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ABSTRACT

In this work spectroscopic and laser characteristic of photo-thermo-refractive (PTR) glasses doped with different concentrations of rare-earth ions (ytterbium-erbium and Neodymium) were comprehensively carried out. Spectroscopic parameters were obtained using some theoretical techniques like Judd-Ofelt theory and Fuchtbauer-Landenburg (F-L) theory. Results show that the optimal concentration of neodymium oxide in PTR is 0.5 mol\%, at which the glass demonstrates the best spectral characteristics. It was found that PTR glass doped with 0.1 mol\% of erbium oxide and co-doped with 2 mol\% of ytterbium oxide also shows good spectral-luminescent properties. Laser action on those two samples was demonstrated. optical losses were found to be 0.34 % for neodymium- doped PTR active element and 0.28 % in the case of erbium and ytterbium co-doped active element. These values are quite low and compared to that obtained in commercial laser glasses. It was concluded that the rare-earth ions doped PTR glass is a promising material that can be a good candidate for producing the DFB lasers.

Keywords: Er-Yb glass laser, Nd glass laser, photo-thermo-refractive glass, diode-pumped laser

1. INTRODUCTION

Monolithic integration of optical devices is one of the challenges of modern photonics and optical information. One of the ways to achieve this miniaturization is producing Distributed-Feedback Lasers (DFB)\textsuperscript{1,2} and Lasers with Distributed Bragg Reflectors (DBR). These types of lasers are one of the most promising devices for narrow-band lasing, in which Bragg grating acts as a selective mirror with a narrow spectral bandwidth reflection, that can provide a narrow spectral emission output. The field of DFB lasers had a lot of developments since 1970s but creating DFB solid-state lasers has not been achieved yet due to the fact that this technology requires a development of a multifunctional optical material that combines a laser generation and the ability to recording a holographic Bragg grating.

Rare-earth ions-doped Photo-thermo-refractive (PTR) glass is one of such multifunctional optical materials. It provides an access to the development of DFB lasers and DBR. In this glass, it is possible to record holographic gratings\textsuperscript{3,4} due to the precipitation of sodium fluoride nano- and microcrystals during photo-thermo-induced crystallization and, at the same time, to obtain laser action on rare-earth ions.

Nowadays, neodymium and erbium lasers are broadly used in different areas such as medical application, LIDAR application, navigation for autonomous cars and optical telecommunications\textsuperscript{5}. 1.5 \textmu m- erbium lasers are a good choice for telecommunication application because the losses in the optical fibers are minimum at this wavelength. Besides, atmosphere demonstrates a high transparency at wavelengths about 1.5 \textmu m, which could serve as an advantage behind using such lasers. Nd-doped laser materials are also frequently used in multiple fields, especially in medical applications.

The main purpose of this work is to investigate the spectral-luminescent properties of PTR glass doped with rare-earth ions and (neodymium, ytterbium and erbium) and to study the laser performance of Nd-PTR and Er/Yb-PTR glasses.

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2. METHODS AND EXPERIMENTS

2.1 Glass synthesis

Fluoride PTR glasses were prepared in a system of ZnO-Na₂O-Al₂O₃-SiO₂-NaF-KBr and Ag₂O, CeO₂, Sb₂O₃ as photosensitive chemical components. Rare-earth oxides were added to the glass system by equimolar substitution of aluminum oxide. Three neodymium-doped PTR samples with a neodymium oxide concentration (0.5, 1, 2.11 mol%) and two ytterbium/erbium- samples – (1.0/0.1 mol% and 2.0/0.1 mol%) were studied. Glasses were synthesized in a high-temperature furnace (1480 °C) for 5 hours and annealed on a temperature of 480 °C. Table 1 shows the chemical composition of the studied samples.

Table 1. The batch composition of studied PTR glasses doped with rare-earth ions.

<table>
<thead>
<tr>
<th></th>
<th>1Nd</th>
<th>2Nd</th>
<th>3Nd</th>
<th>1Yb-01Er</th>
<th>2Yb-01Er</th>
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<tbody>
<tr>
<td>SiO₂</td>
<td>67.02</td>
<td>65.63</td>
<td>62.23</td>
<td>65.48</td>
<td>61.59</td>
</tr>
<tr>
<td>Na₂O</td>
<td>15.65</td>
<td>15.72</td>
<td>15.87</td>
<td>15.77</td>
<td>15.76</td>
</tr>
<tr>
<td>ZnO</td>
<td>4.80</td>
<td>4.70</td>
<td>4.42</td>
<td>4.67</td>
<td>4.39</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>2.41</td>
<td>1.93</td>
<td>1.09</td>
<td>1.76</td>
<td>1.96</td>
</tr>
<tr>
<td>Er₂O₃</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>Yb₂O₃</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1.00</td>
<td>2.00</td>
</tr>
<tr>
<td>Nd₂O₃</td>
<td>0.48</td>
<td>0.96</td>
<td>2.02</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>KBr</td>
<td>1.24</td>
<td>1.19</td>
<td>1.13</td>
<td>1.38</td>
<td>1.30</td>
</tr>
<tr>
<td>NaF</td>
<td>8.34</td>
<td>9.80</td>
<td>13.16</td>
<td>9.77</td>
<td>12.87</td>
</tr>
</tbody>
</table>

2.2 Spectral measurements

Absorption spectra were collected using Perkin-Elmer Lambda 900 spectrophotometer. For neodymium- doped PTR glasses the measurement was obtained within a spectral range of 250 – 900 nm. But for ytterbium/erbium- co-doped glasses measurements were performed within a range of 250–1700 nm. To study the fluorescence characteristics samples were pumped by a 980 nm diode laser (for ytterbium/erbium glasses) and a second harmonic of an Nd:YAG laser model Millennia-Xs (Spectra Physics Corp) for neodymium glasses. And, consequently, the luminescence was collected by a monochromator (model Acton-300 – Acton Research Corp.) and an IR-InGaAs detector and analyzed by a digital oscilloscope (Infinium HP54830 – Agilent Technologies, bandwidth 1.5 GHz). In order to obtain the kinetic measurements a pulsed laser (Solar Laser system LQ129) was used to excite the samples doped with ytterbium and erbium and a second harmonic of pulsed Nd:YAG laser for the neodymium- doped glasses.

2.3 Laser measurements

Experimental setup of the laser oscillator used in current work is shown in Figure 1. A resonator was built with a flat back-rear mirror that perfectly reflects the wavelengths around (1064 nm for Nd and 1530 nm for Er/Yb) and transmits the pump beam at a wavelength of (808 nm for Nd and 976 nm for Er/Yb) and a concave output coupler with a low transmitting coefficient at laser output wavelength. Diode lasers with a fiber output of 100 μm were used to excite the samples. The pump beam was collimated using a telescope system with a magnification of 2.5 and then focused on the back surface of the active medium which means that the spot diameter of the pump beam on the active element surface was 250 μm. The active medium was sandwiched by the copper plates for the cooling process and then placed in the resonator cavity at a 5 mm away from the rear mirror, the cavity length was experimentally optimized.
3. RESULTS AND DISCUSSION

3.1 Spectroscopic properties

3.1.1 Neodymium- doped PTR glasses

Absorption spectra of PTR glasses doped with different concentrations of neodymium ions in a range of 250–900 nm are shown in Figure 2. About 10 intensive absorption bands corresponding to transition from ground state of neodymium ions ($^4I_{9/2}$) to higher excited manifolds are clearly shown in the figure.

From the absorption data the electric dipole linestrengths for transitions illustrated in Figure 2 were calculated using the Judd-Ofelt technique\textsuperscript{6,7}, which depends on the minimization of the deviation between experimentally obtained linestrengths and theoretically calculated ones. These values were used to calculate the Judd-Ofelt intensity parameters $\Omega_{2,4,6}$, which can be utilized to estimate some spectroscopic parameters i.e. radiation lifetimes absorption and stimulated emission cross-sections and other laser parameters.
Figure 3 shows the emission spectrum of Nd-doped PTR glasses from the excited state of Nd$^{3+}$ to the lower manifolds. Emission spectrum consists of three wide bands centered at wavelengths of 885, 1064, 1332 nm related to spontaneous decay from the metastable state of neodymium ions $^4F_{3/2}$ to lower states $^4I_{9/2}$, $^4I_{11/2}$, $^4I_{13/2}$, respectively. From Figure 3 one can see that the transition $^4F_{3/2} \rightarrow ^4I_{11/2}$ is demonstrated that consists with branching ratios obtained using Judd-Ofelt relations and shown in Table 2.

![Figure 3](https://example.com/figure3.png)

**Figure 3.** Fluorescence spectra of PTR glasses doped with Nd$^{3+}$.

Table 2 shows the spectroscopic parameters of the PTR glasses doped with neodymium ions. Judd-Ofelt intensity parameters were used to calculate radiative lifetime and absorption cross-section. Stimulated emission cross-sections ($\sigma$, cm$^2$) were obtained by F-L method$^8$. From the calculation results one can notice that the sample doped with 0.5 mol% of neodymium oxide has the highest value of quantum efficiency and a very low saturation intensity. Therefore, the laser properties were studied on the sample with 0.5 mol% of Nd$_2$O$_3$.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\tau_{\text{rad}}$ ($^4F_{3/2}$), $\mu$s</th>
<th>$\tau_f$ ($^4F_{3/2}$), $\mu$s</th>
<th>$\eta=\tau_f/\tau_{\text{rad}}$</th>
<th>$\sigma$ ($^4F_{3/2}$→$^4I_{11/2}$), $10^{-20}$ cm$^2$</th>
<th>$I_{\text{sat}}$, kW/cm$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1Nd</td>
<td>576</td>
<td>520</td>
<td>90.3</td>
<td>1.743</td>
<td>20.62</td>
</tr>
<tr>
<td>2Nd</td>
<td>532</td>
<td>290</td>
<td>54.5</td>
<td>1.909</td>
<td>33.76</td>
</tr>
<tr>
<td>3Nd</td>
<td>481</td>
<td>126</td>
<td>26.2</td>
<td>2.041</td>
<td>72.66</td>
</tr>
</tbody>
</table>

### 3.1.2 Erbium and Ytterbium co-doped PTR glasses

To study the spectroscopic characteristics of erbium and ytterbium co-doped PTR glass the same strategies used with Nd-doped PTR glasses were curried out. Adsorption spectra were obtained in the range of 350–1600 nm. About nine bands corresponding to transition from the ground state of erbium ions ($^4I_{15/2}$) to higher excited manifolds. The very intense absorption band at 975 nm is related to ytterbium ions promotion from their ground state ($^2F_{7/2}$) to the only excited state of Yb$^{3+}$ ($^2F_{5/2}$), which overlaps with the transition of erbium ions ($^4I_{15/2} \rightarrow ^4I_{11/2}$). This overlap leads to a...
resonant energy transfer from Yb$^{3+}$ to Er$^{3+}$, so, the 980 nm pump leads to an indirect but efficient pump of erbium ions according to the equation:

\[ ^2F_{5/2} [\text{Yb}^{3+}] + ^4I_{15/2} [\text{Er}^{3+}] \rightarrow ^2F_{7/2} [\text{Yb}^{3+}] + ^4I_{11/2} [\text{Er}^{3+}] \]  

(1)

From Figure 4 the electric dipole linestrengths were calculated and used to obtain the Judd-Ofelt intensity parameters by root-square method. The experimentally obtained and radiative lifetimes were calculated for the metastable emitting level of erbium ions ($^4I_{15/2}$). Kinetic studies of both co-doped and single Yb$^{3+}$-doped glasses in order to estimate the efficiency of energy transfer from ytterbium to erbium. Emission spectra of studied samples collected in the range of 1450–1650 nm are shown in Figure 5. In Table 3 some obtained spectroscopic parameters and energy transfer efficiencies $TE$ are shown.

![Absorption spectra of Er and Yb co-doped photo-thermo-refractive glasses.](image)

Figure 4. Absorption spectra of Er and Yb co-doped photo-thermo-refractive glasses.

![Fluorescence spectra of Er and Yb co-doped PTR glasses.](image)

Figure 5. Fluorescence spectra of Er and Yb co-doped PTR glasses.
According to the results obtained above, the sample 2Yb-01Er, which demonstrates the best spectral and luminescent characteristics and quantum efficiency, was chosen to study laser and properties.

### 3.2 Laser properties

Figure 6 shows the laser input-output characteristics of the 0.5 mol% Nd-PTR (a) and the 2Yb-01Er PTR (b) active elements for two output couplers with 0.5% and 1% transmittance (red and black lines, respectively). The collected data were very well fitted to linear functions. Slope efficiencies (energy conversion efficiency) of 13.4%, 16.8% for neodymium and 1.4%, 2.3% for erbium/Ytterbium were achieved by use of the 0.5%, 1% output couplers, respectively.

![Figure 6. Laser performance of Nd-PTR (a) and Er/Yb-PTR (b) with output coupling of 0.5% and 1% transmittance](image)

Round-trip losses ($L$) of the studied active elements were found by Caird plot method using Caird plot relation:

$$\frac{1}{\eta_i} = \frac{1}{\eta_0} \left( 1 - \frac{L}{T_i} \right) \quad (2)$$
where $\eta_i$ is the achieved slope efficiency by use of $T_i$ output coupling mirror ($T_i = 0.5\%$ or $1\%$). Fitting results have shown that the round-trip losses of Nd-doped element are $0.34\%$, this value is quite low, and is comparable to that in a commercially fabricated Nd-YAG crystalline gain medium, which is available in the same length and demonstrates laser action in the same experimental conditions. On the other hand, the round-trip losses in the Er and Yb co-doped element are $0.28\%$, which is a quite low value and comparable to that obtained in a commercially fabricated Yb/Erbium-doped silicate glass gain medium, which is available in the same length and demonstrates laser action in the same experimental conditions. That result may be attributed to a good quality of the glass surface preparation of the active element.

Further studies include the UV irradiation process and its effect on lasing action. During the experiment it was shown that initial steps of the UV irradiation and subsequent heat treatment (photo-thermo induced crystallization process) do not affect the lasing properties of the materials at all. Precipitation of the silver clusters increases the optical losses in the visible region up to 700 nm, which significantly increases threshold of the laser operation. However, thoroughly studies of the treated glasses spectra in the pump and lasing regions do not show either detectable changes in scattering or additional absorption band appearance. Further heat treatment leading to the formation of silver nanoparticles (NP) greatly increases threshold of the laser and may make it impossible to achieve laser action in CW mode. Photodestruction of the silver NP does not decrease the threshold, that make us confident that increased losses are connected neither with the direct absorption of the silver NP nor with molecular silver clusters.

**CONCLUSION**

Photo-thermo-refractive (PTR) glasses doped with rare-earth ions (ytterbium-erbium and Neodymium) were comprehensively studied in this work. Both spectroscopic and laser properties were investigated in order to estimate the optimal concentration of the dopant. Some spectroscopic parameters were obtained using some theoretical approaches like Judd-Ofelt theory and Fuchtbauer-Landenburg (F-L) theory. Emission cross-sections, radiative and experimental lifetimes, quantum efficiencies and energy transfer efficiencies were calculated depending on absorption and fluorescence spectra. Results show that the optimal concentration of neodymium oxide in PTR is $0.5\%$, and $0.1\%$-$2\%$ for erbium/ytterbium co-doped PTR glass. Laser action on those two samples was demonstrate. Caird plot method was applied to calculate the optical losses, which were found to be $0.34\%$ for neodymium- doped PTR active element and $0.28\%$ in the case of erbium and ytterbium co-doping. These values are quite low and compared to that obtained in commercial laser glasses, which demonstrated a laser action in the same experimental conditions. It was concluded that the rare-earth ions doped PTR glass is a promising material that can be a good candidate for producing the DFB lasers.

**REFERENCES**


