

## Optical study of gamma irradiated sodium metaphosphate glasses containing divalent metal oxide MO (ZnO or CdO)



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### ABSTRACT

Sodium metaphosphate glasses containing divalent metal oxide, ZnO or CdO with composition  $50 \text{P}_2\text{O}_5 - (50 - x) \text{Na}_2\text{O} - x \text{MO}$  (ZnO, or CdO) where  $x = 0, 10, 20$  (mol%) were prepared by conventional melt method. UV/visible spectroscopy and FTIR spectroscopy are measured before and after exposing to successive gamma irradiation doses (5–80 kGy). The optical absorption spectra results of the samples before irradiation reveal a strong UV absorption band at ( $\sim 230$  nm) which is related to unavoided iron impurities. The effects of gamma irradiation on the optical spectral properties of the various glasses have been compared. From the optical absorption spectral data, the optical band gap is evaluated. The main structural groups and the influence of both divalent metal oxide and gamma irradiation effect on the structural vibrational groups are realized through IR spectroscopy. The FTIR spectra of  $\gamma$ -irradiated samples are characterized by the stability of the number and position for the main characteristic band of phosphate groups. To better understood the structural changes during  $\gamma$ -irradiation, a deconvolution of FTIR spectra in the range  $650\text{--}1450 \text{cm}^{-1}$  is made. The FTIR deconvolution results found evidence that, the changes occurring after gamma irradiation have been related to irradiation induced structural defects and compositional changes.

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### Introduction

The short range structure of glassy  $\text{P}_2\text{O}_5$  consists of a network of  $\text{Q}^3$  tetrahedral units where three of the oxygens are bridging (P–O–P) and one non-bridging (P=O), with the addition of alkali oxide ( $\text{Na}_2\text{O}$ ) to  $\text{P}_2\text{O}_5$ , the conversion of is P–O–P bridging to P–O–M<sup>+</sup> non-bridging oxygens takes place, which indicate the formation of  $\text{Q}^2$  tetrahedral with two non-bridging oxygen atoms. Thus the addition of alkali oxide to vitreous  $\text{P}_2\text{O}_5$  results in the creation of non-bridging oxygens at the expense of bridging oxygens. As alkali oxide is added to  $\text{P}_2\text{O}_5$ , the phosphate structural groups pass from  $\text{Q}^3$  to  $\text{Q}^2$  to  $\text{Q}^1$  to  $\text{Q}^0$  as the molar ratio of alkali oxide to  $\text{P}_2\text{O}_5$ ,  $R = \text{M}_2\text{O}/\text{P}_2\text{O}_5$ , passes from 0 to 1 to 2 and finally to 3. These modifications are similar to those taking place in a silicate network upon the addition of alkali earth oxides, such as  $\text{Na}_2\text{O}$  to  $\text{P}_2\text{O}_5$ , results in the conversion of the three dimensional network, to linear phosphate chains. This linear chain structure due to the cleavage of P–O–P linkages and the creation of NBOs in the glass [1]. Recently, Phosphate glasses are technologically important because they generally have a high

thermal expansion coefficient and high refractive indices, low melting temperature, low glass transition temperature, low thermo-optical coefficient and large emission. Thus, they are suitable for many applications such as glass to metal seals [2–4], biomedical engineering. However, their relatively poor chemical durability makes them unsuitable for practical applications. The addition of one or more of divalent oxides like ZnO or CdO leads to improvement in the chemical durability, mechanical properties, and thermal expansion by increasing the strength of bonds and the compaction of the phosphate glass system, these cations can enter the glass network either network former or network modifier [5]. It is important to study the interaction of gamma ray with a glass system as it may be lead to various changes in the structural, optical and electrical properties of the glasses [6]. The radiation interaction with the host glass may cause the displacement of lattice atoms or electronic defects which involve changes in the valence state of the lattice or impurity atoms. Also, it may involve ionization and charge trapping and/or radiolysis or photochemical reactions as high energy radiations change in the optical properties of the glasses in a way that the appearance of new absorption bands in visible and ultraviolet part of the spectrum takes place. This is may be due to different kinds of defect centers generated as a result of creation and capture of electron and hole pair during the irradiation process [7,8].

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**Table 1**  
Chemical composition of investigated glass (mol%).

Glass sample	Composition			
	P <sub>2</sub> O <sub>5</sub>	Na <sub>2</sub> O	ZnO	CdO
G1	50	50	–	–
G2	50	40	10	–
G3	50	40	–	10
G4	50	30	20	–

The main objective of this work is to evaluate the effects of ZnO or CdO on optical study and also study the effect of gamma irradiation on the measured spectral properties and justify the induced defects generated, so as to explore its use for nuclear industry.

## Experimental

### Preparation of the glasses

Sodium metaphosphate glasses prepared from P<sub>2</sub>O<sub>5</sub> was introduced in the form of pure ammonium hydrogen phosphate (NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub>), sodium oxide was introduced in the form of its respective anhydrous sodium carbonate Na<sub>2</sub>CO<sub>3</sub> (Analar quality) and MO was added in the form of its respective pure oxide. The composition (50 – x) Na<sub>2</sub>O – 50 P<sub>2</sub>O<sub>5</sub> – x MO (ZnO, or CdO), where x = 0, 10, 20 mol%. The composition of the studied glasses as shown in Table 1. The batches were melted in silica crucibles from 900 to 1100 °C ± 20 °C (depending on composition) for two hours in the electrically heated furnace and each melt was stirred by rotating the crucible several times every 30 min. The homogeneous melts were cast into preheated stainless steel molds. Then the prepared samples were immediately transferred to an annealing furnace regulated at 220 °C. The muffle after one hour was left to cool to room temperature at rate of 25 °C/h.

### UV–visible absorption measurements

The optical absorption of highly polished samples measured against air at room temperature 25 °C. Optical absorption measurements before and after successive gamma irradiation were carried out with a recording double-beam spectrophotometer (type Unicam spectrometer made in England), covering the range from 190 to 1000 nm.

### Infrared absorption spectra measurements

The FTIR absorption spectra were registered at room temperatures using a JASCO FTIR 6200 spectrometer. IR absorption spectra were measured immediately after preparing the disks of KBr and glass samples. The FTIR spectra were recorded in the wavenumber range of 400–4000 cm<sup>-1</sup>. A quantitative analysis of the infrared spectrum has been carried out by a careful deconvolution of the absorption profiles utilizing the “peak fit” program. IR spectra were corrected for the dark current noises and background using two-point baseline correction. Many trials have been carried out using different band shapes, but has fit obtained was been found to be Gaussian shape. The position of each band, its width and intensity are parameters adjusted automatically by the program, on the basis of the minimization of the deviations between experimental and simulated spectrum.

### Irradiation facility

Glasses were subjected for different doses using an Indian <sup>60</sup>Co gamma-rays cell (2000 Ci) with a dose rate 4.26 kGy/h. The

samples were placed in a manner that each glass sample was subjected to the same irradiation dose.

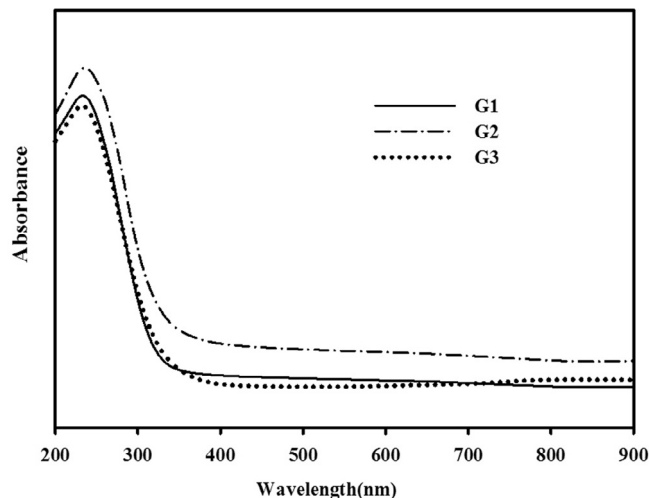
## Results and discussion

### Absorption spectra before irradiation

Fig. 1 illustrate the UV–visible absorption spectrum of samples (G1–G3) before irradiation. The spectra were recorded from (200–800 nm), reveal a strong UV absorption band centered at (230 nm), and no visible absorption is observed. The observed UV absorption band (230 nm) can be assumed to originate from the presence of trace unavoidable iron impurity species (Fe<sup>3+</sup> and Fe<sup>2+</sup>) in the raw materials used to prepare the studied glasses [9–11] Iron can exist in the 2+ and 3+ states in glasses, depending upon the conditions of preparation. The two iron states have strong charge transfer bands with different absorption coefficients, being more than one order of magnitude higher for Fe<sup>3+</sup> than for Fe<sup>2+</sup> [6]. Also, The broadness of the extending band (200 → 300 nm) in the present study is attributed to the possible presence of more than one site of both iron species Fe<sup>2+</sup> and Fe<sup>3+</sup> ion [12]. Thus we can assert the intense charge-transfer absorption near 230 nm in glass is related to Fe<sup>3+</sup> ions. This result is supported by recent investigations done by several publications [13,14]. They have concluded that these glasses exhibit characteristic charge transfer ultraviolet absorption bands because of the presence of transition metal ions (e.g. Fe<sup>3+</sup>, Cr<sup>6+</sup>, etc.) even if present in the ppm level. With careful examination of UV/visible spectra, there is an obvious difference between G2 and G3 is observed in UV–visible spectra of the glasses. Thus, the observed increase in the intensity of G2 than G3 at (250 nm) may be assumed to the function of G2 which increase the absorption trace iron impurities than the G3 which causes retardation of the absorption of iron ions leading to the observed decrease in the intensity retarded the effect of iron impurities so, its intensity decrease.

### Effect of gamma irradiation on absorption spectra of base sodium metaphosphate glass

Fig. 2a Shows the UV–visible spectrum of base sodium metaphosphate glass (G1) after gamma irradiation. The spectrum before irradiation reveals a prominent band at 230 nm in UV region and, no absorption bands is observed in visible region. On



**Fig. 1.** Optical absorption spectra of the sodium metaphosphate glass containing divalent metal oxide.

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