



## Spectroscopic properties and Judd-Ofelt analysis of Dy<sup>3+</sup> ions in molybdenum borosilicate glasses



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### ABSTRACT

Borosilicate glasses containing MoO<sub>3</sub> and Dy<sub>2</sub>O<sub>3</sub> were prepared by the melt quenching technique. The structure and the thermal properties of these glasses have been performed by using infrared (IR) technique at room temperature and DTA. The mechanical properties have been experimentally determined and calculated according to Makishima–Mackenzie model. The optical properties of the prepared glasses have been studied using the UV – Spectroscopy. The glass transition temperature and the optical band gap decrease with increasing the MoO<sub>3</sub> content. The values of the optical band gap lie between 2.25–3.22 eV. The optical properties of the Dy<sup>3+</sup> ion have been investigated using the Judd-Ofelt approach. The oscillator strengths and optical intensity parameters,  $\Omega_t = 2, 4$  and 6 have been evaluated. An interesting behavior of the calculated lifetime versus total angular momentum,  $J$ , has been observed, where, the lifetime values of the <sup>6</sup>F<sub>*J*</sub> states show an exponential increase versus  $J$ .

### 1. Introduction

At the present time, several optical devices are used extensively in the activities of human beings. Permanently, new light source devices have been studied and developed to get a good performance and fair light emission. It was originated that glasses doped with rare earth oxides (REO) are now very interesting materials for the development of optical fibers, optical amplifiers, sensors and laser [1]. Optical studies disclose the radiative properties of REO doped glasses; these properties strongly depend on the host matrix and can be modified by proper network formers as well as modifiers. In addition, the borate based glasses provide interesting optical and structural properties. REO doped glasses are potential candidates for lasers, waveguide, optical fibers, solar concentrators and optical detectors [2]. Dy<sub>2</sub>O<sub>3</sub> doped lead phosphate glasses have various possible applications in several fields, such as solar cells and were characterized by IR, photoluminescence and UV–visible measurements [3]. The glasses containing Dy<sub>2</sub>O<sub>3</sub> can be considered as good paramagnetic materials, which can be utilized as storage media. The principal of this probable use is based on the spread of a unidirectional light in optical materials, which results in high Verdet constant. The concentration of Dy<sub>2</sub>O<sub>3</sub> is largely influenced the values of Verdet constant [4]. Moreover, trivalent dysprosium (Dy<sup>+3</sup>) ion, in particular, act as a potential candidate to absorb UV light and

emit light mainly in the blue spectral region (460–500 nm) and yellow region (560–600 nm). The origin of these emissions was due to the 5d-4f transitions and its 5d state can be easily affected by the crystal field environment. Thus, it is feasible to obtain white light from Dy<sup>3+</sup> doped single phosphor by adjusting the yellow to blue emission intensity ratio (Y/B). Several hosts were reported to develop white light, namely molybdates-, borates-, phosphates-, vanadates- based glasses [5]. The presence of <sup>6</sup>F<sub>11/2</sub>, <sup>6</sup>H<sub>9/2</sub> → <sup>6</sup>H<sub>15/2</sub> transitions in Dy<sup>3+</sup> at about 1.3 μm may be useful for optical amplification and visible up conversion emission required for solid state laser [6]. Furthermore, Dy<sup>3+</sup> ions reveal very strong emission in the visible and NIR regions by providing lasing transitions.

Moreover, glasses containing transition metal oxides (TMO) such as MoO<sub>3</sub> have been studied because of their stimulating semi-conducting properties, which are owing to the springing of “polarons” from the higher to lower valence states, i.e., from Mo<sup>5+</sup> to Mo<sup>3+</sup> [4]. When these glasses are mixed with alkaline earth ions, their chemical resistance and optical transparency, increased at the excitation and lasing wavelengths. They are most compatible with the manufacture process in the development of optical devices [7–9].

The purpose of the current investigation is the study of the effect of the network of alkali borosilicate glasses containing different concentrations of MoO<sub>3</sub> on the optical absorption and the fluorescence

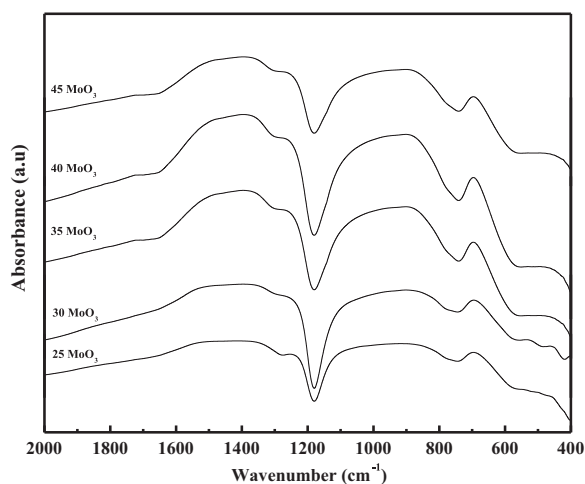
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**Table 1**

The nominal chemical composition, the molar volume ( $V_m$ ), the density ( $\rho$ ), the ultrasonic velocities (transverse ( $v_T$ ) and longitudinal ( $v_L$ )), the elastic moduli (computed Young's modulus ( $Y_{th}$ ), Young's modulus ( $Y$ ), computed bulk modulus ( $K_{th}$ ) and bulk modulus ( $K$ ), the glass transition temperature ( $T_g$ ) and the optical energy gap ( $E_g$ ) of  $x$  MoO<sub>3</sub> – 15 SiO<sub>2</sub> – 20 Dy<sub>2</sub>O<sub>3</sub> – (65- $x$ ) Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub>, ( $25 \leq x \leq 45$ ) glass system.

Chemical composition				$V_m$ (cm <sup>3</sup> mol <sup>-1</sup> )	$\rho$ (g/cm <sup>3</sup> )	$v_T$ (ms <sup>-1</sup> )	$v_L$	$Y_{th}$	$Y$	$K_{th}$	$K$	$T_g$	$E_g$ eV
Na <sub>2</sub> B <sub>4</sub> O <sub>7</sub>	SiO <sub>2</sub>	MoO <sub>3</sub>	Dy <sub>2</sub> O <sub>3</sub>										
40	15	25	20	57.78	2.88	3255	5710	75.5	56.5	130.3	49.3	521	3.3
35	15	30	20	63.12	2.59	3234	5673	71.6	52.6	127.2	45.9	485	3.1
30	15	35	20	66.08	2.43	3207	5627	68.8	49.9	123.7	43.6	473	2.8
25	15	40	20	68.85	2.34	3186	5589	60.8	48.9	98.3	42.7	466	2.4
20	15	45	20	70.56	2.27	3131	5493	56.0	48.1	85.0	41.9	462	2.3



**Fig. 1.** Infrared spectra of the investigated glass system of  $x$  MoO<sub>3</sub> – 15 SiO<sub>2</sub> – 20 Dy<sub>2</sub>O<sub>3</sub> – (65- $x$ ) Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub>, ( $25 \leq x \leq 45$ ) glass system.

spectra of Dy<sup>3+</sup> ions. The characterization of these glasses has been performed by using IR, thermal and ultrasonic techniques along with an application of Judd-Ofelt theory.

## 2. Experimental procedures

In order to prepare glass samples having the listed nominated chemical compositions in Table 1, appropriate amounts of analytically pure grade chemicals SiO<sub>2</sub>, MoO<sub>3</sub>, Na<sub>2</sub>B<sub>4</sub>O<sub>7</sub> and Dy<sub>2</sub>O<sub>3</sub> were thoroughly mixed in an agate mortar and melted in a platinum crucible in an electric furnace at 1150 °C for about 1 h until a bubble free liquid was formed. The melt was revolved in the crucible every 20 min to achieve homogeneity. Then, the resultant melt has been casted in a brass mold and subsequently annealed at 400 °C, in a muffle furnace. After 30 min, the muffle was switched off and the furnace temperature decreased to room temperature with a rate 25 °C/h. The samples were polished to have suitable dimensions (2 × 1 × 0.5 and 2 × 1 × 0.15 cm<sup>3</sup>) for the present ultrasonic and optical measurements. The amorphous state of the glasses was checked by X-ray diffraction using Philips X-ray diffractometer PW/1710 and the IR absorption spectra of the glasses in the wave number range of 2000–400 cm<sup>-1</sup> by an IR spectrophotometer type JASCO, 430 (Japan) were recorded.

The thermal analysis of the explored samples was performed using micro-DTA (SHIMADZU 50) to get the characteristic temperatures of the sample with an accuracy of ± 5 °C. The (15 mg) powdered glass sample was placed in a platinum crucible, examined up to 700 °C in nitrogen medium with a heating rate 10 °C/min. and  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> was used as a reference material. The absorbance  $A(\lambda)$  was recorded at ambient temperature in the range 200–2700 nm using a computerized double beam spectrophotometer, type JASCO V- 670.

The glass densities ( $d$ ) were measured using the well-known Archimedes method with toluene as the standard immersion liquid, for

all studied glass systems. The relationship between the density and the structure of oxide glasses can be expressed in terms of apparent volume ( $V_m$ ) occupied of 1 mol. The molar volume,  $V_m$ , can be got as  $M/d$ .

The longitudinal ( $v_L$ ) and shear ( $v_T$ ) ultrasonic velocities with an uncertainty of ± 10 m/s have been obtained by utilizing a computerized ultrasonic flaw detector (Karl Deutsch Echograph model 1085) and two transducers; x-cut and y-cut (Karl Deutsch) operated at a fundamental frequency of 4 MHz. The two velocities besides the density were used to determine the elastic moduli as previously stated [10].

## 3. Results and discussions

### 3.1. X-ray diffraction measurements

The XRD of the studied glasses (not shown) present no discrete lines, no sharp peaks, and indicate that the glass samples have a high degree of glassy state.

### 3.2. IR spectroscopy

IR spectra of the prepared glass samples are shown in Fig. 1. The spectra reveal five mean absorption broadbands. Manifestation of the broadband indicates that a wide distribution of the structural units within the glasses. The most intense bands of the base glass lie within 777–1118 cm<sup>-1</sup> region and the moderate intensive bands lie between 1200–1500 cm<sup>-1</sup>, while the least intensive bands lie between 1105 and 1200 cm<sup>-1</sup> and 629–793 cm<sup>-1</sup>. There are two small bands at 431 and 529 cm<sup>-1</sup>. The absorption peak at wavenumbers 1500–1430 cm<sup>-1</sup> in these glass samples are due to the vibrational mode of the borate network. The vibrational modes of the borate network are accredited due to the asymmetric stretching reduction in B-O bonds of trigonal BO<sub>3</sub> units. Such categories of vibrations have occurred in the wavenumber range 1200–1500 cm<sup>-1</sup> [11]. The broad absorption band at around 1105–1118 cm<sup>-1</sup> is attributed to a stretching vibration of B-O-Si linkage [12]. The third absorption band is observed within the wavenumber range 808–876 cm<sup>-1</sup> in the IR spectra of all glass samples. This peak remained current owing to the diborate linkage, B-O-B, in the borate glassy net overlapped with the vibrations of a band attributed to Mo–O–Mo bridging bonds in the structure [13]. In this association, both boron atoms are tetrahedrally coordinated with triborate super-structural constituents [14]. The second absorption band is observed within the wavenumber range 600–777 cm<sup>-1</sup> in the IR spectra of all glass samples. An absorption peak at the lower wavenumber side accredited to vibration of metal cation such as Dy<sup>3+</sup> and Si<sup>4+</sup> peak (no. 1) [15,16]. The two small bands at 431 and 529 cm<sup>-1</sup> may be attributed to the vibrations of some types of borate segments and to the bending vibrations of Si–O–Si linkages overlapped with the vibrations of the Mo–O bonds in distorted MoO<sub>4</sub> [13].

The broad FTIR band of the glass was attributed to the overlapping of some individual bands with each other. Each individual band has its center (C), which is related to the vibrations of a specific structural group, and its relative area (A), which is proportional to the

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