

## Article

# Thermal, Optical, and Emission Traits of $\text{Sm}^{3+}$ -Ion-Doped Fluoride/Chloride/Oxide Glass for Red/Orange Laser Fiber Applications

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**Abstract:** This study examined spectroscopic, thermal, and other qualities, such as the lasing parameters, of  $\text{Sm}^{3+}$ -doped glass with the composition  $40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$ . The ellipsometric data were used in a Sellmeier dispersion relation to estimate the refractive index values of the glasses investigated. The measured absorption spectra of the doped glass reveal the presence of various absorption bands assigned to transitions from the  $^6\text{H}_{5/2}$  ground state attributed to  $\text{Sm}^{3+}$ -ion-excited states. We studied the decay of the  $^4\text{G}_{5/2}$  level of the  $\text{Sm}^{3+}$  ions in the doped glass by analyzing its absorption and emission fluorescence spectra. The Judd–Ofelt hypothesis allowed us to determine that the quantum efficiency of the  $^4\text{G}_{5/2}\text{-}^6\text{H}_{7/2}$  transition is high: 96% and 97% for glass doped with  $4.05 \times 10^{19}$  ions/ $\text{cm}^{-3}$  and  $11 \times 10^{19}$  ions/ $\text{cm}^{-3}$ , respectively. Furthermore, this glass exhibits efficient red/orange enhanced spontaneous emission that matches the excitation band of the photosensitizer material used in medical applications.

**Keywords:** thermal stability; absorption cross-section; emission spectra; refractive index; spectroscopic quality parameters; quantum efficiency



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## 1. Introduction

Phosphate glasses possess high thermal stability, relatively low melting temperatures, and unique optical properties, along with a broad range of transparency in the UV-VIS spectral range; however, they have a low refractive index [1]. Therefore, phosphate glasses doped with rare-earth (RE) ions are widely studied for their applications in waveguides, optical detectors, fiber optic amplifiers, and lasers [2]. The addition of various fluorides to RE-ion-doped phosphate glasses enables the creation of host matrices that combine the significant optical properties of fluoride glasses and the high stability of phosphate glasses. Oxides increase the mechanical resistance and thermal stability of the glasses, while low-frequency fluorides reduce the nonradiative decay losses caused by multifamily relaxation [3], improve the quantum efficiency and lifetimes of rare-earth ions' luminescence, and act as flux agents that reduce the melting temperature of the glasses. When samarium ions are doped into fluorophosphate glass, the resulting glass exhibits distinctive optical

behavior due to its emissions in the UV–visible region through various emission channels from the  $^4G_{5/2}$  level of  $Sm^{3+}$  ions [4]. Samarium ions ( $Sm^{3+}$ ) are regarded as excellent luminescent centers among the rare-earth ( $RE^{3+}$ ) ions due to their strong emissions at longer wavelengths in the red/orange region. The strong luminescence in the red/orange region, as well as the high quantum efficiency value of  $Sm^{3+}$ -doped  $P_2O_5$ – $Na_2O$ – $BaO$ – $Al_2O_3$  glasses, makes them a potential candidate for laser emission [5].

This characteristic makes them highly suitable for applications in solid-state lasers, LEDs, and display devices. Additionally, the lowest emitting level of  $Sm^{3+}$  ions,  $^4G_{5/2}$ , exhibits high quantum efficiency and various quenching emission channels [6,7]. The  $^4f_5$  configuration of samarium ions with strong orange-red fluorescence in the visible region is used in underwater communications, color displays, visible semiconductor lasers [8–10], and phosphors for white light-emitting diodes (LEDs) [11,12]. Although extensive research has been conducted on phosphate glasses doped with  $Sm^{3+}$  ions [13–16], there is still a need to explore new and advanced compositions of phosphate glass due to their potential applications.

In [17], the researchers confirmed the formation of defects and the conversion of  $Sm^{3+}$  into  $Sm^{2+}$  ions in  $Sm$ -doped fluorophosphate glasses exposed to X-ray irradiation. Additionally, Hamdy et al. [18] synthesized compositions containing 20, 30, and 50 mol% NaF,  $AlF_3$ , and  $PF_5$  doped with  $Sm^{3+}$  to investigate the impact of  $Sm^{3+}$  on optical transmission and photoluminescence (PL) spectroscopy. The authors of [19] discussed the influence of the chemical composition of  $[(45 - x) P_2O_5 - 10AlF_3 - 45 NaF \times Sm_2O_3]$  (where  $x$  equal 0 to 1 mol%) glasses on their optical properties, mainly their linear and non-linear parameters. The study performed by [20] focused on examining the thermal, structural, and luminescent characteristics of sodium barium metaphosphate glasses doped with  $Sm^{3+}$ . Samarium-doped fluoroaluminate and fluorophosphate glasses studied by Chicilo et al. [21] proved to be excellent candidates for high-resolution, large-dynamic-range microbeam radiation therapy (MRT) dosimetry. It is worth noting that  $Sm^{3+}$  ions are ideal for doping because their excited energy level  $^4G_{5/2}$  has a high quantum efficiency with various quenching emission channels. It emits strong reddish-orange light and possesses sufficient energy to initiate photodynamic reactions. Recently, lasers with excellent directivity and high intensity, as well as LEDs with relatively narrow spectral bandwidths and high fluence rates, have been specifically developed for photodynamic therapy (PDT) treatments. However, scattered or misdirected light from the target area can unintentionally expose large areas of normal tissue to high power densities, potentially leading to side effects such as inflammation, pain, swelling, burns, and scarring. While LEDs can be integrated with optical fibers, their low coupling efficiency limits their use. Additionally, their narrow excitation spectrum restricts the use of multiple photosensitizers, reducing the efficiency of therapy. Amplified spontaneous emission (ASE) fluorescence produced in rare-earth (RE) ion-doped glass channel waveguides offers a broadened bandwidth that can be tuned by adjusting the RE ion concentration and pumping power. This type of light source delivers sufficient intensity, excellent directivity, and high coupling efficiency, making it a promising option for use in photodynamic therapy (PDT) treatments [22].

A fluorophosphate glass channel waveguide produces amplified spontaneous emission (ASE) fluorescence with a broad width of 600–730 nm. This light source has excellent efficiency, strong intensity, and good directivity, thus making it an attractive option for use in photodynamic therapy (PDT) [22,23]. Red light in the 600–730 nm spectral band has 50–200% greater penetrating power than light in the 400–500 nm region, and it possesses more energy to initiate a photodynamic reaction that produces  $^1O_2$  according to most tissue models [22]. A diverse range of light sources, both coherent and incoherent, have demonstrated their effectiveness in achieving anti-tumor effects in PDT for various superficial and interstitial treatment sites [24–26]. Thus, in this paper, we have focused on the lasing parameters of  $Sm^{3+}$ -doped fluorophosphate glasses as a candidate light source for photodynamic therapy (PDT). In the glass compositions, we included  $BaF_2$  ions as network modifier ions with a low field strength and ZnO to improve the mechanical strength,

chemical durability, and hygroscopic nature, which, in turn alter, the optical, electrical, and magnetic properties of phosphate glasses.

## 2. The Experimental Section

The composition of the glasses was chosen as  $40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$  with different concentrations of  $\text{Sm}^{3+}$  ions: sample 1 (SM1) at  $4.05 \times 10^{19} \text{ cm}^{-3}$  and sample 2 (SM2) at  $11 \times 10^{19} \text{ cm}^{-3}$ . The following chemicals were used for batch production: phosphorus oxide ( $\text{P}_2\text{O}_5$ ), zinc oxide ( $\text{ZnO}$ ), lithium chloride ( $\text{LiCl}$ ), barium fluoride ( $\text{BaF}_2$ ), and samarium oxide ( $\text{Sm}_2\text{O}_3$ ). All of the raw materials were carefully mixed. The fluorophosphate glasses were obtained by melting 50 g batches in a gold crucible in an electric furnace at a temperature of  $850^\circ\text{C}$  in an air atmosphere [27]. The densities of the glasses studied were determined using Archimedes' method (Table 1).

**Table 1.** Volume concentration (N), glass matrix composition (mol%), density, and refractive index at 633 nm.

Sample	Composition (mol%)	N ( $10^{19} \text{ cm}^{-3}$ )	Density ( $\text{g/cm}^3$ )	n
SM	$40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$	----	3.872	1.588
SM1	$40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2\text{-}X\text{Sm}_2\text{O}_3$	4.05	3.792	1.585
SM2	$40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2\text{-}Y\text{Sm}_2\text{O}_3$	11.00	3.821	1.579

Their refractive indexes were determined with use of a Woollam M-2000 ellipsometer; the spectra were recorded in the 190–1700 nm spectral range. The transmittance and reflectance spectra were recorded using Jasco V-570 spectrophotometers [27]. Luminescence spectra were measured using the Optron Dong Woo fluorometer system [28]. The luminescence decay curves were measured following short-pulse excitation provided by an optical parametric oscillator with the third harmonic of a Nd YAG laser [29,30]. Changes in the thermal behavior of the  $40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$  glasses were investigated with the DTA/DSC method using the PerkinElmer DTA-7 System.

Under a flowing air environment ( $80 \text{ mL min}^{-1}$ ), the glass samples were ground into powders with grain sizes of 0.1 to 0.3 mm and then heated in platinum crucibles at a rate of  $10^\circ\text{C min}^{-1}$ .

Alumina oxide ( $\text{Al}_2\text{O}_3$ ) was used as a reference material. Several thermal parameters characteristic of the glassy state have been established, i.e., the glass transformation temperature ( $T_g$ ) and the glass crystallization temperature ( $T_c$ ). Using the midpoint of the corresponding transformation phase as the glass conversion temperature ( $T_g$ ) and the start and maximum of the glass crystallization peak as the parameters for the glass melting and solidification processes, we were able to obtain the glass heating and solidification temperatures.

While the midpoint of the corresponding transformation step was used to estimate the glass conversion temperature ( $T_g$ ), the start ( $T_x$ ) and maximum ( $T_c$ ) of the glass crystallization peak were used to calculate the glass crystallization temperature.

The characteristic glass temperature values that appeared in DSC were determined using Proteus Thermal Analysis (Version 5.0.0.). The amorphous nature of the glasses studied was confirmed using the X-ray diffraction (XRD) method and a Philips X'Pert X-ray diffractometer with  $\text{CuK}_\alpha$  radiation. All of the research was conducted at room temperature. The refractive index of the bulk glasses was investigated using transmission mode ellipsometric data. The ellipsometric results were registered in the range of 400–1700 nm, and n was modeled using the Sellmeier dispersion function. Therefore, the Sellmeier refractive index (n)'s normal dispersion, which was determined from our

ellipsometric data collected as a function of the wavelength  $\lambda$  (see Equation (1)) [31,32], was fitted to the low-absorption region, which is 400–1700 nm.

$$n^2 = A + B\lambda^2/(\lambda^2 - C^2) - D\lambda^2 \quad (1)$$

where A, B, C, and D are the fitting parameters.

The refractive index dispersions of the glasses studied are quite similar to each other, as are the values of  $n$  measured at 633 nm (see Table 1). The addition of  $\text{Sm}_2\text{O}_3$  to the glasses led to a decrease in the refractive index compared to the sample SM without a dopant.

It is worth mentioning that ellipsometry is not a direct method for determining the refractive index.

### 3. Results and Discussion

#### 3.1. Thermal Analysis

The broad hump in the XRD profile shows the long-term structural instability (i.e., random atomic arrangements) of the glasses prepared (Figure 1). Therefore, based on XRD analysis of all the glasses, it can be found that they are amorphous. The differential scanning calorimetry (DSC) curves of all of the glasses studied showed distinct endothermic events associated with the glass transition  $T_g$ , as well as the melting point  $T_m$ . The exothermic event  $T_c$  associated with crystallization was observed. During heating, the glasses studied demonstrate several characteristic temperatures. The range of vitreous state transitions ( $T_{g,\text{endset}} - T_{g,\text{onset}}$ ) decreases progressively when the  $\text{Sm}^{3+}$  ion concentration in the glass increases, although the transition temperature increases (Figure 2).

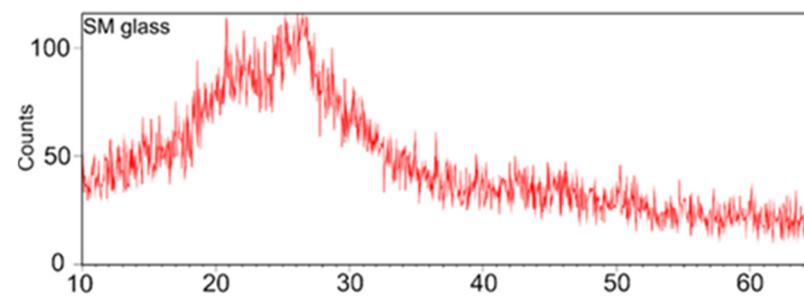


Figure 1. XRD pattern of the reference glass SM.

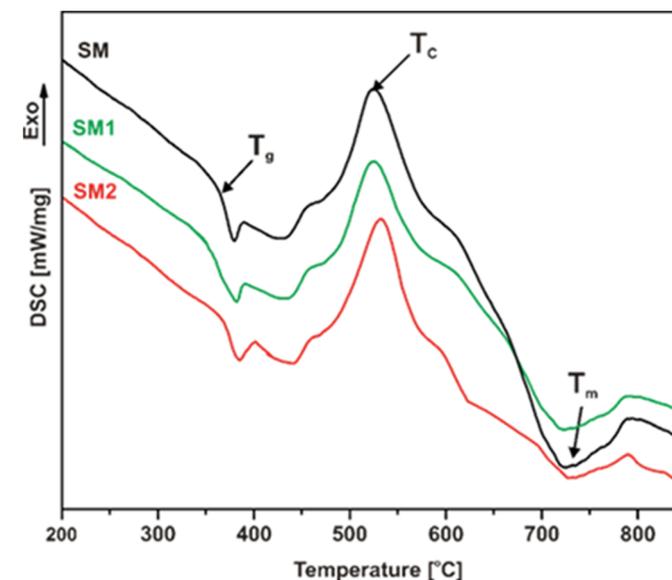


Figure 2. DSC curves of  $\text{Sm}^{3+}$ -doped fluorophosphate glasses.

The increase in  $T_g$  refers to structural changes in the glass network with increasing  $\text{Sm}^{3+}$ ; furthermore, the narrowing of the transition range of the vitreous state (i.e., a reduction in the relaxation time) supports the theory of increased rigidity in the material's internal structure (Table 2). According to [33], this process occurs via the breaking of chemical bonds rather than the displacement of structural units when the time required for structural strain relaxation shortens, maintaining the network's continuity. To determine the thermal behavior of glasses, many parameters have been established, mainly considering the dependencies between values of characteristic temperatures. To evaluate the stability of the glass, the following parameters were utilized: the Hruby ( $K_H = (T_x - T_g)/(T_m - T_x)$ ) [34], the Weinberg ( $K_W = (T_x - T_g)/T_m$ ), the Lu and Liu ( $K_{LL} = T_x/(T_g + T_m)$ ), and the Angell  $K_A = (T_x - T_g)$  parameters [35]. The resistance of the resulting material to crystallization is expressed by the term  $(T_x - T_g)$  in the suggested formula; a narrower gap indicates an increased tendency to crystallize. As the concentration of  $\text{Sm}^{3+}$  ions in the glasses increased, their  $K_A$  was in the 69–73 °C temperature range. Greater  $\text{Sm}^{3+}$  doping results in a higher  $K_A$  value, increasing the glass's thermal stability.

**Table 2.** Thermal characteristics of  $\text{Sm}^{3+}$ -doped fluorophosphate glasses.

Glass ID	$T_g$	$T_g$ range	$T_x$	$T_c$	$T_m$
SM	357	23	426	523	720
SM1	369	14	440	526	725
SM2	372	5	445	540	733

Abbreviations:  $T_g$ —glass transition temperature,  $T_x$ —endothermic onset of the maximum crystallization peak in glass;  $T_c$ —maximum crystallization peak in glass,  $T_m$ —endo thermic peak (glass melting temperature).

The stronger thermal stability of glass against devitrification on heating was further verified by the fact that the values of other thermal stability metrics, such as  $K_W$ ,  $K_H$ , and  $K_{LL}$ , increased with larger  $\text{Sm}^{3+}$  concentrations. Not only can one infer a material's stability from all these variables but one can also infer its glass-forming ability, which is the ease with which a liquid vitrifies upon cooling. Table 3 shows that the stability and glass-forming ability of  $40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$  glasses are enhanced when the  $\text{Sm}^{3+}$  ion concentration in their structure increases. Both Mawlud et al. and Mnjeet et al. [36,37] found similar correlations in their studies.

**Table 3.** Stability parameters of the  $\text{Sm}^{3+}$ -doped fluorophosphate glasses.

Glass ID	Hruby/ $K_H$	Weinberg/ $K_W$	Lu and Liu/ $K_{LL}$	$K_A$
SM	0.234	0.095	0.395	69
SM1	0.249	0.097	0.402	71
SM2	0.253	0.099	0.403	73

Abbreviations:  $K_A$ —Angell parameter,  $K_H$ —Hruby parameter,  $K_W$ —Weinberg parameter,  $K_{LL}$ —Lu and Liu parameter.

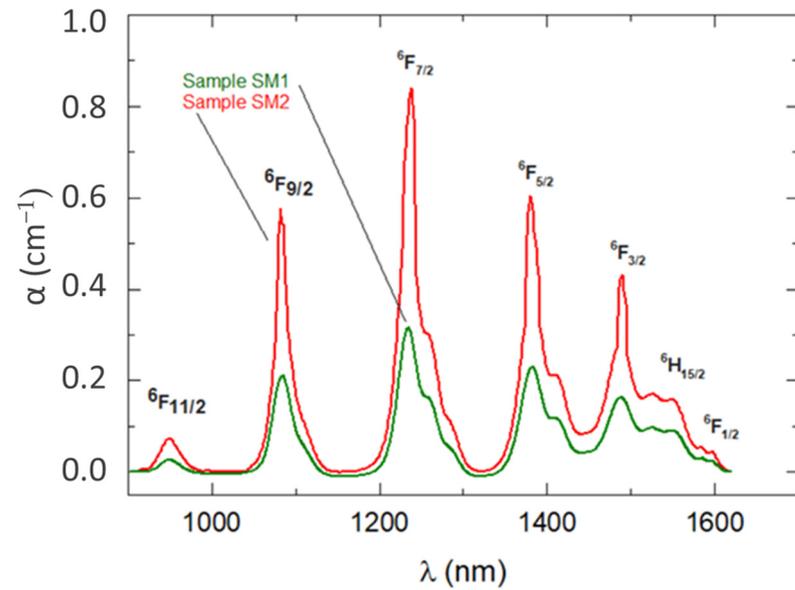
### 3.2. Absorption and Excitation Spectra

The transmittance and reflectance spectra allowed us to calculate the dispersion of the absorption coefficient of the fluorophosphate glasses doped with the  $\text{Sm}^{3+}$  ions. The results are illustrated in Figures 3 and 4.

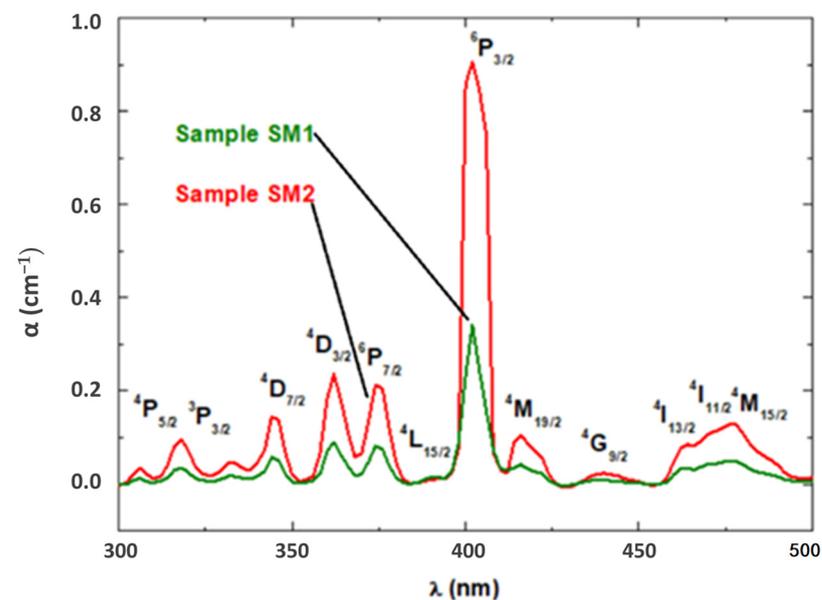
As shown in Figures 3 and 4, the absorption spectra of the  $\text{Sm}^{3+}$ -doped fluorophosphate glasses registered in the UV-VIS-NIR region showed 19 peaks. As can be seen in Figures 3 and 4, these peaks are caused by  $\text{Sm}^{3+}$  ions' transition from the  $^6\text{H}_{5/2}$  ground state to various energy levels [38,39].

The absorption spectra allowed us to observe a large number of energy transitions of the samarium ions in the glass from the system  $\text{P}_2\text{O}_5\text{-ZnO-LiCl-BaF}_2$ . Above 450–500 nm, in the SM1 and SM2 glasses, one may find the high energy manifolds built from the  $^4\text{D}$ ,  $^4\text{G}$ ,  $^4\text{I}$ ,  $^4\text{L}$ , and  $^4\text{M}$  quartets and  $^6\text{P}$  sextet terms. Because many  $^{2\text{S}+1}\text{L}_J$  splitters overlap, the UV-VIS bands that are visible are less intense and difficult to identify. Shifts in the

absorption edge towards the UV region are observed with an increasing  $\text{Sm}^{3+}$  ion content. However, the bands that formed in the near-infrared spectrum are more distinct and strongly differentiated. In the spectral range of 500–250 nm, there is a comparable effect; however, in contrast, intense absorption peaks at 402 nm occurred, and this band is associated with transitions ending in  $^4\text{M}_{19/2}$ ,  $^6\text{P}_{3/2}$ ,  $^4\text{L}_{15/2}$ ,  $^6\text{P}_{7/2}$ ,  $^4\text{D}_{3/2}$ ,  $^4\text{D}_{7/2}$ ,  $^3\text{P}_{3/2}$ , and  $^4\text{P}_{5/2}$  closely spaced multiples.

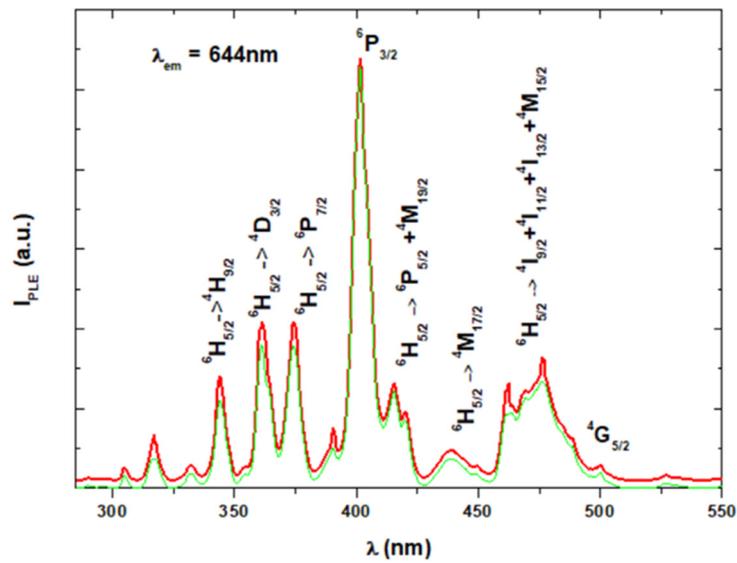


**Figure 3.** Absorption spectra of  $\text{Sm}^{3+}$ -doped fluorophosphate glass samples in the range of 800–1600 nm (NIR).



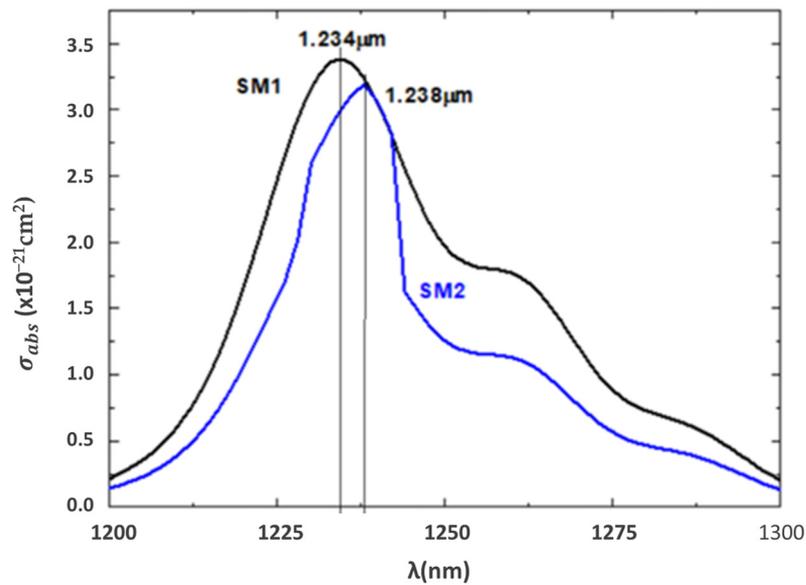
**Figure 4.** Absorption spectra of  $\text{Sm}^{3+}$ -doped fluorophosphate glass samples in the range of 300–500 nm (UV-VIS).

In addition, the absorption spectra of the glasses tested were supplemented by photoluminescent excitation spectra, which are shown in Figure 5.



**Figure 5.** The absorption spectra of phosphate glasses doped with  $\text{Sm}^{3+}$  were observed at 644 nm in the photoluminescence excitation (PLE) mode.

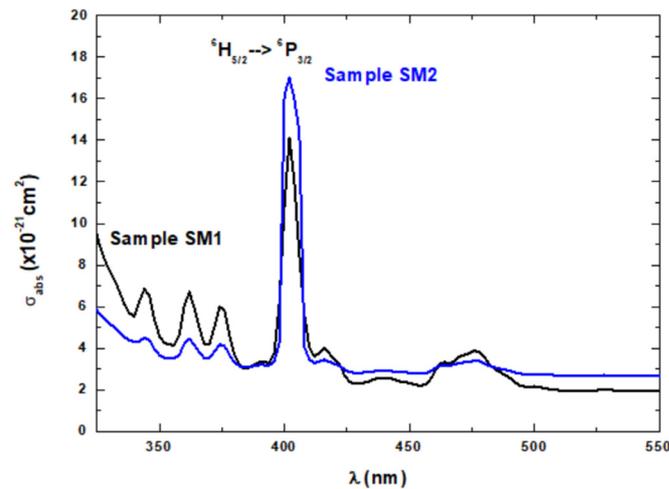
Figure 6 shows the absorption cross-section (ACS) of the  ${}^6\text{H}_{5/2} \rightarrow {}^6\text{F}_{7/2}$  transition, which is fundamental for samarium-doped fiber amplifiers. The ACS is defined as  $\sigma_{\text{abs}} = \alpha/N$  and is easily obtained from the absorption spectra shown in Figure 3.



**Figure 6.** Absorption cross-section  $\sigma_{\text{abs}}(\lambda)$  of investigated glasses for  ${}^6\text{H}_{5/2} \rightarrow {}^6\text{F}_{7/2}$  transition.

The most significant transitions of  $\text{Sm}^{3+}$  ions occur in the range of 1000–1300 nm, corresponding to the  ${}^6\text{H}_{5/2} \rightarrow {}^6\text{F}_{9/2}$  and  ${}^6\text{H}_{5/2} \rightarrow {}^6\text{F}_{7/2}$  transitions of  $\text{Sm}^{3+}$  ions (Figure 6).

Figure 7 presents the absorption cross-section ( $\sigma_{\text{abs}}$ ) calculated for the SM1 and SM2 glasses, and the maximum value at 402 nm was  $\sigma_{\text{abs}} = 17.0067 \times 10^{-21} \text{ cm}^2$  for SM2 and  $\sigma_{\text{abs}} = 14.1206 \times 10^{-21} \text{ cm}^2$  for SM1.



**Figure 7.** The calculated absorption cross-section ( $\sigma_{\text{abs}}$ ) for the investigated glasses for the  ${}^6\text{H}_{5/2} \rightarrow {}^6\text{P}_{3/2}$  transition of  $\text{Sm}^{3+}$  ions.

### 3.3. Judd–Ofelt Analysis

Here, we analyzed the absorption data using the conventional Judd–Ofelt (J–O) theory (Figures 3 and 4). Numerical integration of suitable absorption bands, excluding the background absorption of the glass arrays, was used to establish the experimental oscillator strengths for transitions from the  $\text{Sm}^{3+}$  ions' ground level  ${}^6\text{H}_{5/2}$  to subsequent excited levels.

Many intense transitions can be observed in the PLE spectrum (Figure 5), like those in the absorption spectra. In Table 4, the computed J–O parameters ( $\Omega_2$ ,  $\Omega_4$ , and  $\Omega_6$ ) can be observed. Judd–Ofelt parameters are a useful tool for determining the radiative properties of  $\text{Sm}^{3+}$ -doped glasses, which are strongly dependent on the host matrix. The values of the J–O parameters obtained [40,41] are comparable to values found in other phosphate glasses doped with  $\text{Sm}^{3+}$  and show the same trend, namely  $\Omega_4 > \Omega_6 > \Omega_2$  [42,43]. The values of  $\Omega_4$  and  $\Omega_6$  are associated with structural qualities like the viscosity and stiffness of the material, whereas the value of  $\Omega_2$  indicates the covalency of oxygen atoms and the asymmetry of the RE ion sites, according to J–O theory. The ion sites exhibit less asymmetry, leading to dominating covalent interactions between oxygen ligands and  $\text{Sm}^{3+}$  ions, as shown by the high value of  $\Omega_4$  compared to  $\Omega_2$  and  $\Omega_6$ . We may find out more about the luminescence activators by calculating the spectroscopic quality factor using the  $\Omega_4/\Omega_6$  ratio. The glasses examined show promise as luminescence activators, with computed spectroscopic quality factor values of 1.1 (SM1) and 1.5 (SM2), respectively [44,45].

**Table 4.** Judd–Ofelt intensity parameters ( $\Omega_t$ ) for fluorophosphate glasses doped with  $\text{Sm}^{3+}$  ions.

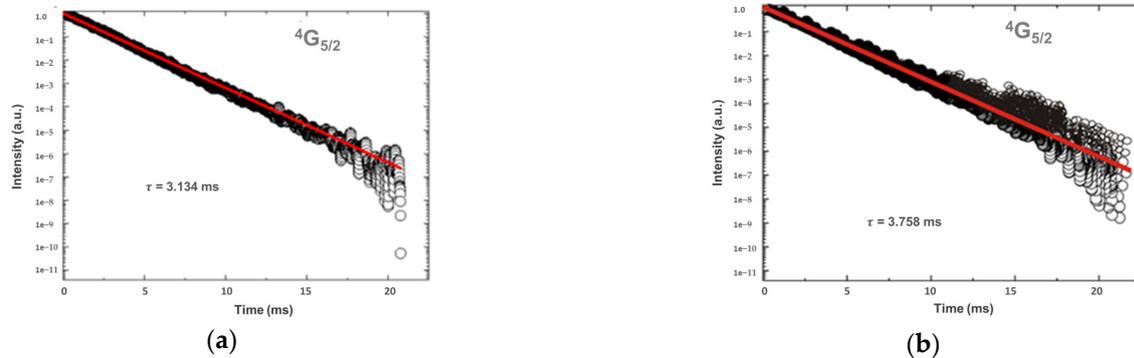
Glass ID	$\Omega_2$ ( $10^{-20} \text{ cm}^2$ )	$\Omega_4$ ( $10^{-20} \text{ cm}^2$ )	$\Omega_6$ ( $10^{-20} \text{ cm}^2$ )	$\sigma_{\text{rms}}$ ( $10^{-6}$ )
SM1	0.4327	1.1224	1.0071	0.354
SM2	0.9897	1.9765	1.4576	0.327

Three phenomenological spectroscopic parameters were calculated. In addition, the value of fitting quality ( $\sigma_{\text{rms}}$ ) estimated from the root mean square (RMS) deviation [46] was determined and is presented in Table 4. This value implies a good match between the measured  $P_{\text{exp}}$  and the calculated  $P_{\text{cal}}$  oscillator strengths.

The radiative transition probability ( $W_r$ ) and the total radiative lifetime ( $\tau_{\text{rad}} = 1/W_r$ ) of the emission level  ${}^4\text{G}_{5/2}$  were estimated using the J–O parameters determined. In the conventional J–O approach, the  $\Omega_t$  parameter values are derived using an averaging process that takes into account all the absorption bands, known as the least-squares method. The samples' observed lifetimes of the excited state  ${}^4\text{G}_{5/2}$  (Figure 8a,b) were considerably lower

than the expected radiation equivalent of  $\tau_{rad}$  (refer to Table 5) and those predicted by the following relationship [42]:

$$\eta = \frac{\tau_{exp}}{\tau_{rad}} \times 100\% \quad (2)$$



**Figure 8.** (a) Luminescence lifetimes of the  ${}^4G_{5/2}$  state of  $Sm^{3+}$  ions for sample SM1. (b) Luminescence lifetimes of the  ${}^4G_{5/2}$  state of  $Sm^{3+}$  ions for sample SM2.

**Table 5.** This study compares the experimentally obtained lifetimes of the  ${}^4G_{5/2}$  emission band in fluorophosphate glasses ( $\tau_{exp}$ ) with the lifetimes estimated using the J-O methods ( $\tau_{rad}^{J-O}$ ) and quantum efficiencies ( $\eta$ ).

Glass ID	${}^4G_{5/2}$ Lifetime (ms)		
	$\tau_{exp}$	$\tau_{rad}^{J-O}$	$\eta$ (%)
SM1	3.134	3.856	96
SM2	3.758	4.865	97

This gives more than 96% of the quantum efficiency of the excited state for the material under study.

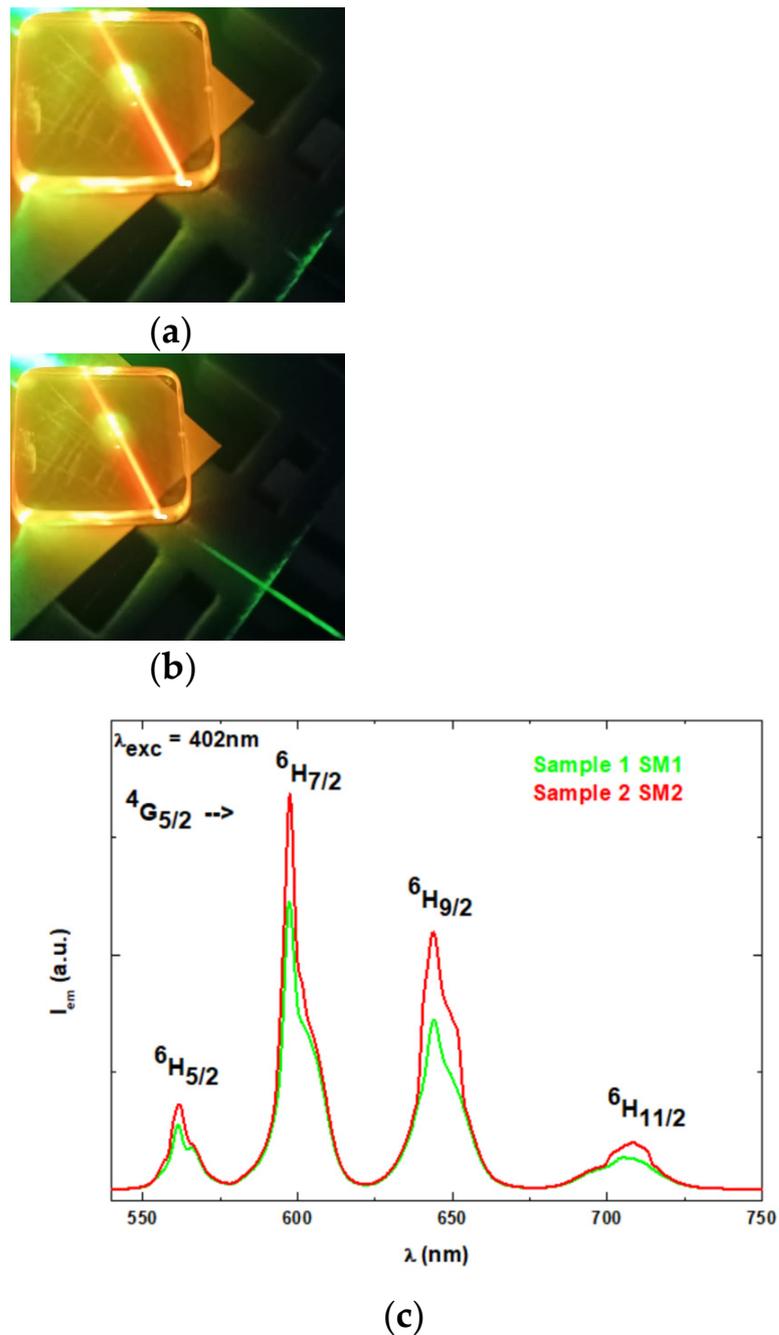
In the case of samples SM1 and SM2, one deals with high-concentration  $Sm^{3+}$  ions, leading to effective cross-relaxation (CR) between them [47]. The CR mechanism causes a decrease in the  $Sm^{3+}:{}^4G_{5/2}$  lifetime for SM1 to 3.134 ms when compared to the J-O radiative lifetime of 3.856 ms; for sample SM2, the lifetime decreases to  $\tau_{exp} = 3.758$  ms, compared to a radiative lifetime of 4.865 ms.

### 3.4. Emission Spectra

The photoluminescence (PL) emission spectra of  $Sm^{3+}$ -doped fluorophosphate glasses are shown in Figure 9c, with images of the glass emission presented in Figure 9a,b.

Figure 9c displays the photoluminescence spectra produced by the glasses stimulated at 402 nm. The emission bands seen at 561.5 nm, 597.5 nm, 643.5 nm, and 708 nm, respectively, are caused by the  ${}^4G_{5/2} \rightarrow {}^6H_{5/2}$ ,  ${}^4G_{5/2} \rightarrow {}^6H_{7/2}$ ,  ${}^4G_{5/2} \rightarrow {}^6H_{9/2}$ , and  ${}^4G_{5/2} \rightarrow {}^6H_{11/2}$  transitions of  $Sm^{3+}$ . The strength of the green and reddish-orange emission bands is significantly affected by the kind of glass host.  ${}^4G_{5/2} \rightarrow {}^6H_{7/2}$  and  ${}^4G_{5/2} \rightarrow {}^6H_{9/2}$  (red/orange) are the two distinct transition peaks of the  $Sm^{3+}$  ions in the visible emission spectrum. A decrease in the higher energy levels of  ${}^4G_{5/2}$  may occur when higher sub-levels of energy are near a  $Sm^{3+}$  ion in the ground state because the energy can be absorbed by the  $Sm^{3+}$  ion.

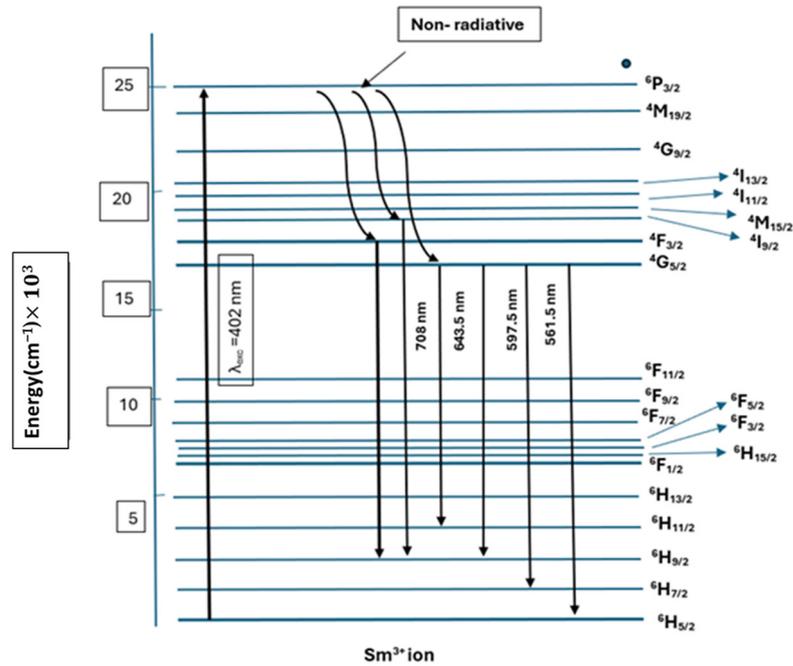
The glasses tested, like glasses in other research, show that the exceptional intensity of the  ${}^4G_{5/2} \rightarrow {}^6H_{7/2}$  transition makes them ideal for various applications, including color displays, high-density optical storage devices, and medical diagnostics. Our data closely align with those on other glasses described in the referenced studies [48–50].



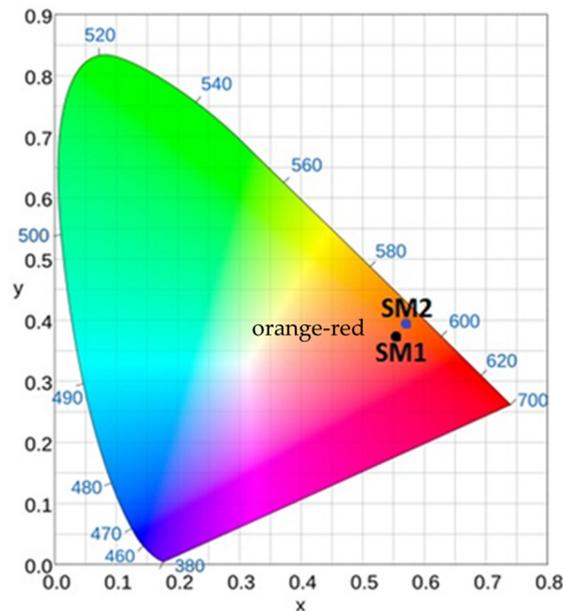
**Figure 9.** (a,b) Photos of glass emissions; (c) emission spectra of the glasses doped with  $\text{Sm}^{3+}$  recorded at room temperature upon 402 nm excitation.

Figure 10 shows a simplified diagram of the emission energy of  $\text{Sm}^{3+}$ . From SM1 and SM2, it can be seen that as the  $\text{Sm}^{3+}$  ions are pumped with a 402 nm excitation wavelength, they are excited to the  $6P_{3/2}$  level, and then some of the  $\text{Sm}^{3+}$  ions relax non-radioactively into lower levels of  $4G_{5/2}$  and  $4F_{3/2}$  and then decay into  $6H_{9/2}$ .

Figure 11 displays a CIE 1931 chromaticity diagram, which illustrates the effect of the emission transitions at a 402 nm excitation wavelength. For the manufactured glasses SM1 and SM2, the color coordinates (x, y) are (0.551, 0.352) and (0.581, 0.398), respectively. Thus, the glasses produced in this study have the potential to serve as materials for optical gain in orange/red laser applications.



**Figure 10.** Schematic energy level diagram of  $\text{Sm}^{3+}$  ion shows ground state absorption and excited state absorption mechanisms.



**Figure 11.** The CIE 1931 chromaticity diagram for the SM1 and SM2 glasses under an excitation wavelength of 402 nm.

#### 4. Conclusions

In this study, glasses with a base chemical composition of  $40\text{P}_2\text{O}_5\text{-}30\text{ZnO-}20\text{LiCl-}10\text{BaF}_2$  doped with  $\text{Sm}^{3+}$  were successfully prepared using a melt quenching technique. The glasses obtained fulfil the criteria to be considered superior materials for channel waveguide applications. They are compositionally and structurally homogeneous to prevent scattering and ensure consistent guiding properties and are free from significant defects and impurities that could introduce additional loss or alter the refractive index profile. From the DCS curve profiles, the glass transition temperature ( $T_g$ ), the onset of the glass crystallization peak ( $T_x$ ), the maximum glass crystallization peak ( $T_c$ ), glass melt-

ing temperature ( $T_m$ ), and thermal stability parameters were analyzed. The range of the vitreous state transition ( $T_{g\_endset} - T_{g\_onset}$ ) was observed to decrease, while the glass transition temperature shifted towards higher temperatures with an increase in the  $Sm^{3+}$  ion concentration. The values of the thermal stability parameters calculated ( $K_w$ ,  $K_H$ ,  $K_{LL}$ ) increased with a greater  $Sm^{3+}$  content, confirming the higher thermal stability of glass against devitrification on heating. Based on thermal studies, we confirmed very good glass stability. The glasses under investigation showed normal refractive index values for fluorophosphate glasses, according to ellipsometric measurements, and the Sellmeier model provided an excellent description of this index's dependency on wavelength. The Judd–Ofelt parameters calculated for the  $40P_2O_5-30ZnO-20LiCl-10BaF_2$  glasses are as follows: SM1 ( $\Omega_2 = 0.4327 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_4 = 1.1224 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_6 = 1.0071 \times 10^{-20} \text{ cm}^2$ ) and SM2 ( $\Omega_2 = 0.9897 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_4 = 1.9765 \times 10^{-20} \text{ cm}^2$ ,  $\Omega_6 = 1.4576 \times 10^{-20} \text{ cm}^2$ ). The large quantum efficiency of  $Sm^{3+}/SM1/{}^4G_{5/2}$  (96%) and SM2 (97%) was reported for all samples. The absorption cross-section data for the  ${}^4G_{5/2} - {}^6H_{7/2}$  transition were determined. The CIE chromaticity coordinate ( $x,y$ ) values corresponded to the orange-red region at the concentrations studied. The prominent transition peaks of the  $Sm^{3+}$  ions in the visible emission spectrum wavelength (red/orange) make our glasses useful as laser fiber sources that could be used in photodynamic therapy (PDT) treatment.

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