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PHYSICAL PROPERTIES OF THE CaF_2–P_2O_5:MoO_3 GLASS SYSTEM

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In this study, ternary glass samples with the composition CaF_2–P_2O_5:MoO_3 were prepared by the conventional melt-quenching method, and their physical properties were investigated. The structural characteristics were examined using X-ray diffraction, which confirmed the amorphous nature of the prepared samples. The densities were determined experimentally using Archimedes' principle. The obtained density and molar volume values were further employed to calculate various physical parameters such as dopant ion concentration, interionic distances, polaron radius, and field strength. Molar and electronic polarizabilities, oxygen packing density, optical band gap, and the metallization criterion were evaluated from the measured refractive indices using the Dimitrov-Komatsu polarizability approach. The results indicate that the incorporation of molybdenum oxide increases chemical durability and density while reducing molar volume. The prepared glasses exhibit metallization parameter values below unity, suggesting reduced insulating behavior.

Keywords: P_2O_5 glass, physical properties, density, refractive index, ternary glass.

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Introduction

Studying the physical properties of glasses makes it possible to understand, control, and optimize their characteristics for various industrial and scientific applications [1]. Investigating the physical properties of glasses with suitable compositions is essential for meeting specific requirements and tailoring their performance. However, unlike crystalline solids, structural characterization of amorphous solids such as glasses is challenging due to their short-range periodicity. The structural parameters of glasses, such as molar volume and metallization factor, provide valuable insights into their behavior and functionality [2].

Compared to borate and silicate glasses, phosphate glasses offer several advantages due to their low melting temperatures, good biocompatibility, thermal expansion coefficients, and UV transparency, despite being water-sensitive, having lower mechanical strength, limited availability, and high cost [3]. Extensive research in recent years has focused on improving the chemical durability and physical properties of phosphate glasses by adding various glass formers and modifiers to the P_2O_5 glass network [4].

One of these oxide semiconductors, MoO_3, shows great potential for use in memory devices, gas sensors, micro-batteries, passive alphanumeric displays, and

optical smart windows [5]. In the glass network, the molybdenum ion exists in at least two stable valence states: Mo(V) and Mo(VI). Depending on the type and composition of the glass, the addition of molybdenum ions alters its electrical and optical properties. Considerable research has been carried out to enhance the chemical and structural properties of phosphate glasses by incorporating various glass formers and modifiers, such as MoO_3, into the P_2O_5 glass network [6].

In recent years, molybdenum phosphate glasses have been extensively studied due to their structural features, which have been shown to be particularly advantageous. The addition of CaF_2 to these glass matrices is expected to significantly reduce the melting temperature and viscosity. It also acts as an efficient mineralizer, enhancing the thermal stability of the glasses. Glasses containing fluoride exhibit several characteristics, including a tendency toward devitrification, chemical reactivity of fluoride melts, and sensitivity to water. In this regard, oxyfluorophosphate glasses appear highly intriguing and promising [7].

The objectives of the present study are to prepare CaF_2–P_2O_5–MoO_3 ternary glasses with varying concentrations of MoO_3 and to investigate how the MoO_3 content affects physical properties, including density, molar volume, and refractive index. Furthermore, the polarizability and metallization criterion were determined using the Dimitrov-Komatsu polarizability approach [8].

Materials and methods

Glass preparation

The glass samples in the present study were prepared using analytical grade (AR) reagents through melting and quenching techniques. The samples were synthesized according to the formula 30 CaF_2–(70–x)P_2O_5–xMoO_3, where x=0, 0.5, 1.0, 1.5, and 2.0 mol.%, and were labeled as A_0, A_1, A_2, A_3, and A_4, respectively. Table 1 presents the detailed chemical composition of the samples along with their corresponding codes.

Appropriate amounts of CaF_2, P_2O_5, and MoO_3 were weighed and ground in an agate mortar. The mixture was melted in a platinum crucible using a proportional-integral-derivative temperature-controlled furnace. The glasses were melted at 900–950°C until a homogeneous, bubble-free liquid was obtained. The resulting melt was then poured onto a rectangular brass mold maintained at room temperature, and the samples were subsequently annealed at 250°C. Figure 1 shows a schematic of the glass sample preparation process.

X-ray diffraction characterization

X-ray diffraction (XRD) analysis was performed on the obtained glass samples using a SEIFERT SO-DEBYE FLUX 202 diffractometer, equipped with a nickel filter and a copper target operating at 40 kV and 30 mA.

Table 1

Detailed composition (in mol.%) of the glass samples and their codes

Sample code	CaF_2	P_2O_5	MoO_3
A_0	30	70	0
A_1	30	69.5	0.5
A_2	30	69	1.0
A_3	30	68.5	1.5
A_4	30	68	2.0

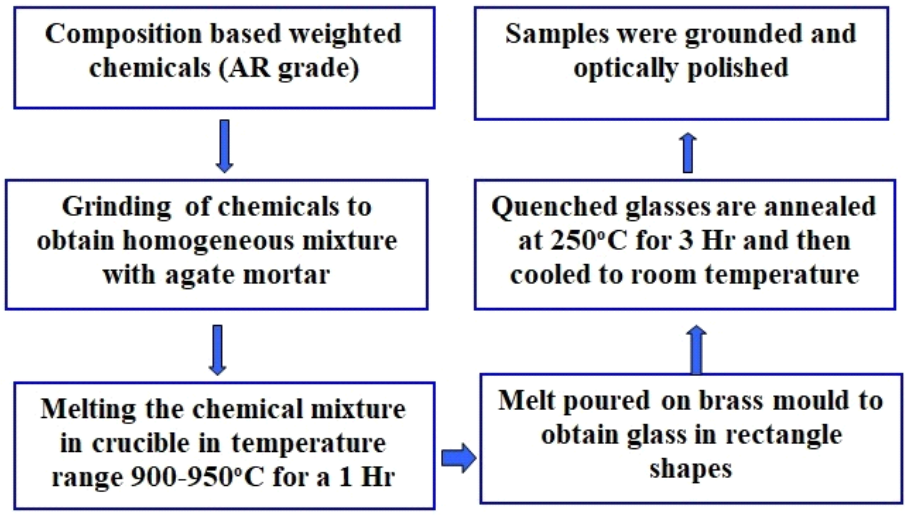

Fig. 1. Schematic representation of the glass preparation process

Physical and structural parameters

The densities of all the glass samples were determined using Archimedes' principle, with measured weights in air and in o-xylene as the buoyant liquid, on an Essae Laboratory electronic digital balance. Various physical parameters of the CaF_2–P_2O_5:MoO_3 glasses were calculated using standard relations and presented in Table 2. The refractive indices of the glass samples were measured using an Abbe refractometer with a light source of wavelength 589.3 nm.

Table 2

Standard relations used to determine physical parameters and detailed explanations of the terms

Parameter	Standard relation used	Explanation of terms
Density, ρ (g/m^3)	$\rho = \frac{W_a}{W_a - W_b}\rho_b$	W_a is the weight of the sample in air; W_b is the weight of the sample in buoyant liquid; ρ_b is the density of the buoyant liquid
Molar volume, V_M (cm^3/mol)	$V_M = \frac{M}{\rho}$	M is the total molecular weight; ρ is the density
Dopant ion concentration, N (ions/cm^3)	$N = \frac{N_A M \rho}{\overline{M}}$	N_A is Avogadro's number; M is the dopant concentration; ρ is the density of the sample; $\overline{M}$ is the average molecular weight
Polaron radius, R_p (Å)	$R_p = \frac{1}{2}\left[\frac{\pi}{6N}\right]^{1/3}$	N is the dopant ion concentration
Interionic distance, R_i (Å)	$R_i = \left[\frac{1}{N}\right]^{1/3}$	N is the dopant ion concentration
Reflection loss, R_L	$R_L = \left[\frac{n-1}{n+1}\right]^2$	n is the refractive index
Molar refraction, R_m	$R_m = \frac{(n^2-1)}{(n^2+2)}V_M$	n is the refractive index; V_M is the molar volume
Molar electronic polarizability, α_m	$\alpha_m = \frac{R_m}{2.52}$	R_m is the molar refraction
Electronic polarizability, α_e	$\alpha_e = \frac{3V_m(n^2-1)}{4\pi N_A(n^2+2)}$	n is the refractive index; V_M is the molar volume; N_A is Avogadro's number
Oxygen packing density, OPD	$OPD = n\left[\frac{\rho}{M}\right]1000$	n is the number of oxygen atom; ρ is the density of the sample; M is the total molecular weight
Energy gap, E_g	$\sqrt{E_g} = 1.23\left[1-\left(\frac{R_m}{V_m}\right)\right]+0.98$	R_m is the molar refraction; V_m is the molar volume
Metallization criterion, M	$M = \left[1-\left(\frac{R_m}{V_m}\right)\right]$	R_m is the molar refraction; V_m is the molar volume

Results and discussion

The XRD patterns of the investigated glasses are shown in Fig. 2. It is well known that an amorphous glass is indicated by a broad curve without sharp peaks. All of the glass samples under study are amorphous, as no sharp peaks are observed in their X-ray diffraction spectra. Furthermore, the XRD patterns of all examined glass samples exhibit a broad hump at $2\theta \approx 25^0 - 28^0$, which confirms the presence of short-range atomic order in the glass structure.

Density is a useful parameter for examining changes in the structural compactness of the glass. Table 3 presents the measured values of density, molar volume, and refractive index for the $CaF_2-P_2O_5$:MoO_3 glass samples [9]. Various physical parameters, such as electronic polarizability, dopant ion concentration, polaron radius, molar refraction, interionic distance, and field strength, were calculated from the measured density, molar volume, and refractive index, and are also presented in Table 3.

The density and molar volume of the samples were only slightly affected by variations in the MoO_3 concentration. As the MoO_3 content in the glasses increases, the density ranges from 2.627 to 2.673 g/cm^3 and shows a slight increase (Fig. 3). This unexpected result can be explained by the replacement of the higher molecular mass P_2O_5 with the lower molecular mass MoO_3 [10]. The incorporation of molybdenum ions into the glass network changes the total molecular weight of the glass. In addition, the small ionic radius of the molybdenum ion (0.65 Å) allows it to occupy interstitial spaces in the glass structure, thereby generating a more compact network. The observed decrease in molar volume is attributed to the short bond lengths and low coordination number in the prepared A_4 glass sample compared with the other glasses, leading to a reduction of free volume within the glass network (Fig. 3). The more open glass network structure can be attributed to the increase in MoO_3 concentration at the expense of P_2O_5.

The refractive index is a significant optical property, and the refractive index values of various glass materials can be used to evaluate their suitability for optical applications [11]. The refractive index values listed in Table 3 increase with increasing MoO_3 concentration. In the present glass samples, the refractive index rises in tandem with an increase in the electronic polarizability of oxide ions (Fig. 4), which results from an increase in the ratio of non-bridging oxygen to bridging oxygen. In materials research, the polarizability approach is well established in glass science and is widely used for the assessment of the electronic polarizability of ions [12].

Analysis of the oxygen packing density (OPD) values of the glass samples provides insight into the stiffness of the oxygen network in the glass. As listed in Table 3 and shown in Fig. 4, the OPD increases with increasing MoO_3 content. The increase in oxygen packing density may lead to a reduction in molar volume and, consequently, an increase in the mass density of the studied glass samples. This suggests that as more linkages form in the matrix, the glass structure becomes more compact [13]. To evaluate the potential for metallization and to study the insulating behavior of the glass system, the metallization criterion of the glass samples was calculated [14]. The metallization parameter values, M, for the examined

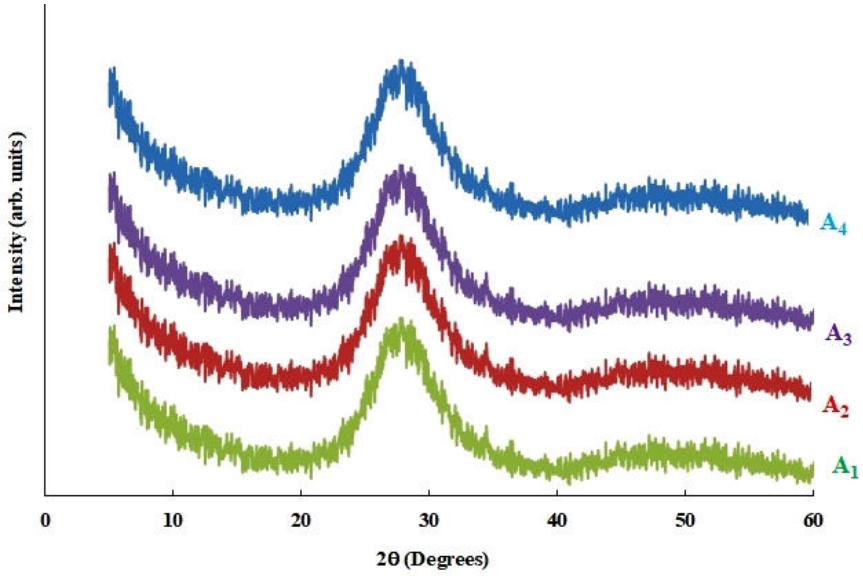

Fig. 2. X-ray diffraction pattern of $CaF_2-P_2O_5$:MoO_3 glasses

glasses are less than 1 and show an increasing trend with MoO_3 content, indicating that the glasses are less insulating in nature [15]. The M values for the present glasses range from 0.366 to 0.369. Figure 5 shows the variation of M with different MoO_3 concentrations.

Conclusions

CaF_2–P_2O_5 glasses with different concentrations of MoO_3 were synthesized and characterized in terms of their physical properties. XRD analysis shows no sharp peaks, indicating that the prepared samples are amorphous. As the MoO_3 concentration increases,

Table 3

Physical parameters of CaF_2–P_2O_5:MoO_3 glasses

Parameter	Value				
	A_0	A_1	A_2	A_3	A_4
Density, ρ (g/m^3)	2.627	2.639	2.650	2.662	2.673
Average molecular weight, $\overline{M}$	222.14	221.44	220.74	220.04	219.34
Refractive index, n_d	1.654	1.655	1.656	1.658	1.660
Molar volume (cm^3/mol)	84.56	83.93	83.30	82.67	82.06
Dopant ion concentration, N·10^{-21} (ions/cm^3)	–	3.588	7.231	10.928	14.680
Polaron radius, R_p (Å)	–	2.632	2.804	1.816	1.646
Interionic distance, R_i (Å)	–	6.532	5.171	4.506	4.084
Reflection loss, R_L·10^{-2}	6.072	6.086	6.100	6.128	6.156
Molar refraction, R_m	30.99	30.80	30.60	30.45	30.29
Molar electronic polarizability, α_m	12.29	12.22	12.14	12.08	12.01
Electronic polarizability, α_e	1.453	1.455	1.457	1.460	1.464
Oxygen packing density, OPD	94.60	95.30	96.04	96.77	97.49
Energy gap, E_g (eV)	3.095	3.093	3.091	3.087	3.083
Metallization criterion, M	0.6335	0.6330	0.6326	0.6317	0.6308

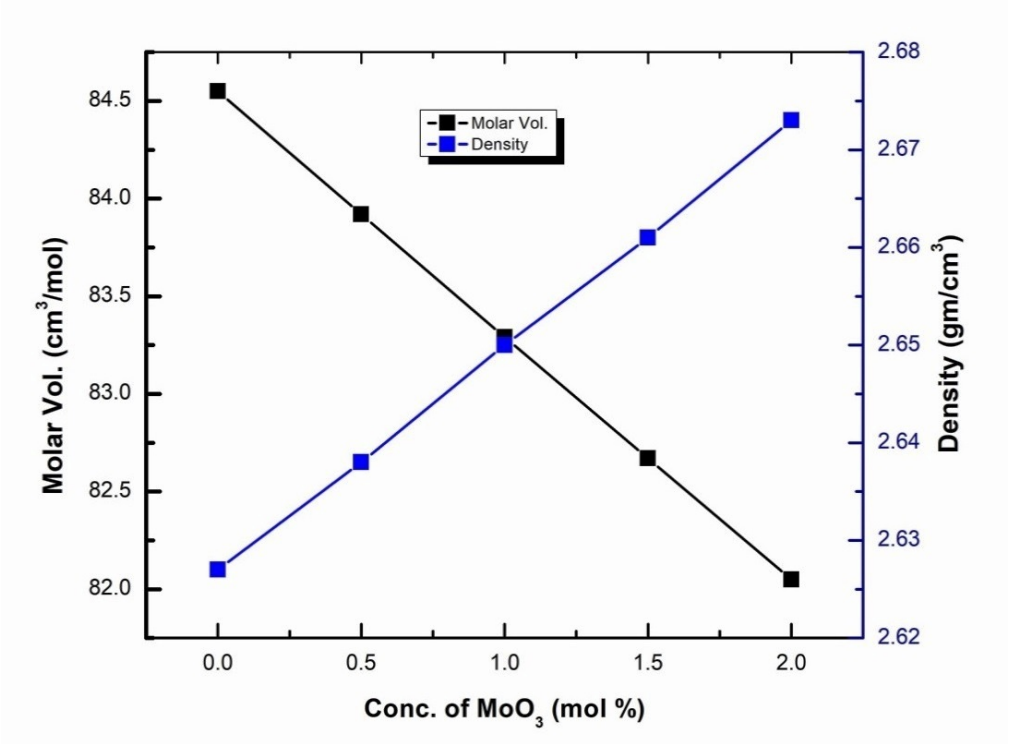

Fig. 3. Variation of density and molar volume with MoO_3 concentration

the density also increases. The refractive index and electronic polarizability rise due to the high polarizability and increasing amount of non-bridging oxygen in the glass network. Both the oxygen packing density (OPD) and polarizability increase with MoO_3 content.

The glasses exhibit metallization parameter values below one (around 0.6), indicating insulating behavior. However, with increasing MoO_3 concentration, the insulating nature slightly decreases, possibly due to a small fraction of bonds changing from ionic to covalent; in other words, the glasses tend toward a slightly less insulating behavior as the dopant concentration increases.

The increasing refractive index-based metallization criterion suggests a considerable potential

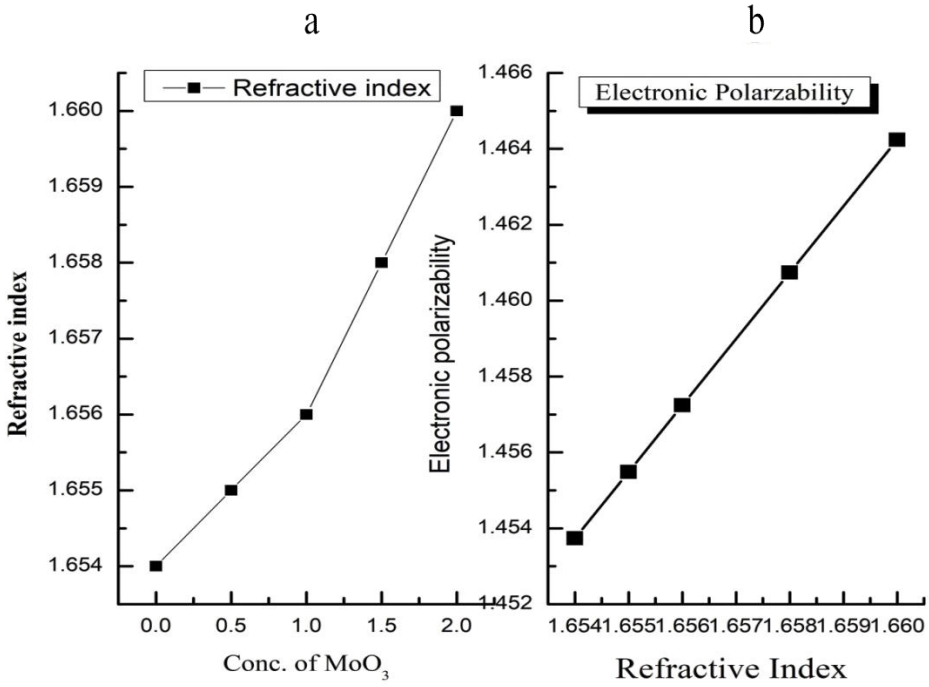

Fig. 4. (a) Variation of refractive index with MoO_3 content; (b) variation of refractive index with electronic polarizability

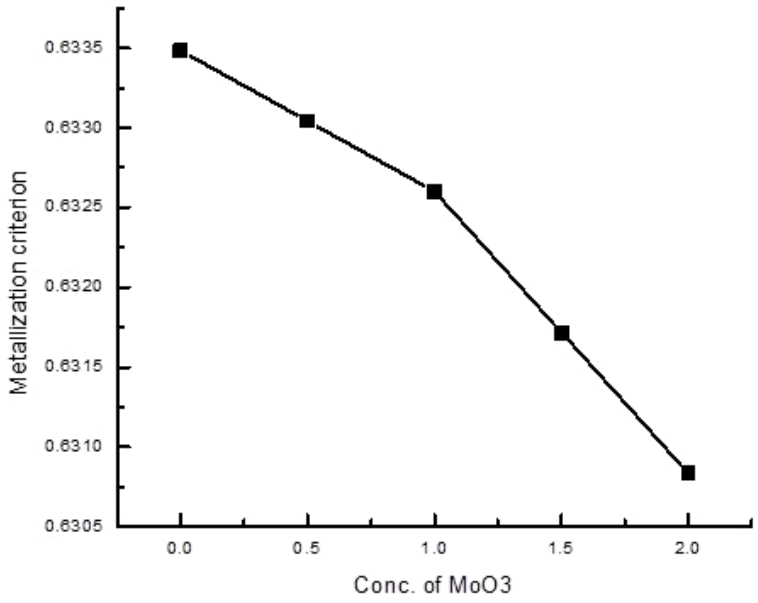

Fig. 5. Variation of the metallization criterion with MoO_3 content

for metallization of the fabricated glasses. The investigated glasses may serve as potential materials for applications such as computing and sealing frits due to their low glass transition temperature and low viscosity, radiation shielding due to high density (≈3 g/cm^3) and the presence of high atomic number metal ions like Mo, and photonic applications due to their increasing electronic polarization and refractive index.

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Conflicts of interest

The authors declare no conflict of interest.

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ФІЗИЧНІ ВЛАСТИВОСТІ СКЛЯНОЇ СИСТЕМИ $CaF_2–P_2O_5$:MoO_3

П. Нареш, Н. Нарасімха Рао, Ґ. Срідіві

У цьому дослідженні були одержані зразки потрійного скла складу $CaF_2–P_2O_5$:MoO_3 методом традиційного швидкого гартування з розплаву, і вивчено їх фізичні властивості. Структурні характеристики досліджували за допомогою рентгенівської дифракції, яка підтвердила аморфну природу одержаних зразків. Густини визначали експериментально, використовуючи принцип Архімеда. Отримані значення густини та молярного об'єму були використані для розрахунку різних фізичних параметрів, таких як концентрація іонів-допантів, міжіонні відстані, радіус полярону та сила поля. Молярна й електронна поляризованості, щільність упаковки кисню, оптична ширина забороненої зони та критерій металізації були оцінені на основі виміряних показників заломлення із застосуванням підходу Димитрова-Комацу щодо поляризованості. Результати свідчать, що введення оксиду молібдену підвищує хімічну стійкість і густину, водночас зменшуючи молярний об'єм. Виготовлені скла характеризуються значеннями параметра металізації меншими за одиницю, що вказує на зниження ізолюючої поведінки.

Ключові слова: скло P_2O_5; фізичні властивості; густина; показник заломлення; потрійне скло.

PHYSICAL PROPERTIES OF THE CaF_2–P_2O_5:MoO_3 GLASS SYSTEM

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Keywords: P_2O_5 glass; physical properties; density; refractive index; ternary glass.

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