

# **UNIVERSITI PUTRA MALAYSIA**

EFFECT OF COBALT OXIDE ON OPTICAL AND DIELECTRIC PROPERTIES OF WILLEMITE-BASED GLASS-CERAMICS USING WHITE RICE HUSK ASH AS SILICA SOURCE

SITI AISYAH BINTI ABDUL WAHAB

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By

SITI AISYAH BINTI ABDUL WAHAB

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirement for the Degree of Doctor of Philosophy

June 2021

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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

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#### SITI AISYAH BINTI ABDUL WAHAB

June 2021

Chairman : Khamirul Amin bin Matori, PhD Institute : Nanoscience and Nanotechnology

Nowadays, researchers are interested in the production of willemite  $(Zn_2SiO_4)$  since it has good phosphor properties in optoelectronic applications. However, still lack of study in the fabrication of cobalt oxide (Co<sub>3</sub>O<sub>4</sub>) doped willemite based glass-ceramics (Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup>) derive from white rice husk ash (WRHA). The Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> were fabricated based empirical formula of  $(Co_3O_4)_v[(ZnO)_{0.55}(WRHA)_{0.45}]_{1-v}$  where v = 0.0, 0.1, 0.5, 1.0 wt.% heat-treated at 700-950 °C using melt-quenching technique. This research focuses on the effect of heat treatment and effect of Co<sub>3</sub>O<sub>4</sub> doping on the physical, structural, optical, and dielectric properties of Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup>. The densities of undoped-Zn<sub>2</sub>SiO<sub>4</sub> and Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> increased from 3.4138 to 3.4659 g/cm<sup>3</sup> as dopant increases, also the linear shrinkage increased from 6.23 to 6.92% when dopant increased. X-ray Diffraction (XRD) shows the formation of  $\beta$ -Zn<sub>2</sub>SiO<sub>4</sub> at 750 °C, then achieve stable state of  $\alpha$ -Zn<sub>2</sub>SiO<sub>4</sub> at 950 °C. Meanwhile, the crystallite size was increased from 74.47 to 74.68 nm then decreased to 73.18 nm as dopant increased. Field Emission Scanning Electron Microscopy (FESEM) shows no obvious changes as the dopant increased. However, at 950 °C, Zn<sub>2</sub>SiO<sub>4</sub>: 0.5 wt.% Co<sup>2+</sup> and Zn<sub>2</sub>SiO<sub>4</sub>: 1.0 wt.% Co<sup>2+</sup> showed a larger grain and less porosity compared to other samples. Fourier Transform Infrared (FTIR) spectroscopy showed eight significant vibrational bands of  $Zn_2SiO_4$  at 750-950 °C for both undoped and doped samples. The presence of SiO<sub>4</sub> and ZnO<sub>4</sub> bands in the FTIR absorption spectrum prove the formation of Zn<sub>2</sub>SiO<sub>4</sub>. The absorption spectra of UV-Visible were recorded in the range of 220-800 nm and the absorption band of undoped- Zn<sub>2</sub>SiO<sub>4</sub> shifted to lower wavelengths (370 and 349 nm) at 900 and 950 °C respectively. When Co<sub>3</sub>O<sub>4</sub> was introduced to Zn<sub>2</sub>SiO<sub>4</sub> two absorption spectra occurred which is at 450 -700 nm and ~325 nm attributed to  ${}^{4}A_{2} \rightarrow {}^{4}T_{1}$  (<sup>4</sup>P) transitions. The optical band gap was increased as dopants were introduced to Zn<sub>2</sub>SiO<sub>4</sub> from ~4.09 eV to 4.57 eV then decreased to 4.29 eV. Photoluminescence spectroscopy (PL) showed blue emissions at ~420, ~444, ~464 and ~485 nm and green emission at ~525 nm under 325 nm excitation which attributed to the transition of  $\text{Co}^{2+}$  from  ${}^{4}\text{A}_{2} \rightarrow {}^{4}\text{T}_{1}({}^{4}\text{P})$ . Besides, the dielectric constant increased from 4.84047 to 5.52423 when  $Co_3O_4$  increase due to



enhancement of the crystallinity and decrement of the polarization at higher temperatures. The dielectric loss remained low with the increase of dopant and AC conductivity showed each sample has different range of frequency cut-off which is at ~1.2, ~1.7, ~1.3 and ~1.4 GHz for undoped-Zn<sub>2</sub>SiO<sub>4</sub>, Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> at 0.1, 0.5 and 1.0 wt.% respectively. The differences of frequency cut-off might occur due to the difference in ability of each sample to transport the electron when electric field is applied. In conclusion, the ability of the Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> to exhibit blue emission and able to operate at higher frequency, also low loss makes it suitable to be used as a phosphor material in optoelectronic applications.



Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk Ijazah Doktor Falsasah

## KESAN KOBALT OKSIDA TERHADAP SIFAT OPTIK DAN DIELEKTRIK WILLEMIT-BERASASKAN KACA-SERAMIK MENGGUNAKAN ABU PUTIH SEKAM PADI SEBAGAI SUMBER SILIKA

Oleh

#### SITI AISYAH BINTI ABDUL WAHAB

Jun 2021

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Pada masa kini, para penyelidik sangat berminat dalam penghasilan willemit  $(Zn_2SiO_4)$ memandangkan ia mempunya sifat fosfor yang baik di dalam aplikasi optoelektronik. Walau bagaimanapun, kajian dalam penghasilan kobalt oksida (Co<sub>3</sub>O<sub>4</sub>) dop willemit berasaskan kaca seramik (Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup>) dihasilkan daripada abu putih sekam padi (WRHA) masih kurang. Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> telah dihasilkan berdasarkan formula emperik  $(Co_3O_4)_{v}[(ZnO)_{0.55}(WRHA)_{0.45}]_{1-v}$  yang mana y = 0.0, 0.1, 0.5, 1.0 wt.% dan dipanaskan pada 700-950 °C menggunakan teknik peleburan-pelindapan. Kajian ini menumpukan kesan rawatan haba dan kesan pendopan Co<sub>3</sub>O<sub>4</sub> terhadap ciri-ciri fizikal, struktur, optik dan dielektrik Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup>. Ketumpatan Zn<sub>2</sub>SiO<sub>4</sub>-tidak didop dan Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> meningkat daripada 3.4138 kepada 3.4659 g/cm<sup>3</sup> apabila dopan meningkat manakala pengecutan linear juga meningkat daripada 6.23 kepada 6.92% apabila dopan meningkat. Pembelaun sinar-X (XRD) menunjukkan pembentukan  $\beta$ -Zn<sub>2</sub>SiO<sub>4</sub> pada suhu 750 °C, kemudian mencapai keadaan stabil (α-Zn<sub>2</sub>SiO<sub>4</sub>) pada 950 °C. Manakala, saiz kristalit meningkat daripada 74.47 kepada 74.68 nm kemudian menurun kepada 73.18 apabila dopan meningkat. Mikroskopi pancaran medan elektron penskanan (FESEM) menunjukkan tiada perubahan ketara apabila dopan meningkat. Walau bagaimanapun, pada 950 °C, Zn<sub>2</sub>SiO<sub>4</sub>: 0.5 wt.% Co<sup>2+</sup> dan Zn<sub>2</sub>SiO<sub>4</sub>: 1.0 wt.% Co<sup>2+</sup> menunjukkan butiran yang lebih besar dan keronggaan yang kurang berbanding sampel lain. Spektroskopi transformasi fourier inframerah (FTIR) menunjukkan bahawa terdapat lapan jalur getaran Zn<sub>2</sub>SiO<sub>4</sub> yang ketara pada suhu 750-950 °C bagi sample yang tidak didop dan didop. Kehadiran jalur SiO4 dan ZnO4 di dalam spektum penyerapan FTIR membuktikan penghasilan Zn<sub>2</sub>SiO<sub>4</sub>. Penyerapan spectra UV cahaya nampak (UV-Vis) dirakam dalam julat 220-800 nm dan jalur penyerapan untuk Zn<sub>2</sub>SiO<sub>4</sub>-tidak didop berganjak kepada julat gelombang yang lebih rendah (370 dan 349 nm) pada suhu 900 dan 950 °C. Apabila Co<sub>3</sub>O<sub>4</sub> diperkenalkan kepada Zn<sub>2</sub>SiO<sub>4</sub>, berlaku dua penyerapan spektra iaitu pada 450 -700 nm dan pada ~325 nm disebabkan peralihan  ${}^{4}A_{2} \rightarrow {}^{4}T_{1}$  (<sup>4</sup>P). Jurang jalur optik

meningkat apabila dopan diperkenalkan kepada Zn<sub>2</sub>SiO<sub>4</sub> daripada ~4.09 eV kepada 4.57 eV kemudian menurun kepada 4.29 eV. Spektroskopi kefotopendarcahayaan (PL) menunjukkan pancaran biru pada ~420, ~444, ~464 dan ~485 nm, dan pancaran hijau pada ~525 nm di bawah pengujaan 325 nm yang mana pancaran tersebut disebabkan oleh peralihan  $\text{Co}^{2+}$  daripada  ${}^{4}\text{A}_{2} \rightarrow {}^{4}\text{T}_{1}({}^{4}\text{P})$ . Selain itu, pemalar dielektrik meningkat daripada 4.84047 kepada 5.52423 apabila Co<sub>3</sub>O<sub>4</sub> meningkat disebabkan oleh peningkatan penghabluran dan pengurangan polarisasi pada suhu yang tinggi. Kehilangan dielektik kekal rendah apabila dopan meningkat dan kekonduksian AC menunjukkan setiap sampel mempunyai julat frekuensi penggal yang berbeza iaitu masing-masing pada ~1.2, ~1.7, ~1.3 and ~1.4 GHz. Perbezaan frekuensi penggal mungkin berlaku kena perbezaan kemampuan setiap sampel untuk membawa elektron apabila medan elektrik dibekalkan. Kesimpulannya, kemampuan Zn<sub>2</sub>SiO<sub>4</sub>: Co<sup>2+</sup> untuk memancarkan pancaran biru dan boleh beroperasi pada frekuensi yang tinggi, serta kehilangan dielektik yang rendah menjadikan ia sesuai untuk digunakan sebagai bahan fosfor di dalam aplikasi optoelektronik.

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This thesis was submitted to the Senate of the Universiti Putra Malaysia and has been accepted as fulfillment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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# LIST OF ABBREVIATIONS

CB	Conduction band
DSC	Differential Scanning Calorimetry
FESEM	Field emission scanning electron microscopy
FTIR	Fourier transform infrared
FWHM	Full width half maximum
LED	Light-emitting diodes
NBOs	Non-bridging oxygen
Oi	Oxygen interstitial
PDPs	Plasma display panels
PL	Photoluminescence
PVA	Polyvinyl alcohol
RE	Rare earth
RH	Rice husk
RHA	Rice husk ash
SRO	Short-range order
T <sub>c</sub>	Crystallization peak temperature
Tg	Glass transition temperature
T <sub>m</sub>	Melting temperature
ТМ	Transition metal
UV-Vis	Ultra-violet Visible
VB	Valance band
Vo	Oxygen vacancies
V <sub>zn</sub>	Zinc vacancy
W-LEDS	White light-emitting diodes

G

WRHA White rice husk ash

XRD X-ray diffraction

 $Zn_2SiO_4: Co^{2+}$  Willemite doped cobalt oxide

Zn<sub>2</sub>SiO<sub>4</sub> Willemite

6



#### **CHAPTER 1**

#### INTRODUCTION

### 1.1 Research background

Nowadays, silicate glasses that have high zinc oxide (ZnO) content are attractive due to their various applications, from both technical glasses and glass-ceramics to highperformance optical glasses (Bondioli et al., 2010). In this research, the fabrication of zinc silicate glasses and glass-ceramics will be done and investigated. The major components that made up zinc silicate glass and glass-ceramics are ZnO and silicon dioxide, SiO<sub>2</sub> (Khaidir et al., 2020). Rice husk (RH) is one of the agricultural by-products that can be used to produce SiO<sub>2</sub>. Burning of RH at a certain temperature can produce white rice husk ash (WRHA) with approximately 87 to 97% silica and a small amount of other metallic impurities (Yalcin & Sevinc, 2001; Tuscharoen et al., 2013). Lee et al., (2017b) reported that burning RH at 1000 °C for 2 hours produced about 95.60% of silica (SiO<sub>2</sub>) without any acid leaching process. This high in silica content from WRHA, making it the most compatible replacer of commercial silica to produce zinc silicate glass and glass-ceramics (Khaidir et al., 2019a).

Zinc silicate or its mineral name willemite  $(Zn_2SiO_4)$  is very familiar amid researchers as the best and most favourable host matrix in glass phosphor for the optoelectronic application due to its phenakite structure (Tarafder et al., 2014). Besides, this rigid lattice of  $Zn_2SiO_4$  also permits it to be utilized as the enhancer for generating the light inside the fluorescent lamp, neon discharged lamps, colour television, black-and-white television. waveguides, laser technology, optical fibre amplifiers, optical communications, oscilloscopes, and light-emitting diodes (LED) (Sarrigani et al., 2015a; Zaid et al., 2015; Effendy et al., 2017). Several studies had been done by previous researchers regarding Zn<sub>2</sub>SiO<sub>4</sub> doped transition metal (TM) and rare earth (RE) (Tarafder et al., 2013; Sarrigani et al., 2015a; Effendy et al., 2016; Samsudin et al., 2016a; Babu et al., 2017; Mohamed et al., 2017; Rasdi et al., 2017a; Omar et al., 2017; Zaid et al., 2017a; Zamratul et al., 2017; Azman et al., 2018; Khaidir et al., 2019b). In this research, fabrication, and characterization of Zn<sub>2</sub>SiO<sub>4</sub> based glass-ceramics derived from WRHA doped with cobalt oxide  $(Co_3O_4)$  by using conventional melt-quenching methods were done.

 $Zn_2SiO_4$  doped  $Co_3O_4$  ( $Zn_2SiO_4$ :  $Co^{2+}$ ) is a promising blue ceramic pigment (Ozel et al., 2010). The deep blue colour of the  $Zn_2SiO_4$ :  $Co^{2+}$  crystals and the characteristic threeband systems around 2632, 1429 and 588 nm in the corresponding absorption spectra are typical for tetrahedral  $Co^{2+}$  (Brunold et al., 1996). As in the case of the well-known spinel pigment, deep blue colour is obtained by doping the  $Zn_2SiO_4$  structure with cobalt, that replaces zinc ions in tetrahedral positions (Ozel et al., 2010) wherein groups of tetrahedral  $Co(II)O_4$ , the first two spins allowed bands fall in the infrared region ~1400 nm and 1600 nm, only the third allowed band available in the visible region and usually triple-band around 540 nm (green region), 590 nm (yellow-orange region) and 640 nm (red region) that gives rise to the blue colour (Babu & Buddhudu, 2014). In terms of emission spectroscopy,  $Zn_2SiO_4$ :  $Co^{2+}$  sol-gel revealed a red emission at 631 nm (Babu & Buddhudu, 2014) while Rasdi et al., (2017b) reported  $Zn_2SiO_4$ :  $Co^{2+}$  prepared by sol-gel methods exhibit blue emission (420 and 480 nm) and green emission (525 nm) when excited at 350 nm. Those peaks were attributed to the d-d transition of  $Co^{2+}$  from  ${}^{4}A_2 \rightarrow {}^{4}T_1$  (<sup>4</sup>P) (Choudhury A & Choudhury B, 2012; Rasdi et al., 2017b). These excellent optical properties of  $Zn_2SiO_4$ :  $Co^{2+}$  make it a good candidate to be used as blue and green phosphors for luminescence optical materials.

Aside from that, dielectric properties also give good information for  $Zn_2SiO_4$  to be a phosphor material. According to Sukriti and co-workers, the material that has a lower dielectric constant and dielectric loss in the higher frequency section will be much suitable to be used in devices that have a high-frequency application such as LED (Sukriti & Chand, 2019b). Therefore, to investigate more on the capability of  $Zn_2SiO_4$ : Co<sup>2+</sup> to be used as a phosphor material, hence its optical and dielectric properties needed to be studied. In this study the  $Zn_2SiO_4$ : Co<sup>2+</sup> will be fabricated and characterized after undergoing heat treatment at several temperatures starting from 750 °C to 950 °C to observe the formation of willemite. After that, the physical, structural, optical, and dielectric properties of  $Zn_2SiO_4$ : Co<sup>2+</sup> based glass-ceramics will be studied.

## 1.2 Problem statement

In recent years, the abundance of rice husk waste had been a huge concern among environmentalists. Thus, various ways had been introduced to utilize the uses of RH such as admixture for cement, the source for energy in rural areas, filler, carbon capture, adsorbent materials, and source of SiO<sub>2</sub> (Pode, 2016). WRHA had been talked by the researchers as its high amount of silica content which can be very beneficial towards the development of current technology (Yalcin & Sevinc, 2001; Tuschareon et al., 2013; Ruengsri et al., 2015; Bakar et al., 2016; Fernandes et al., 2017). According to Bondioli et al., (2010), using SiO<sub>2</sub> from WRHA, the same type of industrial glass can be produced with the same glass characteristic made from commercial silica. SiO<sub>2</sub> from WRHA was acknowledged to be the substituent of commercial silica due to its high silica content. The commercial silica is very expensive (Azman et al., 2018), hence, using WRHA as the substituent, will reduce the cost of production as well as reduce the pollution if the RH were burned in an open space (Bakar et al., 2016).

Glass plays an important role in optical applications such as data transmission, sensor detection, sensor technology and is a good candidate for solid-state lasers (Zaid et al., 2016). ZnO–SiO<sub>2</sub> glasses are among the glasses that had been extensively studied by many researchers due to their great luminescence properties. However, the effect of zinc oxide (ZnO) content on the physical, structural, and optical properties of zinc silicate glasses derived from WRHA of ZnO–SiO<sub>2</sub> glasses are rarely to be found. This is because most of the researchers are focusing on the studies of zinc silicate glass and glass-ceramics doped transition metal or rare-earth. Therefore, in this present study, an extensive discussion regarding the photoluminescence studies of ZnO–SiO<sub>2</sub> glass will be presented.

Apart from that, nowadays glass-ceramics had been great attention among researchers due to their attractive properties. The transformation of glass into glass-ceramics has progressively become an important technique to improve the quality and properties of the zinc silicate glass such as the physical endurance and the luminescene properties. To produce  $ZnO-SiO_2$  glass-ceramics, the zinc silicate glasses need to be heat treated at a certain temperature. Therefore, the heat treatment process needs to be conducted to form the glass-ceramcis. After the heat treatment process,  $ZnO-SiO_2$  glass will form zinc silicate glass-ceramics also known as willemite (Zn<sub>2</sub>SiO<sub>4</sub>). Zn<sub>2</sub>SiO<sub>4</sub> is a promising phosphor for optoelectronic devices (Azman et al., 2018). Among oxide-based phosphor,  $Zn_2SiO_4$  was identified as one of the most compatible host matrixes for many transitions metal (TM) and rare-earth (RE) ions (Rasdi et al., 2017a). The doping of Zn<sub>2</sub>SiO<sub>4</sub> is no longer new (Sariggani et al., 2015; Azman et al., 2018). Among the dopant that had used are europium oxide ( $Eu_2O_3$ ), manganese oxide ( $Mn_2O_3$ ), erbium oxide ( $Er_2O_3$ ), thulium oxide  $(Tm_2O_3)$ , neodymium oxide  $(Nd_2O_3)$  and praseodymium oxide  $(Pr_6O_{11})$  (Tarafder et al., 2013; Sarrigani et al., 2015a; Effendy et al., 2016; Samsudin et al., 2016a; Babu et al., 2017; Mohamed et al., 2017; Rasdi et al., 2017a; Omar et al., 2017; Zaid et al., 2017a; Zamratul et al., 2017; Azman et al., 2018; Khaidir et al., 2019b). These dopants had enhanced the luminescence properties of  $Zn_2SiO_4$ , allowing it to emit various visible colours such as green, yellow, red, and blue.

Green emission can be acquired using Eu<sub>2</sub>O<sub>3</sub>, Mn<sub>2</sub>O<sub>3</sub>, Pr<sub>6</sub>O<sub>11</sub> and Er<sub>2</sub>O<sub>3</sub> as a dopant. In contrast, yellow emission can be obtained by doping  $Zn_2SiO_4$  with Eu<sub>2</sub>O<sub>3</sub> and Mn<sub>2</sub>O<sub>3</sub>, while red emission was usually acquired by doping Zn<sub>2</sub>SiO<sub>4</sub> with Eu<sub>2</sub>O<sub>3</sub>, Mn<sub>2</sub>O<sub>3</sub>, Pr<sub>6</sub>O<sub>11</sub> (Samsudin et al., 2015; Omar et al., 2016d; Zaid et al., 2017a; Khaidir et al., 2019b; Mohamed et al., 2017; Effendy et al., 2016). From here, it can be concluded that the green, yellow, and red emissions of Zn<sub>2</sub>SiO<sub>4</sub> doped TM or RE ions are widespread and possible to get. However, the blue emission of Zn<sub>2</sub>SiO<sub>4</sub> is quite low in number to be found. Other authors reported that Co<sup>2+</sup>-doped ZnO also could give blue emission (Sujinnapram et al., 2009). The same result had been found by Li et al., (2010), where the Co<sup>2+</sup>-doped ZnO also gives arise to blue emission corresponding to the near band edge (NBE) transition and due to oxygen vacancies (Li et al., 2010). Another research that used  $Co_3O_4$  as a dopant had been carried out by Manickam et al., (2016), and the research findings were the visible blue emission had been produced centred at 446 nm. The research regarding the origin of blue emission still had been continued since the blue emission is vital to make white light-emitting diodes (W-LEDs). According to Cho et al., (2017), white light formation includes three primary colours; green, red, and blue (Figure 1.1a and 1.1b). Thus, this shows the importance of blue emission in the progression of current technology that mostly used white light. Zamratul et al., (2017) reported blue emission findings in  $Zn_2SiO_4$  by doping it with Nd<sub>2</sub>O<sub>3</sub>. Still, the blue emission is not the primary emission, and it is just a shoulder of emission accompanied by other emissions such as green and yellow emission. Other than that, Rasdi et al., (2017b) also reported willemite doped  $Co_3O_4$  able to produce blue light at 420 and 480 nm when excited at 350 nm. However, the author is using the sol-gel method, and it is quite expensive since it used pure chemicals. Based on the previous research, the blue emission of any host materials is possible to be produced by using  $Co_3O_4$  as the dopant. Therefore, in this research, the author would like to introduce studies regarding the Zn<sub>2</sub>SiO<sub>4</sub> based glassceramics doped Co<sub>3</sub>O<sub>4</sub> derived from WRHA. Since our aim is also to utilize the uses of WRHA, thus conventional melt and quenching methods are the best ways that can be used.





### 1.3 Objectives

In this research, the study's focus is to fabricate and enhance the optical and dielectric properties of  $Zn_2SiO_4$  based glass-ceramics from WRHA. Thus, this project includes producing the glass phosphor, heat-treating the precursor glass, and doping the  $Zn_2SiO_4$  with  $Co_3O_4$ .

The objectives of the research are:

- 1. To synthesize the zinc silicate glass and Zn<sub>2</sub>SiO<sub>4</sub> based glass-ceramics doped Co<sub>3</sub>O<sub>4</sub> using WRHA as silica source.
- 2. To study the impact of different ZnO content on the thermal, physical, structural, and optical properties of zinc silicate glass.
- 3. To investigate the effect of heat treatment on the physical, structural, optical, and dielectric properties of zinc silicate glass and glass-ceramics.
- 4. To analyze the influence of Co<sub>3</sub>O<sub>4</sub> doping on the physical, structural, optical, and dielectric properties of Zn<sub>2</sub>SiO<sub>4</sub> based glass-ceramics derived from WRHA.

### 1.4 Scope of the study

At first, five series of precursor glasses were fabricated, then the chosen precursor glass will be heat-treated and lastly, the chosen glasses will be doped with  $Co_3O_4$ . Therefore, the scope of the study of this research are as follows:

- 1. A series of ZnO–WRHA glasses were fabricated based on empirical formula  $(ZnO)_x(WRHA)_{1-x}$  where x = 0.50, 0.52, 0.55, 0.57 and 0.60 wt.%
- 2. The precursor glasses were analyzed by using DSC, XRD, FTIR, UV-Vis and PL.
- 3. The precursor glass was subjected to a heat treatment process at 700 °C to 950 °C to produce Zn<sub>2</sub>SiO<sub>4</sub> based glass-ceramics.
- 4. Four series of  $Zn_2SiO_4$  based glass-ceramics derived from WRHA doped  $Co_3O_4$  were fabricated based on the empirical formula  $(Co_3O_4)_y[(ZnO)_{0.55}(WRHA)_{0.45}]_{1-y}$  where  $y = 0.0 \ 0.1, \ 0.5, \ and \ 1.0 \ wt.\%$  by using conventional melt-quenching and control heat treatment method.
- 5. The physical, structural, optical, and dielectric properties of Zn<sub>2</sub>SiO<sub>4</sub> based glassceramics doped Co<sub>3</sub>O<sub>4</sub> were analysed by using bulk density, true density, linear shrinkage, XRD, FESEM, FTIR, UV-Vis, PL and impedance analyzer.

### 1.5 Outline of the thesis

In this study, Chapter 1 gave information about the element that had been used as the silica source which is WRHA. Then, continued with then ZnO-WRHA glasses, glass-ceramics and  $Zn_2SiO_4$  based glass-ceramics doped  $Co_3O_4$ . Meanwhile, in Chapter 2, the RH literature review, types of oxide glasses and previous studies of  $Zn_2SiO_4$  doped glass-ceramics had been referred. Besides, the common characterization of  $Zn_2SiO_4$  dopes glass-ceramics also had been observed. In Chapter 3, the method of carrying out the research, calculation of the result and type of characterization had been discussed. Further discussion regarding the ZnO-WRHA precursor glasses, the effect of heat treatment on the zinc silicate glass-ceramics, and cobalt dopant influence on the  $Zn_2SiO_4$  based glass-ceramics were reported in Chapter 4. Lastly, in Chapter 5, the conclusion from the overall research was done. Some suggested future work is also written in this chapter.

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