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Gamma-ray dosimetry using zinc phosphate glasses

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A series of $ZnO–P_2O_5$ glass systems has been prepared by melting ZnO with anhydrous P_2O_5 in an open crucible. The temperature dependence on the d.c. electrical conductivity measurements for these glasses is studied over the temperature range from room temperature to 573 K. It was found that the d.c. conductivity decreases with increasing mol % ZnO. The induced changes in the d.c. electrical conductivity caused by different doses of gamma irradiations were also studied. Gamma doses up to 5.0 Mrad were used, results were explained and discussed in terms of the interaction of gamma rays with the glass compositions. Results of the present work have shown that the effect of γ-ray on the d.c. conductivity of the glass system is composition dependent and the low ZnO concentration specimen is more sensitive to the γ-irradiations.

1. Introduction

An increasing number of papers have dealt with the electrical properties of amorphous semiconductors. The amorphous form of selenium was one of the first such materials to be studied. More complex amorphous semiconductors were investigated, among them chalcogenide [1–3] and oxide [4–6] semiconducting glasses. In the case of amorphous materials, electronic and/or ionic conductions are involved [7].

Several investigators [8–18] have paid considerable attention to the study of properties of phosphate glasses containing transition metal cations, such as vanadium, cobalt, iron, tungsten, molybdenum, copper, . . . , etc. These cations have a variable valance property and serve as model substances for polaron conduction.

High-energy radiations, such as gamma rays, change the physical properties of the materials they pass through. The changes are strongly dependent on the internal structure of the absorbed substances (such as glasses), and as a result a displacement of the orbital electrons and possibly atoms in the structure will take place. These displaced electrons (photoelectrons) will go back and forth and then become freely or loosely bound to trapping centres somewhere in the glass matrix. These new electronic configurations would cause a change in the electrical conductivity.

In fact, there is an outstanding need for cheap, sensitive radiation dosimeters that are able either to monitor or detect different kinds of radiations. For this purpose, the glassy materials have a wide variety of applications in the field of radiation dosimetry and are of the same importance as many other kinds of currently used radiation detectors, such as some types of polymers. The glass detectors have been extensively used since the production of silver phosphate glasses [19]. Several authors [20–28] have studied the possibility of using different glass compositions as gamma and neutron dosimeters. These studies have shown, more or less, a good success in using such detectors in different domains of radiation doses.

The effects of gamma radiations on the absorption spectra of cobalt-activated silicate glasses have been investigated and given previously [25]. It has been found that the barium-containing silicate glass doped with CoO is sensitive to gamma radiation and can be recommended to be used as a gamma dosimeter in the range 1.67×10^3 to 1.5×10^5 rad. El-Fiki *et al.* [27] have studied the lithium disilicate glass containing neodymium and doped with uranium as gamma and fission fragment track detectors. They have found that such a glass can be used as a gamma-ray detector through optical density measurements and is also suitable for neutron fluence measurements. Sayed *et al.* [28] have studied the effects of pre-neutron exposure on the induced change in the optical density of soda–lime silica glass due to γ-irradiation. They have concluded that low pre-neutron exposure fluence on silicate glass could be used to detect γ-doses to far below 10^3 rad.

In this paper we report experimental measurements of the d.c. conductivity of unirradiated and γ-irradiated $ZnO–P_2O_5$ glass systems and then investigate the possibility of using such glass specimens as gamma-ray dosimeters.

2. Experimental procedure

2.1. Preparation of glasses

$ZnO–P_2O_5$ glasses were prepared by melting the appropriate mixture of Analar phosphorous pentoxide and Analar zinc oxide in alumina crucibles at 850 to 1100° C. The preparation procedure was employed to prepare glasses with a glass formation range from 36 to 70 mol % ZnO (starting composition). Details of the preparation technique and chemical analysis of these glasses are to be presented elsewhere [29, 30].

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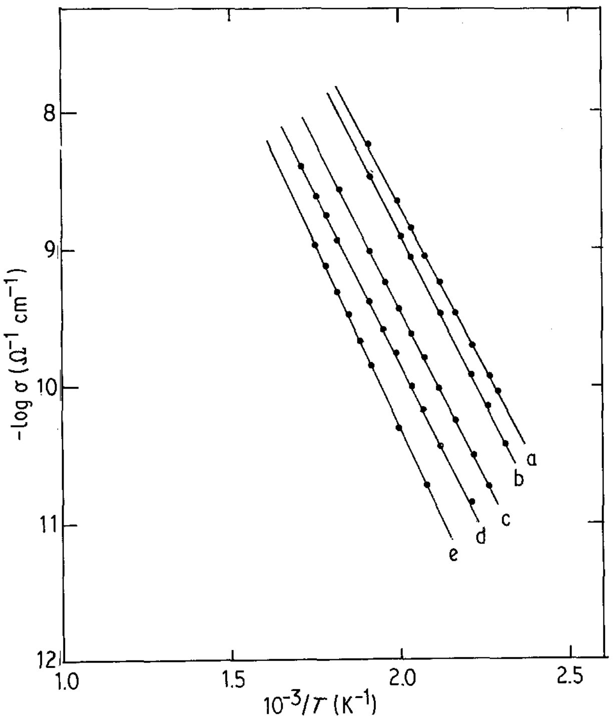

Figure 1 D.c. electrical conductivity of unirradiated $ZnO-P_2O_5$ glasses as a function of inverse temperature, $1{:}T$ (K^{-1}). Glasses a, b, c, d and e represent 43.2, 53.5, 56.32, 60.10 and 70.0 ZnO mol %, respectively.

TABLE I Composition and activation energy values of the studied glass system

Sample no.	ZnO (mol %)	Activation energy* (eV)				
		W_0	$W_{0.5}$	$W_{1.0}$	$W_{2.0}$	$W_{5.0}$
a	43.20	0.950	0.923	0.890	0.844	0.750
b	53.50	0.974	–	–	0.927	0.899
c	56.32	0.981	–	0.943	0.927	0.917
d	60.10	1.009	0.992	0.960	0.951	0.948
e	70.00	1.076	1.070	1.060	1.065	1.020

* W_0 is the activation energy for unirradiated glass specimens; $W_{0.5}$, $W_{1.0}$, $W_{2.0}$ and $W_{5.0}$ are the activation energies for glass samples exposed to 0.5, 1.0, 2.0 and 5.0 Mrad γ-doses, respectively.

The samples were rod-shaped with diameter 1.6 cm and thickness about 5 mm.

2.2. The d.c. electrical conductivity measurements

For the measurements of d.c. electrical conductivity, electrodes were formed by brush painting silver paste. The d.c. conductivity was measured as a function of temperature, using a spring-loaded sample holder in a wire-wound cylindrical furnace. All our d.c. electrical conductivity measurements were made using a Keithley electrometer model 616, with a smoothing adjustable power supply (0 to 1 kV). A fixed voltage of 300 V was applied. The temperature of the specimen was measured by means of a chrome–alumel thermocouple.

The d.c. electrical conductivity (σ) of each specimen was then calculated using the formula

$$\sigma = L/RA$$

where L is the thickness of the sample (cm), A is the cross-sectional area of the electrode (cm^2) and R is the resistance (Ω).

2.3. Irradiation facilities

The glass specimens were exposed in air to different γ-doses using a Co^{60} gamma cell as a source of gamma radiation. The exposure rate of 8.64×10^4 rad h^{-1} was applied at room temperature.

3. Results and discussion

The variation of d.c. electrical conductivity (log σ) with the reciprocal of the absolute temperature (T^{-1})

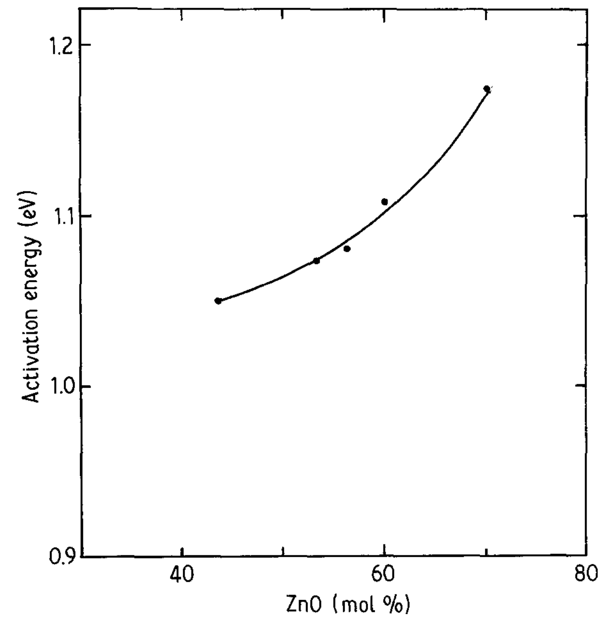

Figure 2 The variation of the activation energy of unirradiated glasses with ZnO mol %.

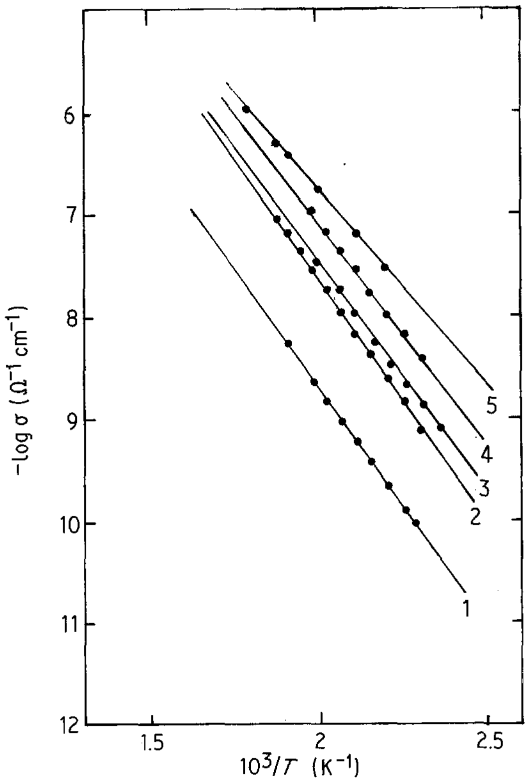

Figure 3 D.c. electrical conductivity of unirradiated and irradiated 43.2 ZnO mol % glass sample a. 1. Unirradiated; 2, 0.5; 3, 1.0; 4, 2; 5, 5.0 Mrad γ-dose.

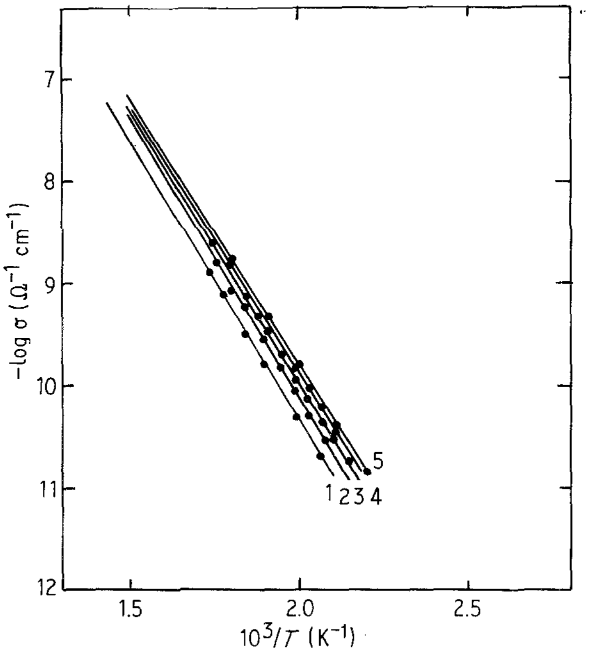

Figure 4 D.c. electrical conductivity of unirradiated and irradiated 70.0 ZnO mol % glass sample e. 1. Unirradiated; 2, 0.5; 3, 1; 4, 2.0; and 5, 5.0 Mrad γ-dose.

was measured and is shown in Fig. 1, for all unirradiated ZnO–P_2O_5 glass samples. It is obvious from Fig. 1 that the d.c. conductivity shows a linearity with temperature which reflects the semiconducting behaviour of the specimens under study. This variation of the d.c. conductivity with temperature could be explained [17] in terms of the mobility activation of charge carriers. Also, Fig. 1 shows that the d.c. electrical conductivity is a composition dependent, where σ

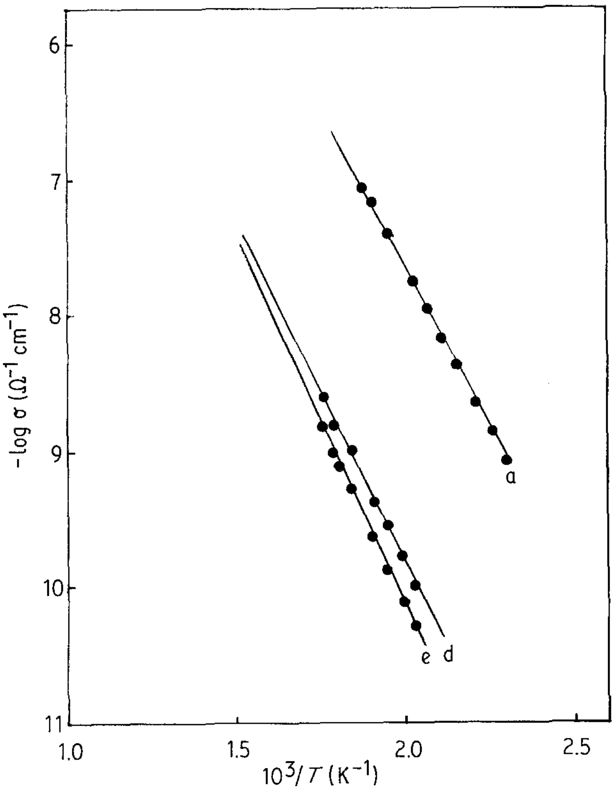

Figure 5 D.c. electrical conductivity of 0.5 Mrad irradiated ZnO–P_2O_5 glasses as a function of inverse temperature, $1/T\,(K^{-1})$. Glasses a, d and e represent 43.2, 60.10 and 70.0 ZnO mol %, respectively.

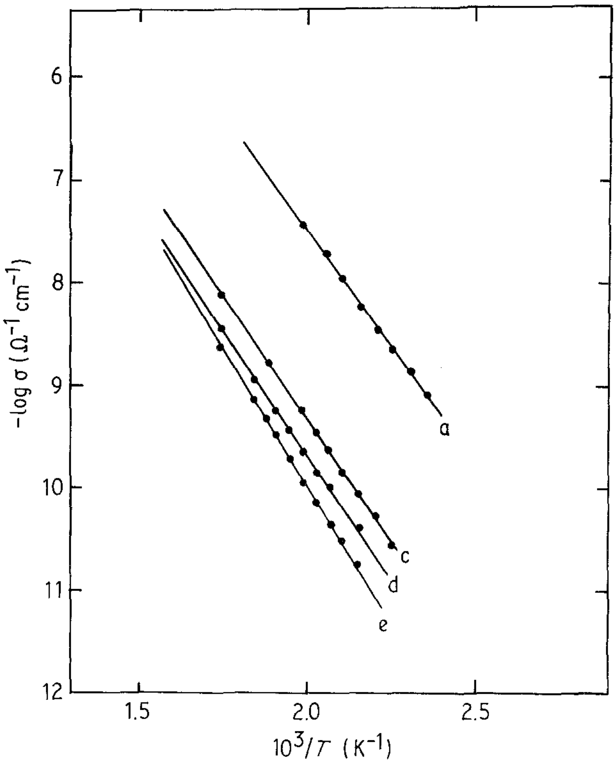

Figure 6 D.c. electrical conductivity of 2.0 Mrad irradiated ZnO–P_2O_5 glasses as a function of inverse temperature, $1/T\,(K^{-1})$. Glasses a, b, c, d and e represent 43.2, 53.5 56.32, 60.1 and 70.0 ZnO mol %, respectively.

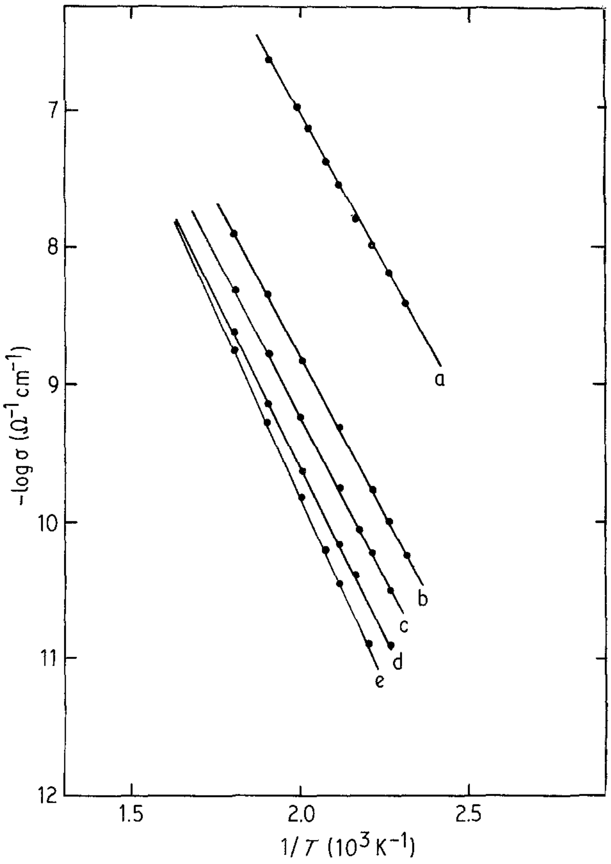

Figure 7 D.c. electrical conductivity of 1.0 Mrad irradiated ZnO–P_2O_5 glasses as a function of inverse temperature, $1/T\,(K^{-1})$. Glasses a, c, d and e represent 43.2, 56.32, 60.1 and 70.0 ZnO mol %, respectively.

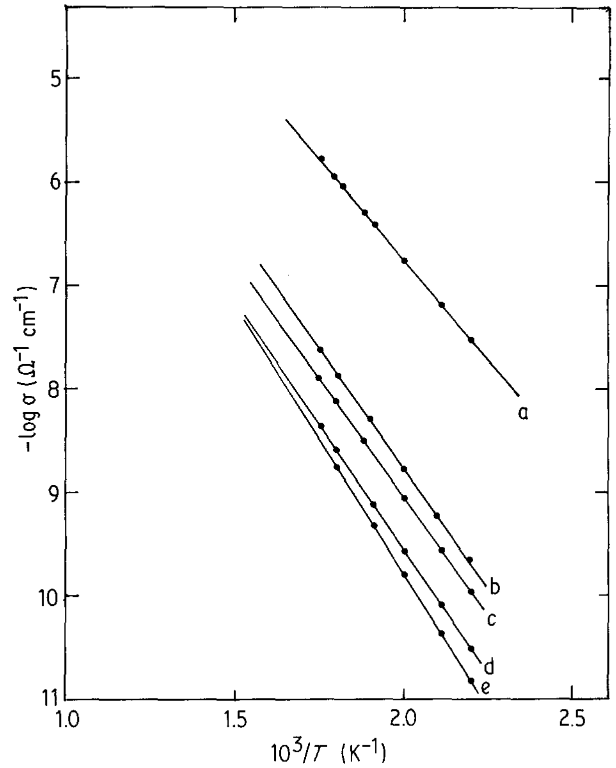

Figure 8 D.c. electrical conductivity of 5.0 Mrad irradiated ZnO–P_2O_5 glasses as a function of inverse temperature, $1/T$ (K^{-1}). Glasses a, b, c, d and e represent 43.2, 53.05, 56.32, 60.1 and 70.0 ZnO mol %, respectively.

decreases with the increase in the ZnO content in the glass specimens.

The conduction activation energy, W, in the present result, was calculated from the slopes of log σ against $1/T$ using a least square fit method; W was found to be temperature independent in the studied temperature range. The activation energy showed an increase with increase in the ZnO concentrations for the glass system (see Table I and Fig. 2). For example, W shows an increase from 0.95 to 1.076 eV as the ZnO content increases from 43.2 to 70 mol %.

The effect of gamma-irradiation on the d.c. electrical conductivity for the ZnO–P_2O_5 glasses was studied under different γ-doses (0.5, 1, 2 and 5 Mrad). Figs 3 and 4 show the dependence of log σ against $1/T$ for unirradiated and irradiated glass specimens a and e, respectively. It is very clear from both figures that a detectable change in the temperature dependence of d.c. conductivity of samples a and e has been obtained as a result of γ-irradiation. It is also interesting to note that the effect of γ-dose on glass sample a has shown (see Fig. 3) a significant increase in the d.c. conductivity, while in case of glass specimen e (see Fig. 4) the induced changes in the conductivity are less pronounced. The induced changes in σ are dose-dependent and can be attributed to the creation of radiation-induced charge carriers in the glass matrix. Actually, the dependence of d.c. conductivity on the γ-dose would lead us to study the possibility of using such a glass system as a dosimeter. Results shown in Figs 3 and 4 indicate that the induced rate of change in σ is greatest for samples irradiated at 0.5 Mrad, while as the dose becomes higher, the rate of induced change in d.c. conductivity becomes smaller. This dependence of the d.c. conductivity on γ-dose might be explained as follows. At the beginning, increasing γ-dose would result in an increase in the number of charge carriers created. This increasing number of carriers will continue to take place as γ-dose increases until we approach a situation at which most of the possible charge carriers are already created. After this threshold dose-limit, we might expect a very slight increase in the d.c. conductivity and a saturation limit might be achieved as the γ-dose exceeds the threshold value. Figs 5 to 8 show the dependence of log σ against $1/T$ for glass specimens irradiated at 0.5, 1, 2 and 5 Mrad, respectively. It can be concluded from Figs 5 to 8 that the d.c. electrical conductivity always shows an increase with increasing γ-dose up to 5 Mrad for all the glass specimens studied. The dependence of the d.c. conductivity on the ZnO concentration remains unchanged as in unirradiated samples.

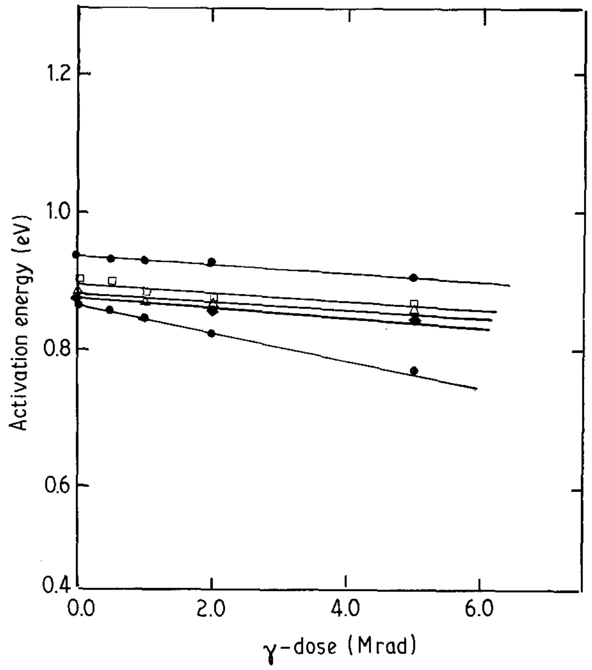

Figure 9 The variation of the activation energy with the absorbed γ-dose. Glasses a, b, c, d and e represent 43.2, 53.5, 56.32, 60.1 and 70.0 ZnO mol %, respectively.

The values of the activation energies of the irradiated glass specimens were calculated from the slopes

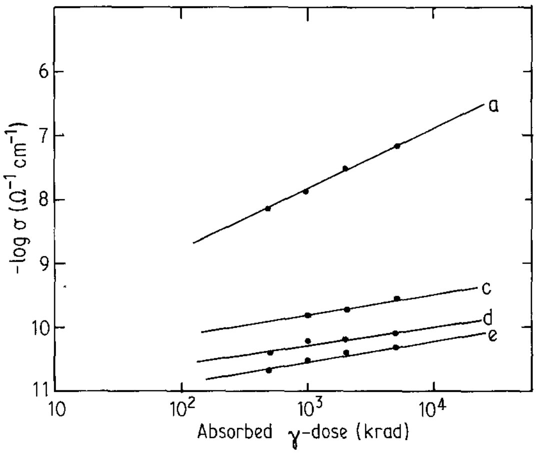

Figure 10 The variation of the d.c. electrical conductivity with the absorbed γ-dose at 473 K. Glasses a, c, d and e represent 43.2, 56.32, 60.1 and 70.0 ZnO mol %, respectively.

of log σ against $1/T$ relationships and are represented in Table I. It is obvious from this table that the activation energy is a function of the absorbed dose; it decreases with increasing γ-dose. Therefore, in all glass specimens studied, W was shown to be a composition and dose dependent. This leads us to a study of the possibility of using such a glass system as a gamma dosimeter through measured values of the activation energies. In case of sample a, W decreases from 0.95 to 0.75 eV after an exposure to 5.0 Mrad γ-dose, while only a change of 0.056 eV is obtained for glass e under the same conditions (see Table I and Fig. 9). Such a linear behaviour obtained in Fig. 9 led to the conclusion that the glass samples studied, especially that of glass a, may be recommended to be used as γ-dosimeter by measuring their activation energies.

On the other hand, the use of such a glass system in measuring the gamma ray doses through a direct measurement (i.e. measuring the d.c. conductivity) can be readily recommended. Fig. 10 represents the variation of the d.c. conductivity with γ-dose at 473 K. The usefulness of the linearity obtained (see Fig. 10) reflects the importance of using such glass samples (particularly glass a) as gamma ray dosimeter in the studied dose range.

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