



**UNIVERSITI PUTRA MALAYSIA**

***EFFECTS OF INCLUSION OF Gd<sub>2</sub>O<sub>3</sub> MICROPARTICLES AND Gd<sub>2</sub>O<sub>3</sub> NANOPARTICLES ON ELASTIC, LINEAR AND NONLINEAR OPTICAL PROPERTIES OF ZINC BOROTELLURITE GLASSES***

**CHUA EE VON**

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**EFFECTS OF INCLUSION OF  $Gd_2O_3$  MICROPARTICLES AND  $Gd_2O_3$   
NANOPARTICLES ON ELASTIC, LINEAR AND NONLINEAR OPTICAL  
PROPERTIES OF ZINC BOROTELLURITE GLASSES**

By

**CHUA EE VON**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,  
in Fulfillment of the Requirements for the Degree of Doctor of Philosophy**

**November 2017**

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

**EFFECTS OF INCLUSION OF Gd<sub>2</sub>O<sub>3</sub> MICROPARTICLES AND Gd<sub>2</sub>O<sub>3</sub> NANOPARTICLES ON ELASTIC, LINEAR AND NONLINEAR OPTICAL PROPERTIES OF ZINC BOROTELLURITE GLASSES**

By

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**November 2017**

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**Faculty : Science**

The purpose of this present work is to study the effect of Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> NPs on elastic, linear and nonlinear optical properties of zinc borotellurite glass system. Zinc borotellurite glass system doped with gadolinium oxide (Gd<sub>2</sub>O<sub>3</sub>) and gadolinium oxide nanoparticles (Gd<sub>2</sub>O<sub>3</sub> NPs) with chemical formula  $\{[(\text{TeO}_2)_{70}(\text{B}_2\text{O}_3)_{30}]_{70}(\text{ZnO})_{30}\}_{1-x}(\text{RE}_2\text{O}_3)_x$  (where  $x = 1.0, 2.0, 3.0, 4.0$  and  $5.0$  mol% and  $\text{RE}_2\text{O}_3 = \text{Gd}_2\text{O}_3$  and  $\text{Gd}_2\text{O}_3$  NPs) were fabricated using conventional melt quenching technique.

XRD results confirmed the amorphousity of the glass samples. The infrared spectra of the glass systems indicate the existence of TeO<sub>3</sub>, TeO<sub>4</sub>, BO<sub>3</sub> and BO<sub>4</sub> vibrational groups. The presence of Gd<sub>2</sub>O<sub>3</sub> NPs was proven from TEM images. The longitudinal ultrasonic velocities vary from 3908 to 4076 m/s and 3883 to 4042 m/s for Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses, respectively while the shear ultrasonic velocities vary from 2222 to 2277 m/s and 2251 to 2282 m/s for Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses, respectively. The observed change in ultrasonic velocities shows that there is a substantial change in the structure of the glass. The elastic moduli (longitudinal modulus (L), shear modulus (G), bulk modulus (K) and Young's modulus (E)) obtained for Gd<sub>2</sub>O<sub>3</sub> doped zinc borotellurite glass system increase from 55.44 to 81.35 GPa, 18.39 to 25.00 GPa, 30.92 to 48.02 GPa and 46.02 to 63.90 GPa, respectively. In addition, the elastic moduli (L, G, K and E) for Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses increase from 55.44 to 79.45 GPa, 18.39 to 26.68 GPa, 30.92 to 43.87 GPa and 46.05 to 66.55 GPa, respectively. The increase elastic moduli indicate that the rigidity of the glass increases. The experimental results showed that the elastic properties system depend on the composition of the glass system and the role of Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> nanoparticles inside the glass network.

The direct and indirect optical band gap energy increases from 3.239 to 3.519 eV and 2.587 to 3.172 eV for Gd<sub>2</sub>O<sub>3</sub> doped zinc borotellurite glasses whereas the direct and indirect optical band gap energy for Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses decrease from 3.301 to 2.985 eV and 2.790 to 2.386 eV, respectively. In addition, the refractive index of Gd<sub>2</sub>O<sub>3</sub> doped glasses decrease from 2.518 to 2.352 while an increasing trend from 2.456 to 2.551 is observed in Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses. It was found that the optical band gap and refractive index of Gd<sub>2</sub>O<sub>3</sub> doped glasses show opposite behaviour to Gd<sub>2</sub>O<sub>3</sub> NPs doped glass systems. This might be attributed to the presence of nanoparticles having large surface area which form high density states that can serve to trap charge carriers.

The nonlinear refractive index of the glass systems showed self-focusing behaviour and reverse saturable absorption (RSA) or two-photon absorption were observed for nonlinear optical absorption. The values of nonlinear refractive index vary from 0.632 to  $0.943 \times 10^{-14}$  cm<sup>2</sup>/W and 0.266 to  $1.515 \times 10^{-14}$  cm<sup>2</sup>/W for Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> NPs doped glass systems, respectively. Moreover, the values of nonlinear absorption coefficient vary from 0.616 to 0.747 cm/GW and 0.660 to 0.729 cm/GW for both Gd<sub>2</sub>O<sub>3</sub> and Gd<sub>2</sub>O<sub>3</sub> NPs doped zinc borotellurite glasses, respectively. The variation of nonlinear optical parameters was due to the formation of TeO<sub>4</sub>, the presence of lone pairs electron in TeO<sub>4</sub> and the effect of large Gd<sup>3+</sup> ion as well as the reduction in size of bulk materials. The results obtained from z-scan suggested that the presently studied glass systems can be used for design of nonlinear optical materials.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

**KESAN PERANGKUMAN MIKROZARAH  $Gd_2O_3$  DAN NANOZARAH  $Gd_2O_3$  KE ATAS SIFAT ELASTIK, OPTIK LINEAR DAN OPTIK TIDAK LINEAR BAGI KACA ZINK BOROTELLURIT**

Oleh

**CHUA EE VON**

**November 2017**

**Pengerusi : Profesor Madya Halimah Mohamed Kamari, PhD**  
**Fakulti : Sains**

Tujuan kajian ini adalah untuk mengkaji kesan  $Gd_2O_3$  dan nanozarah  $Gd_2O_3$  ke atas sifat elastik, optik linear dan optik tidak linear bagi sistem kaca zink borotellurit. Sistem kaca zink borotellurit didopkan dengan gadolinium oksida ( $Gd_2O_3$ ) dan nanozarah gadolinium oksida ( $Gd_2O_3$  NPs) bagi formula kimia  $\{[(TeO_2)_{70}(B_2O_3)_{30}]_{70}(ZnO)_{30}\}_{1-x}(RE_2O_3)_x$  ( $x = 1.0, 2.0, 3.0, 4.0$  and  $5.0$  mol% dan  $RE_2O_3 = Gd_2O_3$  and  $Gd_2O_3$  NPs) telah difabrikasi dengan menggunakan teknik sepuh lindap.

Ciri amorfus bagi sampel kaca telah disahkan oleh hasil XRD. Spektrum inframerah sistem kaca telah menunjukkan kewujudan kumpulan getaran  $TeO_3$ ,  $TeO_4$ ,  $BO_3$  dan  $BO_4$ . Kewujudan  $Gd_2O_3$  NPs telah dibuktikan daripada imej TEM. Halaju gelombang membujur berubah daripada 3908 ke 4076 m/s dan 3883 ke 4042 m/s bagi kaca zink borotellurit didop dengan  $Gd_2O_3$  dan  $Gd_2O_3$  NPs, masing-masing, manakala, halaju gelombang ricih berubah daripada 2222 ke 2277 m/s dan 2251 ke 2282 m/s bagi kaca zink borotellurit didop dengan  $Gd_2O_3$  dan  $Gd_2O_3$  NPs, masing-masing. Perubahan yang diperhatikan bagi halaju gelombang menunjukkan bahawa terdapat perubahan dalam struktur kaca. Modulus kenyal (modulus membujur (L), modulus ricih (G), modulus pukal (K) dan modulus Young (E)) bagi kaca zink borotellurit didop dengan  $Gd_2O_3$  meningkat daripada 55.44 ke 81.35 GPa, 18.39 ke 25.00 GPa, 30.92 ke 48.02 GPa dan 46.05 ke 63.90 GPa, masing-masing manakala modulus kenyal (L, G, K and E) bagi kaca zink borotellurit didop dengan  $Gd_2O_3$  NPs meningkat daripada 55.44 ke 79.45 GPa, 18.39 ke 26.68 GPa, 30.92 ke 43.87 GPa dan 46.05 ke 66.55 GPa. Peningkatan modulus kenyal menunjukkan bahawa ketegaran kaca meningkat. Hasil uji kajian menunjukkan bahawa sifat elastik sistem bergantung pada komposisi sistem kaca dan peranan  $Gd_2O_3$  dan  $Gd_2O_3$  nanozarah dalam rangkaian kaca.

Tenaga jurang jalur langsung dan tidak langsung meningkat dari 3.239 ke 3.519 eV dan 2.587 ke 3.172 eV bagi kaca zink borotellurit didop dengan Gd<sub>2</sub>O<sub>3</sub> manakala tenaga jurang jalur langsung dan tidak langsung bagi kaca zink borotellurit terdop dengan Gd<sub>2</sub>O<sub>3</sub> NPs menurun dari 3.301 ke 2.985 eV dan 2.790 ke 2.386 eV, masing-masing. Tambahan pula, indeks pembiasan bagi kaca didop Gd<sub>2</sub>O<sub>3</sub> menurun dari 2.518 ke 2.352 manakala trend peningkatan dari 2.456 ke 2.551 telah dicerapi bagi kaca zink borotellurit didop dengan Gd<sub>2</sub>O<sub>3</sub> NPs. Didapati bahawa jurang jalur optik dan indeks pembiasan bagi kaca didop Gd<sub>2</sub>O<sub>3</sub> menunjukkan hasil bertentangan dari sistem kaca didop Gd<sub>2</sub>O<sub>3</sub> NPs. Ini mungkin disebabkan oleh kehadiran nanozarah yang mempunyai kawasan permukaan yang besar yang membentuk keadaan ketumpatan tinggi yang boleh bertindak sebagai perangkap pembawa cas.

Indeks pembiasan tidak linear sistem kaca telah menunjukkan hasil swafokus dan penyerapan tepu songsang atau penyerapan dua foton telah dicerapi bagi penyerapan optik tidak linear. Nilai indeks pembiasan tidak linear berubah dari 0.632 ke 0.943 x 10<sup>-14</sup> cm<sup>2</sup>/W dan 0.266 ke 1.515 x 10<sup>-14</sup> cm<sup>2</sup>/W bagi sistem kaca terdop Gd<sub>2</sub>O<sub>3</sub> dan Gd<sub>2</sub>O<sub>3</sub> NPs masing-masing. Malahan, nilai pekali penyerapan tidak linear berubah dari 0.616 ke 0.747 cm/GW dan 0.660 ke 0.729 cm/GW bagi kedua-dua kaca zink borotellurit terdop Gd<sub>2</sub>O<sub>3</sub> dan Gd<sub>2</sub>O<sub>3</sub> NPs masing-masing. Variasi optik parameter tidak linear adalah disebabkan oleh pembentukan TeO<sub>4</sub>, kehadiran pasangan tunggal elektron dalam TeO<sub>4</sub> dan kesan Gd<sup>3+</sup> ion yang besar serta pengurangan saiz bahan pukal. Hasil ujikaji daripada imbasan-Z mencadangkan bahawa sistem kaca yang diujikaji ini boleh digunakan sebagai reka bentuk bagi bahan optik tidak linear.

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I certify that a Thesis Examination Committee has met on 2 November 2017 to conduct the final examination of Chua Ee Von on her thesis entitled "Effects of Inclusion of Gd<sub>2</sub>O<sub>3</sub> Microparticles and Gd<sub>2</sub>O<sub>3</sub> Nanoparticles on Elastic, Linear and Nonlinear Optical Properties of Zinc Borotellurite Glasses" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

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## LIST OF ABBREVIATIONS

Symbols	Description	Units
Gd <sub>2</sub> O <sub>3</sub>	Gadolinium oxide	-
Gd <sub>2</sub> O <sub>3</sub> NPs	Gadolinium oxide nanoparticles	-
TeO <sub>2</sub>	Tellurium oxide	-
B <sub>2</sub> O <sub>3</sub>	Boron oxide	-
ZnO	Zinc oxide	-
XRD	X-ray diffraction	-
FTIR	Fourier transform infrared spectroscopy	-
TEM	Transmission electron microscopy	-
NBO	Non-bridging oxygen	-
BO	Bridging oxygen	-
tbp	Trigonal bipyramids	-
tp	Trigonal pyramids	-
OPD	Oxygen packing density	-
V <sub>e</sub>	Excess volume	m <sup>3</sup> /mol
V <sub>o</sub>	Oxygen molar volume	m <sup>3</sup> /mol
V <sub>c</sub>	Crystalline volume	m <sup>3</sup> /mol
ρ	Density	kg/m <sup>3</sup>
V <sub>m</sub>	Molar volume	m <sup>3</sup> /mol
v <sub>L</sub>	Longitudinal ultrasonic velocity	m/s
v <sub>S</sub>	Shear ultrasonic velocity	m/s
L	Longitudinal modulus	GPa
S/ G	Shear modulus	GPa

$K$	Bulk modulus	GPa
$E$	Young's modulus	GPa
$\sigma$	Poisson's ratio	-
$v_m$	Mean ultrasonic velocities	m/s
$\theta_D$	Debye temperature	K
$T_s$	Softening temperature	K
$V_t / C_t$	Packing density	-
$G_t$	Dissociation energy	kJ/cm <sup>3</sup>
$n_b$	Number of bonds per unit volume	m <sup>-3</sup>
$l$	Atomic ring size	nm
$\langle n_c \rangle$	Average cross-link density	-
$x_c$	Peak position	cm <sup>-1</sup>
$\lambda_c$	Cut-off wavelength	nm
$\alpha$	Absorption coefficient	cm <sup>-1</sup>
$E_{opt}$	Optical band gap energy	eV
$\Delta E$	Urbach energy	eV
$n_0$	Refractive index	-
$R_m$	Molar refraction	cm <sup>3</sup> /mol
$\alpha_m$	Electronic polarizability	cm <sup>3</sup>
$\alpha_{O^{2-}}$	Oxide ion polarizability	Å <sup>3</sup>
$\Lambda$	Optical basicity	-
$n_2$	Nonlinear refractive	cm <sup>2</sup> /W
$\beta$	Nonlinear absorption coefficient	cm/GW
$\chi^{(3)}$	Third order nonlinear susceptibility	esu

FOM	Figure of merit	-
UVB	Ultraviolet broadband light	-
ESA	Excited state absorption	-



# CHAPTER 1

## INTRODUCTION

### 1.1 Preamble

The studies of materials with high optical nonlinearity have attracted the interests of many researcher due to their important application in fabricating nonlinear optical devices for advanced telecommunication system (Nanda et al., 2015b; Said Mahraz et al., 2013; Paz et al., 2016). Thus, it is important to explore the linear and nonlinear optical behaviour of the materials by estimating the parameters such as optical band gap energy, linear and nonlinear refractive index as well as nonlinear absorption coefficient for desired practical applications. The linear and nonlinear optical properties of different type of materials have been investigated and reported. Among these materials, glasses have been widely received due to their range of composition, the variety in shapes, easy and low cost in mass production, fast response time as well as high optical nonlinearities that are important for fabricating photonic devices (Zaman et al., 2016). In addition, glasses can be doped with wide variety of modifiers or dopants such as heavy metals or rare earth oxides to achieve better linear and nonlinear optical properties for desirable practical applications (Nanda et al., 2015b).

Lately, the glasses doped with lanthanide ions have attracted much more attention due to their potential applications in the design of laser, sensors, optical amplifiers, optoelectronics devices, optical switches and many other applications (Zaman et al., 2016; Liang et al., 2014; Said Mahraz et al., 2013). The presence of 4f electrons in rare earth ions can greatly enhance the nonlinear optical properties of the amorphous material. Besides that, rare earth doped glasses have important properties such as thermal resistant, mechanically strong and chemically stable (Marzouk, 2010; Mahani and Marzouk, 2013; Ami Hazlin et al., 2017).

Amongst all the glasses, tellurite and borate glasses have been widely used as host matrix due to their outstanding characteristics which are attractive for specific applications. TeO<sub>2</sub> glasses possess high refractive index ( $n_0 > 2$ ), high rare earth ion solubility, wide transparent range, low loss phonon energy and low melting point (Halimah et al., 2017; Ami Hazlin et al., 2017; Aziz et al., 2017; Paz et al., 2016). Furthermore, TeO<sub>2</sub> glasses can also be applied as nonlinear optical materials due to their excellent optical properties. On the other hands, B<sub>2</sub>O<sub>3</sub> glass system has low melting point, high transparency, high thermal stability and good solubility of rare earth ions. Borate glasses have potential applications in laser and photonic devices for development of optical technologies (Ami Hazlin et al., 2017; Halimah et al., 2017; Paz et al., 2016; Aziz et al., 2017; Nanda et al., 2015a). Other than that, glasses doped with ZnO are also attractive materials due to their low cost, non-toxicity and non-hygroscopic nature. ZnO can acts either as glass former or as a modifier or both in the vitreous network. The addition of ZnO to borotellurite glass system will produce low rate crystallization, reduce the melting point and improve the glass forming ability



(Ami Hazlin et al., 2017; Said Mahraz et al., 2013). Moreover, the presence of ZnO give rise to good nonlinear optical properties.

Gd<sub>2</sub>O<sub>3</sub> is chosen as a dopant for this study because of its high permittivity and wide energy band gap (5.4 eV) along with good thermal stability that make it as a promising material. Gd<sub>2</sub>O<sub>3</sub> has high luminescence efficiency which make it good host material for luminescence application (Maalej et al., 2015; Singh et al., 2015b). The role of Gd<sub>2</sub>O<sub>3</sub> in oxide glasses is of particular interests because it may serve as a network modifier (Mahani and Marzouk, 2013). The addition of Gd<sub>2</sub>O<sub>3</sub> to the glass system will leads to optically more dispersive with higher refractive index. Besides that, the inclusion of Gd<sub>2</sub>O<sub>3</sub> will increase the nonlinear refractive index and third-order susceptibility of the glass system (Marzouk et al., 2013).

Gd<sub>2</sub>O<sub>3</sub> nanoparticles are also used in this study because glasses doped with rare earth oxide nanoparticles have wide range of resonant absorption frequencies and ultrafast response time which are promising materials for all-optical switching devices. Besides that, it can integrate the large third-order nonlinearity of rare earth ions, excellent thermal and chemical stability of the vitreous network, which associate with the surface plasmon resonance (SPR) for the enhancement effect to electric or light field around the nanoparticles. Meanwhile, the addition of rare earth oxide nanoparticles to the glass system can greatly improve the third-order nonlinear optical response of the amorphous material (Zhong et al., 2014; Aziz et al., 2017).

## 1.2 Problem statements

The search for new material as alternative for nonlinear optical materials has been reported. Nonlinear optical materials are important in the development of optical fibres. The silica optical fibres have been widely used for all optical switching due to their low loss and long inter-action length. However, the low value of third-order nonlinear susceptibility,  $\chi^{(3)}$ , of silica ( $\chi^{(3)} = 2.8 \times 10^{-14}$  esu) requires high switching power and a very long length of fibre to achieve appreciable optical switching application (Senthil Murugan et al., 2006). Besides that, silica-based fibres have been deployed due to zero dispersion and low loss at the wavelength of 1.3  $\mu\text{m}$  which have important impact on telecommunications. Nonetheless, it is difficult to attain efficient gain at 1.3  $\mu\text{m}$  in silicate glasses. It is suggested that glass hosts with lower characteristics phonon energies are expected to yield an environment with considerably lower non-radiative decay rates, which is more favourable than for 1.3  $\mu\text{m}$  emission. Thus, by using glass host other than silica glasses can enhance excited-state absorption (ESA) or non-radiative decay and 1.3  $\mu\text{m}$  devices may be feasible. Hence, a wide range of glass hosts such as silica phosphates and fluorophosphates, sulphides and other chalcogenides glasses have been investigated but had shown generally low performance (Wang et al., 1994).

It is known that tellurite based glasses are potential amorphous materials for the development of photonic materials application. This is because tellurite based glass have good optical switching response, excellent optical nonlinearity, low optical loss, excellent chemical durability and thermal stability (Yousef et al., 2012). Furthermore, it is reported that tellurite glasses have high refractive index ( $\sim 1.8-2.3$ ), large nonlinear refractive index (about 25 times higher than silica based glasses) and low phonon energy (the highest phonon energy about  $800 \text{ cm}^{-1}$ ). The high index is desirable for the increment of the local field correction at a rare earth site which lead to large radiative transition probabilities (Wang et al., 1994). In addition, tellurite glasses have low melting points and high solubility to rare earth ions. The addition of rare earth oxide and rare earth oxide nanoparticles to the tellurite based glass systems are able to enhance the optical properties of the glass systems. Gadolinium oxide ( $\text{Gd}_2\text{O}_3$ ) is significant rare-earth element because of its compounds having excellent electric, optical and magnetic properties (Singh et al., 2015b). Besides that, it also has high luminescence efficiency, acts as attractive role as active ions in optical materials and useful for the development of photonic devices. The inclusion of metal nanoparticles into vitreous matrices can also further improve the third-order nonlinear optical response of the glassy material (Zhong et al., 2014).

However, there are limited information on the effect of gadolinium oxide and gadolinium oxide nanoparticles on elastic, linear and nonlinear optical properties. Therefore, the investigation on gadolinium oxide and gadolinium oxide nanoparticles doped zinc borotellurite glass system are still needed since there are limited data such as elastic, linear and nonlinear optical properties to support their future optical applications. Hence, the purpose of this work is to study the effect of gadolinium oxide and gadolinium oxide nanoparticles on elastic, linear and nonlinear optical properties of zinc borotellurite glass system.

### **1.3 Scope of study**

The purpose of the scope of study is to limit the field of study in this work and to obtain specific parameters that are required for the application of nonlinear optic. The scope of the study in this work is limited to structural properties (XRD, FTIR and TEM), physical properties (density and molar volume), elastic properties (experimental elastic moduli and theoretical elastic moduli calculated using Makishima-Mackenzie model, Rocherulle model, bond compression model and ring deformation model), linear optical properties (optical band gap, urbach energy, refractive index, electronic polarizability, oxide ion polarizability, optical basicity and metallization criterion) and nonlinear optical properties (nonlinear refractive index, nonlinear absorption coefficient, third-order nonlinear susceptibility and figure of merit). The elastic, linear and nonlinear optical properties obtained are for required for the application of nonlinear optic material.

## 1.4 Research objectives

1. To synthesize gadolinium oxide and gadolinium oxide nanoparticles doped zinc borotellurite glass system with chemical formula,  $\{[(\text{TeO}_2)_{70}(\text{B}_2\text{O}_3)_{30}]_{70}(\text{ZnO})_{30}\}_{1-x}(\text{Gd}_2\text{O}_3)_x$  and  $\{[(\text{TeO}_2)_{70}(\text{B}_2\text{O}_3)_{30}]_{70}(\text{ZnO})_{30}\}_{1-x}(\text{Gd}_2\text{O}_3 \text{ NPs})_x$ , where  $x = 1.0, 2.0, 3.0, 4.0$  and  $5.0$  mol% by using conventional melt-quenching technique.
2. To study the physical and elastic properties of gadolinium oxide and gadolinium oxide nanoparticles doped zinc borotellurite glass system by using pulse-echo technique as well as to correlate and compare the experimental values and calculated theoretical elastic moduli using Makishima-Mackenzie model, Rocherulle model, Bond compression model and ring deformation model.
3. To analyse the effect of gadolinium oxide and gadolinium oxide nanoparticles on linear and nonlinear optical properties of zinc borotellurite glass system.

## 1.5 Hypothesis

### 1.5.1 Structural properties

The addition of  $\text{Gd}_2\text{O}_3$  and  $\text{Gd}_2\text{O}_3$  NPs to the glass system does not leads to any distinguishable sharp peak but denote a broad hump implying the characteristic of amorphous materials (Ami Hazlin et al., 2017; Faznny et al., 2017; Kaur et al., 2016).

The expected structural units of the glass system present in the FTIR spectra are  $\text{TeO}_3$ ,  $\text{TeO}_4$ ,  $\text{BO}_3$  and  $\text{BO}_4$  (Azlan et al., 2013; Said Mahraz et al., 2013; Rada et al., 2008b).

### 1.5.2 Physical properties

The density of the glass system is expected to increase with  $\text{Gd}_2\text{O}_3$  and  $\text{Gd}_2\text{O}_3$  NPs content. This is because  $\text{Gd}_2\text{O}_3$  and  $\text{Gd}_2\text{O}_3$  NPs has higher molecular weight as compared to  $\text{TeO}_2$ - $\text{B}_2\text{O}_3$ - $\text{ZnO}$  (Pavani et al., 2012; Kundu et al., 2014; Veeranna Gowda, 2015).

The molar volume is expected to have the opposite trend to density of the glass system. This is due to the fact that density is inversely proportional to the molar volume (Kaur et al., 2010).

### 1.5.3 Elastic properties

The elastic moduli of the glass system are expected to increase with  $\text{Gd}_2\text{O}_3$  and  $\text{Gd}_2\text{O}_3$  NPs concentration. This is because the increase in density leads to increase in compactness and rigidity of the glass as well as increasing elastic moduli (Kanappan et al., 2009; Krishna et al., 2008; Yousef et al., 2016; Saddeek et al., 2010b).

#### 1.5.4 Linear optical properties

The addition of  $Gd_2O_3$  and  $Gd_2O_3$  NPs is expected to enhance the linear optical properties of the glass system. The optical band gap is expected to increase with increasing  $Gd_2O_3$  and  $Gd_2O_3$  NPs concentration. This can be attributed to the higher bond energy of Gd-O bond as compared to Te-O bond and B-O bonds which results in higher average bond energy and thus, increasing optical band gap (Luo, 2007; Abdel-Baki et al., 2012).

The addition of  $Gd_2O_3$  and  $Gd_2O_3$  NPs to zinc borotellurite glass systems will enhance the refractive index. This is due to high polarization of large size  $Gd^{3+}$  cations (El-Mallawany et al., 2008).

#### 1.5.5 Nonlinear optical properties

The inclusion of  $Gd_2O_3$  and  $Gd_2O_3$  NPs into zinc borotellurite glass system is expected to cause increment in the nonlinear refractive index. The nonlinear refractive index is directly proportional to the linear refractive index of the glass system. Hence, the higher value of linear refractive index will result in higher nonlinear refractive index (Nanda et al., 2015b; Chen et al., 2008).

The addition of  $Gd_2O_3$  and  $Gd_2O_3$  NPs to the glass system will decrease the nonlinear absorption coefficient. This is because the presence of free carriers in the 4f shell transition in  $Gd^{3+}$  ions will cause reduction of nonlinear absorption coefficient (Van Stryland and Sheik-Bahae, 1998).

### 1.6 Outline of thesis

The overall presentation of this thesis consists of the overview of the research work, literature review of different types of glasses related to the current studies, the theoretical description underpinning this work, the fabrication of rare earth oxide ( $Gd_2O_3$  and  $Gd_2O_3$  NPs) doped zinc borotellurite glass series, the experimental procedure for each characterization and experimental results taken as well as analysing the effect of  $Gd_2O_3$  and  $Gd_2O_3$  NPs on structural characteristic, physical properties, elastic properties, linear and nonlinear optical properties of zinc borotellurite glasses. The thesis is divided into 6 chapters and each chapter is sub-divided into sub-sections.

Chapter 1 gives an overview to the research, a brief introduction of the glass material and rare earth oxide ( $Gd_2O_3$  and  $Gd_2O_3$  NPs) used. It also emphasized the core of the research, the problem statement and the research objectives.

Chapter 2 of this thesis gives a brief literature review on tellurite glass system and gadolinium doped glass system. Besides that, the effect of composition on structural, physical, elastic, linear and nonlinear optical properties were reviewed.

Chapter 3 of this thesis highlights the theoretical aspects of this work, including the definition of glass, basic theory of glass formation, the structure of tellurite and borate glasses. Apart from that, the theoretical model used for calculated theoretical elastic moduli such as Makishima-Mackenzie model, Rocherulle's model, Bond compression model as well as ring deformation model are also included in this chapter. In addition, this chapter also describe briefly the equations and important parameters used to obtained linear and nonlinear optical properties.

Chapter 4 discussed the method used for glass fabrication and the characterization techniques for structural, physical, optical and elastic properties. Conventional melt-quenching technique is used to fabricate glass samples. The structural properties characterization includes X-ray diffraction (XRD), Fourier transform infrared (FTIR) and Transmission electron microscopy (TEM). Pulse-echo technique was employed to obtain the elastic properties characterization. The physical properties characterization involves density measurements and molar volume calculation. UV-Vis spectroscopy and Z-scan technique are used for linear and nonlinear optical properties characterization.

Chapter 5 highlighted the experimental results taken and data analyses for each characterization. The results obtained for structural properties, physical properties, elastic properties, linear and nonlinear optical properties were discussed. The comparative studies between the experimental and theoretical elastic moduli as well as the comparatives studies between the current glass samples and the previous studies were also covered in this chapter.

Chapter 6 summarized the important outcome of the overall research and suggestions of the future studies.

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