

STRUCTURAL, LUMINESCENCE PROPERTIES AND JUDD-OFELT  
ANALYSIS OF RARE-EARTH DOPED CALCIUM-SULFOBOROPHOSPHATE  
AND BARIUM-SULFOBOROPHOSPHATE GLASSES

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## **DEDICATION**

This thesis is dedicated to my beloved

Father, Alhaji Abdullahi Yamusa, Mother, Hafsat Abdullahi, Wife, Rahma Sani Yamusa, Children, Aisha Yamusa Abdullahi and Hafsat Yamusa Abdullahi for their support and encouragement.

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## ABSTRACT

Glass samples of undoped calcium-sulfoborophosphate and barium-sulfoborophosphate with chemical composition of  $x\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$  and  $x\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$  with  $15 \leq x \leq 35$  mol% were prepared using melt quenching method. A series of glass samples doped with rare earth (RE =  $\text{Dy}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$  and  $\text{Eu}_2\text{O}_3$ ) with the chemical compositions of  $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{RE}$  and  $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{RE}$  with  $0.1 \leq y \leq 1.0$  mol% were also prepared by melt quenching method. The amorphous phase of the glass samples were characterized by X-Ray diffraction (XRD) method, while the structural features of the samples were measured using Fourier transform infrared (FTIR) spectroscopy and Raman spectroscopy. The optical properties of glass samples were characterized by ultraviolet-visible-near infrared (UV-Vis-NIR) spectroscopy and luminescence spectroscopy. The amorphous phase of the glass samples was confirmed by the diffuse broad XRD pattern. The infrared spectral measurements revealed the presence of vibrational groups of P-O linkage,  $\text{BO}_3$ ,  $\text{BO}_4$ , P-O-P, O-P-O, S-O-B (sulfoborate network) groups and the bending B-O-B units in sulfoborophosphate structural network of glass samples. The Raman spectra also revealed the coexistence of structural units of  $\text{BO}_4$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , and P-O-P in sulfoborophosphate glass samples. The luminescence spectra of  $\text{Dy}^{3+}$  ions doped glass samples exhibit four emission bands at around 482 nm, 572 nm, 662 nm and 685 nm, which correspond to the  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{15/2}$ ,  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$ ,  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{11/2}$  and  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{9/2}$  transitions, respectively. The emission spectra of glass samples doped with  $\text{Sm}^{3+}$  ions show dominant peaks at around 559 nm, 596 nm, 642 nm and 709 nm which correspond to the transitions of  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{5/2}$ ,  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{7/2}$ ,  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{9/2}$  and  $^4\text{G}_{5/2} \rightarrow ^6\text{H}_{11/2}$ , respectively. Meanwhile, glass samples doped with  $\text{Eu}^{3+}$  ions show emission spectra peaks around 589 nm, 611 nm, 651 nm and 701 nm which correspond to the transitions of  $^5\text{D}_0 \rightarrow ^7\text{F}_1$ ,  $^5\text{D}_0 \rightarrow ^7\text{F}_2$ ,  $^5\text{D}_0 \rightarrow ^7\text{F}_3$  and  $^5\text{D}_0 \rightarrow ^7\text{F}_4$ , respectively. Absorption and emission spectra are used to evaluate the Judd-Ofelt intensity parameters and radiative transition probabilities, branching ratios and stimulated emission cross-sections of the three rare-earth ions ( $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ , and  $\text{Eu}^{3+}$ ) doped glass system. Based on this study, it can be concluded that the structural network features of calcium sulfoborophosphate and barium sulfoborophosphate glasses are similar, despite of different modifier. The incorporation of sulphate and rare-earth ions into the glass network show enhancement of chemical and physical stability, in addition to improving optical properties performance of the prepared glasses such as having high value of branching ratio, stimulated cross-section, gain bandwidth and optical gain. In view of this, calcium sulfoborophosphate and barium sulfoborophosphate glasses could be suggested as promising luminescent host material for solid-state lighting device application.

## ABSTRAK

Sampel kaca tanpa dop kalsium sulfoborofosfat dan barium sulfoborofosfat dengan komposisi kimia  $x\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$  dan  $x\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$  dengan  $15 \leq x \leq 35$  mol% telah disediakan melalui kaedah lindap-kejut leburan. Satu siri sampel kaca didop dengan nadir bumi (RE =  $\text{Dy}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$  dan  $\text{Eu}_2\text{O}_3$ ) dengan komposisi kimia  $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{RE}$  dan  $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{RE}$  dengan  $0.1 \leq y \leq 1.0$  mol% telah juga disediakan melalui kaedah lindap-kejut leburan. Fasa amorfus sampel kaca telah dicirikan melalui kaedah pembelauan sinar-X (XRD), sementara ciri-ciri struktur sampel telah diukur menggunakan spektroskopi transformasi Fourier inframerah (FTIR) dan spektroskopi Raman. Sifat optik sampel kaca dicirikan melalui spektroskopi ultraungu-nampak-inframerah dekat (UV-Vis-NIR) dan spektroskopi luminesens. Fasa amorfus sampel kaca telah disahkan oleh corak XRD memburu yang lebar. Pengukuran spektrum inframerah menunjukkan kewujudan kumpulan getaran P-O,  $\text{BO}_3$ ,  $\text{BO}_4$ , P-O-P, O-P-O, S-O-B (rangkaian sulfoborat) dan unit pembengkokan B-O-B dalam rangkaian struktur sampel kaca sulfoborofosfat. Spektrum Raman juga menunjukkan kewujudan unit struktur  $\text{BO}_4$ ,  $\text{SO}_4^{2-}$ ,  $\text{PO}_4^{3-}$ , dan P-O-P dalam sampel kaca sulfoborofosfat. Spektrum luminesens sampel kaca didop dengan ion  $\text{Dy}^{3+}$  mempamerkan empat jalur pancaran pada sekitar 482 nm, 572 nm, 662 nm dan 685 nm, yang masing-masing berpadanan dengan peralihan  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{15/2}$ ,  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{13/2}$ ,  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{11/2}$  dan  ${}^4\text{F}_{9/2} \rightarrow {}^6\text{H}_{9/2}$ . Spektrum pancaran bagi sampel kaca didop dengan ion  $\text{Sm}^{3+}$  menunjukkan puncak dominan pada sekitar 559 nm, 596 nm, 642 nm dan 709 nm yang masing-masing berpadanan dengan peralihan  ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{5/2}$ ,  ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{7/2}$ ,  ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{9/2}$  dan  ${}^4\text{G}_{5/2} \rightarrow {}^6\text{H}_{11/2}$ . Sementara itu, sampel kaca didop dengan ion  $\text{Eu}^{3+}$ , menunjukkan puncak spektrum pancaran sekitar 589 nm, 611 nm, 651 nm dan 701 nm yang masing-masing bersesuaian dengan peralihan  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_1$ ,  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_2$ ,  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_3$  dan  ${}^5\text{D}_0 \rightarrow {}^7\text{F}_4$ . Spektrum serapan dan pancaran telah digunakan untuk menilai parameter keamatan Judd-Ofelt dan kebarangkalian peralihan pancaran, nisbah cabangan dan keratan rentas pancaran rangsangan bagi tiga sistem kaca didop dengan ion nadir bumi ( $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$ , and  $\text{Eu}^{3+}$ ). Berdasarkan kepada kajian ini, boleh disimpulkan bahawa ciri rangkaian struktur kaca kalsium sulfoborofosfat dan kaca barium sulfoborofosfat adalah sama walaupun berbeza pengubahsuai. Penambahan ion sulfat dan ion nadir bumi ke dalam rangkaian kaca telah meningkatkan kestabilan kimia dan fizik di samping meningkatkan prestasi sifat optik sampel kaca yang telah disediakan seperti mempunyai nilai yang tinggi bagi nisbah cabangan, keratan rentas rangsangan, jalur lebar gandaan dan gandaan optik. Oleh itu, kaca kalsium sulfoborofosfat dan kaca barium sulfoborofosfat boleh dicadangkan sebagai bahan hos pendaraahaya bagi aplikasi peranti pencahayaan keadaan pepejal.

## TABLE OF CONTENTS

	TITLE	PAGE
	<b>DECLARATION</b>	<b>ii</b>
	<b>DEDICATION</b>	<b>iii</b>
	<b>ACKNOWLEDGEMENT</b>	<b>iv</b>
	<b>ABSTRACT</b>	<b>v</b>
	<b>ABSTRAK</b>	<b>vi</b>
	<b>TABLE OF CONTENTS</b>	<b>vii</b>
	<b>LIST OF TABLES</b>	<b>x</b>
	<b>LIST OF FIGURES</b>	<b>xv</b>
	<b>LIST OF ABBREVIATIONS</b>	<b>xxiv</b>
	<b>LIST OF SYMBOLS</b>	<b>xxv</b>
	<b>LIST OF APPENDICES</b>	<b>xxvi</b>
<b>CHAPTER 1</b>	<b>INTRODUCTION</b>	<b>1</b>
	1.1 Background of the Research	1
	1.2 Problem Statements	6
	1.3 Objectives of the Research	7
	1.4 Scopes of the Research	8
	1.5 Significance of the Research	9
	1.6 Outlines of Thesis	9
<b>CHAPTER 2</b>	<b>LITERATURE REVIEW</b>	<b>11</b>
	2.1 Introduction	11
	2.2 Amorphous phase of glass	11
	2.3 Thermal properties of glass system	15
	2.4 The structure of phosphate glasses	18
	2.5 Structural studies of Phosphate glasses by IR and Raman	20
	2.6 Structural Studies of Borate Glasses by IR and Raman	25

2.7	Structural Studies of Sulfophosphate and Sulfoborate Glasses by IR and Raman	27
2.8	Ultraviolet Visible Near Infrared (UV-Vis-NIR) Absorption Spectroscopy	33
2.9	Physical and Optical Properties of glass	34
2.9.1	Absorption Analysis	39
2.10	Judd-Ofelt Theory of glass	46
2.11	Judd-Ofelt Studies of Glass System	49
2.11.1	Doped Dy <sup>3+</sup> ions	50
2.11.2	Doped Eu <sup>3+</sup> ions	51
2.11.3	Doped Sm <sup>3+</sup> ions	52
2.12	Luminescence Studies of Glass System	54
2.12.1	Doped Dysprosium Ions	54
2.12.2	Doped Europium Ions	60
2.12.3	Doped Samarium Ions	66
<b>CHAPTER 3</b>	<b>RESEARCH METHODOLOGY</b>	<b>75</b>
3.1	Introduction	75
3.2	Sample Preparation	75
3.3	Density Measurement	79
3.4	Differential Thermal Analyzer	80
3.5	X-Ray Diffraction	81
3.6	Fourier Transform Infrared Spectroscopy	81
3.7	Raman Spectroscopy	82
3.8	Absorption Spectroscopy	83
3.9	Photoluminescence Spectroscopy	84
<b>CHAPTER 4</b>	<b>RESULTS AND DISCUSSION</b>	<b>87</b>
4.1	Introduction	87
4.2	Amorphous Phase	87
4.3	Thermal Properties	90
4.4	Density and Molar Volume Analysis	94
4.5	Structural Studies by IR	102
4.5.1	Doped Dy <sub>2</sub> O <sub>3</sub>	102

4.5.2	Doped Sm <sub>2</sub> O <sub>3</sub>	107
4.5.3	Doped Eu <sub>2</sub> O <sub>3</sub>	112
4.6	Structural Studies by Raman	116
4.6.1	Doped Dy <sub>2</sub> O <sub>3</sub>	116
4.6.2	Doped Sm <sub>2</sub> O <sub>3</sub>	120
4.6.3	Doped Eu <sub>2</sub> O <sub>3</sub>	125
4.7	Optical Properties Analysis	128
4.7.1	Absorption Spectra Analysis of Dy <sup>3+</sup> , Sm <sup>3+</sup> and Eu <sup>3+</sup> ions	128
4.8	Energy Band Gap	138
4.9	Refractive index and molar refractivity	152
4.10	Urbach Energy	160
4.11	Judd-Ofelt Analysis	166
4.11.1	Doped Dysprosium Ions	166
4.11.2	Doped Samarium Ions	172
4.11.3	Doped Europium Ions	177
4.12	Luminescence Properties Analysis	182
4.12.1	Excitation and Emission Spectra Analysis	182
4.13	Radiative Properties Analysis	197
<b>CHAPTER 5</b>	<b>CONCLUSION AND RECOMMENDATIONS</b>	<b>205</b>
5.1	Introduction	205
5.2	Conclusions	205
5.3	Recommendation	209
<b>REFERENCES</b>		<b>211</b>
<b>LIST OF PUBLICATIONS</b>		<b>267</b>



## LIST OF TABLES

TABLE NO.	TITLE	PAGE
<b>Table 2.1</b>	IR band assignment of borate, phosphate, sulphate and borophosphate glasses.	31
<b>Table 2.2</b>	Raman band assignment of borate, phosphate, sulphate and borophosphate glasses	32
<b>Table 3.1</b>	The undoped chemical composition for $x\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$ where $15 \leq x \leq 35$ mol%, and $y\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}y\text{)P}_2\text{O}_5$ where $15 \leq y \leq 35$ mol% glasses.	76
<b>Table 3.2</b>	Glass composition for $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}x\text{)P}_2\text{O}_5\text{-}x\text{Dy}_2\text{O}_3$ , RE = Dy, Sm and Eu where $0.1 \leq x \leq 1.0$ mol%, $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{Sm}_2\text{O}_3$ where $0.1 \leq y \leq 1.0$ mol% and $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}z\text{)P}_2\text{O}_5\text{-}z\text{Eu}_2\text{O}_3$ , where $0.1 \leq z \leq 1.0$ mol% glasses.	77
<b>Table 3.3</b>	Glass composition for $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}x\text{)P}_2\text{O}_5\text{-}x\text{Dy}_2\text{O}_3$ , RE = Dy, Sm and Eu where $0.1 \leq x \leq 1.0$ mol%, $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{Sm}_2\text{O}_3$ where $0.1 \leq y \leq 1.0$ mol% and $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}z\text{)P}_2\text{O}_5\text{-}z\text{Eu}_2\text{O}_3$ , where $0.1 \leq z \leq 1.0$ mol% glasses.	78
<b>Table 4.1</b>	$\text{CaSO}_4$ concentration dependent thermal properties of $x\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}x\text{)P}_2\text{O}_5$ with $15 \leq x \leq 35$ mol% glasses	92
<b>Table 4.2</b>	$\text{BaSO}_4$ concentration dependent thermal properties of $y\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}70\text{-}y\text{)P}_2\text{O}_5$ with $15 \leq y \leq 35$ mol%.	92
<b>Table 4.3</b>	Physical parameters of $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}x\text{)P}_2\text{O}_5\text{-}x\text{Dy}_2\text{O}_3$ glasses.	96
<b>Table 4.4</b>	Physical parameters of $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{Dy}_2\text{O}_3$ glasses.	97
<b>Table 4.5</b>	Physical parameters of $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}x\text{)P}_2\text{O}_5\text{-}x\text{Sm}_2\text{O}_3$ glasses.	98
<b>Table 4.6</b>	Physical parameters of $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{Sm}_2\text{O}_3$ glasses.	99
<b>Table 4.7</b>	Physical parameters of $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}x\text{)P}_2\text{O}_5\text{-}x\text{Eu}_2\text{O}_3$ glasses.	100
<b>Table 4.8</b>	Physical parameters of $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-(}45\text{-}y\text{)P}_2\text{O}_5\text{-}y\text{Eu}_2\text{O}_3$ glasses.	101

<b>Table 4.9</b>	IR band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses.	104
<b>Table 4.10</b>	IR band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses.	106
<b>Table 4.11</b>	IR band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses.	109
<b>Table 4.12</b>	IR band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses	111
<b>Table 4.13</b>	IR band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses	113
<b>Table 4.14</b>	IR band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses	115
<b>Table 4.15</b>	Raman band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$	118
<b>Table 4.16</b>	Raman band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$	119
<b>Table 4.17</b>	Raman band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$	122
<b>Table 4.18</b>	Raman band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$	124
<b>Table 4.19</b>	Raman band assignment and the reported values for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$	126
<b>Table 4.20</b>	Raman band assignment and the reported values for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$	127
<b>Table 4.21</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters ( $\delta$ ) of the $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	130
<b>Table 4.22</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters ( $\delta$ ) of the $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	130
<b>Table 4.23</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters ( $\delta$ ) of the $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses series.	133
<b>Table 4.24</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters and ( $\delta$ ) of the $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses series.	133

<b>Table 4.25</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters ( $\delta$ ) of the $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses series.	137
<b>Table 4.26</b>	Observed band positions ( $\text{cm}^{-1}$ ), Nephelauxetic ratio ( $\bar{\beta}$ ) and bonding parameters ( $\delta$ ) of the $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses series.	138
<b>Table 4.27</b>	Indirect and direct energy gap of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	141
<b>Table 4.28</b>	Indirect and direct energy gap of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	143
<b>Table 4.29</b>	Indirect and direct energy gap of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses series.	145
<b>Table 4.30</b>	Indirect and direct energy gap of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	147
<b>Table 4.31</b>	Indirect and direct energy gap of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses series.	149
<b>Table 4.32</b>	Indirect and direct energy gap of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	151
<b>Table 4.33</b>	Refractive index and molar refractivity of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses.	153
<b>Table 4.34</b>	Refractive index and molar refractivity of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	156
<b>Table 4.35</b>	Refractive index and molar refractivity of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	158
<b>Table 4.36</b>	Urbach's energy of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses.	161
<b>Table 4.37</b>	Urbach's energy of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	163
<b>Table 4.38</b>	Urbach's energy of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	164
<b>Table 4.39</b>	Experimental and calculated oscillator field strength ( $\times 10^{-6}$ ) and root mean square deviation ( $\delta_{\text{rms}}$ ) for $25\text{CaSO}_4$	

	– 30B <sub>2</sub> O <sub>3</sub> – (45-x)P <sub>2</sub> O <sub>5</sub> – xDy <sub>2</sub> O <sub>3</sub> with 0.1 ≤ x ≤ 1.0 mol% glasses series.	168
<b>Table 4.40</b>	Experimental and calculated oscillator field strength (x 10 <sup>-6</sup> ) and root mean square deviation (δ <sub>rms</sub> ) for 25BaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-y)P <sub>2</sub> O <sub>5</sub> – yDy <sub>2</sub> O <sub>3</sub> with 0.1 ≤ y ≤ 1.0 mol% glasses series.	168
<b>Table 4.41</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> , Ω <sub>4</sub> and Ω <sub>6</sub> (cm <sup>2</sup> x 10 <sup>-20</sup> ) of the 25CaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-x)P <sub>2</sub> O <sub>5</sub> – xDy <sub>2</sub> O <sub>3</sub> glasses series.	169
<b>Table 4.42</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> , Ω <sub>4</sub> and Ω <sub>6</sub> (cm <sup>2</sup> x 10 <sup>-20</sup> ) of the 25BaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-y)P <sub>2</sub> O <sub>5</sub> – yDy <sub>2</sub> O <sub>3</sub> with (0.1 ≤ y ≤ 1.0 mol%) glasses series.	169
<b>Table 4.43</b>	Experimental and calculated oscillator field strength (x 10 <sup>-6</sup> ) and root mean square deviation (δ <sub>rms</sub> ) for 25CaSO <sub>4</sub> –30B <sub>2</sub> O <sub>3</sub> –(45-x)P <sub>2</sub> O <sub>5</sub> –xSm <sub>2</sub> O <sub>3</sub> with 0.1 ≤ x ≤ 1.0 mol% glasses.	174
<b>Table 4.44</b>	Experimental and calculated oscillator field strength (x 10 <sup>-6</sup> ) and root mean square deviation (δ <sub>rms</sub> ) for 25BaSO <sub>4</sub> –30B <sub>2</sub> O <sub>3</sub> –(45-y)P <sub>2</sub> O <sub>5</sub> –ySm <sub>2</sub> O <sub>3</sub> with 0.1 ≤ y ≤ 1.0 mol% glasses	174
<b>Table 4.45</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> , Ω <sub>4</sub> and Ω <sub>6</sub> (cm <sup>2</sup> x 10 <sup>-21</sup> ) of the 25CaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-x)P <sub>2</sub> O <sub>5</sub> – xSm <sub>2</sub> O <sub>3</sub> glasses series.	175
<b>Table 4.46</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> , Ω <sub>4</sub> and Ω <sub>6</sub> (cm <sup>2</sup> x 10 <sup>-21</sup> ) of the 25BaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-y)P <sub>2</sub> O <sub>5</sub> – ySm <sub>2</sub> O <sub>3</sub> with (0.1 ≤ y ≤ 1.0 mol%) glasses series.	175
<b>Table 4.47</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> and Ω <sub>4</sub> (cm <sup>2</sup> x 10 <sup>-20</sup> ) of the 25CaSO <sub>4</sub> – 30B <sub>2</sub> O <sub>3</sub> – (45-x)P <sub>2</sub> O <sub>5</sub> – xEu <sub>2</sub> O <sub>3</sub> with (0.1 ≤ x ≤ 1.0 mol%) glasses series.	179
<b>Table 4.48</b>	The Judd-Ofelt intensity parameters Ω <sub>2</sub> and Ω <sub>4</sub> (cm <sup>2</sup> x 10 <sup>-20</sup> ) of the 25BaSO <sub>4</sub> –30B <sub>2</sub> O <sub>3</sub> –(45-y)P <sub>2</sub> O <sub>5</sub> –yEu <sub>2</sub> O <sub>3</sub> with (0.1 ≤ y ≤ 1.0 mol%) glasses series.	179
<b>Table 4.49</b>	Emission band position (λ <sub>p</sub> , nm), radiative transition probability (A <sub>rad</sub> , s <sup>-1</sup> ), total radiative transition probability (A <sub>T</sub> s <sup>-1</sup> ), fluorescence branching ratio (β <sub>r</sub> , %), calculated lifetime (τ <sub>cal</sub> x 10 <sup>-4</sup> ), emission cross section (σ x 10 <sup>-22</sup> cm <sup>2</sup> ), gain bandwidth (σ x FWHM x 10 <sup>-28</sup> cm <sup>2</sup> ) and optical gain (σ x τ <sub>cal</sub> x 10 <sup>-25</sup> cm <sup>2</sup> s <sup>-1</sup> ) for 25CaSO <sub>4</sub> 30B <sub>2</sub> O <sub>3</sub> – (45-x)P <sub>2</sub> O <sub>5</sub> – xDy <sub>2</sub> O <sub>3</sub> glasses.	199
<b>Table 4.50</b>	Emission band position (λ <sub>p</sub> , nm), radiative transition probability (A <sub>rad</sub> , s <sup>-1</sup> ), total radiative transition probability (A <sub>T</sub> s <sup>-1</sup> ), fluorescence branching ratio (β <sub>r</sub> , %), calculated	

lifetime ( $\tau_{\text{cal}} \times 10^{-4}$ ), emission cross section ( $\sigma \times 10^{-22} \text{ cm}^2$ ), gain bandwidth ( $\sigma \times \text{FWHM} \times 10^{-28} \text{ cm}^2$ ) and optical gain ( $\sigma \times \tau_{\text{cal}} \times 10^{-25} \text{ cm}^2 \text{ s}^{-1}$ ) for  $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$  glasses 199

**Table 4.51** Emission band position ( $\lambda_p$ , nm), radiative transition probability ( $A_{\text{rad}}$ ,  $\text{s}^{-1}$ ), total radiative transition probability ( $A_T \text{ s}^{-1}$ ), fluorescence branching ratio ( $\beta_r$ , %), calculated lifetime ( $\tau_{\text{cal}} \times 10^{-3}$ ), emission cross section ( $\sigma \times 10^{-22} \text{ cm}^2$ ), gain bandwidth ( $\sigma \times \text{FWHM} \times 10^{-28} \text{ cm}^2$ ) and optical gain ( $\sigma \times \tau_{\text{cal}} \times 10^{-25} \text{ cm}^2 \text{ s}^{-1}$ ) for  $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$  glasses 200

**Table 4.52** Emission band position ( $\lambda_p$ , nm), radiative transition probability ( $A_{\text{rad}}$ ,  $\text{s}^{-1}$ ), total radiative transition probability ( $A_T \text{ s}^{-1}$ ), fluorescence branching ratio ( $\beta_r$ , %), calculated lifetime ( $\tau_{\text{cal}}$ , ms), emission cross section ( $\sigma \times 10^{-22} \text{ cm}^2$ ) gain bandwidth ( $\sigma \times \text{FWHM} \times 10^{-28} \text{ cm}^2$ ) and optical gain ( $\sigma \times \tau_{\text{cal}} \times 10^{-25} \text{ cm}^2 \text{ s}^{-1}$ ) for  $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$  glasses. 201

**Table 4.53** Emission band position ( $\lambda_p$ , nm), radiative transition probability ( $A_{\text{rad}}$ ,  $\text{s}^{-1}$ ), total radiative transition probability ( $A_T \text{ s}^{-1}$ ), fluorescence branching ratio ( $\beta_r$ , %), calculated lifetime ( $\tau_{\text{cal}}$ , ms), emission cross section ( $\sigma \times 10^{-22} \text{ cm}^2$ ), gain bandwidth ( $\sigma \times \text{FWHM} \times 10^{-27} \text{ cm}^2$ ) and optical gain ( $\sigma \times \tau_{\text{cal}} \times 10^{-25} \text{ cm}^2 \text{ s}^{-1}$ ) for  $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$  glasses 202

**Table 4.54** Emission band position ( $\lambda_p$ , nm), radiative transition probability ( $A_{\text{rad}}$ ,  $\text{s}^{-1}$ ), total radiative transition probability ( $A_T \text{ s}^{-1}$ ), fluorescence branching ratio ( $\beta_r$ , %), calculated lifetime ( $\tau_{\text{cal}}$ , ms), emission cross section ( $\sigma \times 10^{-22} \text{ cm}^2$ ), gain bandwidth ( $\sigma \times \text{FWHM} \times 10^{-27} \text{ cm}^2$ ) and optical gain ( $\sigma \times \tau_{\text{cal}} \times 10^{-25} \text{ cm}^2 \text{ s}^{-1}$ ) for  $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$  glasses 203

## LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
<b>Figure 2.1</b>	Schematic mechanism of X-ray diffractometer (Cullity, 1978)	12
<b>Figure 2.2</b>	Principle of X-ray Diffraction	13
<b>Figure 2.3</b>	XRD pattern of $15\text{Li}_2\text{O}-30\text{B}_2\text{O}_3-15\text{SO}_3-(14-x)\text{P}_2\text{O}_5-x\text{Dy}_2\text{O}_3$ glasses (Bulus <i>et al.</i> , 2017).	14
<b>Figure 2.4</b>	XRD patterns of pure ZBP and (0.9 mol%) $\text{Eu}^{3+}$ : ZBP glasses along with 1.0 mole% $\text{Sm}^{3+}$ : ZBP glass, (Hima Bindu <i>et al.</i> , 2016).	14
<b>Figure 2.5</b>	XRD patterns of borophosphate glasses doped $\text{Mn}^{2+}$ (Wan <i>et al.</i> , 2014).	15
<b>Figure 2.6</b>	DTA traces of $\text{Na}_2\text{SO}_4-\text{B}_2\text{O}_3-\text{P}_2\text{O}_5: \text{MoO}_3$ glasses (Ravi Kumar <i>et al.</i> , 2012).	17
<b>Figure 2.7</b>	DTA traces of lithium borate glasses doped with $\text{Dy}_2\text{O}_3$ glasses (Pawar, Munishwar and Gedam, 2017).	17
<b>Figure 2.8</b>	Tetrahedral Phosphate glasses (Brow, 2000).	20
<b>Figure 2.9</b>	FTIR spectra of glass samples from $x\text{BaO}-(20-x)\text{CaO}-32\text{Fe}_2\text{O}_3-48\text{P}_2\text{O}_5$ ( $x = 2,4,6,8,10,12,14,16,18$ mol%) (a) $x = 2,6,10,14,18$ ; (b) $x = 4,8,12,16$ (Li <i>et al.</i> , 2016).	23
<b>Figure 2.10</b>	Raman spectra of $(x\text{ZnO}-10\text{Fe}_2\text{O}_3-(90-x)\text{P}_2\text{O}_5)$ (Li <i>et al.</i> , 2013).	24
<b>Figure 2.11</b>	Structural groups postulated for borate (Yano <i>et al.</i> , 2003).	25
<b>Figure 2.12</b>	FTIR spectra of glass samples from $(60-x)\text{P}_2\text{O}_5-20\text{MgO}-20\text{ZnSO}_4-x\text{Sm}_2\text{O}_3$ ( $x = 0.0,0.5,1.0,1.5$ and $2.0$ mol%) (Ahmadi, Hussin and Ghoshal, 2017).	29
<b>Figure 2.13</b>	Variation of density and molar volume with $\text{Dy}_2\text{O}_3$ (Pawar, Munishwar and Gedam, 2017).	37
<b>Figure 2.14</b>	Urbach tail, conduction and valence band (Choudhury, Dey and Choudhury, 2013)	39
<b>Figure 2.15</b>	Optical absorption spectrum of 2 mol% of dysprosium ions doped borophosphate glasses in Vis- NIR region (Vijayakumar, Venkataiah and Marimuthu, 2015a).	40

<b>Figure 2.16</b>	Optical absorption spectrum of 0.9 mol% of europium ions doped zinc borophosphate glass in UV-Vis region (Hima Bindu <i>et al.</i> , 2016).	41
<b>Figure 2.17</b>	Optical absorption spectrum of 0.9 mol% of europium ions doped zinc borophosphate glass in NIR region (Hima Bindu <i>et al.</i> , 2016).	41
<b>Figure 2.18</b>	Optical absorption spectrum of 2 wt% of Samarium ions doped borophosphate glass in UV-Vis and NIR region (Vijayakumar and Marimuthu, 2015).	42
<b>Figure 2.19</b>	Tauc plots of indirect allowed transitions of Dy <sup>3+</sup> doped borophosphate glasses [inset shows Tauc's plot for indirect allowed transitions (Vijayakumar, Venkataiah and Marimuthu, 2015a).	43
<b>Figure 2.20</b>	Tauc plots of the indirect and direct bandgap of (0.9 mol%) of Eu <sup>3+</sup> doped borophosphate glasses (Hima Bindu <i>et al.</i> , 2016).	44
<b>Figure 2.21</b>	Tauc plots of indirect allowed transitions of Sm <sup>3+</sup> doped borophosphate glasses [inset shows Tauc's plot for indirect allowed transitions. (Vijayakumar, Marimuthu and Sudarsan, 2015).	45
<b>Figure 2.22</b>	Emission spectra of calcium fluoroborate doped Dy <sup>3+</sup> glasses (Suresh Kumar <i>et al.</i> , 2010).	57
<b>Figure 2.23</b>	Luminescence spectra of the Dy <sup>3+</sup> doped oxyfluoroborophosphate glasses [insert shows the excitation spectrum of the 0.5 mol% of Dy <sup>3+</sup> doped oxyfluoroborophosphate glasses (Vijayakumar and Marimuthu, 2015)	58
<b>Figure 2.24</b>	Luminescence spectra of 2 mol% of dysprosium ions doped oxyfluoro-borophosphate glass (Vijayakumar and Marimuthu, 2015).	59
<b>Figure 2.25</b>	Excitation spectrum of Dy <sup>3+</sup> doped phosphate glass (Sreedhar <i>et al.</i> , 2013).	60
<b>Figure 2.26</b>	Excitation spectra for 0.2 mol% of europium ions doped borate glasses (Venkateswarlu and Rudramadevi, 2015).	62
<b>Figure 2.27</b>	Emission spectra for 0.2 mol% of europium ions doped borate glass (Venkateswarlu and Rudramadevi, 2015).	63
<b>Figure 2.28</b>	Emission spectra of the europium ions doped zinc borophosphate glasses (Hima Bindu <i>et al.</i> , 2016).	64
<b>Figure 2.29</b>	Excitation spectra for 0.9 mol% of europium ions doped zinc borophosphate glasses (Hima Bindu <i>et al.</i> , 2016).	65

<b>Figure 2.30</b>	Photoluminescence spectra of the $\text{Eu}^{3+}$ doped lead borotelluro-phosphate glasses [insert shows the excitation spectrum for the 1.0 mol% of $\text{Eu}^{3+}$ doped lead borotelluro-phosphate glass (Selvi <i>et al.</i> , 2016)	66
<b>Figure 2.31</b>	Excitation spectrum of the 0.5 mol% $\text{Sm}^{3+}$ glass. (Vijayakumar and Marimuthu, 2015).	69
<b>Figure 2.32</b>	Luminescence spectra of $\text{Sm}^{3+}$ doped borophosphate glasses (Vijayakumar and Marimuthu, 2015).	70
<b>Figure 2.33</b>	Luminescence spectra of the $\text{Sm}^{3+}$ doped lead fluoroborophosphate glasses [insert shows the excitation spectrum for the 0.5 mol% of $\text{Sm}^{3+}$ doped leadfluoroborophosphate glass] (Vijayakumar, Marimuthu and Sudarsan, 2015)	71
<b>Figure 2.34</b>	Photoluminescence spectra of $\text{Sm}^{3+}$ doped lead borotelluro-phosphate glasses (Selvi, Marimuthu and Muralidharan, 2015).	72
<b>Figure 2.35</b>	Excitation spectra for different concentration of samarium ions doped borate glasses (Swapna <i>et al.</i> , 2014)	73
<b>Figure 2.36</b>	Emission spectra for different concentration of $\text{Sm}^{3+}$ ions in borate glasses (Swapna <i>et al.</i> , 2014)	74
<b>Figure 3.1</b>	The flowchart of sample preparation for Calcium Sulfo borophosphate glass and Barium Sulfo borophosphate glass.	79
<b>Figure 3.2</b>	Schematic diagram of DTA (El-Mallawany, 2002).	80
<b>Figure 3.3</b>	X-ray diffractometer	81
<b>Figure 3.4</b>	Schematic diagram for FTIR spectroscopic instrument.	82
<b>Figure 3.5</b>	Schematic diagram for Raman spectroscopic instrument.	83
<b>Figure 3.6</b>	Schematic diagram of UV-Vis-NIR spectroscopy.	84
<b>Figure 3.7</b>	Schematic diagram photoluminescence spectroscopy.	85
<b>Figure 4.1</b>	XRD pattern of un-doped $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-}45\text{P}_2\text{O}_5$ glass.	88
<b>Figure 4.2</b>	XRD pattern of un-doped $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-}45\text{P}_2\text{O}_5$ glass.	88
<b>Figure 4.3</b>	XRD pattern of $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-}(45-x)\text{P}_2\text{O}_5\text{-}x\text{Dy}_2\text{O}_3$ ( $0.1 \leq x \leq 0.5$ mol% ) glasses.	89
<b>Figure 4.4</b>	XRD pattern of $25\text{CaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-}(45-x)\text{P}_2\text{O}_5\text{-}x\text{Sm}_2\text{O}_3$ ( $0.1 \leq x \leq 0.5$ mol% ) glasses.	89
<b>Figure 4.5</b>	XRD pattern of $25\text{BaSO}_4\text{-}30\text{B}_2\text{O}_3\text{-}(45-y)\text{P}_2\text{O}_5\text{-}y\text{Eu}_2\text{O}_3$ ( $0.1 \leq y \leq 0.5$ mol% ) glasses.	90



<b>Figure 4.6</b>	DTA spectra of $x\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (70-x)\text{P}_2\text{O}_5$ ( $15 \leq x \leq 35$ mol%) glasses.	91
<b>Figure 4.7</b>	DTA spectra of $y\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (70-y)\text{P}_2\text{O}_5$ ( $15 \leq y \leq 35$ mol%) glasses.	92
<b>Figure 4.8</b>	Relationship between $T_g$ , $T_c$ and $(T_c-T_g)$ of $x\text{CaSO}_4-30\text{B}_2\text{O}_3-(70-x)\text{P}_2\text{O}_5$ with $15 \leq x \leq 35$ mol% glass as a function of $\text{CaSO}_4$ concentration.	93
<b>Figure 4.9</b>	Relationship between $T_g$ , $T_c$ and $(T_c-T_g)$ of $y\text{BaSO}_4-30\text{B}_2\text{O}_3-(70-y)\text{P}_2\text{O}_5$ with $15 \leq y \leq 35$ mol% glass as a function of $\text{BaSO}_4$ concentration.	93
<b>Figure 4.10</b>	Variation of glass density and molar volume of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses.	96
<b>Figure 4.11</b>	Variation of glass density and molar volume of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses.	97
<b>Figure 4.12</b>	Variation of glass density and molar volume of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses.	98
<b>Figure 4.13</b>	Variation of glass density and molar volume of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	99
<b>Figure 4.14</b>	Variation of glass density and molar volume of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses.	100
<b>Figure 4.15</b>	Variation of glass density and molar volume of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	101
<b>Figure 4.16</b>	The IR spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	103
<b>Figure 4.17</b>	The IR spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	105
<b>Figure 4.18</b>	The IR spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	108
<b>Figure 4.19</b>	The IR spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	110
<b>Figure 4.20</b>	The IR spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	113
<b>Figure 4.21</b>	The IR spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	114
<b>Figure 4.22</b>	The Raman spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	117
<b>Figure 4.23</b>	The Raman spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	118

<b>Figure 4.24</b>	The Raman spectra of $25\text{CaSO}_4-30\text{B}_2\text{O}_3-(45-x)\text{P}_2\text{O}_5-x\text{Sm}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	121
<b>Figure 4.25</b>	The Raman spectra of $25\text{BaSO}_4-30\text{B}_2\text{O}_3-(45-y)\text{P}_2\text{O}_5-y\text{Sm}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	123
<b>Figure 4.26</b>	The Raman spectra of $25\text{CaSO}_4-30\text{B}_2\text{O}_3-(45-x)\text{P}_2\text{O}_5-x\text{Eu}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	126
<b>Figure 4.27</b>	The Raman spectra of $25\text{BaSO}_4-30\text{B}_2\text{O}_3-(45-y)\text{P}_2\text{O}_5-y\text{Eu}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	127
<b>Figure 4.28</b>	Absorption spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	129
<b>Figure 4.29</b>	Absorption spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	129
<b>Figure 4.30</b>	Absorption spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ ( $0.1 \leq x \leq 1.0$ mol%) glasses.	132
<b>Figure 4.31</b>	Absorption spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ ( $0.1 \leq y \leq 1.0$ mol%) glasses.	132
<b>Figure 4.32</b>	Absorption spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses series in UV-Visible regions ( $0.1 \leq x \leq 1.0$ mol%).	135
<b>Figure 4.33</b>	Absorption spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses series in NIR regions ( $0.1 \leq x \leq 1.0$ mol%).	136
<b>Figure 4.34</b>	Absorption spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses series in UV-Visible regions ( $0.1 \leq y \leq 1.0$ mol%).	136
<b>Figure 4.35</b>	Absorption spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses series in NIR regions ( $0.1 \leq y \leq 1.0$ mol%).	137
<b>Figure 4.36</b>	Graph of $(\alpha h\nu)^{1/2}$ against photon energy ( $h\nu$ ) for indirect allowed transitions of the $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	140
<b>Figure 4.37</b>	Graph of $(\alpha h\nu)^2$ against photon energy ( $h\nu$ ) for direct allowed transitions of the $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	141
<b>Figure 4.38</b>	Variation of indirect and direct energy gap of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses.	142
<b>Figure 4.39</b>	Graph of $(\alpha h\nu)^{1/2}$ against photon energy ( $h\nu$ ) for indirect allowed transitions of the $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	142

<b>Figure 4.40</b>	Graph of $(\alpha hv)^2$ against photon energy (hv) for direct allowed transitions of the $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - yDy_2O_3$ glasses series.	143
<b>Figure 4.41</b>	Variation of indirect and direct energy gap of $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - yDy_2O_3$ glasses.	144
<b>Figure 4.42</b>	Graph of $(\alpha hv)^{1/2}$ against photon energy (hv) for indirect allowed transitions of the $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xSm_2O_3$ glasses series.	144
<b>Figure 4.43</b>	Graph of $(\alpha hv)^2$ against photon energy (hv) for direct allowed transitions of the $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xSm_2O_3$ glasses series.	145
<b>Figure 4.44</b>	Variation of indirect and direct energy gap of $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xSm_2O_3$ glasses.	146
<b>Figure 4.45</b>	Graph of $(\alpha hv)^{1/2}$ against photon energy (hv) for indirect allowed transitions of the $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - ySm_2O_3$ glasses series.	146
<b>Figure 4.46</b>	Graph of $(\alpha hv)^2$ against photon energy (hv) for direct allowed transitions of the $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - ySm_2O_3$ glasses series.	147
<b>Figure 4.47</b>	Variation of indirect and direct energy gap of $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - ySm_2O_3$ glasses.	148
<b>Figure 4.48</b>	Graph of $(\alpha hv)^{1/2}$ against photon energy (hv) for indirect allowed transitions of the $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xEu_2O_3$ glasses series.	148
<b>Figure 4.49</b>	Graph of $(\alpha hv)^2$ against photon energy (hv) for direct allowed transitions of the $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xEu_2O_3$ glasses series.	149
<b>Figure 4.50</b>	Variation of indirect and direct energy gap of $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xEu_2O_3$ glasses.	150
<b>Figure 4.51</b>	Graph of $(\alpha hv)^{1/2}$ against photon energy (hv) for indirect allowed transitions of the $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - yEu_2O_3$ glasses series.	150
<b>Figure 4.52</b>	Graph of $(\alpha hv)^2$ against photon energy (hv) for direct allowed transitions of the $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - yEu_2O_3$ glasses series.	151
<b>Figure 4.53</b>	Variation of indirect and direct energy gap of $25BaSO_4 - 30B_2O_3 - (45-y)P_2O_5 - yEu_2O_3$ glasses.	152
<b>Figure 4.54</b>	Variation of refractive index and molar refractivity of $25CaSO_4 - 30B_2O_3 - (45-x)P_2O_5 - xDy_2O_3$ glasses.	154

<b>Figure 4.55</b>	Variation of refractive index and molar refractivity of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses	154
<b>Figure 4.56</b>	Variation of refractive index and molar refractivity of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses.	156
<b>Figure 4.57</b>	Variation of refractive index and molar refractivity of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	157
<b>Figure 4.58</b>	Variation of refractive index and molar refractivity of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses.	159
<b>Figure 4.59</b>	Variation of refractive index and molar refractivity of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	159
<b>Figure 4.60</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses	162
<b>Figure 4.61</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses	162
<b>Figure 4.62</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses	163
<b>Figure 4.63</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses	164
<b>Figure 4.64</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses	165
<b>Figure 4.65</b>	The graph of $\ln\alpha$ as a function of photon energy to determine the $E_{\text{urb}}$ of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses	165
<b>Figure 4.66</b>	J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses.	170
<b>Figure 4.67</b>	J-O parameters as a function of dopant concentration for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses.	171
<b>Figure 4.68</b>	Variation of J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses.	171
<b>Figure 4.69</b>	J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses.	176

<b>Figure 4.70</b>	J-O parameters as a function of dopant concentration for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	176
<b>Figure 4.71</b>	Variation of J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses.	177
<b>Figure 4.72</b>	J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses.	180
<b>Figure 4.73</b>	J-O parameters as a function of dopant concentration for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	180
<b>Figure 4.74</b>	Variation of J-O parameters as a function of dopant concentration for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glasses and $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glasses.	181
<b>Figure 4.75</b>	Excitation spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series .	184
<b>Figure 4.76</b>	Excitation spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	184
<b>Figure 4.77</b>	Emission spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	185
<b>Figure 4.78</b>	Emission spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	185
<b>Figure 4.79</b>	Energy level diagram of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Dy}_2\text{O}_3$ glasses series.	186
<b>Figure 4.80</b>	Energy level diagram of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Dy}_2\text{O}_3$ glasses series.	186
<b>Figure 4.81</b>	Excitation spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses series.	189
<b>Figure 4.82</b>	Excitation spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses series.	189
<b>Figure 4.83</b>	Emission spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses series.	190
<b>Figure 4.84</b>	Emission spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses series.	190
<b>Figure 4.85</b>	Energy level diagram of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Sm}_2\text{O}_3$ glasses series.	191
<b>Figure 4.86</b>	Energy level diagram of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ glasses series.	191

<b>Figure 4.87</b>	Excitation spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glass series.	194
<b>Figure 4.88</b>	Excitation spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glass series.	194
<b>Figure 4.89</b>	Emission spectra of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ glass series.	195
<b>Figure 4.90</b>	Emission spectra of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ glass series.	195
<b>Figure 4.91</b>	Energy level diagram of $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (45-x)\text{P}_2\text{O}_5 - x\text{Eu}_2\text{O}_3$ with glass series.	196
<b>Figure 4.92</b>	Energy level diagram of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Eu}_2\text{O}_3$ with glass series.	196

## LIST OF ABBREVIATIONS

$B_2O_3$	-	Borate
$CaSO_4$	-	Calcium Sulfate
$P_2O_5$	-	Phosphorus Pentoxide
$BaSO_4$	-	Barium Sulfate
$Dy_2O_3$	-	Dysprosium Oxide
$Eu_2O_3$	-	Europium Oxide
$Sm_2O_3$	-	Samarium Oxide
$Dy^{3+}$	-	Dysprosium Ion
$Eu^{3+}$	-	Europium Ion
$Sm^{3+}$	-	Samarium Ion
$f_{exp}$	-	Experimental oscillator strengths
$f_{cal}$	-	Calculated oscillator strengths
IR	-	Infrared
$S_{ed}$	-	Electric dipole line strength
$S_{md}$	-	Magnetic dipole line strength
$A_{rad}$	-	Radiative transition probability
$\tau_{rad}$	-	Radiative lifetime
$\beta_r$	-	Branching ratio
$\lambda_p$	-	Emission band position
FTIR	-	Fourier Transform Infrared
KBr	-	Potassium bromide
XRD	-	X-Ray Diffraction
UV	-	Ultraviolet
RE	-	Rare Earth
PL	-	Photoluminescence

## LIST OF SYMBOLS

$h$	-	Planck's constant
$^{\circ}\text{C}$	-	Degree Celsius
$\nu$	-	Frequency
$c$	-	Speed of light
$\alpha(\nu)$	-	Absorption coefficient
$\beta$	-	Nepheleuxetic ratios
$\Omega_2$	-	Judd-Ofelt parameter
$\Omega_4$	-	Judd-Ofelt parameter
$\Omega_6$	-	Judd-Ofelt parameter
$J$	-	Total angular momentum
$\theta$	-	Diffraction angle of the X-Ray beam
$\lambda$	-	Wavelength
$n$	-	Refractive Index



## LIST OF APPENDICES

APPENDIX	TITLE	PAGE
Appendix A	Example of the Calculation of mol% of the compounds involve in a glass system	231
Appendix B	Calculation of experimental and calculated oscillator strength, root mean square deviation and Judd–Ofelt for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - 44.7\text{P}_2\text{O}_5 - 0.3\text{Sm}_2\text{O}_3$ glass	233
Appendix C	Calculation of radiative properties for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - 44.7\text{P}_2\text{O}_5 - 0.3\text{Sm}_2\text{O}_3$ glass	242
Appendix D	Calculation of Judd-Ofelt for $25\text{CaSO}_4 - 30\text{B}_2\text{O}_3 - (44.7)\text{P}_2\text{O}_5 - 0.3 \text{Eu}_2\text{O}_3$ glass	247
Appendix E	Linear fit equation for indirect bandgap of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ with $0.1 \leq y \leq 1.0$ mol% glasses	251
Appendix F	Linear fit equation for direct bandgap of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ with $0.1 \leq y \leq 1.0$ mol% glasses	254
Appendix G	Linear fit equation for Urbach energy of $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (45-y)\text{P}_2\text{O}_5 - y\text{Sm}_2\text{O}_3$ with $0.1 \leq y \leq 1.0$ mol% glasses	257
Appendix H	Calculation of uncertainties for indirect and direct bandgap, Urbach energy, refractive index and molar refractivity for $25\text{BaSO}_4 - 30\text{B}_2\text{O}_3 - (44.9)\text{P}_2\text{O}_5 - 0.1 \text{Sm}_2\text{O}_3$ glass.	262
Appendix I	Calculation of Physical parameters and uncertainties for $25\text{BaSO}_4-30\text{B}_2\text{O}_3-44.9\text{P}_2\text{O}_5-0.1\text{Eu}_2\text{O}_3$	264

# CHAPTER 1

## INTRODUCTION

This chapter outlines the fundamental background knowledge of the study. This includes the statement of research problem which led to the present research, objectives of the study, scope, significance of the research and outlines of the thesis.

### 1.1 Background of the Research

Glass is any solid that has an amorphous structure in nature and displays a glass transition when heated. Precisely, it is a solid formed by rapid melt quenching. It is hard, breakable and optically transparent. Diverse type of materials is used for making glass such as polymers, alloys of metals, aqueous solution, molecular liquids, ionic melts, etc. However, other elements are usually added to the ordinary glasses to change their physical and chemical properties.

The composition of materials used significantly contributes to the physical and chemical properties of glasses. Oxide glasses have three classes of components: the network formers, the intermediates, and modifiers. The network formers constitute a system of highly cross-linked chemical bond, while the intermediates and modifiers that are usually present as ions alter the network structure by being counterbalance by nonbridging oxygen atoms that are covalently bonded by the glass network (Saidu *et al.*, 2014).

In recent years, Lanthanide (Ln)-doped inorganic materials such as Ln-doped glasses, crystals and phosphors have gained rapid research interest due to their technological importance in the development of various optical and optoelectronic devices such as lasers, display devices, LEDs, fiber optic amplifiers and optical sensors (Srinivasulu *et al.*, 2013).

Borate ( $B_2O_3$ ) is one of the essential glass forming oxides and has been incorporated into various kinds of glass system to attain the desired physical and chemical properties (Wan *et al.*, 2014). Borate glasses have been of scientific interest for many years because of their potential applications like electro-optic switches, electro-optic modulators, solid-state laser materials and non-linear optical parametric converters. It possess excellent transparency, thermal stability and excellent rare earth ion solubility but it has higher phonon energy ( $\approx 1300\text{ cm}^{-1}$ ) which reduces the rare earth emission intensity due to their higher nonradiative decay (Swapna *et al.*, 2013). The structure of vitreous  $B_2O_3$  consists of a random network of boroxol rings and  $BO_3$  triangles connected by B-O-B linkages. However, metal oxides like MgO, CaO, SrO, BaO, ZnO and  $Al_2O_3$ , etc. have been added to  $B_2O_3$  and were found to be excellent stabilizers of borate glasses (Sumalatha *et al.*, 2013).

Phosphate ( $P_2O_5$ ) is another good glass forming oxides due to their favourable properties such as reasonably low liquid and glass transition temperatures, low viscosity, high thermal expansion coefficient, high electrical conductivity and high ultraviolet transmission, it's found application in a wide range of fields. For example, phosphate glasses are used in lasers, a solid electrolyte, bio-medical devices and nuclear waste immobilization (Joseph *et al.*, 2015). However, practical forming characteristics of phosphate glasses is limited because of their hygroscopic nature and relatively poor chemical durability (Jha and Jayasimhadri, 2016).

Therefore, to overcome the difficulties and limitations of both borate and phosphate glasses, the two host are combined to form a new glassy material called "Borophosphate glass" which gives a better advantageous as they exhibit different properties. However, borophosphate glasses are promising host materials for optical applications because of their excellent optical properties, low refractive indices, low dispersion and good transparency from ultraviolet to the near-infrared regions. Furthermore, the combination of  $B_2O_3$  and  $P_2O_5$  in the same matrix with additional oxides resulted in properties enhancements (Wan *et al.*, 2014). The role played by  $B_2O_3$  and  $P_2O_5$  in the glass structure, and the interaction with other elements in the glass network is an interesting subject of glass science. Hence, the combination of

the two network formers enables considerable modifications of the properties of the materials compared to pure borate and phosphate networks alone (Pang *et al.*, 2014). For instance, the chemical durability can be increased, or volume nucleation can be controlled by mixing the borate and phosphate groups.

Borophosphate along with modifiers (sulfate) is a fascinating area of study. In these glasses, the basic units of pure borate glasses are trigonal  $\text{BO}_3$  groups, whereas those of pure phosphate glasses are  $\text{PO}_4$  tetrahedra linked through covalent bridging oxygen. The addition of a modifier to borate and phosphate networks has different effects. In borate network, the addition of a modifier in some concentration ranges increases the degree of polymerization. The boron coordination changes from trigonal ( $\text{BO}_3$ ) to tetrahedral ( $\text{BO}_4$ ), whereas in phosphate network, an ultra-phosphate network consisting of  $\text{Q}^2$  and  $\text{Q}^3$  tetrahedra may form with  $\text{O/P} < 3.0$  (Ravi Kumar *et al.*). However, in the development of glass material, the stability and efficiency can be tailored by introducing a modifier.

Calcium oxide and barium oxide are two useful modifiers in modifying the phosphate properties (Li *et al.*, 2016). Calcium oxide and barium oxide served as a modifier to reduce the hygroscopic properties while sulfur was added into borophosphate as intermediate to enhance the host network. The influence of calcium oxide on iron phosphate could improve glass chemical durability, especially the alkaline resistance of glass fibre reinforced concrete (Brow, 2008). Barium oxides work as the modifier that could strengthen the glass network, restraining glass from crystallization, leading to P-O-Ba bands and improving the glass thermal stability, low melting temperature and wide glass forming region (Lu *et al.*, 2015). Moreover, as a divalent network modifying oxide, BaO increases density, refractive index and vitreous luster of the glass, slightly promote the melting process and enhance the ability to absorb radiation (Lu *et al.*, 2015).

Rare earth (RE) doped glasses have become an important class of optical systems due to their applicability as a solid-state laser, waveguides lasers and optical amplifiers. Most of the studies of glasses focus on explaining the structure properties regarding non-bridging oxygen (NBO) and boron coordination number and a few

studies were concerned about understanding the role of rare earth doping in the structure of glasses (Dias *et al.*, 2016).

Luminescence intensity and lifetime of Ln<sup>3+</sup> ions in glasses would depend on the excitation wavelength, environment, symmetry or nature of ligands, i.e., the covalence between rare earth ions and the ligands around them and cross-linking of the f-f transitions (Dias *et al.*, 2016). Thus, borophosphate-based glasses are expected to be a promising host material for RE ions because of its excellent optical properties, low refractive indices, low dispersion and good transparency from the ultraviolet to the near-infrared regions (Yao *et al.*, 2017).

Among the trivalent Lanthanide: Eu<sup>3+</sup> ions are found to be an essential ion to probe deep into the local environment around RE<sup>3+</sup> ions in different matrices and to have the potential applications. This useful information about the local structure around Eu<sup>3+</sup> ions can be obtained quite easily from its f-f transition spectra. In Eu<sup>3+</sup> ions, the ground state <sup>7</sup>f<sub>0</sub> level and the first excited <sup>5</sup>D<sub>0</sub> level are non-degenerate (J=0) under any symmetry and the local environment of Eu<sup>3+</sup> ions depends only on the splitting of <sup>5</sup>D<sub>0</sub> → <sup>7</sup>f<sub>j</sub> (J = 0-4) transitions (Hima Bindu *et al.*, 2016). Among all the rare earth ions, Eu<sup>3+</sup> doped borate materials are prepared as red luminophores for display applications as well as for red LED's due to the host matrix chemical, mechanical durability and broad spectral transparency (Swapna *et al.*, 2014).

Furthermore, the trivalent europium ions are well-established as a spectroscopic probe to get an insight into the structure and nature of chemical bonds. This is mainly because of the simple energy level scheme of Eu<sup>3+</sup> ions and the site-selective nature of intensities between <sup>7</sup>f<sub>j</sub> and <sup>5</sup>D<sub>0</sub> energy levels. Also, the relative variation of emission intensities within the orange-red region due to site-selective nature of hypersensitive and non-hypersensitive <sup>5</sup>D<sub>0</sub> → <sup>7</sup>f<sub>j</sub> and <sup>7</sup>f<sub>2</sub> transitions are of particular interest for device applications (Swapna *et al.*, 2014).

Sm<sup>3+</sup> ions containing glasses are fascinating to study due to strong luminescence and high quantum efficiency of the <sup>4</sup>G<sub>5/2</sub> level. Therefore, glasses doped with Sm<sup>3+</sup> ions have attractive applications as optical devices ( e.g., optical

data storage, colour displays e.t.c.) (Ramteke *et al.*, 2015).  $\text{Sm}^{3+}$  ions give very strong fluorescence in the orange-red spectral region in a variety of lattices, leading to potential high-power lasers, both in compact fiber and planar geometries. But only a few attempts have been made to explore the possibility of using orange-red luminescence of  $\text{Sm}^{3+}$  ions for the development of LED's in the visible spectral region as well as visible optical devices such as visible lasers and fluorescent devices.

Basavapoornima and Jayasankar (2014) noted that the main reason for not conducting several spectral studies on  $\text{Sm}^{3+}$  ions doped in glasses is connected to its  $4f^6$  complicated structure. Many energy levels lying close to each other interpret the absorption spectrum of this ion somewhat tricky for the determination of essential intensity parameters needed in the calculation of various radiative properties which otherwise require a suitable and skilful calculation technique.

$\text{Dy}^{3+}$  ion is another promising rare earth ion for white light applications due to the transitions between  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{15/2}$  and  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$  energy levels corresponding to the dominant emission bands at blue and yellow region respectively. The  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$  emission band is due to the electric dipole transition and is profoundly affected by the ligand field and the  $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{15/2}$  emission is due to magnetic dipole transition (Vijayakumar *et al.*, 2014). The linked between a blue and yellow region in CIE 1931 chromaticity diagram usually passes through the white light region. Furthermore, white light can be produced from the glass materials by adjusting the yellow to blue (Y/B) intensity ratio by varying the glass composition, RE ion concentration and excitation wavelengths (Vijayakumar *et al.*, 2014).

The optical homogeneity of glassy matrices makes available RE ions to exhibit different latent laser transitions. Spectroscopic study of RE ions in glasses suggests information with considering transition probabilities, lifetimes, branching ratios of excited states, which are vital in the design and growth of various electro-optic and optical devices. To understand the quantitative optical phenomena of rare-earth ions in glasses, it is essential to evaluate radiative and non-radiative decay process of related  $4f$  levels. The Judd-Ofelt theory parameterizes the induced electric

dipole transitions. The intensity of induced electric dipole transitions can be described regarding three phenomenological intensity parameters  $\Omega_\lambda$  ( $\lambda = 2, 4, 6$ ). The Judd-Ofelt theory is usually adopted to obtain the radiative transition probabilities including emission by utilizing the data of absorption cross section of several f-f electric dipole lines. The physical and chemical implement of three  $\Omega_\lambda$  parameters ( $\lambda = 2, 4, 6$ ) is becoming more evident by combining information of the local ligand field of doped ions by other spectroscopic technique and give the information about the rare-earth environment in glass such as bond covalency and symmetry. The non-radiative decay rate can be evaluated experimentally by combining the lifetime measurements, which includes contributions of multiphoton decay, energy transfer such as cross relaxation and frequency up conversion properties of rare earth doped glasses (Madhukar Reddy *et al.*, 2015).

Selection of borophosphate glass in this study as a glass former is due to they provide interesting optical and structural properties such as low refractive indices, low dispersion and good transparency from the ultraviolet to the near-infrared regions (Yao *et al.*, 2017). An exciting characteristic of borate glasses is the appearance of variations in its structural properties when different modifier oxides are introduced. The addition of alkali earth metal into the glass structure leads to disruption of the glass network and promotes the formation of non-bridging oxygen groups, which is in contrast to alkaline earth oxides (Balakrishna *et al.*, 2017).

Therefore, due to the increasing demands on the distinct types of visible lasers and light sources. Also, the studies of these rare-earth ions' materials have become an interesting topic in the field of material science because of its essential properties, so, more findings need to be done to determine the efficiency of rare earth in new material.

## **1.2 Problem Statements**

The problem confronting phosphate glasses research is the hygroscopic and volatile nature of most metaphosphate, ultra-phosphate and polyphosphate materials.

Furthermore, phosphate glasses have limitations in their optical performance. However, improving the optical performance for a new efficient luminescent material remained the most challenging task in the industry for solid-state laser applications. Therefore, the incorporation of sulphate ions into the phosphate network improved the rare earth optical performance of borophosphate glasses. Furthermore, lasers are based on specific active materials that needs to satisfy good doping levels. Therefore, an effort on new materials and new dopants concentrations is required to achieve progress in the field. The Judd-Ofelt parameters, radiative properties, optical properties, physical properties, luminescence properties and structural features of RE ( $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$ ) doped calcium sulfoborophosphate glass and barium sulfoborophosphate glass are rarely investigated. Additionally, since there is a lack of report on these glasses, it is of important to study these glasses in order to give more information on the influence of  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions on the glasses.

### **1.3 Objectives of the Research**

The main objective of this research is to develop a new luminescent host material that can exhibit a substantial enhancement of the optical and luminescence properties via doping of calcium sulfoborophosphate glass and barium sulfoborophosphate glass with different concentrations of  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  ions. The specific objectives of this research are:

- (a) To determine and compare the influence of doped  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  of different concentration on calcium sulfoborophosphate glass and barium sulfoborophosphate glass in terms of their structural features and regarding their luminescence characteristics enhancements.
- (b) To analyze and compare the impact of emission and absorption data between calcium sulfoborophosphate glass and barium sulfoborophosphate glass doped varies concentrations of  $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$  in terms of radiative properties using Judd-Ofelt analysis.



## 1.4 Scopes of the Research

- (a) In this study, the samples of undoped calcium sulfoborophosphate glass and barium sulfoborophosphate glass with the chemical composition of  $x\text{CaSO}_4-30\text{B}_2\text{O}_3-(70-x)\text{P}_2\text{O}_5$  with  $15 \leq x \leq 35$  mol% and  $y\text{BaSO}_4-30\text{B}_2\text{O}_3-(70-y)\text{P}_2\text{O}_5$  with  $15 \leq y \leq 35$  mol% were prepared by conventional melt quenching method. The series of glass samples doped with rare earth (RE =  $\text{Dy}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$  and  $\text{Eu}_2\text{O}_3$ ) with the chemical compositions of  $25\text{CaSO}_4-30\text{B}_2\text{O}_3-(45-x)\text{P}_2\text{O}_5 - x\text{RE}$  with  $0.1 \leq x \leq 1.0$  mol% and  $25\text{BaSO}_4-30\text{B}_2\text{O}_3-(45-y)\text{P}_2\text{O}_5 - y\text{RE}$  with  $0.1 \leq y \leq 1.0$  mol% were also been prepared by conventional melt quenching method.
- (b) The amorphous nature of the glass sample was ascertained by X-ray Diffraction (XRD) and thermal stability of the undoped prepared glass samples were determined using Differential Thermal Analyzer (DTA).
- (c) The structural features of the host materials are investigated using Infrared and Raman spectroscopic techniques and excitation, emission and absorption of (RE= $\text{Dy}^{3+}$ ,  $\text{Sm}^{3+}$  and  $\text{Eu}^{3+}$ ) doped calcium sulfoborophosphate glass and barium sulfoborophosphate glass were determined using photoluminescence and ultraviolet visible spectroscopy respectively.
- (d) The optical absorption parameters like optical band gap, refractive index, electronic polarizability and Urbach energy are measured from the data of UV-Vis spectroscopy.
- (e) The radiative parameters on the luminescence properties of (RE= $\text{Dy}_2\text{O}_3$ ,  $\text{Sm}_2\text{O}_3$  and  $\text{Eu}_2\text{O}_3$ ) doped calcium sulfoborophosphate glass and barium sulfoborophosphate glass were determined using Judd-Ofelt theory.

## **1.5 Significance of the Research**

Glasses are exceptionally significant optical materials. The rapid development of laser research has led to get much attention about theoretical treatment of RE ions in glass compare to other luminescence center. Conducting details and comprehensive research on the proposed glass host samples and the rare earth ions as dopant would contribute in perspective and investigative studies regarding their structural and optical properties of the glass samples. The Judd-Ofelt results provides information on the absorption and emission of the newly luminescence host. The new material can, therefore, provide a baseline data for future research and can be used as an alternative material for solid-state laser applications such as colour displays, optical fibre and amplifiers.

## **1.6 Outlines of Thesis**

This thesis is classified into five different chapters. Chapter 1 describes the background of the research, problem statement, objective of the research, outlines of the thesis and significance of the research aimed to highlight the introduction aspect of the research work. In Chapter 2, an extensive literature review regarding the host structure used in the current study were made. The review includes description of structural features, luminescence properties as well as Judd-Ofelt and radiative parameters of other host materials. Chapter 3 describes the experimental procedures which encompass the methodology in preparing the glass samples with the analytical techniques used. Furthermore, detailed information about the types of spectroscopic methods used and the working principle of X-ray diffraction (XRD), FTIR and Raman spectroscopy, luminescence and UV-Visible-NIR spectrometer. Chapter 4 presents the results and discussion regarding the different characterization, measurements and evaluations of the prepared samples. Chapter 5 presents the conclusions and recommendations for future work based on the research vacuums acknowledge during this study.

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