

MCCUMBER ANALYSIS AND GAIN CROSS-SECTION OF Er^{3+} ACTIVATED GADOLINIUM PHOSPHATE OXYFLUORIDE GLASSES FOR BROADBAND COMMUNICATION MATERIALS

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Abstract

Er^{3+} activated gadolinium phosphate oxyfluoride glasses have been prepared via melt-quenching process and measured the absorption and luminescence spectra. The absorption spectra of glasses have shown in UV-VIS and Near-infrared region. The absorption intensity of glasses was enhanced as a function of Er_2O_3 concentrations. With laser diode excitation at 980 nm, the emission in near infrared region has shown a prominent peak at 1536 nm ($^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$) which is important peak for transmission windows. The absorption and emission measurements led to calculate the stimulated emission cross-sections by McCumber analysis. It was found to be $2.66 \times 10^{-20} \text{ cm}^2$ for 2.0 mol% of Er_2O_3 . Furthermore, the positive gain cross-section of glass is identified when the population inversion (P) is more than 0.4. It is pointed out the flat gain bandwidth in wavelength range 1400 - 1700 nm will be achieved when the P exceeds 40% which covered S, C and L bands.

Keywords: Erbium; Gain Cross-Section; McCumber Analysis; Phosphate Oxyfluoride Glasses

Introduction

In modern society, the rapid development of computer networks and optical telecommunication has heightened the demand for enhanced transmission capacity in Wavelength Division-Multiplexing (WDM) systems. Many researchers

have investigated the development of integrated optical amplifiers with wide bandwidths, which are crucial for WDM applications (Hau *et al.*, 2013). The incorporation of rare earth (RE) ions into glass plays a significant role in advancing

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various photonic applications, including solid-state lasers, sensors, light converters, 3D displays, optical amplifiers, and optical fiber communications (El-Maaref *et al.*, 2021). Among RE-doped materials, Er^{3+} -doped glass has garnered significant research interest. For example, Alvarez-Ramos *et al.* reported the optical and structural properties of Er^{3+} -doped tellurite-titanium-zinc glasses. Aliyu *et al.* studied the spectroscopic properties of Er_2O_3 -doped silica-borotellurite glasses (Aliyu *et al.*, 2021), and Prabhu *et al.* presented spectroscopic studies of Er^{3+} -doped borate glass systems. The strong interest in Er^{3+} arises from its emission wavelength around 1.53–1.55 μm , which is applicable in optical fiber amplifiers, infrared lasers, optical waveguides, and LIDAR systems (Guo *et al.*, 2012). Among host glasses, Er^{3+} -doped silicate glass faces limitations such as a restricted amplification bandwidth of about 35 nm, low doping concentration, and a non-uniform gain spectrum (Kesavulu *et al.*, 2014). On the other hand, borate glass has high phonon energy compared to phosphate glass. Phosphate glass offers several advantages, including low melting temperature, high Er^{3+} dopant concentration, excellent optical properties, a high thermal expansion coefficient, and high transparency. However, the hygroscopic nature of phosphate glass is a notable drawback. To address this limitation, alkali fluoride is added to the phosphate glass network, resulting in oxyfluorophosphate glass. This combination takes advantage of the fluoride reacting with hydroxyl groups to form hydrofluoric acid, thereby mitigating the hygroscopic nature. Oxyfluorophosphate glass is an attractive host material because it combines the beneficial properties of phosphate and fluoride glasses. It enables the fabrication of compact and highly efficient fiber amplifiers while providing excellent fracture toughness, making it suitable for solid-state laser applications. Additionally, alkali ions in the glass matrix increase the solubility of Er^{3+} ions and reduce the melting temperature (Kesavulu *et al.*, 2014; Rajagukguk *et al.*, 2019).

This study investigates PKAYGER glasses by measuring their density, molar volume, refractive index, absorption spectra, and near-infrared (NIR) emission spectra. Furthermore, McCumber's theory is applied to evaluate the emission cross-section and gain bandwidth of the glasses.

Materials and Methods

PKAYGER glasses with the chemical composition $(64-x)\text{P}_2\text{O}_5-15\text{KF}-10\text{AlF}_3-5\text{YF}_3-6\text{Gd}_2\text{O}_3-x\text{Er}_2\text{O}_3$ (where $x = 0.1, 0.5, 1.0, 1.5, 2.0$, and 4.0 mol%)

have been prepared by the melt quenching technique. In an agate mortar, 20 g of batch constituents were completely mixed, and the homogenous mixture was placed in an alumina crucible and maintained in an electric furnace at a temperature close to 1200°C , for 3 h. Melted poured into a warmed brass mold, then annealed at 500°C , 3 h to reduce thermal stress and strains before cooling to ambient temperature. Glasses were then buffed to provide good transparency and smooth surfaces for optical tests.

Archimedes measured the density of glasses (ρ) by weighing samples of glasses in air and water. The absorption spectra were measured with a UV-Vis-NIR spectrophotometer (Shimadzu UV-3600) in the 300–2400 nm wavelength range. The near-infrared (NIR) emission spectra with stimulating at 521 and 980 nm were measured using spectrofluorometer Quanta Master (QM)-300 made by Photon Technology International (PTI) Horiba. The Judd-ofelt theory was used for analysis of the symmetry as equation and quality factor (χ), while emission cross-section and gain bandwidth obtain by McCumber's theory.

Results and Discussion

Figure 1 presents the density, molar volume, and refractive index of PKAYGER glasses. The density of the glasses increases with higher Er_2O_3 concentrations, primarily due to the higher molecular weight of Er_2O_3 (382.5182 g/mol) compared to P_2O_5 (191.9445 g/mol). Similarly, the molar volume of the glasses increases with rising Er_2O_3 concentrations. This is attributed to Er^{3+} ions acting as network modifiers, where the addition of Er_2O_3 reduces the number of bridging oxygens (BOs) and increases the number of non-bridging oxygens (NBOs). Meanwhile, the refractive index also increases with higher Er_2O_3 concentrations due to the corresponding rise in glass density (Kesavulu *et al.*, 2014).

Figure 2 illustrates the absorption spectra of PKAYGER glasses in the wavelength range 300–1800 nm at room temperature. Ten peaks because of Er^{3+} ions (Aliyu *et al.*, 2021). The $^4\text{I}_{15/2}$ to $^4\text{G}_{11/2}$, $^2\text{H}_{11/2}$ transitions show the highest absorbance among all transitions. Both transitions are hypersensitive transition and comply with the selection rule of $|\Delta S| = 0$, $|\Delta L| \leq 2$ and $|\Delta J| \leq 2$ (Sailaja *et al.*, 2021). However, the absorbance of present glass is steadily rising since increasing Er_2O_3 concentrations. The JO parameters of PKAYGER2 glass are analyzed by Judd-Ofelt theory and showed a trend to be $\Omega_2 > \Omega_6 > \Omega_4$. It indicated more covalent and asymmetry of the

ligand field around Er^{3+} site. χ value of PKAYGER2 glass is calculated by the ratio between Ω_4 and Ω_6 . It shows 0.51 which is more than Er^{3+} : YAG (Linganna *et al.*, 2015) and higher than other reports as shown in Table 1. The larger χ value conveys high fluorescence performance of the $^4\text{I}_{13/2}$ to $^4\text{I}_{15/2}$ transition in PKAYGER2 glass.

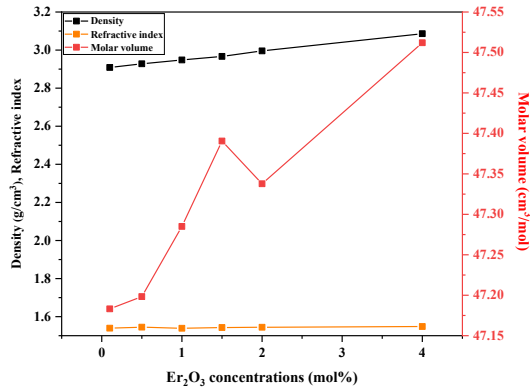


Figure 1. Density, molar volume, and refractive index of PKAYGER glasses for different concentrations

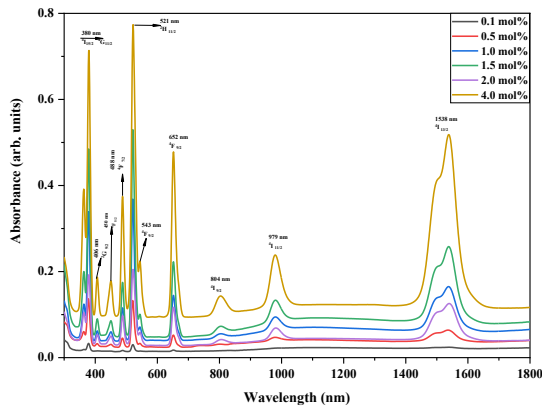


Figure 2. Absorption spectra of PKAYGER glasses for different concentrations

The near-infrared emission spectra of PKAYGER glasses under excitation at 521 nm and 980 nm are shown in Figure 3 and Figure 4. Both excitations produced a dominant peak at 1536 nm, corresponding to the $^4\text{I}_{13/2}$ to $^4\text{I}_{15/2}$ transition, which is suitable for optical communication, infrared laser applications, and eye-safe lasers. When comparing the emission intensity at 1536 nm across different Er_2O_3 concentrations, the intensity increased progressively up to 2.0 mol% but decreased at 4.0 mol% due to the concentration quenching effect (Basavapoornima *et al.*, 2017). Furthermore, the

emission intensity at 1536 nm under 980 nm excitation was higher than that under 521 nm excitation. Another notable parameter is the broad emission range, which spans three optical communication bands: the S-band, C-band, and L-band. These bands are critical for WDM applications and infrared laser technologies (Basavapoornima *et al.*, 2017).

Table 1. Comparison of Judd-Ofel parameters ($\Omega_\lambda \times 10^{-20} \text{cm}^2$) and spectroscopic quality factor (χ) of PKAYGER2 glasses.

Glass	Ω_2	Ω_4	Ω_6	Trends	χ
PKAYGER2 [present work]	3.61	0.66	1.29	$\Omega_2 > \Omega_6 > \Omega_4$	0.51
PKSAE10 (Linganna <i>et al.</i> , 2015)	6.14	0.61	1.40	$\Omega_2 > \Omega_6 > \Omega_4$	0.44
OCBTER1.0 (Sailaja <i>et al.</i> , 2021)	2.45	0.87	3.91	$\Omega_6 > \Omega_2 > \Omega_4$	0.22
PBG (Kassab <i>et al.</i> , 2005)	0.95	2.01	4.30	$\Omega_6 > \Omega_4 > \Omega_2$	0.47
0.5Er (Rayappan, and Marimuthu, 2013)	4.98	2.15	2.53	$\Omega_2 > \Omega_6 > \Omega_4$	-
LiEr 2.0 (Guo <i>et al.</i> , 2013)	9.18	1.53	1.65	$\Omega_2 > \Omega_6 > \Omega_4$	-
ErNAP (Kaewnuam <i>et al.</i> , 2018)	2.97	0.49	0.54	$\Omega_2 > \Omega_6 > \Omega_4$	-
Tellurite (Mariyappan <i>et al.</i> , 2019)	4.93	1.30	1.31	$\Omega_2 > \Omega_6 > \Omega_4$	-
ZBLAY (Tian <i>et al.</i> , 2011)	3.08	1.46	1.69	$\Omega_2 > \Omega_6 > \Omega_4$	-

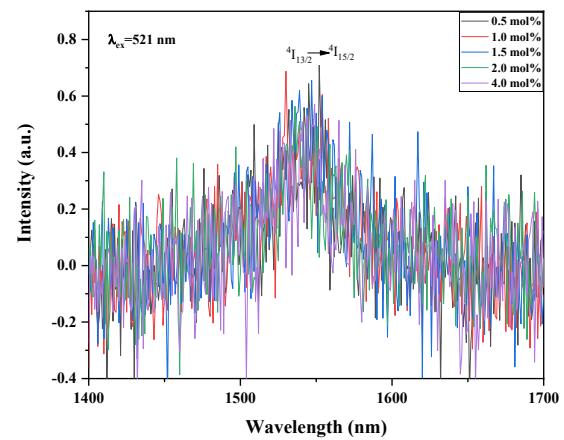


Figure 3. Absorption spectra of PKAYGER glasses for different concentrations

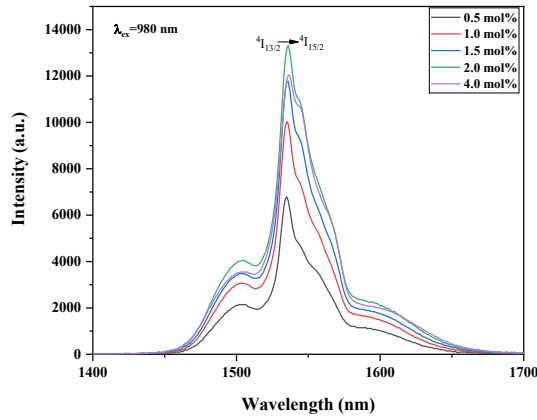


Figure 4. The emission spectra of PKAYGer glasses for different concentrations

The PKAYGer2 glass ($\lambda_{ex} = 980$ nm) was evaluated using the JO parameters obtained from absorption spectra measurements, as shown in Table 2. A_R represents the radiative transition probabilities, β_R denotes the luminescence branching ratios, and σ_e is the stimulated emission cross-section, which reflects the energy extraction rate from the optical material. These parameters are crucial for identifying an effective fluorescent host material. A high σ_e value indicates that the glass can be stimulated to emit laser light using low power while generating a significant amount of laser output. Moreover, the value for the emission wavelength of 1536 nm exceeds 0.5, demonstrating the material's capability to emit high-power laser light. In conclusion, the PKAYG glass doped with 2 mol% Er₂O₃ shows promising potential for further development as a material for green lasers (Ouannes *et al.*, 2014).

Table 2. Emission peak wavelength (λ_P (nm)), A_R (s⁻¹), β_R and σ_e for the observed emission transition of PKAYGer

Transition parameter	λ_P	A_R	β_R	σ_e
$^4I_{13/2} \rightarrow ^4I_{15/2}$	1536	227.77	1	8.38

From McCumber's theory, the absorption (σ_{abs}) and emission cross section (σ_{em}) play an essential role in evaluating the efficiency of the device. The absorption spectrum of $4I_{15/2}$ to $4I_{13/2}$ transition of PKAYGer2 glass was used for the calculation of the absorption cross section, which is shown in Figure 5. According to Beer-Lambert's equation, the σ_{abs} is given as (Meejitpaisan *et al.*, 2023).

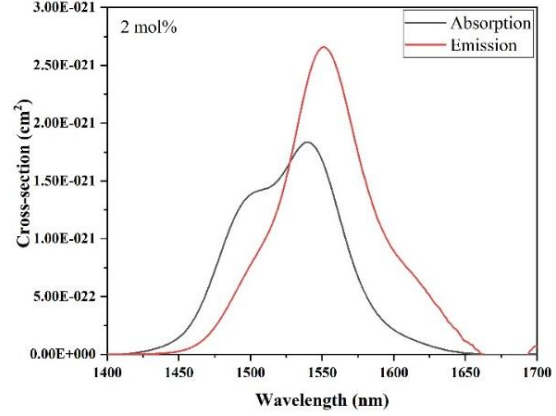


Figure 5. Absorption and emission cross-section of PKAYGer2 glass

$$\sigma_{abs} = \frac{2.303 \log \left[\frac{I_0}{I} \right]}{Nl} \quad (1)$$

in which I_0 parameter is the incident optical intensity, I parameter is the output optical intensity, N parameter is the Er³⁺ ion concentration (ion/cm³), and l parameter are the thickness of PKAYGer2 glass. Meanwhile, the emission cross-section of PKAYGer2 glass can be received by McCumber theory as equation (Meejitpaisan *et al.*, 2023).

$$\sigma_{em} = \sigma_a \exp \left(\frac{\epsilon - hv}{kT} \right) \quad (2)$$

Another crucial factor in the efficiency of laser media is the optical gain coefficient. From the absorption cross-section ($\sigma_{abs}(\lambda)$) and emission cross-section ($\sigma_{emi}(\lambda)$) values, by using the follow equation (3).

$$G(\lambda) = \gamma \sigma_{emi}(\lambda) - (1 - \gamma) \sigma_{abs}(\lambda) \quad (3)$$

The gain cross-section values for $\gamma = 0.0, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9$ and 1.0 are shown in Figure 6 for the current glass sample. As illustrated in Figure 6, the gain becomes positive when the population inversion value exceeds 0.4 for all these glasses. This indicates that, with a population inversion greater than 40% , the sample exhibits a smooth increase in gain bandwidth within the 1400 - 1700 nm range, which corresponds to the C-band (1530 - 1565 nm) and L-band (1565 - 1625 nm) used in optical communication systems (Meejitpaisan *et al.*, 2023).

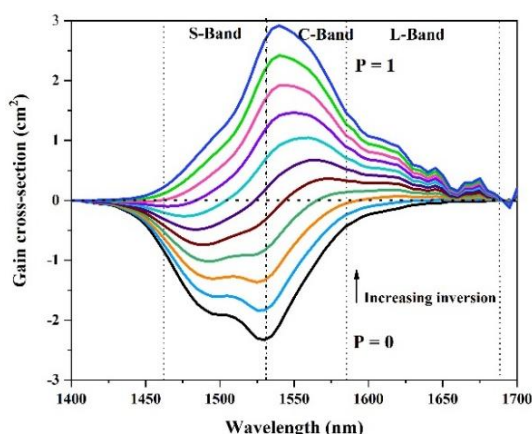


Figure 6. Gain spectra of PKAYGe2 glass for different values of the population inversion factor of P, ranging from 0.0 to 1.0

Conclusions

The results showed that increasing the concentration of Er_2O_3 in the glass led to increases in glass density, molar volume, and refractive index. Higher Er_2O_3 concentrations enhanced the glass's ability to absorb photons in the UV-VIS and NIR regions. NIR emission under excitation at 521 nm and 980 nm exhibited a dominant peak at 1536 nm, corresponding to the $^4\text{I}_{13/2}$ to $^4\text{I}_{15/2}$ transition. This transition has potential applications in broadband communication materials. Comparing emission intensities at different Er_2O_3 concentrations revealed that the intensity at 1536 nm increased progressively up to 2.0 mol% of Er_2O_3 but decreased at 4.0 mol%. The J-O intensity parameters calculated for the glass exhibited a trend of $\Omega_2 > \Omega_6 > \Omega_4$ for 2PKAYGe glass. Using J-O theory, the luminescence branching ratios and the emission cross-section at 1536 nm were determined to be 1 and $8.38 \times 10^{-21} \text{ cm}^2$, respectively. Additionally, McCumber's theory was applied to calculate the stimulated emission cross-section for the $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ NIR transition at 1536 nm, yielding a value of $2.66 \times 10^{-21} \text{ cm}^2$. The gain cross-section of 2PKAYGe glass, as derived from McCumber's theory, supported an inverse population threshold of $\gamma \geq 0.4$. In conclusion, the analysis indicates that PKAYGe2 glass is well-suited for communication applications, particularly as a material for broadband communication systems.

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