SPECTROSCOPY PROPERTIES IN Dy^{3+} ION DOPED ZINC BARIUM TELLURITE OXYFLUORIDE GLASSES FOR WHITE LED APPLICATIONS

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Received: July 11, 2019; Revised: December 20, 2019; Accepted: January 03, 2020

Abstract

Dysprosium (Dy^{3+}) ion doped tellurite glasses were prepared by the normal melt quenching technique with the compositions (55-x)TeO_2-10ZnF_2-35BaO-xDy_2O_3 (where x = 0.00, 0.05, 0.10, 0.50 1.00, and 1.50 mol%) to study the physical and optical properties. All the absorption bands have been occurred from the ground state $^6H_{15/2}$ to various excited states $^6F_{3/2}$, $^6F_{5/2}$, $^6F_{7/2}$, $^6F_{9/2}$, $^6F_{11/2}$, and $^6H_{11/2}$ at wavelengths 750, 804, 901, 1,095, 1,277, and 1,681 nm respectively. The J-O intensity parameters, assess from the absorption spectra and emission spectra to lead to the calculation of values transition probability (A_R), branching and ratio (β_R) for the prominent emission transitions of Dy^{3+} ion in tellurite glasses. The highest intensity of the emission spectra of Dy^{3+} ion doped tellurite glasses is at 575 nm that corresponding to the $^4F_{9/2}\rightarrow{}^6H_{13/2}$ transitions and also indicate that the intense white region appear when excited at 452 nm. Luminescence results represent that these glasses can be further modified for white LED and display device. The lifetime values, corresponding to the $^4F_{9/2}$ excited state, inversely vary with the increase of Dy^{3+} ion concentration. These obtained results were discussed and reported in the present works.

Keywords: Tellurite glasses, Dy^{3+}, optical properties, Luminescence

Introduction

White light emitting diodes (W-LED) are more interesting because of the potential application in lasers., back light and general illumination (Nakamura and Fasol, 1997). Therewith, they have high luminous efficiency, long operation lifetime, reliability, safety and environmental friendly characteristics (Lin *et al.*, 2008). Normally, the W-LEDs are fabricated by the combination of blue light emitting diode (LED) chip with fluorescence phosphors (Park *et al.*, 2003). However, the individual degradation of the blue LED chip and the phosphors coated on it could cause chromatic aberration and poor white luminescence. An alternative is essential to develop novel materials that could emit bright white light by ultraviolet (UV) excitation (Liang *et al.*, 2008; Chang and Chen, 2007).

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Tellurite glasses are the alternative glass with the properties such low energy of phonon, high refractive index, low melting temperature, higher rare earth ion solubility, good chemical stability (Wang *et al.*, 1994; El-Mallawany, 2002), good optical transmission at infrared (Sidkey *et al.*, 1997; Saddeek, 2005) and visible wavelengths that due to the good qualities of TeO_2. Some of the alkaline earth oxides (BaO), when added to glasses, will result in the creation of non-bridging oxygens at the expense of bridging oxygens and the mentioned alkaline earth cations are also solely acting as modifiers (Brow, 2000). The addition of modifiers such as ZnF_2 to the tellurite glass or the substitution of fluorine ions for oxide ions is expected to increase the glass forming range, glass stability, lower viscosity and improve the transparency substantially (Nazabal *et al.*, 2003; El-Desoky and Abo-Naf, 2004).

Luminescence properties of rare earth ions have drawn abundant interest among the researchers. Rare earth doped glasses have potential applications due to their emission efficiencies of 4f-4f and 4f-5d electronic transitions in the RE ion (Liang *et al.*, 2007; Azizan *et al.*, 2014; Ramteke and Gedam, 2015; Pawar *et al.*, 2016). Rare earth doped glasses are potential candidate for laser hosts, waveguide, optical fibers, solar concentrators, plasma display panel, optical amplifiers, semiconductor light-emitting diodes, and optical detectors (Selvi *et al.*, 2014). Among RE ions, the Dy^{3+} ion is a most interesting ion to studied for luminescence properties, color display phosphors and white light emissions since the Dy^{3+} ion typically emitting two intense emissions in the (~576 nm) and (~482 nm) regions that are attributed to the $^4F_{9/2} \rightarrow {}^6H_{13/2}$ and $^4F_{9/2} \rightarrow {}^6H_{15/2}$ transitions, respectively (Merino *et al.*, 1997; Mahato *et al.*, 2005; Boruc *et al.*, 2012; Rao *et al.*, 2013; Vijaya *et al.*, 2013; Ci *et al.*, 2014; Pisarski *et al.*, 2014; Caldiño *et al.*, 2015; Devi *et al.*, 2016; Ramteke *et al.*, 2016; Zhou *et al.*, 2016a; 2016b; Kaewnuam *et al.*, 2017; Srihari and Jayasankar, 2017). So the white light emission Dy^{3+} doped in tellurite glasses have been applications in lasers medium and white LED applications (Tingqiao *et al.*, 2016).

The objective in this work of the tellurite glasses doped with Dy^{3+} ion for studies on the characterization of physical properties, optical properties, and luminescence. The applicability of these glasses are used for display devices and white LEDs applications.

Experimental

Glasses samples have been prepared from the composition $(55\text{-}x)TeO_2\text{-}10ZnF_2\text{-}35BaO\text{-}xDy_2O_3$ (where x = 0.00, 0.05, 0.10, 0.50, 1.00, and 1.50 mol%). Used chemicals of 99.99% like tellurium dioxide (TeO_2), zinc fluoride (ZnF_2), barium carbonate ($BaCO_3$), and dysprosium oxides (Dy_2O_3), with high purity, were mixed in mortar and pestle thoroughly in a porcelain crucible for 15 g total weight. Melting by electrical furnace at 950°C, for 1 h. After complete melting, after that poured into a graphite mold and annealed at 350°C for 3 h and cooled down to room temperature and then to glasses samples were cut and polished to a measure of (Width × Depth × High) 1.0 cm × 0.3 cm × 1.5 cm.

The Archimedes method is used to evaluate density by the densitometer HR-200 weighing balance. The refractive index of these samples were measured at room temperature by using Reflectivity Meter (PRM). The excitation spectra (wavelength of 300-500 nm) and emission spectra (wavelength of 400-800 nm) of Dy^{3+} ion doped tellurite glasses were registered Cary Eclipse Fluorescence Spectrophotometer. The optical absorption spectra of glasses samples were recorded on the UV-Vis-NIR regions in the range of 500-2,000 nm using UV-3600 Shimadzu UV-VIS-NIR spectrophotometer. The decay curve were investigated by using a Cary Eclipse Fluorescence Spectrophotometer. The J-O analysis was used to investigate the glass potential for using as a laser medium. Several parameters such as oscillator strength (*f*), J-O parameter (Ω_λ) where λ = 2, 4, 6, radiative transition possibility (A_R), stimulated emission cross section (σ) and branching ratio (β_R) were calculated (Kaewnuam *et al.*, 2017).

Table 1. Physical properties of the Dy^{3+} dope tellurite glasses

Physical properties	Dy 0.00 mol%	Dy 0.05 mol%	Dy 0.10 mol%	Dy 0.50 mol%	Dy 1.00 mol%	Dy 1.50 mol%
Density (g/cm^3)	5.2791±0.0029	5.2572±0.0181	5.2579±0.002	5.2583±0.0033	5.2869±0.0056	5.2700±0.0330
Molar volume (cm^{3}mol)	28.7515	30.0460	30.0595	30.2010	30.2166	30.4928
Refractive index	1.609	1.833	1.852	1.865	1.871	1.876

Results and Discussion

Physical Properties

Glass samples are shoe in Figure 1. The color of glasses are yellow, because of Te component. Table 1. Shows the physical properties of the Dy^{3+} ion doped tellurite glasses. The value of densities are in the range 5.2572±0.0181-5.2869±0.0056 g/cm^3. The densities of glasses are not depended on Dy_2O_3 concentration. The molar volume and refractive index increased with increasing of Dy_2O_3 concentration that results when the amount of concentration Dy_2O_3 increased the glasses structure also expanded, resulting in non-bridging oxygen has been increased. The value of molar volume are in the range 28.7515-30.4928 cm^3/mol. The value of refractive index are in the range 1.609-1.876.

Optical Properties

The absorption spectra of Dy^{3+} ion doped tellurite glass is shown in Figure 2 in the wavelength range 500-2,000 nm. The spectrum has six absorption peaks which observed in NIR region. All the absorption bands have been occurred from the ground state $^6H_{15/2}$ to various excited states $^6F_{3/2}$, $^6F_{5/2}$, $^6F_{7/2}$, $^6F_{9/2}$, $^6F_{11/2}$, and $^6H_{11/2}$ at wavelengths 750, 804, 901, 1,095, 1,277, and 1,681 nm respectively. The transition of Dy^{3+} ion is $^6H_{15/2}\rightarrow{}^6F_{11/2}$ centered at 1,277 nm and found to be high intense more than the other transitions.

Oscillator Strengths and Judd–Ofelt Intensity Parameters

The radiative transitions associated within the $^4f_{9/2}$ configuration of the Dy^{3+} ion. These have been analysed using the Judd-Ofelt (JO) theory (Judd, 1962; Ofelt, 1962). According to JO theory that calculated oscillator strengths (f_{cal}), induced electric-dipole transitions from an initial state (ΨJ) to the final state ($\Psi' J'$) depends on three parameters Ω_λ (λ=2, 4 and 6) as

$$f_{cal} = \left[\frac{8\pi^2 mcv}{3h(2J+1)}\right]\left[\frac{(n^2+2)^2}{9n}\right]\times\sum_{\lambda=2,4,6}\Omega_\lambda(\psi J\|U^\lambda\|\psi' J')^2 \quad (1)$$

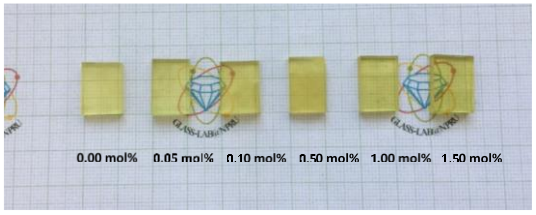

Figure 1. Glasses samples of the Dy^{3+} ion doped tellurite glasses (Not mentioned in the article)

where m is the electron mass, λ is the mean wavelength of the observed transition, c is the light velocity, *n* is the refractive index, h is Planck's constant and the doubly reduced square matrix elements of the unit tensor operator which are evaluated from the intermediate coupling approximation for a transition from to (Jayasimhadri *et al.*, 2006). The Lorentz local field correction factor which indicates that the ions are calculated in a dielectric medium but not in a vacuum (Carnall *et al.*, 1978). Integrating each absorption bands of the Dy^{3+} ion doped tellurite glasses use to calculate the experimental oscillator strength of the $^4F_{9/2}$ level to the various exciting levels follow as

$$f_{\exp} = 4.318\times10^{-9}\int\varepsilon(\nu)d\nu \quad (2)$$

The branching ratio (β_R) can be obtained from the equation given below

$$\beta_R(\psi J,\psi' J) = \frac{A(\psi J,\psi' J)}{A_T\psi J} \quad (3)$$

The relative values of the branching ratios can be obtained from the areas under the emission curves. The peak stimulated emission cross-section (σ_P^E) can be calculated using the expression (Maheshvaran *et al.*, 2011).

$$\sigma_P^E = \frac{\lambda_P^4 A}{8\pi cn^2\Delta\lambda_{eff}} \quad (4)$$

Where λ_p is the emission transition peak wavelength and $\Delta\lambda_{eff}$ is the effective line width of the transition, given by

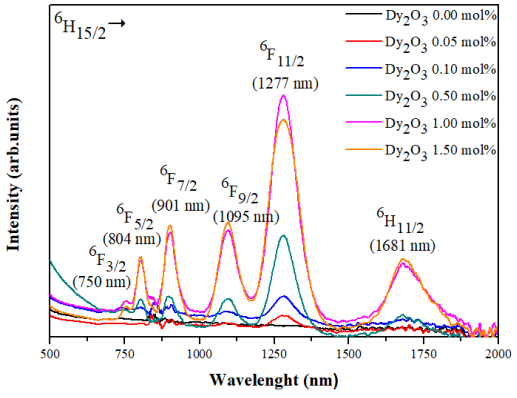

Figure 2. The absorption spectra of the Dy^{3+} doped tellurite glasses

$$\Delta\lambda_{eff} = \frac{1}{I_{max}}\int I(\lambda)d\lambda \quad (5)$$

where I is the fluorescence intensity and I_{max} is the intensity at band maximum.

The f_{exp} and f_{cal} parameters together with σ_{rms} including and excluding hypersensitive transition ($^6H_{15/2}\rightarrow{}^6F_{11/2}$) are displayed in Table 2. For 0.50 mol% is highest luminescence. The niceness of fit between f_{exp} and f_{cal} as well as the accuracy of J-O intensity parameters Ω_λ (λ=2, 4, 6) are established by considerably small σ_{rms} values, when hypersensitive transition is excluded. For the titled glasses the J-O intensity parameters are following the same trend ($\Omega_2 > \Omega_6 > \Omega_4$). Table 3. represents the evaluated J-O intensity parameters for the as prepared glasses under investigation along with comparison of J-O intensity parameters in various other glass matrices (Kesavulu and Jayasankar, 2011; Swapna *et al.*, 2013; Babu and Jayasankar; 2000). The magnitudes of Ω_λ (λ=2, 4, 6) suggest that the observed trend of the intensity parameters is due to the ligand environment around the RE ion.

The Ω_2 parameter are covalent nature of metal-ligand bond and asymmetry of ion sites in the neighbourhood of RE ion. The structure dependent parameters Ω_4 and Ω_6 are related to the rigidity and viscosity of the medium in which ionsare situated (Jørgensen and Reisfeld, 1983).

Table 2. Assignment of absorption bands, experimental f_{exp}(10^{-6}), calculated oscillator strengths f_{cal}(10^{-6}) of 0.5 mol% Dy^{3+} ion in tellurite glasses

Transition $^6H_{15/2}\rightarrow$	λ_p (nm)	Dy_2O_3 0.5 mol%	
		f_{exp}	f_{cal}
$^6F_{3/2}$	750	0.690	1.304
$^6F_{5/2}$	804	5.250	5.171
$^6F_{7/2}$	901	2.190	2.328
$^6F_{9/2}$	1095	2.550	2.222
$^6F_{11/2}$	1277	2.610	1.104
$^6H_{11/2}$	1681	2.840	0.209

Luminescence Properties

Figure 3 shown the excitation spectra of tellurite glasses doped with Dy^{3+} ion have been recorded by monitoring the emission at 575 nm. The excitation spectra seven prominent bands were observed at 326, 351, 365, 388, 427, 452, and 473 nm in all the glasses corresponding to levels excited from ground state $^6H_{15/2}$ to excited states $^6P_{3/2}$, $^6P_{7/2}$, $^6P_{5/2}$ $^4F_{7/2}$, $^4G_{11/2}$, $^4I_{15/2}$, and $^4F_{9/2}$ transitions, serially. The excitation observed at 452 nm is most intense and choose as excitation wavelength for measuring the emission spectra.

The emission spectra of tellurite glasses are in the range of 460-800 nm by exciting at 452 nm which is present in Figure 4 the emission spectra it is clear that emission intensities increases with the increase of Dy_2O_3 concentration up to 0.50 mol% and then decreases with increasing of Dy_2O_3 concentration. This behavior directly related to the concentration quenching due to an increase number of Dy^{3+} ions in the glass matrix. Also as an inset is a photograph of the light output from the glass while excited at 452 nm. The corresponding to the transition of emission peak, $^4F_{9/2} \rightarrow {}^6H_{13/2}$ at 575 nm has the most intensity than the peaks corresponding to the other transitions. The spectra show four significant peaks. The observed emission peaks are centered at 482 nm, 575 nm, 663 nm, and 752 nm.

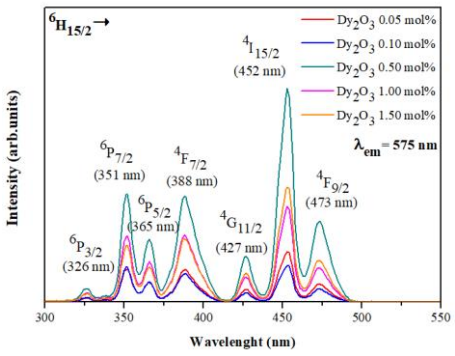

Figure 3. The excitation spectra of the Dy^{3+} ion doped tellurite glasses

Table 3. J-O intensity parameters of the Dy^{3+} ion doped tellurite glasses and other reported Dy^{3+} ion doped glasses

Glass composition	J–O intensity parameters ($\times10^{-20}$ cm^2)			Tends	References
	Ω_2	Ω_4	Ω_6		
0.5 mol% Dy_2O_3	4.45	0.99	2.12	$\Omega_2>\Omega_6>\Omega_4$	Present work
PYBDy10	7.75	2.31	2.70	$\Omega_2>\Omega_6>\Omega_4$	Davis and Mott (1970)
D0LBE	5.319	0.54	1.54	$\Omega_2>\Omega_6>\Omega_4$	Agarwal *et al.* (2003)
DZTFB	5.28	2.54	1.97	$\Omega_2>\Omega_4>\Omega_6$	Suthanthirakumar and Marimuthu (2016)
PKMAFDy	7.04	1.73	1.57	$\Omega_2>\Omega_4>\Omega_6$	Basavapoornima *et al.* (2009)
LiTeDy	1.46	2.32	3.60	$\Omega_6>\Omega_4>\Omega_2$	Rai *et al.* (2006)
LiLaBDy	3.00	6.00	3.44	$\Omega_4>\Omega_6>\Omega_2$	Wantana *et al.* (2016)

These peaks are correspond to $^4F_{9/2} \rightarrow {}^6H_{15/2}$, $^4F_{9/2} \rightarrow {}^6H_{13/2}$, $^4F_{9/2} \rightarrow {}^6H_{13/2}$, and $^4F_{9/2} \rightarrow {}^6H_{13/2}$.

Lifetime

The lifetime of the $^4F_{9/2}$ energy level for the different mol% of Dy_2O_3 was measured and the experimental decay curves are shown in Figure 5. The lifetime curves obtained exciting the sample with 452 nm wavelength and by monitoring the emission at 575 nm. The lifetime decreases with increasing of Dy_2O_3 concentration. This may be due to the energy transfer between excited ions and ground state ion via cross relaxation (Suhasini *et al.*, 2009). The lifetime is the decreases level time from $^4F_{9/2}$ decreasing level as a result of energy transfer process from Te back to Dy^{3+}ion. Even more Dy^{3+} ion increase the energy transfer rate affect to decrease time of luminescence.

CIE Chromaticity Coordinates

In order to identify the dominant emission color of the present Dy^{3+} ion doped tellurite glasses, emission spectra were characterized through CIE (Commission International d'Eclairage) 1931 color chromaticity diagram. The CIE 1931 diagram is the universally accepted system to represent the composition of any color by means of three primary colors. To describe the color produced by any light source, three color matching functions such as $\bar{x}(\lambda)$, $\bar{y}(\lambda)$, $\bar{z}(\lambda)$ are essential and the degree of simulation essential to match the color of the given spectral power density $P(\lambda)$ (Krishnaiah *et al.*, 2013; Shanmugavelu and Kumar, 2014). The color coordinates of the tellurite glasses doped with Dy^{3+} at 0.50 mol% are x = 0.39 and y = 0.43 (as show in Figure 6) which is very near to the white light emission. The 0.50 mol% Dy^{3+} ion doped glass has exhibited excellent color tunability of white light emission.

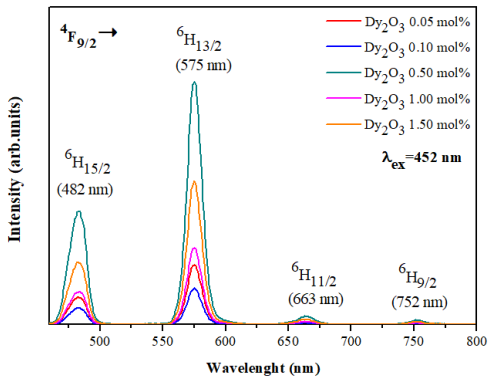

Figure 4. The emission spectra of the Dy^{3+} ion doped tellurite glasses

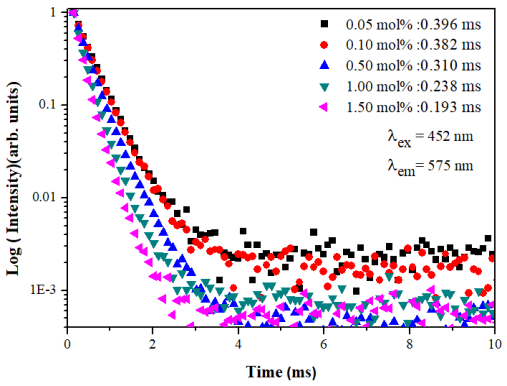

Figure 5. The decay time of Dy^{3+} doped the tellurite glasses

Conclusions

The dysprosium ion doped tellurite glasses to studied physical and optical properties. The molar volume and refractive index were increased with increasing of Dy_2O_3 concentration. The absorption spectra have six peaks which observed in NIR region. The observed trend in the variation of the J-O parameter was $\Omega_2 > \Omega_6 > \Omega_4$. The strongest white emission at 575 nm and emission peak corresponding to the transition, $^4F_{9/2} \rightarrow {}^6H_{13/2}$. The excitation spectra of Dy^{3+} ion doped tellurite glasses sample have seven peaks the highest peaks at 452 nm. We can excited the tellurite glasses by 452 nm for the best luminescence application. The lifetime values corresponding to the $^4F_{9/2}$ excited state is found to decrease with the increase in Dy^{3+} ion concentration. The CIE 1931 chromaticity coordinate (0.39, 0.43) it can be observed that the

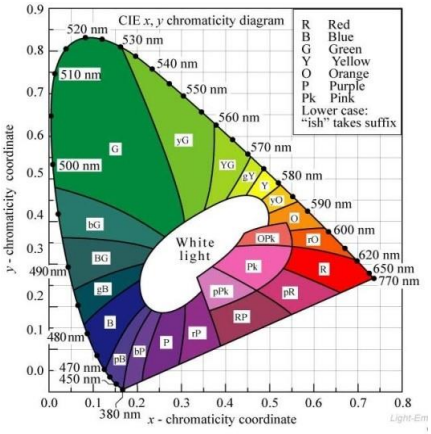

Figure 6. CIE chromaticity diagram of Dy^{3+} in tellurite glasses (Not mentioned in the article.)

white region. Dy^{3+} ion doped tellurite glasses are suitable for application in a White LED.

Acknowledgements

The authors would like to thank the Center of Excellence in Glass Technology and Materials Science (CEGM), Faculty of Science and Technology, Nakhon Pathom Rajabhat University for instrument and facilities. Thanks are also due to Research and Development Institute, NPRU for facilities.

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