OPTICAL SPECTRA AND JUDD-OFELT ANALYSIS OF DYSPROSIUM AND SAMARIUM IMPURITIES ACTIVATED STRONTIUM MAGNESIUM BORATE GLASSES

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DEDICATION

This thesis is dedicated to God Almighty, the Ichojas' and the Ebiegas' for His divine mercy and favour and their unflinching support. To my beloved angel wife, Mrs. Justina Enuwa Ichoja for all the pains, loneliness, understanding, prayers, endurance, encouragement, steadfastness and carrying the burden of raising the children alone in the course of this academic pursuits

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ABSTRACT

New types of strontium magnesium borate glasses namely Dy^{3+}– and Sm^{3+}– singly activated and Dy^{3+} + Sm^{3+}– co-doped with the nominal compositions of 20SrO – 10MgO – (70 – x) B_2O_3 – $x Dy_2O_3$ ($0.1 \leq x \leq 0.8$ mol%); 20SrO – 10MgO – (70 – y) B_2O_3 – $y Sm_2O_3$ ($0.5 \leq y \leq 2.5$ mol%) and 20SrO – 10MgO – (70 – z) B_2O_3 – $0.7Dy_2O_3$ – $z Sm_2O_3$ ($0.2 \leq z \leq 1.0$ mol%) were prepared by the melt-quenching method. The structural properties of the quenched glass samples were investigated using X-Ray diffraction (XRD), Fourier Transform Infrared (FTIR), Energy Dispersive X-ray (EDX) analyses and Field Emission Scanning Electron Microscope (FESEM) respectively. The Ultraviolet-Visible-near-IR Spectroscopy (UV-Vis-NIR) spectra of the glasses exhibited characteristic absorption transitions of Dy^{3+} and Sm^{3+} respectively. The nephelauxetic effect on the absorption transitions has been used to elucidate the bonding nature of the doping ions with the surrounding ligands revealing the predominant ionic nature. The photoluminescence (PL) emission spectra analysis of the Dy^{3+}-doped glasses revealed three peaks including blue at 483 nm, yellow at 575 nm and red at 664 nm attributed to 4f-4f transitions. The glass made with Dy_2O_3 content of 0.7 mol% revealed optimum PL intensity and this composition was chosen for co-doping with various Sm_2O_3 contents. The PL spectra for Dy^{3+} / Sm^{3+} co-doped glass system exhibited five emission bands due to the $^4F_{9/2}\rightarrow{}^6H_{15/2}$ (Dy^{3+}), $^4F_{9/2}\rightarrow{}^6H_{13/2}$ (Dy^{3+}), $^4G_{5/2}\rightarrow{}^6H_{7/2}$ (Sm^{3+}), $^4G_{5/2}\rightarrow{}^6H_{9/2}$ (Sm^{3+}) and $^4G_{5/2}\rightarrow{}^6H_{11/2}$ (Sm^{3+}) transitions in Dy^{3+} and Sm^{3+}, respectively. The luminescence spectra of Dy^{3+} / Sm^{3+} co-doped glasses revealed that the successive addition of Sm^{3+} to Dy^{3+}-doped strontium magnesium borate glasses has enhanced the emission intensity of Dy^{3+} with decreased emission intensity of Sm^{3+} at 0.4 mol% in Dy^{3+} + Sm^{3+} co-doped glasses due to strong migration of Sm^{3+} excitation energy to Dy^{3+}. From the optical absorption measurements and based on Judd–Ofelt theory (J–O), the influence of Dy^{3+} and Sm^{3+} on the three J–O intensity parameters (Ω_2, Ω_4, Ω_6) were evaluated. The achieved values of intensity parameters were used to calculate the J–O radiative properties including the branching ratio, stimulated emission cross–section, optical bandwidth and optical gain. The achieved high values of the branching ratio (> 60% and 74%) and stimulated emission cross-section ($> 10 \times 10^{-22}$ cm^2) recorded at $^4F_{9/2}\rightarrow{}^6H_{13/2}$ and $^4G_{5/2}\rightarrow{}^6H_{7/2}$ electronic transitions showed an excellent lasing and optical energy harnessing potentials of the proposed glass compositions. The observed increase in optical gain envisages an effective lasing efficiency and optical amplification power of the glass samples for low threshold amplifier design and solid-state laser development.

ABSTRAK

Kaca strontium magnesium borat baharu iaitu Dy^{3+} – dan Sm^{3+} – teraktif tunggal dan ko-dop $Dy^{3+} + Sm^{3+}$ – dengan komposisi nominal 20SrO – 10MgO – (70 – x) B_2O_3 – xDy_2O_3 ($0.1 \leq x \leq 0.8$ mol%); 20SrO – 10MgO – (70 – y) B_2O_3 – ySm_2O_3 ($0.5 \leq y \leq 2.5$ mol%) dan 20SrO – 10MgO – (70 – z) B_2O_3 – $0.7Dy_2O_3$ – zSm_2O_3 ($0.2 \leq z \leq 1.0$ mol%) telah disediakan dengan kaedah pelindap-kejutan leburan. Sifat struktur sampel kaca yang dilindap kejut masing-masing disiasat menggunakan Pembelauan Sinar-X (XRD), Transformasi Fourier Inframerah (FTIR), Tenaga Sinar-X Terserak (EDX) dan Mikroskop Pengimbas Elektron Pancaran Medan (FESEM). Spektrum Spektroskopi Ultralembayung-Cahaya Nampak-Hampir-Inframerah (UV-Vis-NIR) kaca masing-masing menunjukkan ciri peralihan penyerapan Dy^{3+} dan Sm^{3+}. Kesan nefelauksetik pada peralihan penyerapan telah digunakan untuk menjelaskan sifat ikatan ion doping dengan ligan sekitarnya yang mendedahkan sifat ionik yang predominan. Analisis spektrum pancaran kefotopendarcahayaan (PL) dari kaca Dy^{3+} menunjukkan tiga puncak termasuk biru pada 483 nm, kuning pada 575 nm dan merah pada 664 nm yang dikaitkan dengan peralihan 4f-4f. Kaca yang dibuat dengan kandungan 0.7 mol% Dy_2O_3 menunjukkan keamatan PL yang optimum dan komposisi ini dipilih untuk ko-doping dengan kandungan Sm_2O_3 yang berbeza. Spektrum PL untuk sistem kaca ko-dop menunjukkan lima jalur pancaran disebabkan oleh peralihan $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (Dy^{3+}), $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (Dy^{3+}), $^4G_{5/2} \rightarrow {}^6H_{7/2}$ (Sm^{3+}), $^4G_{5/2} \rightarrow {}^6H_{9/2}$ (Sm^{3+}) dan $^4G_{5/2} \rightarrow {}^6H_{11/2}$ (Sm^{3+}) yang masing-masing dalam Dy^{3+} dan Sm^{3+}. Spektrum PL kaca ko-dop Dy^{3+} / Sm^{3+} mendedahkan bahawa penambahan Sm^{3+} secara berturutan pada kaca strontium magnesium borat terdop-Dy^{3+} telah meningkatkan keamatan pancaran Dy^{3+} dengan penurunan keamatan pancaran Sm^{3+} pada 0.4 mol% kaca ko-dop $Dy^{3+} + Sm^{3+}$ disebabkan oleh migrasi tenaga pengujaan Sm^{3+} ke Dy^{3+}. Dari pengukuran penyerapan optik dan berdasarkan teori Judd-Ofelt (J–O), pengaruh ion Dy^{3+} dan Sm^{3+} pada tiga parameter keamatan J–O (Ω_2, Ω_4, Ω_6) dinilai. Nilai parameter keamatan yang dicapai digunakan untuk mengira sifat menyinar J–O termasuk nisbah mencabang, keratan rentas pancaran dirangsang, lebar jalur optik dan gandaan optik. Nisbah mencabang (> 60% dan 74%) dan keratan rentas pancaran dirangsang ($> 10 \times 10^{-22}$ cm^2) yang tinggi dicatatkan pada peralihan elektronik $^4F_{9/2} \rightarrow {}^6H_{13/2}$ dan $^4G_{5/2} \rightarrow {}^6H_{7/2}$ menunjukkan keupayaan mengabah tenaga las dan optik bagi komposisi kaca yang dicadangkan. Peningkatan gandaan optik yang dicerap membayangkan kecekapan las dan kuasa penguat optik sampel kaca yang berkesan untuk reka bentuk penguat ambang rendah dan pengembangan laser keadaan pepejal.

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LIST OF ABBREVIATIONS

B_2O_3	Borate
NBO	Non–Bridging Oxygen
MgO	Magnesium Oxide
SrO	Strontium Oxide
Dy_2O_3	Dysprosium Oxide
Sm_2O_3	Samarium Oxide
Dy^{3+}	Dysprosium Ion
Sm^{3+}	Samarium Ion
f_{exp}	Experimental oscillator strength
f_{cal}	Calculated oscillator strength
IR	Infrared
S_{ed}	Electric dipole field strength
S_{md}	Magnetic dipole field strength
A_{rad}	Radiative transition probability
β_R	Branching ratio
τ_{rad}	Radiative lifetime
λ_p	Emission band position
FTIR	Fourier Transform Infrared
KBr	Potassium Bromide
XRD	X-Ray Diffraction
UV	Ultraviolet
RE	Rare Earth
PL	Photoluminescence
EDX	Electron Dispersive X-ray
FESEM	Field Emission Scanning Electron Microscope

LIST OF SYMBOLS

h	Planck's constant
°C	Degree Celsius
ν	Frequency
c	Speed of light
$\alpha(\nu)$	Absorption coefficient
β	Nephelauxetic ratio
Ω_2	Judd–Ofelt parameter
Ω_4	Judd–Ofelt Parameter
Ω_6	Judd–Ofelt parameter
δ	Bonding parameters
δ_{rms}	Root Means Square Deviation
J	Total angular momentum
θ	The diffracted angle of the X–Ray beam
n	Refractive index
λ	Wavelength
R_M	Molar Refractivity
R	Reflection Loss
α_M	Molar Polarizability
d	Thickness
$\|\|U\|\|$	Reduced Matrix Element
N_A	Avogadro's Number
E_{opt}	Optical Band Gap
M_{av}	Molecular Weight Average
M	Metallization Criterion
F	Field Strength
r_i	Inter-Nuclear Distance
r_p	Polaron Radius
N	Ion Concentration

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CHAPTER 1

INTRODUCTION

1.1 Research Background

Since ancient times, different kinds of light sources ranging from burnfire, touch, candle, incandescent and fluorescent lamps, lasers and light-emitting diodes (LEDs) have been used for different purposes. Among all the existing light sources offered for human comfort, lasers remain attractive due to their high emission intensity and precise light beam (Kaewnuam et al., 2017). The ease of fabricating glasses into a variety of shapes and sizes, low cost of production, high optical transparency and ability to accommodate high concentration of dopants without destroying their network structures are the reasons for the choice of glass-based materials over the expensive crystalline materials for laser development (Pye et al., 1972; Rajaramakrishna et al., 2014). By definition, the glass may be defined as an amorphous substance (structureless solid or without long-range order). According to the American Society for Testing and Materials (ASTM), this lustrous, transparent and vitreous substance (Glass) can be defined as an inorganic product of fusion that is cooled to a rigid condition without crystallization. Meanwhile, Shelby defines glass as a non-crystalline, non-equilibrium and condensed state of matter that exhibits glass transition. Glass is also considered as an amorphous solid completely lacking in long range order, periodic atomic structure and exhibiting a region of glass transformation behaviour (Shelby, 2005). Glass can be developed from different materials. The formation of any glass system from either polymer, alloys of metals, aqueous solution, molecular liquids and ionic melts required the appropriate composition of different components to attract specific features (Sokolov et al., 2009). The glass network former; a system of the highly cross-linked chemical bond, network modifier; that distorts the structural network and intermediate are the major constituents in glass formation. Glass formers are regarded as the backbone of glass because altering the element or compound will automatically change the properties of the final materials while modifiers and intermediate are added to give special property like stability to the glass. Among the various techniques available for synthesizing glass, the cheapest, time-saving and most convenient means of producing glass is the melt quenching technique (Bansal & Doremus, 2013; Uhlmann & Yinnon, 1983). This method involves rapid cooling of the melt to a low temperature in the order of 10^7 degrees per second (fast cooling) to avoid crystallization and is used in this work.

Glass forming materials are increasing in number. Among the various glass formers (Borate, Silicate, Tellurite, Phosphate, Germanium) studied so far, borate-based glasses are well known to possess excellent properties such as low melting point, dominant thermal and mechanical strengths, chemical durability, higher bond strengths and solubility to many rare-earth ions (Pawar et al., 2016). Doping ions either rare-earth or transition metal ions are added to the glass matrix primarily to enhance its luminescence efficiency for technological applications (Elliot, 1984). But the technical know-how in the improvement of optical and luminescence properties of dopants activated inorganic glasses is a daunting task currently confronting the material science research community. The enhancements in the absorption, emission or luminescence properties of rare earth (RE) doped glasses may be achieved through variation of glass composition alongside their selected modifiers and doping ions. In glass technology, low phonon energy-based glasses are of great importance for infrared to visible up-conversion lasers. The minimization of non-radiative losses in glasses is critical to enhancing the glass luminescence efficiency. Consequently, the selection of the host matrix where low phonon energy is maximized is a necessary pre-condition for improving the luminescence quality of the glass network for maximum output. The existence of energy losses due to non-radiative relaxation (multi-phonon energies) differ from one glass host to another, in phosphate glass network (P–O) 1200–1350 cm^{-1} , borate matrix (B–O) 1340–1480 cm^{-1}, germanium host (Ge–O) 800–975 cm^{-1}, silicate glass (Si–O) 1000–1200 cm^{-1} and tellurite lattice (Te–O) 600–750 cm^{-1} (Abdel-Baki & El-Diasty, 2016; Lakshminarayana et al., 2017; Pal et al., 2013; Shen et al., 2015; Zmojda et al., 2014). The choice of borate as the network former in this research is owed to their intriguing optical and basic properties that are relevant for cutting edge innovations. Generally, the network structure of pure glass formers is so tight due to the existence of bridging oxygens (BOs) which makes the incorporation of a small quantity of doping ions into the glass composition very difficult. However, the inclusion modifier cations such as BaO, CaO, SrO, K_2O, Li_2O, MgO, and Bi_2O are intended to disrupt the glass structural networks through the formation of non-bridging oxygens (NBOs) to reduce the hygroscopic effect of the glass host and accommodate rare-earth ions. The modifiers are expected to suppress the phonon energy of borate via systemic transformation of trigonal sp^2 to a more optically stable tetrahedra sp^3 and increase the energy-releasing potency which is optically beneficial to the radiative energy transfer mechanism. It is equally anticipated that the incorporated modifiers will improve the glass preparation conditions, creates non-bridging oxygen, weakens the bond strength, lowers the glass viscosity level and increases their mechanical stability (Alajerami et al., 2013). Interestingly, borate based-glasses have demonstrated their potentials for lasing and nonlinear optical applications due to their variety of compositional possibilities and a large amount of REIs solubilities in the host matrix (Krishnaiah et al., 2013). The B_2O_3 glasses comprised of symmetric stretching vibration of sp^2 planar BO_3 trigonal, sp^3 tetrahedra BO_4 units and B–O–B bonds (Maheshvaran & Marimuthu, 2011) which is required for the enhancement of the glass rigidity. The high emission efficiency of REs in borate glasses is key to the development of more environmentally friendly, long lifetime, low energy consumption, high brightness and low-temperature performance solid-state based lighting materials which could serve as a possible replacement for conventional incandescent lamps (Arunkumar et al., 2015; Dutta et al., 2013; Krishnaiah et al., 2013; Murthy et al., 2010; Pawar et al., 2016). Again borate glasses possess an exceptional quality of producing interesting structures of the form B_xO_y where the BO_3 and BO_4 can form unique structural units such as di-borate, tri-borate, tetra-borate, Penta-borate, boroxyl ring (Pawar et al., 2017; Yao et al., 2016). Despite the aforementioned numerous advantages of borate glasses, the practical applications of these glasses are limited due to hygroscopic effects, luminescence attenuation and high phonon energy that inhibits the glass emission efficiency (Rao et al., 2017).

Glasses doped with lanthanide ions (Ln^{3+}) are often considered potential luminescent materials not only because of the occurrence of sharp fluorescence in ultraviolet, visible and infrared regions but as a result of shielding effects of the outer 5s and 5p orbitals on the 4f electrons. The emission efficiencies of rare-earth-doped glasses of 4f-4f and 4f-5d electron transitions portray the glass as good candidates for various technological applications such as optoelectronics, solid-state lasers, solar concentrators, optical fibre for fluorescent colour displays and communications (Liang et al., 2007; Nishiura et al., 2011). The current research focuses on Ln^{3+} embedded glasses that are not just constrained to infrared optical devices alone but as a result of increasing curiosity in visible optical systems (Rüter & Bauhofer, 1996; Tsuboi, 2004; Turnbull et al., 1996). The rare earth energy levels are the deciding factors in the lasing power of lanthanide ions in solid-state materials and are greatly affected by the host lattice. The radiative characteristics of RE ions-doped glasses are equally remarkably dependent on the glass host. Therefore, the modification of the host matrix through an appropriate choice of glass formers, network modifiers or dopants has potential effects on the radiative properties of the glass system (Murugesan & Bergman, 2007).

Among the existing rare-earth ions, Dy^{3+} and Sm^{3+} have continuously demonstrated their potential relevance in advanced technology due to their strong emission intensities in the visible region. Dy^{3+} can easily be excited because of the effective shielding of the 4f-4f electrons by the 5p and 5s shells. The emission spectrum of Dy^{3+} containing two strong bands attributed to the $^4F_{9/2} \rightarrow H_{15/2}$ (blue) and $^4F_{9/2} \rightarrow H_{13/2}$ (yellow) transitions and another weak band located at $^4F_{9/2} \rightarrow H_{11/2}$ (red). Hence, the study of Dy^{3+} activated glasses is essential for white light generation via an appropriate combination of these band intensities and primary-coloured luminescent materials. Sm^{3+} ion, on the other hand, has shown its usefulness as a structural probe emitting orange emissions for colour displays, optical devices, data storage systems (Ramteke et al., 2016) due to strong orange colour in the visible region emanating from the $^4G_{5/2} \rightarrow {}^6H_{7/2}$ and $^4G_{5/2} \rightarrow {}^6H_{9/2}$ (Ramteke et al., 2016). Also, when Sm^{3+} is used as a co-dopant together with other rare earth (Eu^{3+}, Er^{3+}, Pr^{3+} etc) it serves as a donor (Sensitizer) by transferring its energy to other activators called acceptors thereby improving their luminescence qualities (Naresh et al., 2015). Given this, more scientific research is required for exploring the reddish-orange luminescence of Sm^{3+} ions to potentially utilized them for the advancement of LEDs' invisible lasers and fluorescent devices. According to (Basavapoornima & Jayasankar, 2014), the complex energy structure of Sm^{3+} where many energy levels are closely packed to each other is responsible for the limited studies on Sm^{3+} doped glasses. The existence of complicated energy levels makes the interpretation of absorption spectra tricky and problematic for the determination of essential intensity parameters required for the calculation of radiative properties in Judd–Ofelt theory.

According to the literature, many scientific works have been carried out and reported on borate glasses. But these extensive studies by several authors centres primarily on the physical and optical properties of different dopants activated in glasses but without co-doping technique. Dawaud et al. (2014) studied the optical and structural properties of lithium sodium borate doped-Sm^{3+}. Mhareb et al. (2014) investigated the physical and optical properties of Li_2O-MgO-B_2O_3 doped-Dy^{3+}. Mhareb et al. (2014) reported the impact of Nd^{3+}on physical and optical properties of lithium magnesium borate glass. Maheshvaran et al. (2013) carried out "structural and optical studies on Eu^{3+}-doped boro-tellurite glasses". Azizan et al. (2014) investigated the physical and optical properties of Li_2O-K_2O-B_2O_3 glasses. Alajerami et al. (2012) investigated the optical properties of Li_2O-MgO-B_2O_3-doped dysprosium and samarium ions. "Optical absorption and photoluminescence of Dy^{3+}-doped zinc alumino bismuth borate glass for lasing materials and white LEDs" has been studied by (Swapna et al., 2013). Sreedhar et al. (2013) investigated the optical properties of zinc fluorophosphate glass doped-Dy^{3+}. Kumar et al. (2017) studied structural optical and thermoluminescence properties of Dy^{3+}-doped sodium strontium borate glasses. Balakrishna et al. (2012) studied "structural and photoluminescence properties of Dy^{3+}-doped different modifier oxide-based lithium borate" glass. Ali (2009) investigated the optical properties of Sm^{3+} doped CaF_2 bismuth borate glasses. Meanwhile, the Judd–Ofelt investigation of the radiative properties of the current glass composition via the co-doping method is grossly lacking. Luminescence quenching has had a significant impact on the luminescence properties of materials and this has aroused the desire for vigorous scientific activities on the quenching effect of oxide-based glasses to enhance the commercialization of improved optical materials for technological applications. According to the literature, this drawback can be addressed through the combination of two dopants of different identities (co-doping method) or the introduction of nanoparticles for enhancement of their optical and luminescence values (Reza Dousti et al., 2013).

1.2 Problem Statement

The present era of technological revolution demands the synthesis of new optical materials through probably a simple technique but with remarkable features and versatile applications. Previous studies of rare-earth-doped glass have been focussed majorly on the characterization of singly activated rare-earth ions in various host glass systems. To pursue this area, a well-designed new glass composition should be developed and their physical, structural, optical and luminescence properties investigated.

The use of a higher concentration of REIs in glasses to increase luminescence efficacy is yet to yield any positive outcome. This is because emission intensity easily gets quenched at higher concentrations owed to losses which are stimulated by the de-excitation of different energy levels. Meanwhile, the co-activation mechanism where two non-identical rare-earth ions (REIs) are doped in the host glass matrix has been reported to be one of the successful ways to improving the absorption cross-section or luminescence intensity of the glass network due to energy migration from the sensitizer doping ion to the acceptor ions and local field effects. The enhancement of the luminescence intensity and the avoidance of the luminescence quenching effect of optical glasses with sufficiently high luminescent yield is still a great concern. The development and successful use of rare earth compounds as solid-state laser materials and their effectiveness in modern technology as optical devices have stimulated interest in rare-earth optical spectroscopy. Because of this, several investigations are ongoing to optimize new glasses matrices containing different dopant ions. Optimization of new or improved optical glass qualities of rare-earth ions has been characterized by absorption and emission transition probabilities which are influenced by the ligand field of the surrounding rare-earth ions. To identify new optical devices for specific functions or devices with enhanced performance, active work is being carried out by selecting appropriate new hosts doped with different rare-earth ions. At the moment, the most study is centred on the individual doping of Dy^{3+}, Er^{3+}, Tm^{3+}, Pr^{3+}, Sm^{3+}, Nd^{3+} and Eu^{3+} with various glass hosts. However, the co-doping mechanism of Dy^{3+} and Sm^{3+} on strontium magnesium borate to study their influence on PL intensity, absorption features, structural and radiative properties is grossly inadequate. In sequence, the co-doping of Dy^{3+} with optimum concentration into borate glass host containing individual rare earth especially Sm^{3+} should be critically emphasized.

Furthermore, the Judd-Ofelt intensity parameters and radiative properties of Dy^{3+} / Sm^{3+} co-doped strontium magnesium borate glasses are rarely investigated since there is little literature available on Dy^{3+} / Sm^{3+} co-doped glass systems. The investigation on these glass systems would provide vital information on the influence of the REIs on the physical, structural, optical properties as well as the radiative characteristics of the studied glass compositions to assessing the lasing potential of the materials for advanced technological applications.

1.3 Objectives of the Research

The general objective of this study is to develop new glass materials called strontium magnesium borate glasses and probing their physical, optical, structural and luminescence characteristics dependence on rare-earth ions. But, the specific objectives of the research include:

(i) To synthesis three series of glass samples containing Dy^{3+}, Sm^{3+} and Dy^{3+} / Sm^{3+} ions with the following chemical formula by using melt quenching technique;

   (a) Series I: $20SrO - 10MgO - (70 - x) B_2O_3 - xDy_2O_3$ ($0.1 \leq x \leq 0.8$ mol%);
   (b) Series II: $20SrO - 10MgO - (70 - y) B_2O_3 - ySm_2O_3$ ($0.5 \leq y \leq 2.5$ mol%)
   (c) Series III: $20SrO - 10MgO - (70 - z) B_2O_3 - 0.7Dy_2O_3 - zSm_2O_3$ ($0.2 \leq z \leq 1.0$ mol%)

(ii) To characterize the physical, structural and optical features of dysprosium (Dy^{3+}), samarium (Sm^{3+}) and dysprosium/samarium (Dy^{3+} / Sm^{3+}) co-doped strontium magnesium borate glasses

(iii) To calculate the Judd–Ofelt intensity and radiative parameters of the prepared glasses as a function of individually doped Dy^{3+}-, Sm^{3+} and Dy^{3+} + Sm^{3+} co-doping contents to assessing the lasing potency of the as-prepared glass samples.

1.4 Scope of the Research

In this research, three series of various concentrations of rare earth ions activated strontium magnesium borate glass networks with chemical compositions $20SrO - 10MgO - (70 - x)B_2O_3 - xDy_2O_3$ ($0.1 \leq x \leq 0.8$ mol%); $20SrO - 10MgO - (70 - y)B_2O_3 - ySm_2O_3$ ($0.5 \leq y \leq 2.5$ mol%) and $20SrO - 10MgO - (70 - z)B_2O_3 - 0.7Dy_2O_3 - zSm_2O_3$ ($0.2 \leq z \leq 1.0$ mol%) were fabricated using standard melt-quenching method and characterized by means of different analytical techniques.

For the determination of physical properties of the as-quenched glasses, firstly, Archimedes' principle was employed to evaluate the physical properties such as density, molar volume, ion concentration, molar refractivity, refractive index, optical dielectric constant, polaron radius, reflection loss, dielectric constant, inter-nuclear distance, metallization and molar polarizability of glass samples. The elemental composition of the glass network was established by using Energy Dispersive X-ray (EDX) spectroscopy. The glass surface morphological structure was analysed by Field Emission Scanning Electron Microscopy (FESEM). But, for the characterization of the as-prepared glass samples, different kinds of analytical techniques were performed. X-ray diffraction (XRD) technique was used to check the non-crystalline nature of the as-prepared glasses used in this research. Photoluminescence (PL) analysis was carried out on glass samples using a photoluminescence spectrophotometer to determine the glass luminescence features. The optical absorption of the prepared samples was investigated using a UV-Vis-NIR spectrophotometer. Fourier transform infrared (FTIR) was used as a structural probe to determine glass local structures. Energy level diagrams and cross-relaxation channels were provided to observe the luminescence enhancement of the glass system. To improve the optical properties of our glass structures for higher luminescence efficiency, the co-doping technique was used to assess the influence of co-dopants on luminescence quenching and lasing potency of the glasses. Furthermore, based on the Foster-Dexter theory, the possibility of energy transfer is strongly dependent on the overlap of absorption spectra and emission spectra owed to acceptor and donor ions relations and the separating distance between them. Therefore, the occurrence of migration of excitation energy in REs doped glass is only made possible due to narrowed and well shielded electronic structure of the 4f-4f configuration by 5s and 5p electronic orbitals which helps in the enhancement of the optical quality of the glass systems which guaranteed their participation in solid-state laser applications.

Finally, the theoretical calculations of Judd–Ofelt theory were explored to determine the J–O intensity parameters (Ω_2, Ω_4, Ω_6), radiative properties and lasing potential of the glass samples.

1.5 Significance of the Research

The development of glassy materials is receiving great attention now at both commercial and technological levels due to their potential applications in various fields such as solid-state lasers, optical fibres, telecommunication, modern lightening technology, and photovoltaics. Glass incorporated with different rare-earth ions (REIs) exhibits huge advantages like standardized light-emitting capacity, modest manufacturing measures, low cost of production and good mechanical strength. REIs doped in various host matrices (borate) have demonstrated some interesting properties considered advantageous for advanced technology. The enclosure of modifiers in the present glass composition is expected to minimize the hygroscopic nature of borate and increase the mechanical resistivity of the glass against atmospheric hydrolysis via the formation of metal cation networks. The reddish-orange colour exhibited by Sm^{3+} ion originated from $^4G_{5/2} \rightarrow {}^6H_{7/2}$, and $^4G_{5/2} \rightarrow {}^6H_{9/2}$ emission transitions could be a good source of colour display. The high values of branching ratio and stimulated emission cross-section of Sm^{3+} emission transitions are useful conditions for low threshold, high gain optical fibres, and amplifiers development. Dy^{3+}–doped borate glass former is also characterized by two remarkable emission spectral $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (blue) and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (yellow) in the visible region. These prominent emission transitions can be used for the development of photonic devices like white light-emitting diodes (W-LEDs) and light amplification by stimulated emission radiation (LASER) which could serve as a replacement for fluorescent lamps and incandescent bulbs.

The W-LEDs are commonly known for high brightness, good reliability, small size, safety, low power consumption, and environmentally friendly. These luminescence features can improve benefits with regards to the environment and reducing the consumption of global energy. This study will equally provide fundamental knowledge and basic information required on optical features of the studied glass for potential applications in advanced technology. The information obtained from this research can be used for further studies and the development of enhanced luminescence host materials for solid-state lasers. The results from Judd–Ofelt analysis using emission and absorption data will provide basic but vital information regarding the radiative transition parameters, radiative branching ratio, and radiative lifetime of luminescence characteristics that portray the glass system as a potential candidate for advanced technological applications.

Due to quenching effects, the need to co-dope different uncommon REIs or transition metals in any glass network structure emerges when REIs or transition metals ions in the glass compositions display weak emission because of its incapacity in absorbing excitation energy or reduction in emission intensity due to concentration quenching arising from multi-phonon relaxation or cross-relaxations. In this way to defeat such circumstances, the co-doping mechanism where dissimilar REIs or transition metals are dually added to already existing fluorescence REs for improving its luminescence process through a process of sensitization is a welcome development. The achieved optimization mechanism for an effective co-doping process in the under-investigated glass samples is optically beneficial for advanced technological applications and the determination of the lasing potential of the studied materials.

1.6 Thesis Outline

This thesis is shared into five chapters with each chapter containing the following description. Chapter 1 contains the research background alongside the problem statement, objectives of the research, scope, and significance of the research.

Chapter 2 provides a review of relevant literature. This is segmented into five different parts. The first part includes scientific insights into the glass mechanism such as Glass formation theory and Dynamical features of borate-based glasses. The second part is dedicated to modifiers (Strontium and Magnesium) inclusion in borate containing glasses and dopants (Dysprosium and Samarium) effects on borate and borate related glasses. The third part is allocated to structural (X-ray diffraction and Fourier transform Infrared) and physical properties (Density, Molar volume, Ion concentration, Polaron radius, Inter-nuclear distance, and Field strengths). The Ultraviolet Near-Infrared Region (Glass optical properties, Absorption spectra characteristics, Direct and Indirect allowed optical band gaps), Urbach energy and Judd–Ofelt theory are discussed in part four while in part five the discussion is cantered on photoluminescence (Emission and Excitation) analysis of the glass samples as well as luminescence theory in non-crystalline solids.

Chapter 3 presents the various experimental procedures employed in the present study. This category of demonstration includes the method of sample preparation and spectroscopic techniques used in the current work. The spectroscopic instrument characterizations are X-ray Diffraction (XRD), Fourier transforms infrared (FTIR) techniques, Energy Dispersive X-ray (EDX) and Field Emission Scanning Electron Microscope (FESEM). Optical absorption, as well as information on emission and excitation, were provided via UV-Vis-NIR and Photoluminescence (PL) spectroscopy respectively.

Chapter 4 contains the results and discussions of the experimental procedure described in chapter 3 as well as tables and figures as used in the present study. These include outputs from physical and optical measurements and characterization techniques, EDX and FESEM outputs, luminescence properties, optical analysis, Judd–Ofelt evaluation of radiative parameters of separately doped Dy^{3+}, Sm^{3+}, and Dy^{3+} + Sm^{3+} co-doped strontium magnesium borate glasses.

Chapter 5 is dedicated to conclusions and recommendations for future works.

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Appendix A Batch calculation of the compound used in the glass matrix

The sample calculation for 20SrO – 10MgO – (70 – y) B_2O_3 – $y$$Sm_2O_3$; for $0.5 \leq y \leq 2.5$ mol%

SrO	MgO	(70 – y) B_2O_3	$y$$Sm_2O_3$
20	10	69.5	0.5
20	10	69.0	1.0
20	10	68.5	1.5
20	10	68.0	2.0
20	10	67.5	2.5

The molecular weight of the compound used

Molar mass of SrO = 103.62 g mol^{-1}

Molar mass of MgO = 40.3044 g mol^{-1}

Molar mass of B_2O_3 = 69.63 g mol^{-1}

Molar mass of Sm_2O_3 = 348.72 g mol^{-1}

Weight system of 20SrO – 10MgO – (70 – y) B_2O_3 – $y$$Sm_2O_3$

$$\left(\frac{20}{100}\times 103.62\right)+\left(\frac{10}{100}\times 40.3044\right)+\left(\frac{69.5}{100}\times 69.63\right)+\left(\frac{0.5}{100}\times 348.72\right)$$

$$= 20.724+4.030+48.393+1.744$$

$$= 74.891\,g$$

To produce 25g of 20SrO – 10MgO –(70 – y) B_2O_3 – 0.5Sm_2O_3

$$SrO = \frac{20.724}{74.891}\times 25g$$

$$SrO = 6.92\,g$$

To produce 25g of MgO

$$MgO = \frac{4.030}{74.891}\times 25g$$

$$MgO = 1.35\,g$$

To produce 25g of B_2O_3

$$B_2O_3 = \frac{48.393}{74.891}\times 25g$$