



# STRUCTURAL, INTENSITY PARAMETERS AND LUMINESCENCE ANALYSIS OF rGO-/ CNTs-COATED TELLURITE **GLASS DOPED WITH ERBIUM** NANOPARTICLES



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# UNIVERSITI PENDIDIKAN SULTAN IDRIS

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## THESIS SUBMITTED IN FULFILLMENT OF THE REQUIREMENT FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

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### ABSTRACT

This study aimed to improve the optical properties of reduced graphene oxide- and carbon nanotubes-coated tellurite glass doped with erbium oxide nanoparticles denoted as ZBTEr(NPs)-rGO and ZBTEr(NPs)-CNTs. Two sets of glass series were synthesized by melt-quenched technique with chemical composition of  $(0.47(1-y))TeO_2+(0.2(1$ y)) $B_2O_3+(0.29(1-y))+ZnO+Er_2O_3(y)$  wherein, y = 0.005, 0.01, 0.02, 0.03, 0.04, and 0.05 mol% Er<sub>2</sub>O<sub>3</sub> nanoparticles. Tellurite-based glass was deposited with rGO and CNTs onto the glass surfaces by spray-coated technique. The physical, morphological, structural, and optical properties of ZBTEr(NPs)-rGO and ZBTEr(NPS)-CNTs glasses were characterized via densimeter, scanning electron microscopy (SEM), energy dispersive X-ray (EDX), transmission electron microscopy (TEM), X-ray diffraction (XRD), Fourier transformmicro-Raman ultraviolet-visible infrared (FT-IR), spectroscopy, (UV-Vis) spectrophotometer, and photoluminescence. SEM micrographs revealed morphological structures of rGO and CNTs on the glass surfaces meanwhile, EDX spectra disclosed elemental composition of rGO and CNTs. TEM images proved existence of Er(NPs) with average size (~23.53 nm) in glass matrix. XRD pattern verified amorphous nature of glasses. FT-IR spectra evaluated the presence of non-bridging oxygens (NBOs) with TeO<sub>4</sub>. TeO<sub>3</sub>, and BO<sub>3</sub> functional groups meanwhile, Raman spectra demonstrated good quality of rGO and CNTs. The refractive index value was increased (2.402-2.775) for ZBTEr(NPs)rGO meanwhile, (2.432-2.542) for ZBTEr(NPs)-CNTs. The optical bandgap energy value was improved (1.913-2.931 eV) for ZBTEr(NPs)-rGO and (2.513-2.875 eV) ZBTEr(NPs)-CNTs meanwhile, non-linear trend of Urbach energy (0.118-0.408 eV) for ZBTEr(NPs)rGO and (0.158-0.375 eV) for ZBTEr(NPs)-CNTs. Judd-Ofelt's intensity parameters showed  $\Omega_2 > \Omega_6 > \Omega_4$  trend for ZBTEr(NPs)-rGO whilst,  $\Omega_2 > \Omega_4 > \Omega_6$  trend for ZBTEr(NPs)-CNTs. Radiative parameters and branching ratio proved  ${}^{2}H_{11/2} \rightarrow {}^{4}I_{15/2}$  transition showed highest radiative transition value, resulting in a strong green emission. Luminescence spectra exhibited two emission peaks assigned to  ${}^{2}H_{11/2} \rightarrow {}^{4}I_{15/2}$  and  ${}^{4}S_{3/2} \rightarrow {}^{4}I_{15/2}$  transitions. In conclusion, the rGO and CNTs deposition enhanced the optical properties of glass materials. Implication of this study offers a new milestone in the glass coatings field for improving current fiber optics.

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### STRUKTUR, PARAMETER KEAMATAN, DAN ANALISIS PENDARCAHAYAAN BAGI KACA TELLURIT DISALUT rGO/CNTs TERDOP DENGAN NANOZARAH ERBIUM

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#### ABSTRAK

Kajian ini bertujuan untuk meningkatkan sifat optik kaca tellurit disalut grafin oksida terkurang (rGO)/nanotiub karbon (CNTs) terdop dengan nanozarah erbium oksida (ZBTEr(NPs)-rGO/-CNTs). Dua set siri kaca berasaskan tellurit telah disintesis dengan teknik lebur lindap konvensional berdasarkan komposisi kimia (0.47(1-y))TeO<sub>2</sub>+(0.2(1y)) $B_2O_3+(0.29(1-y))+ZnO+Er_2O_3(y)$  iaitu, y = 0.005, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%nanozarah Er2O3. Kaca berasaskan tellurit telah disalut dengan rGO dan CNTs atas permukaan kaca dengan teknik salutan semburan. Sifat fizikal, morfologi, struktur, dan optik bagi kaca ZBTEr(NPs)-rGO/-CNTs dicirikan dengan menggunakan densimeter, mikroskop pengimbasan elektron (SEM), penyebaran tenaga sinar-X (EDX), mikroskop penghantaran elektron (TEM), pembelauan sinar-X (XRD), transformasi Fourier inframerah (FT-IR), spektroskopi mikro-Raman, spektrofotometer cahaya nampak ultralembayung (UV-Vis), dan kefotopendarcahayaan. Mikrograf SEM mendedahkan struktur morfologi rGO dan CNTs pada permukaan kaca manakala, spektrum EDX mendedahkan komposisi unsur rGO dan CNTs. Imej TEM membuktikan kewujudan Er(NPs) dengan saiz purata (~23.53 nm) dalam matriks kaca. Corak XRD megesahkan sifat amorfus bagi kaca. Spektrum FT-IR mengesahkan kehadiran oksigen bukan penitian (NBO) dengan kumpulan berfungsi TeO<sub>4</sub>, TeO<sub>3</sub>, dan BO<sub>3</sub> manakala, spektrum Raman menunjukkan kualiti rGO dan CNTs yang baik. Nilai indeks biasan meningkat (2.402-2.775) bagi ZBTEr (NPs)-rGO manakala, (2.432-2.542) bagi ZBTEr(NPs)-CNTs. Nilai tenaga jurang jalur optik meningkat (1.913-2.931 eV) bagi ZBTEr(NPs)-rGO dan (2.513-2.875 eV) bagi ZBTEr(NPs)-CNTs manakala, trend tidak linear tenaga Urbach (0.118-0.408 eV) bagi ZBTEr (NPs)-rGO dan (0.158-0.375 eV) bagi ZBTEr(NPs)-CNTs. Parameter keamatan Judd-Ofelt menunjukkan trend  $\Omega_2 > \Omega_6 > \Omega_4$  bagi ZBTEr(NPs)-rGO manakala, trend  $\Omega_2 > \Omega_4 > \Omega_6$  bagi ZBTEr(NPs)-CNTs. Parameter sinaran dan nisbah percabangan membuktikan peralihan  ${}^{2}H_{11/2} \rightarrow {}^{4}I_{15/2}$  mempunyai nilai peralihan sinaran tertinggi yang menghasilkan pancaran hijau yang kuat. Spektrum pendarcahayaan menunjukkan dua puncak pancaran yang diberikan kepada peralihan  ${}^{2}H_{11/2} \rightarrow {}^{4}I_{15/2}$  dan  ${}^{4}S_{3/2} \rightarrow {}^{4}I_{15/2}$ . Kesimpulannya, salutan rGO/CNTs meningkatkan sifat optik bahan kaca. Implikasi kajian ini adalah menawarkan pencapaian yang baru dalam bidang salutan kaca untuk penambahbaikan gentian optik semasa.





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## LIST OF SYMBOLS AND ACRONYMS

α	-	Absorption Coefficient	cm <sup>-1</sup>
$\alpha_0^{2-}$	-	Oxide Ion Polarizability	Å
А	-	Area	m <sup>2</sup>
A <sub>ed</sub>	-	Electric Dipole Transition Probability	s <sup>-1</sup>
A <sub>md</sub>	-	Magnetic Dipole Transition Probability	s <sup>-1</sup>
$\beta_R$	-	Branching Ratio	%
С	-	Speed of Light	ms <sup>-1</sup>
d 🕐 pustaka.up	os <del>i.</del> edu.my	Thickness stakaan Tuanku Bainun DistakaTBai	n <b>m O</b> ptbup
$\delta_{rms}$	-	Root Mean Square Deviation	-
е	-	Electronic Charge	С
3	-	Molar Extinction Coefficient	Lmol <sup>-1</sup> cm <sup>-1</sup>
E	-	Dielectric Constant	-
E <sub>opt</sub>	-	Optical Bandgap Energy	Joule (J)/eV
$\Delta E$	-	Urbach Energy	Joule (J)/eV
F	-	Field Strength	cm <sup>3</sup>
f <sub>cal</sub>	-	Calculated Oscillator Strength	-
f <sub>exp</sub>	-	Experimental Oscillator Strength	-
h	-	Plank's Constant	m <sup>2</sup> kgs <sup>-1</sup>
ħω	-	Photon Energy	Joule (J)/eV

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J	-	Angular Momentum	kgm <sup>2</sup> s <sup>-1</sup>
$M_W$	-	Molecular Weight	g/mol
т	-	Mass of Electron	g
N <sub>A</sub>	-	Avogadro's Number	-
Ν	-	Ionic Concentration of Rare-earth	ions/cm <sup>3</sup>
η	-	Refractive Index	-
OPD	-	Oxygen Packing Density	g.atom/L
ρ	-	Density	kg/m <sup>3</sup>
$arOmega_t$	-	Judd-Ofelt's Intensity Parameter	-
R <sub>i</sub>	-	Inter-ionic Distance	Å
$R_L$	-	Reflection Loss	-
R <sub>P</sub> pustaka.up	os <u>i</u> edu.my	Polaron Radius Tuanku Bainun Abdul Jalil Shah	in Å 🚺 ptbur
S <sub>ed</sub>	-	Electric Dipole Line Strength	-
S <sub>md</sub>	-	Magnetic Dipole Line Strength	-
$S aj'\rangle ^2$	-	Manifold State of Elements	-
Т	-	Transmission Coefficient	-
t <sub>r</sub>	-	Radiative Lifetime	ms
$  ^{2}$	-	Reduced Matrix Elements	-
$V_m$	-	Molar Volume	m <sup>3</sup> /mol
ν	-	Wavenumber	cm <sup>-1</sup>
ω	-	Radian Frequency	Fm <sup>-1</sup>
λ	-	Wavelength	m
χ	-	Electronegativity	-

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B <sub>2</sub> O <sub>3</sub>	-	Boron (III) Oxide
BOs	-	Bridging Oxygens
CNTs	-	Carbon Nanotubes
EDX	-	Energy Dispersive X-ray
Er <sub>2</sub> O <sub>3</sub>	-	Erbium (III) Oxide
FESEM	-	Field Emission Scanning Electron Microscopy
FT-IR	-	Fourier Transform Infrared
GO	-	Graphene Oxide
HR-TEM	-	High Resolution Transmission Electron Microscopy
NBOs	-	Non-bridging Oxygens
rGO	-	Reduced Graphene Oxide
REIs pustaka.up	s <del>i.</del> edu.my	Rare-earth Ions Tuanku Bainun Kampus Sultan Abdul Jalil Shah
SDS	-	Sodium Dodecyl Sulphate
TeO <sub>2</sub>	-	Tellurium (II) Oxide
tbp	-	Trigonal Bipyramidal
tp	-	Trigonal Pyramidal
XRD	-	X-ray Diffraction
UV-Vis	-	Ultraviolet Visible
ZnO	-	Zinc Oxide



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## LIST OF APPENDICES

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- В Presentations
- С Awards



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This chapter outlines a brief introduction on the research studies and background. The objectives, research problems, scopes, limitations, and significance of the research work are presented in this chapter. The last part of this chapter elucidates the summary of the thesis.



#### 1.2 **Research Background**

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The discovery of a breakthrough in glass-making was initiated in 5000 BC during the Mesopotamia era when they found a natural glass known as "fulgurites" that has been formed from the strike of lightning. The early efforts of glassmaking were inspired from the Egyptian era when glass production was initially invented from a substantial amount of sand-based silica. Then, it was followed by the evolution of glass manufacturing during the Roman Empire via the glass blowing technique and has been extended to the production of stained-glass windows. The commercial products from glasses in this era were mainly covered the ornaments and small containers for cosmetics.

In the early 1900s, glass development and production began to increase rapidly since the invention of machinery and thereafter the revolution of glass production was initiated by Sir Alastair Pilkington. In addition, the innovation of glass manufacturing is undoubtedly one of the most extensively high-demanding due to the continuous research in the field of glass science and technology. Consequently, the widespread use and broad commercialization in glass applications were utilized in optical lenses, light-emitting devices, optical fiber amplifiers, optoelectronics, solid-state high-power lasers for telecommunications, and many other fields. The rare-earth ions (REIs) doped glasses recently have been a great interest in the field of photonic materials in terms of multifunctional applications particularly, for upconversion luminescence temperature sensors.



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Commonly, glass belongs to solid materials which can be classified into crystalline and amorphous solids. In Greek, the term glass can be defined as an amorphous or "shapeless" or non-crystalline solid material from a fusion process. Moreover, glass can be formed from silica sand, soda ash, limestone, dolomite, lead oxide, and borate oxide. The amorphous glasses possess a short-range periodic arrangement and does not consist of a well-shaped or geometrical shape in contrast with the crystalline solids. Typically, glass materials have several essential properties, such as high transparency, excellent refractive index, high hardness, rust resistance, and low manufacturing costs. In addition, glass can be easily customized to different shapes or sizes and altered its properties especially to produce high-quality optical glasses (Al-Hadeethi et al., 2019; Gupta et al., 2020; Sayyed et al., 2020).

Glass system is occupied by a continuous random network due to the disappearance of any crystalline structure meanwhile, the amorphous silica consists of two tetrahedral that shows the relation between the Si-O and O-O atoms and bridging oxygen existing between the two tetrahedral, as presented in Figure 1.1. The network structure showed the short-range order which is compatible with the absence of long-range order.

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Figure 1.1. Basic atomic structure of an amorphous silica (a) Schematic representation of two tetrahedra displaying nominal distance between neighboring Si-O and O-O atoms, and a bridging oxygen (BO) between two tetrahedra and (b) Subsection of an amorphous silica simulation which shows the random arrangement of silica tetrahedral unit (Lunt et al., 2018)

05-4506832 😵 pustaka.upsi.edu.my 🕈 Perpustakaan Tuanku Bainun Kampus Sultan Abdul Jalil Shah The formation of glass usually occurs by a rapid cooling when the molten continuously hardens into the solid state without the crystallization of materials. According to the studies on glass formation by Zachariasen, natural glass can be formed based on the following rules as outlined below (Zachariasen, 1932):

- 1. No oxygen atom may be linked to more than two cations
- 2. Cation coordination number is small either 3 or 4
- 3. Oxygen polyhedral share corners, not edges, or faces
- 4. For 3D networks, at least three corners must be shared





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The constituents of glass can be comprised to three components which are glass former, glass intermediate, and glass modifier. The glass former forms a highly crosslinked network of chemical bonds and possesses high electronegativity which can produce good glass-forming. Commonly, silicon oxide has been used as a network-forming in glass network rather than the other oxides such as germanium and boron oxides. Meanwhile, the glass modifier can be added in the glass network as it may alter the glass properties and break up all the bonds in the glass structure which attributes to the formation of nonbridging oxygen (NBO) atoms in the glassy matrix.

Additionally, the glass modifier can only participate in glass network modification which form highly ionic bonds with oxygen and reducing network connectivity. The glass intermediate usually possesses the intermediate electronegativity value between the glass former and the glass modifier. Due to that, the glass intermediate is not able to form the glass network by itself however, can be formed when the glass intermediate is mixed with another glass former. This behavior depends on the single bond strength value. Moreover, a high single bond strength value may potentially improve the glass formation (Dimitrov & Komatsu, 2013).

The extensive research on silicate glasses is significantly received good attentions when trivalent rare-earth ions is introduced in silicate glass network which gives excellent fluorescence efficiency in photoluminescence properties (Wang et al., 2020). The use of borate glasses are demanding due to high chemical durability and thermal shock resistance which usually has been added to silicate glasses (Bengisu, 2015). In addition, recent studies







suggested that germanate glasses possess low-phonon energy, and strong chemical and mechanical stability which is useful for optical fiber drawing (Pisarski et al., 2020). Meanwhile, the phosphate glasses captured an interest among the researchers due to their intense green light emission which is suitable to be used as optical fiber amplifiers (Taherunnisa et al., 2019). Fortunately, the tellurite-based glasses are non-crystalline solids that shows many advantages which have been extensively studied for their numerous optical, photonics, and laser applications (Azlan et al., 2019; Umar et al., 2017).

In comparison with these glasses, tellurite-based glass provides superior properties such as low melting point, high refractive index, high dielectric constant, and excellent infrared transmission (Pepe et al., 2019; Usman et al., 2018; Yuliantini et al., 2019; Zaitizila et al., 2018). Furthermore, tellurite glasses possess good chemical durability, excellent mechanical strength, and allow a wide spectral in the range of 3-18 µm which attribute to the high possibility of fabricating stable glass (Gomes et al., 2017).

Tellurium oxide, TeO<sub>2</sub> and zinc oxide, ZnO can be a good combination and excellent compatibility to form stable glasses. Thus, these glasses have been widely explored for potential applications in optical fiber. The inclusion of ZnO in tellurite-based glasses may provide superior mechanical properties, low ability to crystallize, low optical bandgap energy, as well as high refractive index than other oxide materials (Faznny et al., 2016). Additionally, the addition of borate oxide,  $B_2O_3$  in tellurite-based glass matrix had been developed that might potentially enhance the glass transparency, possess good stability, and rare-earth ion solubility.







Rare-earth ions (REIs) doped glasses have grabbed attention due to their outstanding characteristics to improve the optical properties. In this research, the lanthanide oxide nanoparticles have been selected to be doped in the glass system to achieve higher optical performance and specifically, obtain a good impact on the fiber optics technology. Moreover, erbium, Er<sup>3+</sup> nanoparticles are often chosen among researchers which act as the most remarkable candidate in the development of broadband erbium-doped fiber amplifier (EDFA) due to its emission of about 1.53  $\mu$ m and thus, suitable to be used as a lasing glass material.

The technology of carbon-based nanomaterials (CBNs) is growing rapidly in recent years. CBNs have extraordinary characteristics and multifunctionality which importance in many applications such as optoelectronics, biomedical, and photonics devices. Owing to such characteristics, the CBNs sparked immense interest among the researchers for upcoming applications in optical sensors (Li et al., 2019), ultrafast lasers (Ma et al., 2019), and optoelectronic devices (Wang et al., 2019). Hence, due to these properties, the CBNs have high potential to enhance the optical properties of the glass materials.

Considerable research studies on glass coatings have been continuously investigated by utilizing the CBNs as coating materials on the glass surfaces. Notably, the fiber optics are usually made from glass materials. The deposition of graphene oxide (GO) and carbon nanotubes (CNTs) on glass materials may give insights the effect of these materials on optical fiber. Based on the current studies, the optical fiber can be coated with graphene oxide via simple and low-cost technique. The previous research on optical fiber





coated with GO shows outstanding optical properties and can be applied for multifunctional fiber devices, highly sensitive sensors, and flexible optical electronics (Fang, Xiong, Hao, Zhang, & Wang, 2019; Ishtiaq et al., 2022). The studies of CBNs coated-glass fibers are still on-going and may contribute to wide optical applications such as light-weight electromagnetic shielding, optical sensors, and optoelectronics device performances.

#### 1.3 **Research Problems**

The applications of glass materials especially in fiber optics were initiated many years ago. Cao et al., (2016) investigated the spectroscopic properties of rare-earth ions (REIs) namely,  $Er^{3+}/Ho^{3+}$  co-doped silicate glasses, which is useful in improving the Ho<sup>3+</sup> 2.0 µm fiber laser performance and mid-infrared laser materials. Taking over the advantages of silicate glasses, they have achieved an excellent transmittance, which acts as a key to fast communications. The silicate glasses was significantly used in the production of highly non-linear fibers, high-concentration rare-earth ions doped fibers, ultrafast lasers, and optical fiber communications (Liang et al., 2021).

Nevertheless, the problems encountered with these glasses are due to the relatively high loss and high melting temperature of around 2100 °C, as proposed in the previous study (Kasik et al., 2020). In addition, the low optical loss of glass materials is aimed especially for mid-IR applications. Therefore, these limitations need to be overcome and







progressive research work is still required to find a suitable glass material with lower melting point and low transmission loss.

Over the years, tellurite-based glasses have been extensively introduced due to their unique advantages over silicate glasses, as reported in previous studies (Azlan et al., 2019). Shen, et al. (2020) proposed that tellurite glass fiber possesses an efficient low-loss and outstanding candidate for mid-infrared laser emission applications. Subsequently, the tellurite glass has good solubility with rare-earth ions (REIs). Furthermore, the tellurite glasses are well-known as the most stable glass former than the other types of glass, such as germanate, borate, phosphate, and silicate glasses.

In addition, the lanthanide oxide nanoparticles such as erbium nanoparticles can be used as a dopant to improve the optical properties of tellurite-based glass system which is beneficial to be applied in photonic devices. Despite extensive studies on tellurite glasses doped with erbium ions, the optical properties of tellurite glasses are still limited. Tellurite glass with refractive index (>2.0) is aimed to meet the criteria for optical fiber.

Therefore, the research on glass coatings can be adopted to overcome these limitations. The deposition of carbon-based nanomaterials (CBNs) on glass surfaces is expected to achieve excellent optical properties. The investigation of reduced graphene oxide (rGO) and carbon nanotubes (CNTs) coated on glass surfaces is still limited in number. Therefore, rGO and CNTs deposited on the tellurite-based glass surfaces via a simple spray coating technique can be a first step to achieve high functionality of optical





fiber. Hence, the outcome of this research is believed to contribute to the new glass technology especially, in the application of fiber optics. In summary, the research problems proposed in the present study are summarized in Table 1.1.

### Table 1.1

Summary of the research problems and research solutions that relate to this study

Research problems	Research solutions
• The commercial fiber optics used •	Tellurite-based glasses has been
silicate glasses as a key to fast	introduced to reduce high loss and
communication however, the problems	melting temperature; and possess
encountered with these glasses are due	high-solubility of rare-earth (RE)
to the high loss and high melting	u Bainun ul jons ah PustakaTBainun O ptbupsi
temperature of around 2100 °C (Kasik	
et al., 2020)	
• The extensive studies on tellurite- •	Novel research on glass coatings can
based glasses doped with erbium, Er <sup>3+</sup>	be adopted to enhance the optical
ions nevertheless, these glasses are still	efficiency and overcome the
lacking to achieve a significant	limitations of current glass materials
refractive index value (>2.0)	
especially, for fiber optics application	

(Continue)

### Table 1.1 (Continue)

	Research problems		Research solutions
•	A recent study proposed the GO	•	The rGO and CNTs can be
	coating prominently improves the		deposited on the tellurite-based
	optical properties by balancing the		glass surfaces via a simple spray
	oxygen functional (Wang et al., 2020)		coating technique to compare the
	however, the investigation of rGO and		optical properties of the glass
	CNTs towards glass coatings has not		materials
	been reported yet		

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The objectives of this work are outlined as follows:

- i. To synthesize the two-glass series by using melt-quenched and spray coating techniques with composition of  $\{(0.47(1-y)) \text{ TeO}_2 + (0.2 (1-y)) \text{ B}_2\text{O}_3 + (0.29 (1-y))\}$  $ZnO + (y) Er_2O_3$  (nanoparticles)} -rGO/-CNTs (coated), (y = 0.005, 0.01, 0.02, 0.03, 0.04, and 0.05 mol%) denoted as ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs.
- ii. To investigate the surface morphology, particle size, and bonding parameters of ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses by using FESEM, FT-IR, TEM, XRD, Raman, and EDX spectroscopy.

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- iii. To determine the absorption spectra, refractive index, optical bandgap energy, and Urbach energy of ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses by using UV-Vis spectroscopy.
- iv. To analyze the intensity parameters, emission lifetime, and luminescence properties of ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses by using Judd-Ofelt theory and luminescence spectroscopy.

#### 1.5 **Scope and Limitations of Study**

This study attempts to identify the structural and optical properties of erbium nanoparticles doped tellurite-based glasses coated with reduced graphene oxide (rGO) and carbon nanotubes (CNTs). The two sets of glass series with chemical composition of  $\{(0.47(1$ y)) $TeO_2 + (0.2(1-y))B_2O_3 + (0.29(1-y))ZnO + (y)Er_2O_3 (nanoparticles)\} -rGO/-CNTs$ (coated) were prepared via conventional melt-quenching technique. Subsequently, the rGO and CNTs were deposited on the tellurite-based glasses surface using a simple and low-cost spray coating technique.

The physical properties of the prepared glasses including, density, molar volume, rare-earth inter-ionic distance, polaron radius, field strength, and oxygen packing density (OPD) were calculated by Archimedes' principle. Then, several analysis methods were carried out for glass characterizations such as Field Emission Scanning Electron



Microscopy (FESEM), High-Resolution Transmission Electron Microscopy (HR-TEM), X-ray Diffraction (XRD), Fourier Transform Infrared Spectrometer (FT-IR), micro-Raman, and X-ray Energy Dispersive X-ray (EDX) spectroscopy to investigate the structural and morphological properties of the glasses. The FESEM analysis was conducted to confirm the existence of rGO and CNTs morphology on the glass surfaces meanwhile, the elemental compositions that exists in rGO and CNTs were evaluated by using EDX analysis. HR-TEM analysis was performed to determine the particles' size of erbium oxide in the amorphous matrix system. Next, XRD was analyzed to confirm amorphous nature of the tellurite-based glasses. FT-IR analysis was used to determine the structural bonding, functional groups, stretching vibrational modes, and the presence of non-bridging oxygens (NBOs). Lastly, the micro-Raman analysis was utilized to determine the characteristics vibrational modes of GO, rGO, and CNTs that attached on the glass surfaces.

Meanwhile, the study on optical properties of ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses were identified via Ultra-violet Visible (UV-Vis) spectrophotometer to obtain the optical absorption spectra, optical bandgap energy, Urbach energy, refractive index, reflection loss, transmission coefficient, dielectric constant, and electronegativity. The theoretical analysis of optical parameters was performed by using Judd-Ofelt's theory to identify the intensity spectral parameters ( $\Omega_2$ ,  $\Omega_4$ ,  $\Omega_6$ ), oscillator strength ( $f_{exp}$ ), radiative transition probabilities ( $A_{rad}$ ), branching ratio ( $\beta_R$ ), as well as excited-state radiative lifetime  $(\tau_r)$ . The investigation of luminescence spectra of of ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses was studied by using photoluminescence spectroscopy to analyze the emission and excitation state.







Understanding the CBNs characteristics is important to determine the suitability of CBNs as coating materials on glass surfaces. The association between the CBNs and glass materials promotes a novel glass especially to be used in fiber optics. The current focus of this study is to determine the optical properties of coated glasses.

#### 1.6 Significance of Research

In this study, the characterization of glasses can be determined based on their physical, structural, and optical properties. Moreover, these properties are essential for optical fiber applications and optoelectronic devices. Hence, the main purpose of this work is to improve the optical properties of the tellurite-based glass via coating the GO and CNTs on the glass surfaces. The fabrication and spectroscopic studies of the ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses may attribute to a novel approach to improve the optical properties of the tellurite-based glass.

Taking advantage of CBNs in the glass technology development, rGO and CNTs are potential candidates as the coating materials due to high demand in industry. In this work, optical glasses can be significantly improved by using rGO and CNTs as the coating materials.

To the best of our knowledge, this work will lead to excellent improvement, especially for fiber optics technology. It has been known that broadband of the optical fiber





transmits the optical data via infra-red spectrum at the "speed of light" about  $3.0 \times 10^8$  m/s. This research will upgrade the current fiber optics with "coated-fiber optics" which may allow the optical data to transmit faster from one destination to another. Thus, the present research was completely done to explore more understanding of the glass coatings whereby, a clearer understanding of the effects of CBNs on the glass surfaces was employed. Particularly, the interest in rGO and CNTs coating layer was absolutely exploited which is closely associated with the attached oxygens functionalization on the glass surfaces.

#### **Thesis Overview** 1.7

This thesis consists of five chapters that describe the overall presentation of the present study which comprises two glass series namely, erbium nanoparticles doped tellurite glass coated with reduced graphene oxide (ZBTEr (NPs)-rGO) and erbium nanoparticles doped tellurite glass coated with carbon nanotubes (ZBTEr (NPs)-CNTs). Therefore, the outlines of the thesis in this study are described as follows:

Chapter 1 introduces the research purpose including the background of the study, research problems, research objectives, scopes, limitations, and significance of the research. The summarization of the thesis has been well described, with provided research aims, respective analysis, and the main contribution in this work.





Chapter 2 presents a discussion of the literature review, including tellurite-based glass structure, the role of the dopant in the glass matrix, as well as the history of the carbon-based materials (CBNs) with their properties and synthesis techniques. Importantly, this chapter highlights the fundamental theory and calculation of the optical properties, including optical bandgap, Urbach energy, refractive index, Judd-Ofelt's theoretical study, as well as other optical parameters.

The research methodology, procedures, and experimental techniques which involved the glass fabrication, synthesis of rGO and CNTs, and the spray coating technique are comprehensively presented in Chapter 3. Furthermore, the physical, structural, and optical properties characterizations are briefly described, and this chapter summarizes the research work in flowchart diagram. Proustakaan Tuanku Bainun Destaka Bainun Destaka Bainun Destaka Bainun

Chapter 4 discusses the results of the physical, morphological, structural, and optical properties of the ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses. Taking advantage of the deposition of rGO and CNTs on the glass surfaces, this chapter presents in-depth discussion on the effects and interaction between the coating materials and ZBTEr (NPs) glasses. Theoretical optical parameters of the ZBTEr (NPs)-rGO and ZBTEr (NPs)-CNTs glasses are carried out by using Judd-Ofelt's analysis. In addition, the luminescence analysis is analyzed to support the Judd-Ofelt's analysis.

Finally, Chapter 5 summarizes the conclusion in this study and provides several future works.

