

**Effect of Gamma Irradiation on the Physical Properties of Some  
Oxide Glasses used in Radiation Dosimeter**

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**Abstract**

In this work the effect of gamma irradiated on the structure, electric and magnetic properties of some borophosphate glasses containing iron were studied in a trial to decide if it can be used as radiation dosimeter. The change in the studied properties was presented as functions of the chlorine ions content and radiation dose. Samples of alkali borophosphate glasses containing iron oxide of the base composition 0.30 mol % B<sub>2</sub>O<sub>3</sub>, 0.30 mol % P<sub>2</sub>O<sub>5</sub>, 0.25 mol % Na<sub>2</sub>O, 0.25 mol % NaCl and 0.15 mol % FeO were prepared and irradiated by gamma rays with different doses (5, 10, 20 kGy). ME spectroscopic analysis was done to the prepared samples and the results showed two oxidation states of the iron Fe<sup>2+</sup> and Fe<sup>3</sup>. Also the analysis indicate that with increasing radiation doses and chlorine ion Fe<sup>3+</sup> content increase while Fe<sup>2+</sup> content decrease. The dielectric constant and the dielectric loss showed gradually increase as the radiation doses was increased.

**Keywords:** Phosphate glasses, radiation dosimeter, gamma irradiation, radiation doses

## INTRODUCTION

Phosphate glasses are the most common type of glasses has a wide range of application because of their unique properties. They have a very high transmission in the ultraviolet region compared to silicate glasses. Many applications were found to depend on the low transition temperature of some alkali-aluminum phosphite glasses. Other applications are also dealt with connection with organic polymers (**Mogus-Milankovi, 2001**).

Borophosphate glasses (**Wakabayash, 1996**) are multi-component glasses of high durability comparing with pure borate or phosphate glasses. They have found industrial appealing applications as solid electrolytes and glass solders. The basic building units of these glasses are BO<sub>4</sub>, PO<sub>4</sub> tetrahedral and BO<sub>3</sub> triangles. These glasses offer the possibility to change their structure and properties on changing their composition.

### Effect of Radiation on Glasses

The effect of gamma irradiation on the physical properties and chemical properties of glass and glass-ceramics are studied by (**Asia H. Al-Mashhadani 2013**). This study showed that the value of glass density decreased after irradiation but the density of glass-ceramic increases after irradiation with gamma doses, after that the densities remain constant with increasing radiation dose.

There are two reasons for the interest in the effect of radiation on the glass (**Levy, 1964**). Firstly the effect of radiation is in itself important because the glass is much as widely used as a material of construction and secondly it is a useful method for investigating the glass structure. When the glass is exposed to radiation, numerous changes in its physical properties can take place, such as optical absorption bands may be induced, density of the glass can change, disruption of the glass structure can take place, paramagnetic defect centers can be formed, macroscopic phase separation can be induced and also the physical strength of the glass can be changed, just to mention a few (**Clark L. Allred 2003**).

There are two principle interactions of radiation with glass (**Donnell, 1968**) the ionization of electrons and the direct displacement of the atoms by elastic scattering when the glass is subjected to ionizing radiation (X or  $\gamma$  - rays, photons, ultraviolet light or charge particles such as electron or protons), the electrons are ionized from the valence band if the energy of radiation is greater than the band gap and the excess energy is converted to kinetic energy. Thus, the electrons move through the glass matrix and will either be trapped by pre-existing flaws to form defect centers in the glass structure, recombine with the positively charged holes or at high energy Compton electrons produce a secondary electron cascade by knock on collisions with the bound electrons. The secondary electrons continue traveling through the matrix, ionizing additional bound electrons by coulombs interactions, losing approximately 20eV of energy for each ionization. Finally, the electrons will have too low energy to ionize additional

electrons and they will either become trapped at defect sites or recombine with the positive holes in the matrix, 4.25eV must be transferred to an atom to remove it from its normal position in the lattice. The actual color (**Wilson, 1974**) of the irradiated glass depends on the impurities present, the irradiation history, the temperature of irradiation and the thermal history of the glass sample. When the glass is irradiated, strong optical absorptions in the visible spectral region are generated in addition to the intrinsic bands. Several bands seem to be related to aluminum and/or the alkali impurities. The optical absorption bands induced by the irradiation depends on the number of parameters including the type of glass former ( $\text{SiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{P}_2\text{O}_5$ ... etc), the type and the concentration of the alkali or the alkaline earth present, the concentration of the OH group, the redox conditions during the glass melting and the temperature at which the glass is irradiated and measured. The behavior of the multi component glasses expose to radiation is important since these glasses are used as lenses, windows and other optical elements in a large number of different optical devices. These glasses are typically much more sensitive to the radiation than pure silica unless they have been initially doped with radiation protecting elements.

## EXPERIMENTAL WORK

### Sample preparation

A sodium borophosphate glass system containing iron oxide was prepared according to the basis of percentage molecular weight composition. In these samples Cl content was increased with constant concentrations of  $\text{B}_2\text{O}_3$ ,  $\text{P}_2\text{O}_5$  and FeO according to the following

formula:-

(0.30) mol % of  $\text{B}_2\text{O}_3$ , (0.30) mol %  $\text{P}_2\text{O}_5$ , (0.25-X) mol %  $\text{Na}_2\text{O}$ , (X)mol % NaCl, (0.15) mol % FeO

Where X= 0.0, 0.05, 0.10, 0.15, 0.20, and 0.25, as shown in table (1).

**Table 1.** Samples composition

Sample No.	$\text{B}_2\text{O}_3$	$\text{P}_2\text{O}_5$	$\text{Na}_2\text{O}$	NaCl	FeO
1	0.30	0.30	0.25	0.00	0.15
2	0.30	0.30	0.20	0.05	0.15
3	0.30	0.30	0.15	0.10	0.15
4	0.30	0.30	0.10	0.15	0.15
5	0.30	0.30	0.05	0.20	0.15
6	0.30	0.30	0.00	0.25	0.15

### Irradiation procedures

All the glasses under investigation were exposed to gamma radiation using an Indian  $^{60}\text{Co}$  gamma ray cell (1000 Ci) with dose rate using 1.2 Gy/ sec. In which each glass sample was placed in a manner that it was subjected to the same radiation dose. This has been achieved by arranging the glass samples around a cylinder placed inside the chamber. The required dose achieved by calculating the desired overall dose.

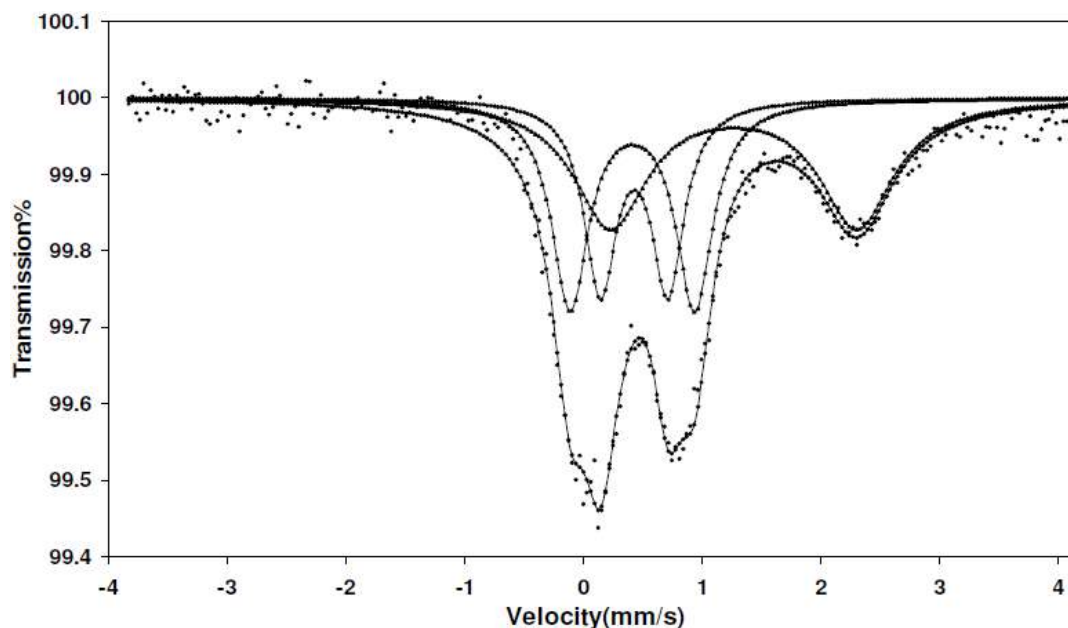
## RESULT AND DISCUSSION

### Effect of gamma irradiation

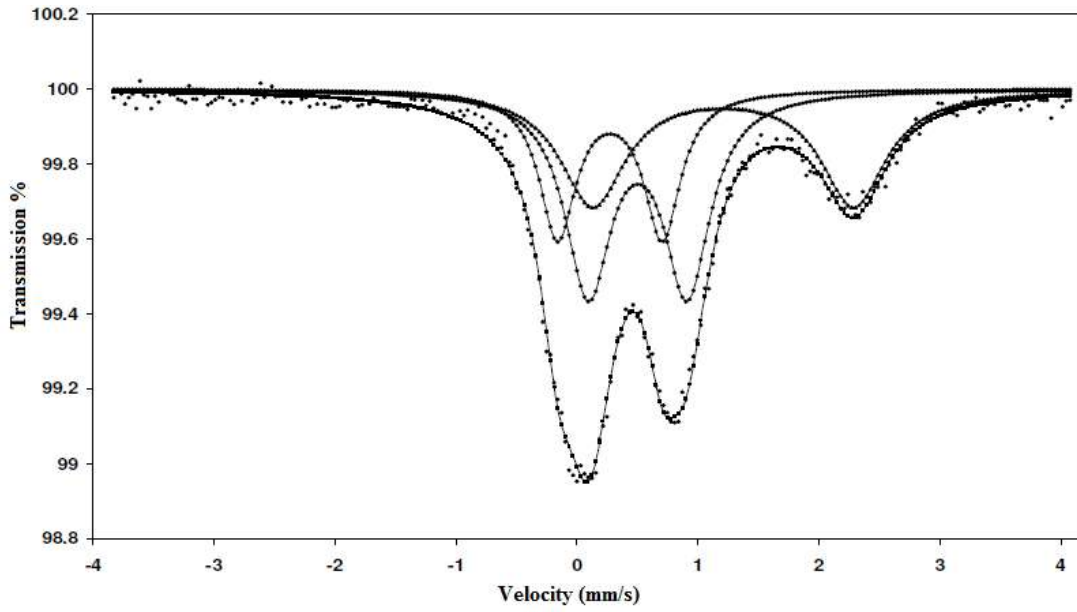
The effect of gamma irradiated on the structure, electric and magnetic properties of some borophosphate glasses containing iron were studied in a trial to decide if it can be used as radiation dosimeter.

### Effect of gamma irradiation on the structure of alkali borophosphate glasses

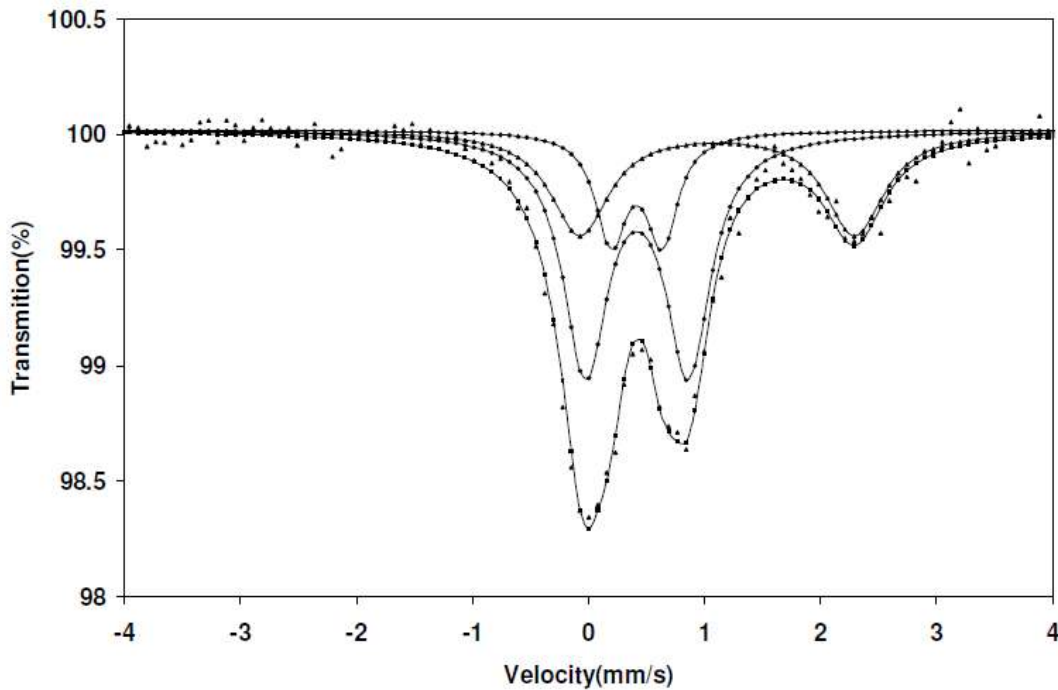
When some selected alkali borophosphate glasses containing iron oxide of the base composition:- (0.30) mol %  $\text{B}_2\text{O}_3$ , (0.30) mol %  $\text{P}_2\text{O}_5$ , (0.25-X) mol %  $\text{Na}_2\text{O}$ , (X) mol %  $\text{NaCl}$ , (0.15) mol %  $\text{FeO}$ , where  $X = 0.0, 0.25$  were subjected to gamma radiation dose 5,10 and 20kGy, ME spectra was found to be changed as gamma radiation. Dose was gradually increased as given in the following.



**Fig.1.** ME Spectra at  $X = 0.00$  (0.00 mol %  $\text{NaCl}$ ) Subjected to radiation dose = 0 kGy



**Fig.2.** ME Spectra at X= 0.00 (0.00 mol % NaCl) Subjected to radiation dose = 5 kGy



**Fig.3.** ME Spectra at X=0.00 (0.0 mol % NaCl) Subjected to radiation dose = 10 kGy

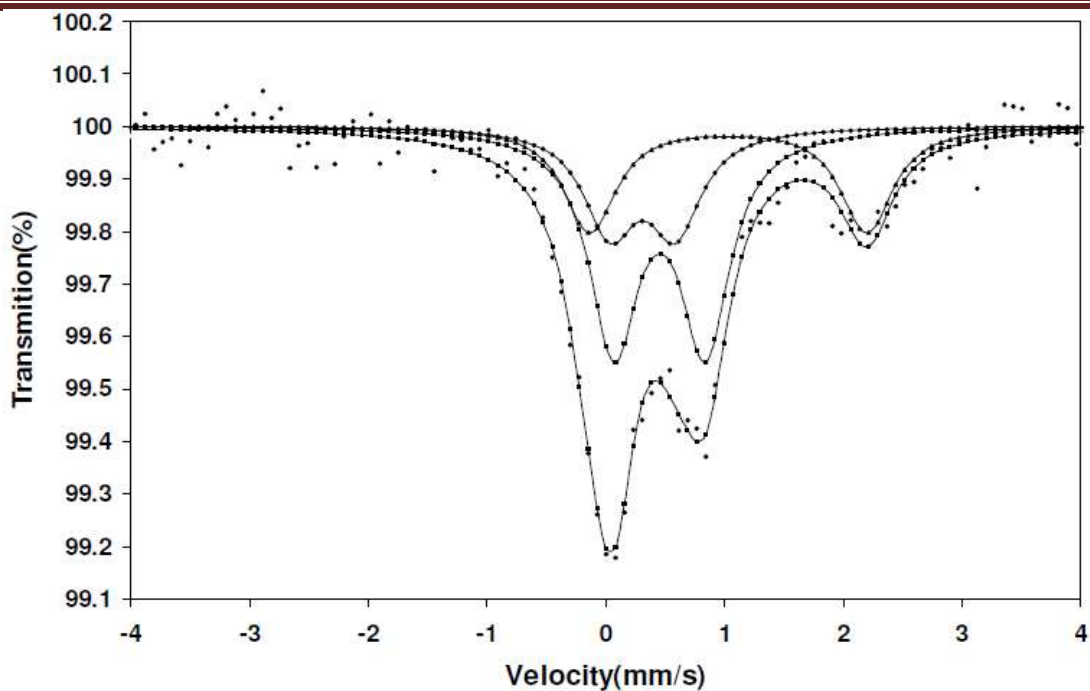


Fig.4. ME Spectra at X= 0.00 (0.0 mol % NaCl) Subjected to radiation dose= 20 kGy

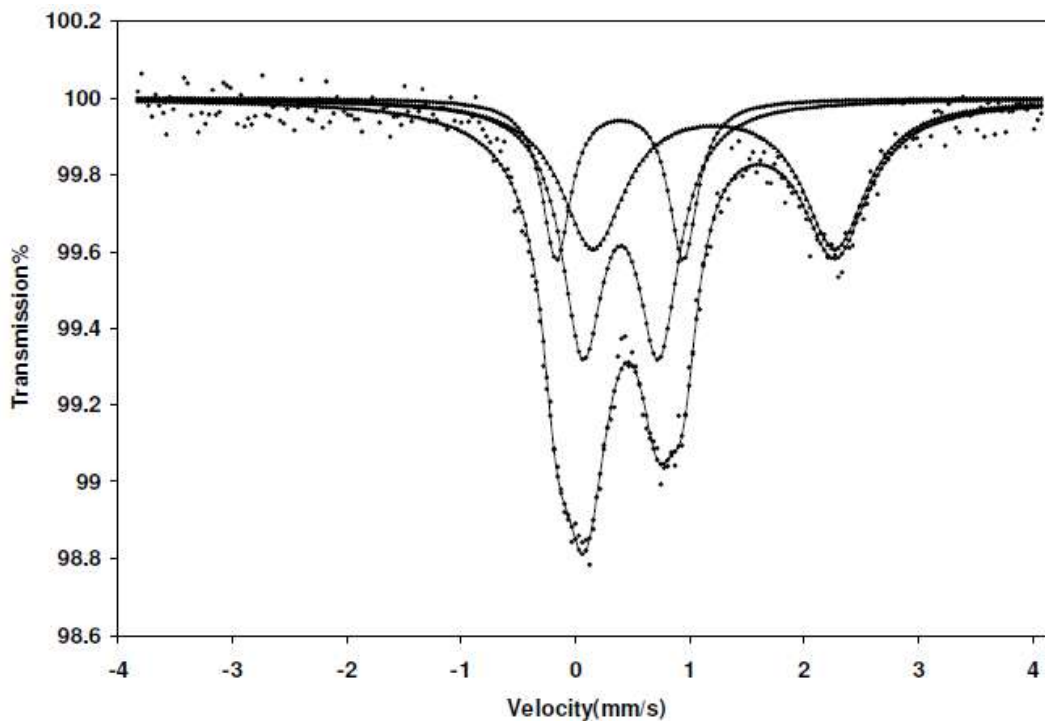


Fig.5. ME Spectra at X=0.25 (0.25 mol% NaCl) Subjected to radiation dose= 0.0 kGy

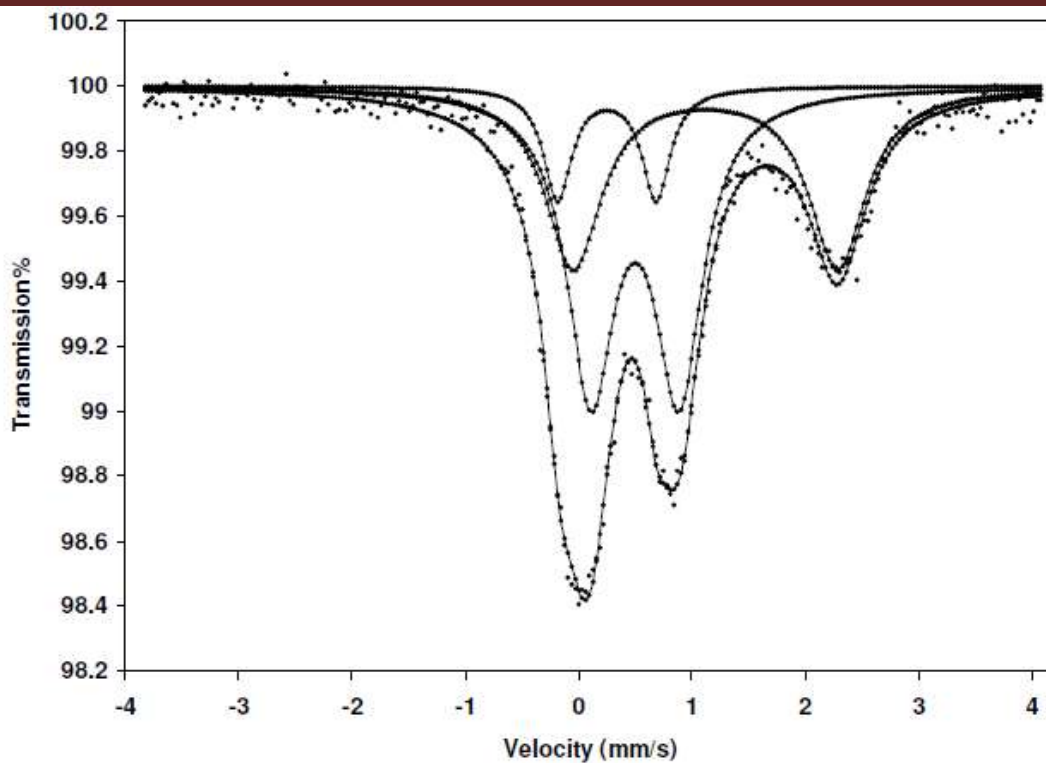


Fig.6. ME Spectra at X = 0.25 (0.25 mol % NaCl) Subjected to radiation dose = 0.5 kGy

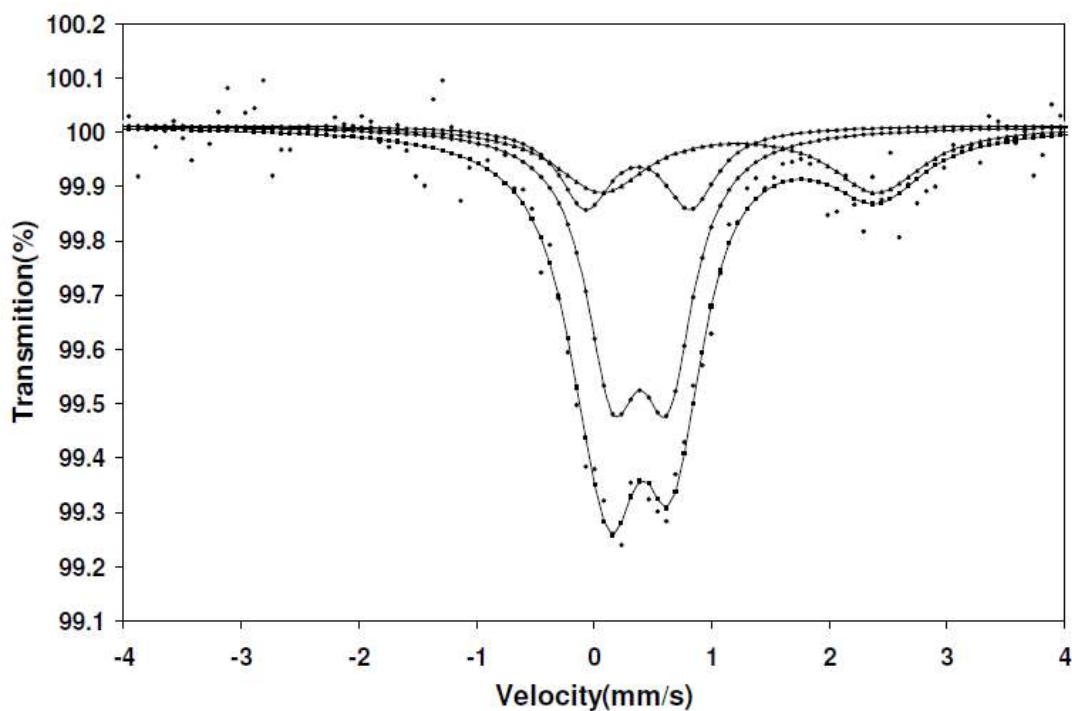


Fig.7. ME Spectra at X = 0.25 (0.25 mol % NaCl) Subjected to radiation dose = 10 kGy

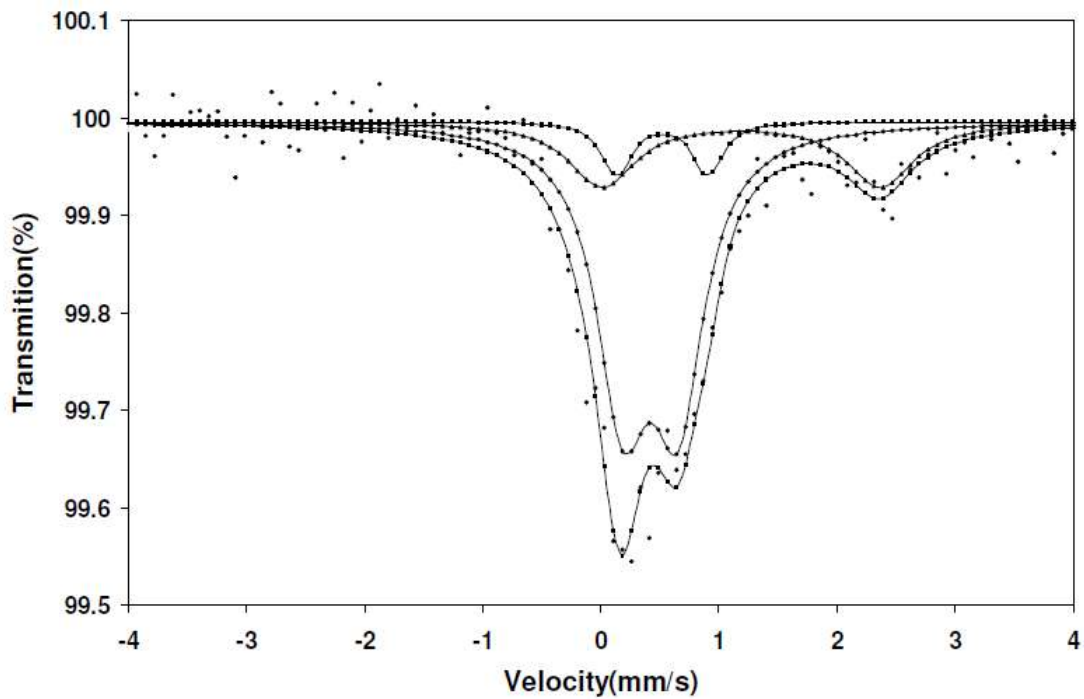


Fig.8. ME Spectra at X = 0.25 (0.25 mol % NaCl) Subjected to radiation dose = 20 kGy

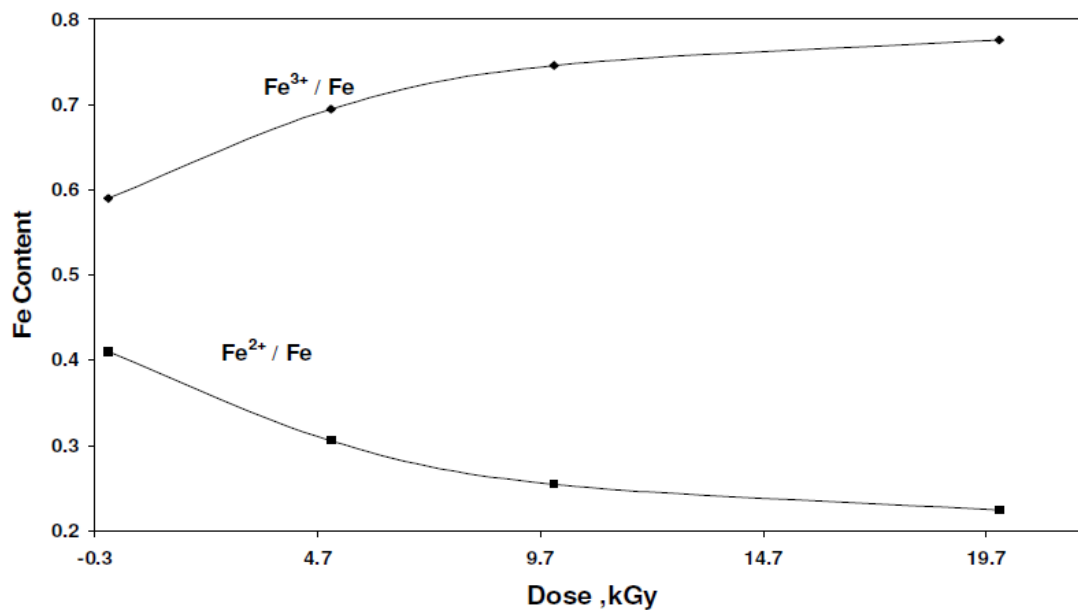


Fig.9.  $Fe^{2+}/Fe$  and  $Fe^{3+}/Fe$  as function of radiation dose at NaCl and X= 0.00



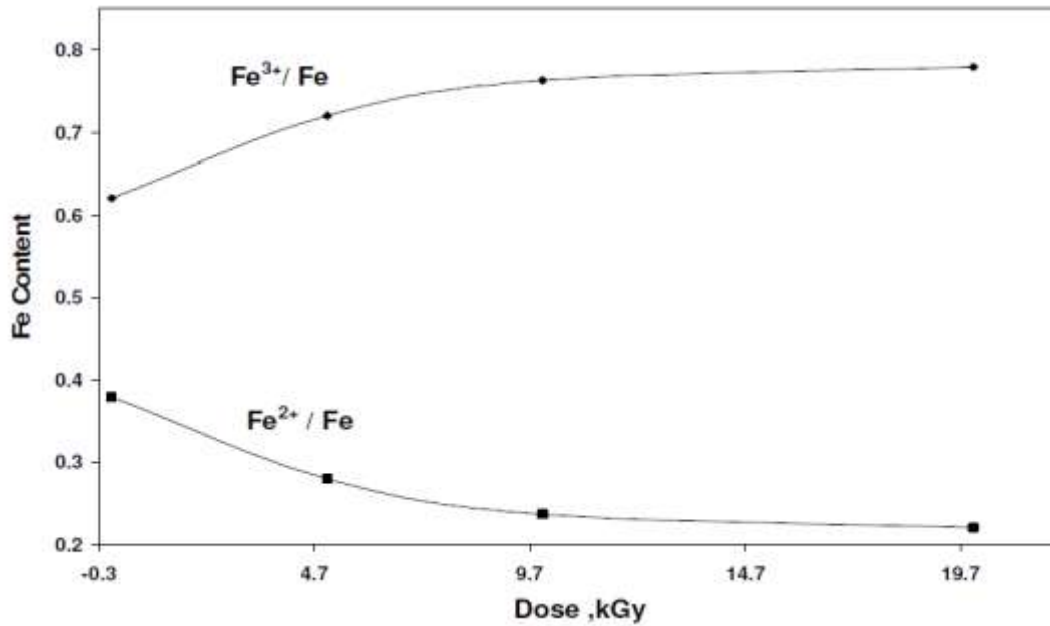


Fig.10. Fe<sup>2+</sup>/Fe and Fe<sup>3+</sup>/Fe as function of radiation dose at Na<sub>2</sub>O and X= 0.25

From figure ( 9) and figure ( 10), Fe<sup>3+</sup> content showed an increase while Fe<sup>2+</sup> content showed a decrease as the radiation doses was increased and this is attributed to the ionization act due to gamma irradiation.

#### Effect of gamma irradiation on the AC conductivity

When some selected alkali borophosphate glasses containing iron oxide of the base composition: - (0.30) mol % B<sub>2</sub>O<sub>3</sub>, (0.30) mol % P<sub>2</sub>O<sub>5</sub>, (0.25-X) mol % Na<sub>2</sub>O, (X) mol % NaCl, (0.15) mol % FeO.

Where X= 0.0, 0.25 were subjected to gamma radiation dose 5, 10 and 20 kGy.

The AC conductivity showed gradual increase as the radiation doses were increased. The effects of exposing the studied glasses to gamma irradiation (El-Alaily 1988, El-Hadi 1993, Kumar 2007).were attributed to irradiation-induced changes in the configuration of the glass network, including the formation of matrix defects. Gamma irradiation of glass (El-Hadi 1993, Kumar 2007, Cornelius 1998) is reported to lead mainly to surface damage, unstable charging, and migration of mobile (nonnetwork) cations. The electron hole trapping sites in borate glass are believed to arise from the local non-stoichiometry, which is present in the glass as a result of either fabrication or radiation induced atomic displacement. the effect of increasing the electrical conductivity of the glass irradiated with gamma rays at a dose 5, 10 and 20kGy may be attributed to the initial formation of the induced color centers and the decrease in the number of the intrinsic defects that have no trapped electrons or holes in the glass network structure. The increased electrical conductivity of the glasses subjected to the higher gamma irradiation doses may also be due to the decreased density of the glass,

which permits higher and continuous running of ions through holes in the glass network. Alternatively, this decreased density may offer an easier path for current flow through the specimens.

Ac conductivity was found to change as gamma radiation dose was gradually increased as given in the following figures:-

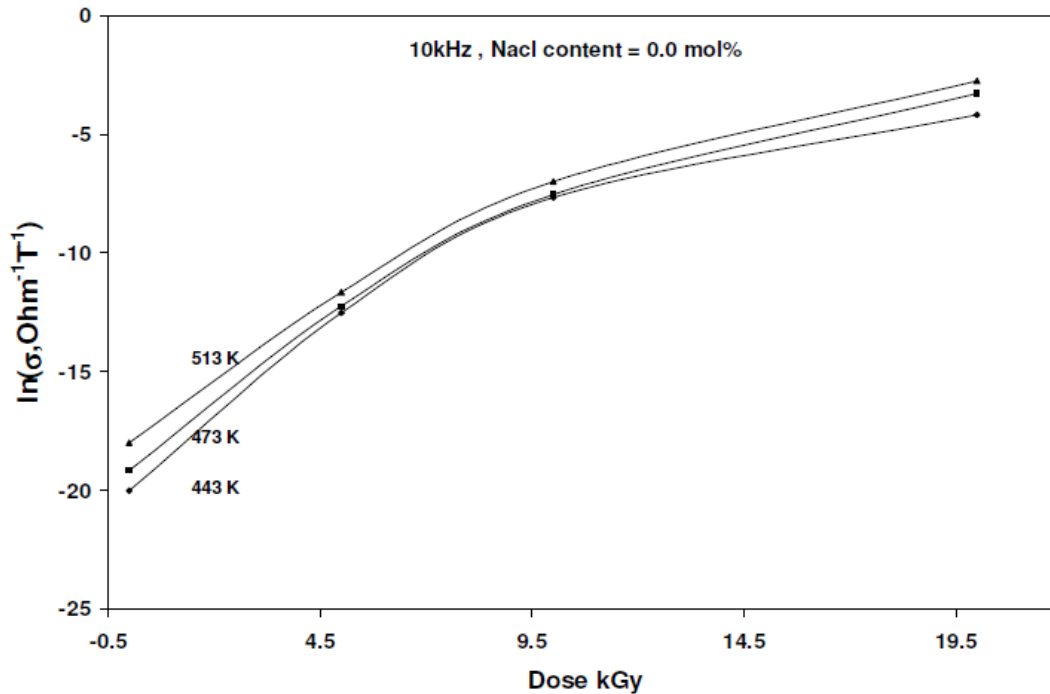


Fig.11. Ac conductivity as function of radiation dose at NaCl = 0.00 mole %

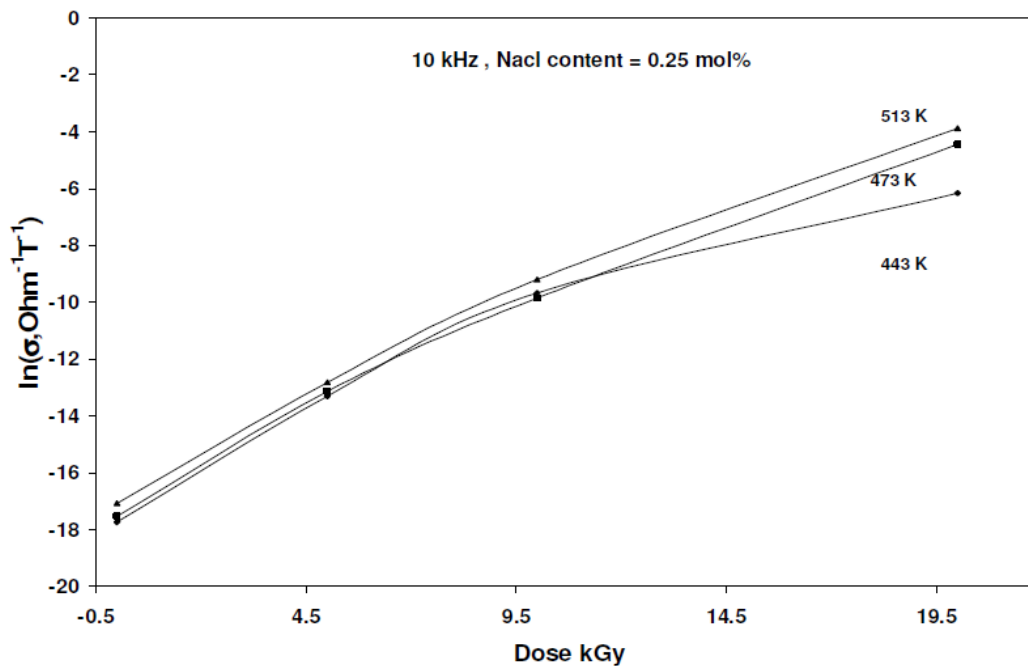
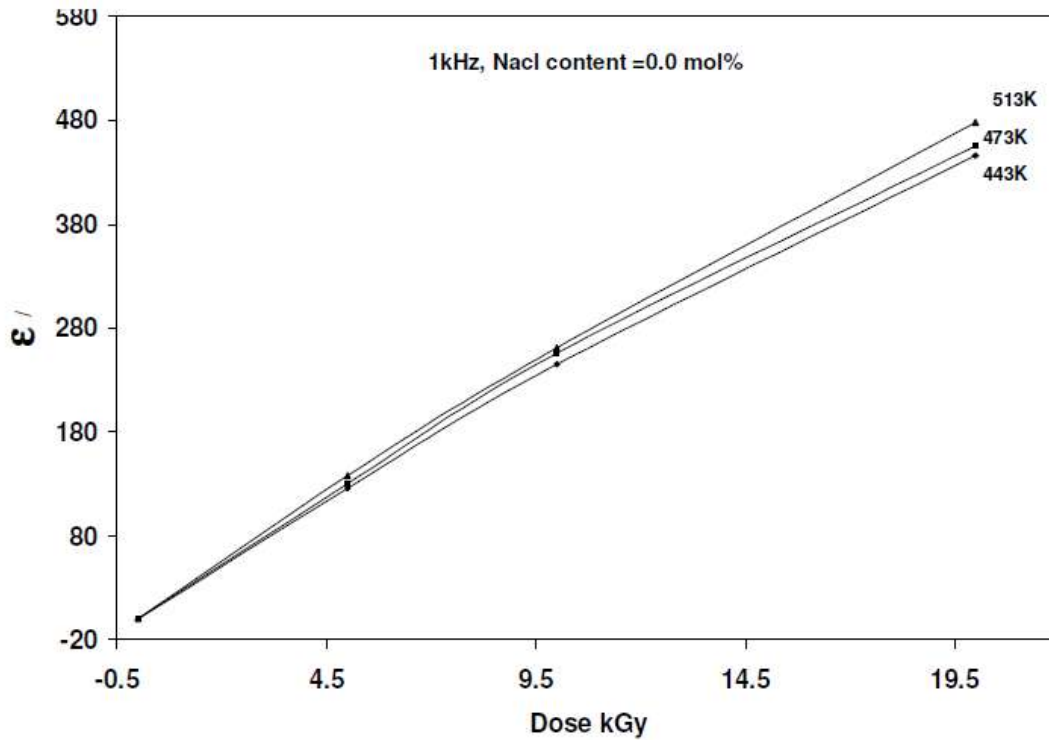


Fig.12. Ac conductivity as function of radiation dose at NaCl = 0.25 mole %

**Effect of gamma irradiation on the dielectric constant ( $\epsilon$ )**

When some selected alkali borophosphate glasses containing iron oxide of the base composition: - (0.30) mol %  $B_2O_3$ , (0.30) mol %  $P_2O_5$ , (0.25-X) mol %  $Na_2O$ , (X) mol %  $NaCl$ , (0.15) mol %  $FeO$ ,  $X=0.0, 0.25$  were subjected to gamma radiation dose 5, 10 and 20 kGy. The dielectric constant ( $\epsilon$ ) showed gradually increase as the radiation doses were increased, and this is attributed to the gamma radiation ability for ionization and induced structural defects and color centers formation as given in the following figures:-.



**Fig.13.** Dielectric constant ( $\epsilon$ ) as function of radiation dose at NaCl = 0.00 mole

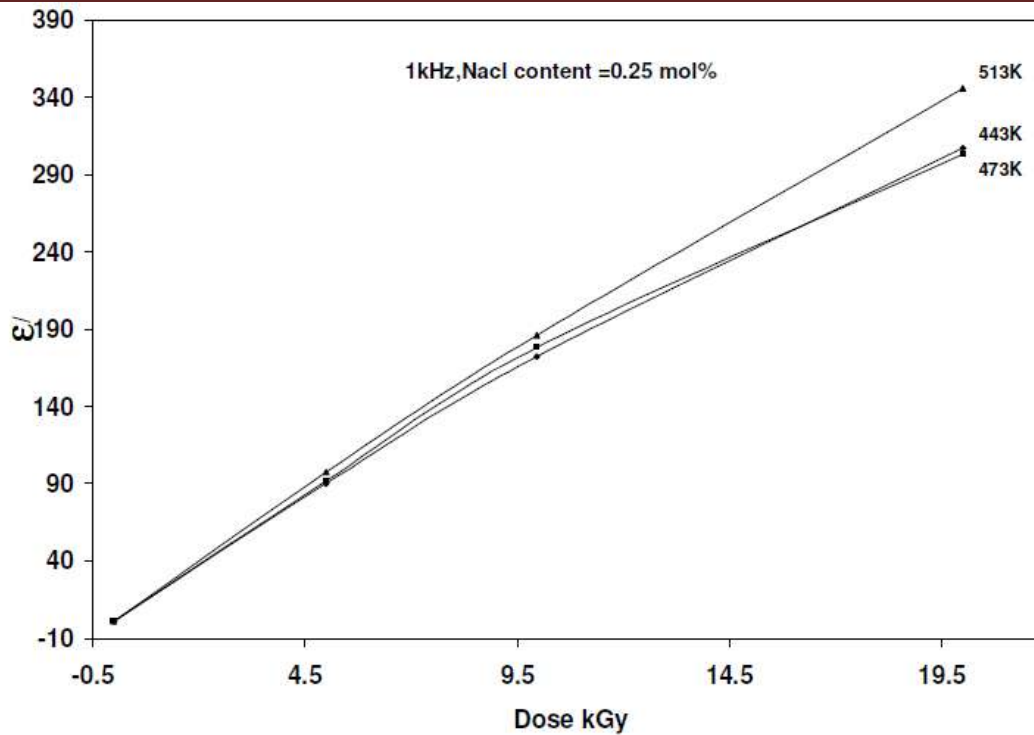


Fig.14. Dielectric constant ( $\epsilon'$ ) as function of radiation dose at  $\text{Na}_2\text{O} = 0.00$  mole

#### Effect of gamma irradiation

When some selected alkali borophosphate glasses containing iron oxide of the base composition: - (0.30) mol %  $\text{B}_2\text{O}_3$ , (0.30) mol %  $\text{P}_2\text{O}_5$ , (0.25-X) mol %  $\text{Na}_2\text{O}$ , (X) mol %  $\text{NaCl}$ - (0.15) mol %  $\text{FeO}$ . X = 0.0, 0.25 were subjected to gamma radiation dose 5, 10 and 20 kGy. The dielectric loss ( $\epsilon''$ ) showed gradually increase as the radiation doses were increased, and this is attributed to the gamma radiation ability for ionization and induced a structural defects and color centers formation as given in the following:-.

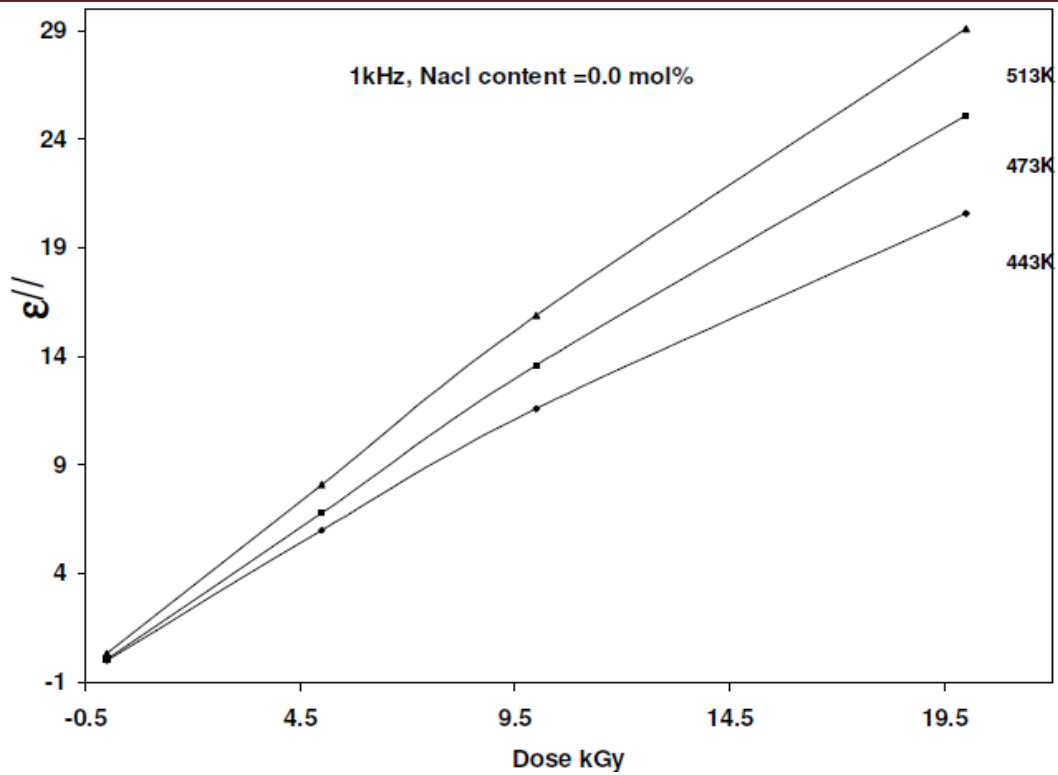


Fig.15. Dielectric loss ( $\epsilon''$ ) as function of radiation dose at NaCl = 0.00 mole

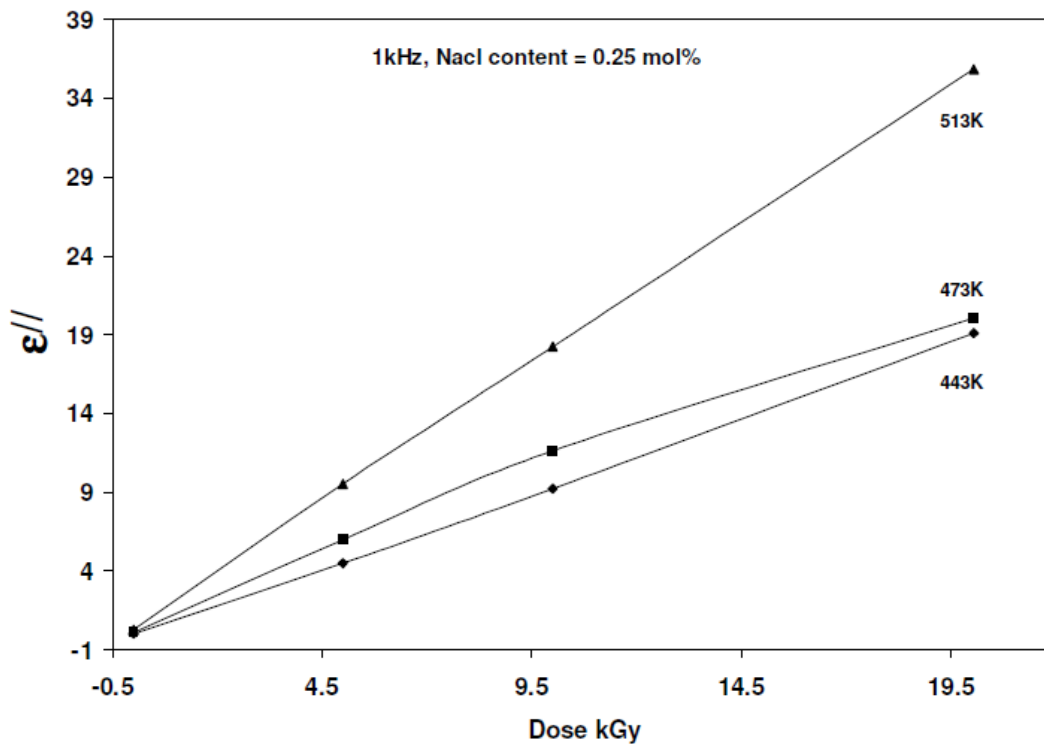


Fig.16. Dielectric loss ( $\epsilon''$ ) as function of radiation dose at Na<sub>2</sub>O = 0.00 mole

### CONCLUSION

All the investigated glasses appeared to be in good homogenous and glassy state. ME spectroscopic analysis showed the presence of the two oxidation states of the iron  $Fe^{2+}$  and  $Fe^{3+}$ . It indicated also that all  $Fe^{2+}$  ions occupied octahedral coordination states, while  $Fe^{3+}$  ions occupied tetrahedral and octahedral coordination states,  $Fe^{3+}$  content showed an increase while  $Fe^{2+}$  content showed a decrease as chlorine content and radiation doses were increased and this is attributed to the oxidation effect of both chlorine and gamma irradiation. The AC conductivity showed gradually decrease as the chlorine content was increased. The effect of gamma irradiation on the conductivity and dielectric properties of these glasses shows an increase as well as a slight response in ME parameters, so it can be used in radiation dosimetry especially in food irradiation processes and in dose mapping of gamma irradiation cells for flux symmetry checking.

### REFERENCES

- Asia H. Al-Mashhadani; Thoria Amer Al-Dafae; Hayder S. Hussain, Iraqi Journal of Physics, Vol.11, No. 21, (2013) 75-83.
- Bih. L, Al Omari M, Reau J. M. , J. Non-Cryst. Solids, 15 (2006) 374.
- Donnell I. H. and San Oster D. F., "Principles of Radiation Chemistry", Edward Arnold Pup. Ltd., London, 23 (1968) 138.
- Cornelius N. A. and Moynihan T. J., solid state ionics , 75 (1998) 105.
- Clark L. Allred, Ph.D thesis " Effect of Radiation on Silicon and Borosilicate Glass", Massachusetts Institute of Technology, (2003).
- El-Alaily N.A.; Mohamed R.M., Al-Azhar Bull.Sci. , 3 (5) (1988) 155.
- El - Hadi Z. A. , Commun . Fac. Sci. Univ. Ank. Series B , 39 (1993) 45.
- Heba A.S. SOP Transactions on Physical Chemistry, Vol. 1, no.1, Feb., 2014 .
- ICRU, "Radiation Quantities and Units", Int. Ceram. Radiat., Units Meas., Washington, (1971).
- Karam A. A.; Ahmed G. Truk J Phys. 27, (2003).
- Kumar S. , Hsain M. and Zulfequar M. , Physica B 387 (2007) 400.
- Leong P.M.; Eeu T. Y.; Sains Malaysiana 43 (6) (2014): 915–922.
- Levy P. W., "Symposium on Effect of Radiation on Inorganic Soc., 12 (1964) 359.
- Mogus-Milankovi A. , J. Non Cryst. Solid, 57 ( 2001)296.
- Sudarsan V. and Kulshreshtha S. K. , J. Non-Cryst. Solids, 320 (2005) 1050.
- Wakabayash H.; J. Non Cryst Solids , 203 (1996) 274.
- Wilson I. E., "Radiation Chem. of Monomers, Polymers and Plastics", Marcel Dekker Inc., New York, 31(1974) 80.
- Yasser B. S; Abo-Naf S. M., PAN-IPPT, Vol. 37, no.3 (2012) 341-347 .