub>4</sub> -CdSO <sub>4</sub> -B <sub>2</sub> O <sub>3</sub>	2.421	2.089	[29]
Na <sub>2</sub> SO <sub>4</sub> -CdSO <sub>4</sub> -B <sub>2</sub> O <sub>3</sub>	2.420	2.086	[29]
K <sub>2</sub> SO <sub>4</sub> -CdSO <sub>4</sub> -B <sub>2</sub> O <sub>3</sub>	2.420	2.089	[29]
CaSO <sub>4</sub> -CdSO <sub>4</sub> -B <sub>2</sub> O <sub>3</sub>	2.421	2.086	[29]
Li <sub>2</sub> CO <sub>3</sub> -BaCO <sub>3</sub> -H <sub>3</sub> BO <sub>3</sub>	2.284	2.053	[30]
Na <sub>2</sub> CO <sub>3</sub> -BaCO <sub>3</sub> -H <sub>3</sub> BO <sub>3</sub>	2.262	2.049	[30]
K <sub>2</sub> CO <sub>3</sub> -BaCO <sub>3</sub> -H <sub>3</sub> BO <sub>3</sub>	2.259	2.048	[30]
Li <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -PbO-TeO <sub>2</sub>	2.323	2.067	[31]
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -PbO-TeO <sub>2</sub>	2.293	2.056	[31]
K <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -PbO-TeO <sub>2</sub>	2.291	2.051	[31]
Li <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -TeO <sub>2</sub>	2.310	2.063	[31]
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -TeO <sub>2</sub>	2.303	2.060	[31]
K <sub>2</sub> B <sub>4</sub> O <sub>7</sub> -TeO <sub>2</sub>	2.294	2.056	[31]
LiRbB <sub>4</sub> O <sub>7</sub>	2.445	2.0561	[32]
NaRbB <sub>4</sub> O <sub>7</sub>	2.443	2.0505	[32]
KRbB <sub>4</sub> O <sub>7</sub>	2.421	2.0590	[32]
20 LiF-09.6 Sb <sub>2</sub> O <sub>3</sub> -70 B <sub>2</sub> O <sub>3</sub> :0.4 CuO	2.456	2.065	Present study

The optical absorption spectra of CuO doped PbO-B<sub>2</sub>O<sub>3</sub> glasses exhibited a broad absorption band at about 765 nm. This is the characteristic of distorted octahedral symmetry. Thus the band can be assigned to  ${}^2B_{1g} \rightarrow {}^2B_{2g}$  transition [22,23]. As the concentration of CuO is increased there is a gradual increase in the concentration of structural defects such as dangling bonds, non-bridging oxygens (NBO's) in the glass matrix that lead to increase of the disorder in the glass network. Such increase of disorder in the network results decrease in the optical bandgap and shifts absorption edge towards higher wavelength side as observed in Fig. 3 for the glasses C<sub>1</sub> to C<sub>4</sub>. The increase in the intensity of the band due to vibrations of BO<sub>3</sub> structural units with the increase of CuO may also be explained based on the similar conclusions.

## References

1. M. Nalin, M. Poulain, M. Poulain, J.L. Ribeiro, Y. Messaddeq, *J. Non-Cryst. Solids*. 284(2001) 117.
2. B.V. Raghavaiah, C. Laxmikanth, N. Veeraiah, *J. Optic Comm.* 235 (2004) 341

3. A. Mori, Y. Ohishi, M. Yamada, H. Ono, Y. Nishida, K. Oikawa, S. Sudo, *Proc. O F C*, 97 (1997) 1.
4. Sung-Ping Szu, Chung-yilin, *Mate. Chem. phy.* 82 (2003) 295.
5. R. V. S. S. N Ravikumar, R. Komatsu, K. Ikeda, A. V. Chandraseker, B.J Reddy, Y. P. reddy, P.S. Rao *J. Phys. Chem. Solids* 64 (2003) 261; *Physica B*, 334 (2003) 398.
6. Lucian Baia, Razvan Stefan, Wolfgang Kiefer, Jugen Popp, Simion Simon *J.Non-Cryst. Solids* 303 (2002) 379.
7. A. Chahine, M. Et-tabirou, M. Elbenaissi, M. Haddad, J. L. Pascal, *Materials. Chem. Phy.* 84 (2004) 341.
8. R. P. Sreekanth Chakradhar, A. Murali, J. Lakshmana Rao *J. Alloys and Compounds*, 265 (1998) 29.
9. I. Ardelean, M. Peteanu, V. Simon, O. Cozar, F. Ciorcas, S. Lupsor *J. Magnetism and Magnetic Materials*, 196 (1999) 253.
10. I. Bratu, I. Ardelean, A. Barbu, D. Maniu, C. Borsa *J. Molecular Structure* 482 (1999) 675.
11. A. Chahine, M. Et-tabirou, M. Elbenaissi, M. Haddad, J. L. Pascal, *J. Mat. Res. Bulliten.* 37 (2002) 1973.
12. A. Duran, J.M. Fernandez Navarro, *Phys. Chem. Glasses* 26 (1985) 126.
13. R.P. Tandon, S. Hotchandani, *Phys. Stat. Sol. (a)* 185 (2001) 453.
14. V. Sudarsan, S.K. Kulshreshtha, *J. Non-Cryst. Solids.* 286 (2001) 99.
15. A. Bishay, C. Maghrabi, *Phys. Chem. Glasses.* 10 (1969) 1.
16. D. Chakravarty, H. Satou, H. Sakata, *J. Appl. Phys.* 82 (1997) 5520.
17. P. Subbalakshmi, N. Veeraiah, *J. Non-Cryst. Solids* 298 (2002) 89.
18. D.K. Durga, N. Veeraiah, *J. Mater. Sci.* 36 (2001) 5625.
19. A. Paul. Chemistry of glasses, Chapman & Hall, London (1982).
20. S. R. Elliot. Physics of amorphous materials, (Longman, London (1990).
21. J. F. Shackl Ford. Introduction to materials science for Engineers (Macmillan, New York, 1985).
22. D. Kivelson and R. Neiman, *J. Chem. Phys.* 35 (1961) 149.
23. A. Klonski, *Phys. Chem. Glasses* 24 (1983) 166.
24. A. Abragam, M.H.L. Pryce. *Proc. Roy. Soc.* 205A (1951) 135.
25. H. Imagawa, *Phys. Stat. Sol. (b)*, 30 (1968) 469.
26. F. E. Mabbs and P. J. Maclin, Magnetism Transition Metal Complexes (Chpman and Hall, London 1973) 154.
27. V.P. Seth, A. Yadav, *J. Mater. Sci.* 22 (1987) 239.
28. A. Yadav, V.P. Seth, S.K. Gupta, *J. Non-Cryst. Solids*, 101 (1988) 1.
29. A.S. Rao, J. Laxma Rao, S.V.J. Laxman, *J. Phys. Chem. Solids* 53 (1992) 1221.
30. I. Ardelesn, M. Peteanu, V. Simon, S. Filip, G. Gyosffy *Solid State Comm.* 102 (1997) 341.
31. A. Murali, J. Lakshmana Rao *J. Phys. Condens. Matter* 11(1999) 7921.
32. R. V. S. S. N Ravikumar, R. Komatsu, K. Ikeda, A. V. Chandraseker, B.J Reddy, Y. P. reddy, P.S. Rao *J. Phys. Chem. Solids* 64 (2003) 261; *Physica B*, 334 (2003) 398.