EFFECT OF ACTIVATED CARBON TO THE STRUCTURAL PROPERTIES OF COPPER ZINC FERRITE PREPARED VIA MECHANOSYNTHESIS

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Thesis submitted in fulfillment of the requirements
for the award of the degree of
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This thesis is dedicated to my parents

For their endless love, support and encouragement.

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This research on "The Effect of Activated Carbon to the Structural Properties of Copper Zinc Ferrite Prepared via Mechanosynthesis" has been signed to me as part of the curriculum in my 4 years of Bachelor Degree.

I have tried my best to present this research as clearly as possible using scientific terms that I hope will be comprehended by the widest spectrum of researchers, analyst and students for further studies.

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Thank you.

ABSTRACT

In this research, copper zinc ferrite nanoparticles, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ were successfully produced by mechanosynthesis method. $Cu_{0.2}Zn_{0.8}Fe_2O_4$ nanoparticle precursor were produced dried Fe_2O_3, CuO and ZnO were mixed with a molar ratio of 1:0.1:0.4 to form the mixture. After ball milled for 8 hours at 600 rpm with ball to powder ratio of 10:1, the brownish powder of copper zinc ferrite nanoparticles, dried at 120 °C in air and then were calcined at 750 °C for an hour. The activated carbon were produced from sawdust via pyrolysis method using nitrogen gas. Copper zinc ferrite nanoparticles were added with activated carbon with four different weight percentage of copper zinc ferrite and activated carbon and then were mixed homogenously by sol-gel method with the presence of absolute ethanol as fuel. The amorphous precursor of carbon copper zinc ferrite then characterized by Fourier Transform Infrared Spectroscopy (FTIR) Study , X-Ray Powder Diffraction (XRD) Analysis and Scanning Electron Microscopy (SEM) for the structural measurement of carbon copper zinc ferrite nanoparticle. The effect of activated carbon to the $Cu_{0.2}Zn_{0.8}Fe_2O_4$ were investigated. It was found that the activated carbon does not interrupt the magnetic properties of the $Cu_{0.2}Zn_{0.8}Fe_2O_4$ but the structure of the magnetite become lighter and more porous as the percentage weight of carbon is increasing. From the XRD data, at the weight percentage of 40%wt of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and 60% wt of activated carbon, the peak of magnetite at (3 1 1) was the highest and most sharp. Proving the by adding carbon to the copper zinc ferrite with that respective percentage weight does not affect the properties of copper zinc ferrite but does affect its structure.

ABSTRAK

Sebuah kajian mengenai nanopartikal kuprum zinc ferrite telah dijalankan menggunakan kaedah sintesis kimia mekanik. $Cu_{0.2}Zn_{0.8}Fe_2O_4$ nanopartikal disintesis daripada bahan kering Fe_2O_3, CuO dan ZnO daripada pelbagai berat molekular dengan nisbah 1:0.1:0.4 untuk menghasilkan campuran kuprum zinc ferrite nanopartikal. Campuran itu kemudian di hancurkan menggunakan mesin ball mill pada 600 rpm selama 8 jam dengan nisbah campuran bahan kepada bola penghancur 10:1. Campuran homogene itu kemudiannya dikeringkan di dalam oven pada suhu 750 °C selama sejam untuk pembuangan sebarang molekul air yang mungkin terdapat dalam campuran $Cu_{0.2}Zn_{0.8}Fe_2O_4$. Selain itu, carbon aktif yang telah diperbuat menggunakan serbuk kayu melalui proses pirolisis telah dilakukan dengan kehadiran nitrogen gas supaya pembakaran tanpa udara dapat dilakukan. Karbon aktif yang bewarna hitam dan ringan telah dihasilkan mengikut keperluan. Karbon aktif kemudiannya dicampur bersama $Cu_{0.2}Zn_{0.8}Fe_2O_4$ mengikut persen berat yang telah dipersutujui. Campuran itu kemudiannya melalui proses sol-gel dengan kehadiran ethanol tulen sebagai pelincir dan dikacau dengan laju dengan suhu 100 °C selama 2 hingga 3 jam sehingga gel terhasil. Gel yang terhasil kemudiannya dikeringkan selama sejam di dalam oven pada suhu 120 °C untuk pengeringan dan kemudiannya dibakar selama 3 jam di dalam suhu 800 °C untuk pengaktifan ciri-ciri nanopartikel karbon kuprumr zink ferrite. Kesemua campuran kemudiannya dianalysis menggunakan Fourier Transform Infrared Spectroskopi (FTIR), X-Ray Powder Diffraction (XRD) Analysis dan Scanning Electron Microscopy (SEM) untuk mengkaji kesan karbon kepada kuprum zink ferrite.

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LIST OF SYMBOLS

~	Approximately
%	Percent
λ	Wavelength
μ	Micron (10-6)
η	Coulombic efficiency
2θ	Bragg angle
^{o}C	Degree celcius
Å	Angstrom (10-10)
g	Grams
h	Hour
t	Time
=	Equal to
nm	Nano metre
L	Litre
rpm	Revolution per minute
Cu	Copper
Zn	Zinc
Fe	Iron
O	Oxygen
Mn	Manganese

LIST OF ABBREVIATIONS

FTIR	Fourier Trasnform Infrared Spectroscopy
IR	Infrared Spectroscopy
KBr	Potassium bromade
SEM	Scanning Electron Microscopy
XRD	X-ray Diffraction

CHAPTER 1

INTRODUCTION

1.1 Background of the Study

Ferrites possess both of magnetic and electrical properties, which makes ferrites useful in many technological application. The basic magnetic and electrical properties of ferrites can be modified to the desired application. The modification of ferrites' properties come in various ways and one of the crucial way in modifying its properties is to use different synthesis methods by optimizing the synthesis parameters (Shinde, 2013).

Magnetite (Fe_3O_4) and maghemite (γ-Fe_2O_3) are presently the fundamental intrigue contrasted with other iron oxides and ferrites. The spinel ferrites can be portrayed by the recipe AB_2O_4, where A and B signify divalent and trivalent cations, separately. $ZnFe_2O_4$ is a typical spinel with all the Fe^{3+} particles in the B destinations and all the Zn^{2+} particles in the A locales, while $CuFe_2O_4$ has an opposite spinel structure with the Cu^{2+} particles basically in the B locales and Fe^{3+} particles disseminated similarly between the A and the B site (Singh, et al., 2016).

Meanwhile, the enacted carbon from sawdust were created in a tube heater with the nearness of nitrogen gas. Actuated carbon is imperative microporous adsorbents as a result of its brilliant adsorptive properties, a proclivity for assortment of disintegrated organics and capacity to be specially delivered taking after fancied properties for particular applications (Ismadji et al., 2005).

As of late, activated carbons with extensive particular surface territory are created by synthetic enactment utilizing KOH as initiation reagent. These enacted carbons are relied upon to be a valuable material for the vitality gadget, for example, a gas capacity and electric twofold layer capacitor (Okuma & Horikawa, 2015).

In this research, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ were synthesized by mechanosynthesis method using ball mill machine with suitable powder to ball ratio. In order to mixed copper zinc ferrite nanoparticle and the sawdust, sol gel method were used with absolute ethanol was used as fuel because of its better completing ability and low ignition temperature (200 – 250 °C) than other fuels used in wet chemical methods (Raut et al., 2014). These synthesized ferrite nanoparticles were then undergoes characterization analysis to study about their structural and magnetic properties.

1.2 Problem Statement

In recent years, the interest in an investigation of nano-size materials has increased due to their u physical and chemical properties which are often differ from the bulk counterpart. Nanocrystalline ferrites are as of now the subject of interest on the grounds that a wide application in modern and examination ranges (Sonal Singhal, et al., 2015). The difference in properties of nano-size materials is attributed to increase in surface area along with decreased particle size and various size effect. (Raut et al., 2014).

Due to the large electronegativity of oxygen, the ionic type of bonds prevails in almost all oxide spinels. Soft spinel ferrite (MFe_2O (where M = Ni, Zn and Mn), Ni, Zn, Mn,) nanoparticles have been intensively investigated due to their remarkable magnetic and electrical properties and wide practical applications in ferro fluids, magnetic drug delivery, and magnetic high density information storage (Sharma, et al., 2014).

Enacted carbon is a standout amongst the most vital micro porous adsorbents because of its colossal adsorptive limit, a proclivity for assortment of broke up organics and ability to be especially custom-made to suit particular application (Ismadji et al., 2005). Many sorts of materials are utilized as a part of delivering actuated carbon are agribusiness squanders, for example, coconut shell, pistachio shell, saw clean, walnut shell, tropical wood and almond shell which are the most normally utilized (Adinata, 1998). The common field that connected initiated carbon are in waste, water and gas refinement, desulphurization, mercury evacuation and water treatment.

Therefore, in this research, the carbon obtained from sawdust were used as a dopant of copper zinc ferrite nanoparticles. The structural, dielectric and magnetic properties of nano-crystalline carbon dopped copper ferrites were investigated.

1.3 Objectives of Research

Objectives of this research are:

1. To synthesis zinc substituted copper ferrite nanoparticles $Cu_{0.2}Zn_{0.8}Fe_2O_4$ where ($x = 0.2$) by mechanosynthesis method.
2. To produce activated carbon from sawdust with nitrogen gas where carbon acted as dopant to copper zinc ferrite nanoparticle via pyrolysis method.
3. To investigate the structural, dielectric and magnetic properties of nanocrystalline carbon dopped copper ferrites.
4. To investigate the effect of carbon to copper zinc ferrite nanoparticles.

1.4 Scope of The Study

In this research, nano-crystalline copper zinc ferrite was prepared by mechanosynthesis method, using the 99.9% pure AR grade Iron (III) Oxide (Fe_2O_3), AR grade Zinc Oxides (ZnO) purchased from R&M Chemicals and Copper (II) Oxide (CuO) to produce copper zinc ferrite nanoparticles ($Cu_xZn_{1-x}Fe_2O_4$) where absolute ethanol was use as a fuel. The loose powder of copper zinc ferrite will be characterized by using Fourier Transform Spectroscopy (FTIR), X-ray diffractometer (XRD) and scanning electron microscopy (SEM).The carbon produced from the sawdust then were mixed with copper zinc ferrite nanoparticles via sol-gel method with absolute ethanol acted as fuel where the carbon is a dopant. The magnetic property of carbon doped copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ was investigated.

CHAPTER 2

LITERATURE REVIEW

2.1 Introduction

The nanotechnology is rapidly growing and ensure that the substantial changes that will impose a significant ad vantages economically and scientifically that are applicable to a wide range of areas. Nanocrystalline spinel ferrites are imperative attractive materials on account of their fascinating attractive electrical properties with high chemical and thermal stabilities (Godbole et al., 2013). Spinel ferrites have the general sub-atomic equation A $[B]_2O_4$, where A and B are the molecules involving the tetrahedral and octahedral interstitial locales of the FCC cross section framed by the anions (oxygen particles). The most famous super paramagnetic nanoparticles are those in the light of iron oxide nanoparticles with a specific end goal to alter their attractive properties, $M_xFe_{3-x}O_4$ blended ferrites where (M = Co,Ni, Mn,Cu ...) could be gotten by inserting paramagnetic molecules in the magnetite crystal structure supplanting some of Fe^{2+} cations of the octahedral or tetrahedral gaps. These ferrites display opposite spinel-sort structure, where O^{2-} particles are masterminded in a cubic close pressing and the cations are conveyed in 1/8 of the tetrahedral (A) and 1/2 of the octahedral (B) sites (Lasheras et al., 2016).

The frameworks made up of nanoparticles are seriously examined both practically and theoretically, for all intents and purposes because of their dielectric, electric, and magnetic properties affectability sensibly not quite the same as that of the bulk materials and their conceivable applications in different fields (Masrour et al., 2013). Magnetic

nanoparticle carriers comprise of three utilitarian parts which are a magnetic core, a surface coating and a functionalized outer coating (Vatta et al., 2006).

2.2 Copper Zinc Ferrite

Non-nano sized range copper ferrites have certain catalytic applications, such as, for example, for CO conversion to CO_2 (Lou, 2006). In a difference of pure nickel ferrites, copper ferrite NPs are applied in organic catalysis in more uniform particle size (mainly about 20 nm). Thus, copper ferrite nano-material (20 nm) was used as reusable heterogeneous initiator in the synthesis of 1, 4-dihydropyridines. The reaction of substituted aromatic aldehydes, ethyl acetoacetate and ammonium acetate was achieved in the presence of copper ferrite nano-powders at r.t. (room temperature) in ethanol. The nano-catalyst was easily recovered and its reusability was confirmed (Kasi Viswanath and Murthy, 2013).

Copper ferrite nanomaterial is a reusable heterogeneous initiator in the synthesis of α-aminonitriles. The nano-catalyst is easily recovered and its reusability is recorded.

2.3 Activated Carbon from Sawdust

Initiated carbons have their very much created porosity and big surface area. Accordingly, they are utilized as adsorbent, as well as impetus and impetus bolster. They are the helpful material and are utilized as a part of the different fields, for example, the detachment/separation procedure, the cleaning process and the water treatment handle (Okuma & Horikawa., 2015). According to Viswanathan et al., (2009), activated carbon is a material with exceedingly porosity comprising of hydrophobic graphene layer and in addition hydrophilic surface useful gatherings making them valuable for sorption and reactant applications.

In addition, carbon nanomaterials have novel electrical and structural properties accordingly making them helpful and fundamental in vitality change and capacity gadgets (as cathode material), reactant process (as bolster material) and purifying environments advancements (Viswanathan et al., 2009).

There are two (2) sorts of strategy in creating enacted carbon which are chemical and physical initiation. In physical initiation, the crude material is carbonized and along these lines is gasified by CO_2 (Yang and Lua 2003) or steam (Bacaoui et al 2001). In concoction J.Hayashi, O. Okuma, T. Horikawaactivation, the crude material is impregnated with initiation reagent, for example, H_3PO_4 (Haimour and Emeish 2006) and $ZnCl_2$ (Tsai et al., 1997) and the impregnated crude material is carbonized (Okuma & Horikawa, 2015).

2.4 Methods in Producing Nanoparticles

2.4.1 Mechanosynthesis Synthesis of Copper Zinc Ferrite

A number of chemical routes have been used for the synthesis of ferrite nanoparticles. These methods includes sol-gel, micro emulsion, chemical co-precipitation and many other methods. The size and the properties of spinel ferrite nanoparticles can be greatly depends on pH, fuel, stirring time and speed, metal nitrates to fuel ratio and preparative parameters (Shinde, 2013). The combination conditions have firmly impacted the nanoparticles crystal structure, crystallite measure, microstructure, attractive and photo catalytic properties.

Mechanochemistry is the subject that arrangements with the concoction and physicochemical changes of substances prompted by mechanical constrain. Mechanochemistry has been broadly utilized as a part of many fields, for example,

materials designing, extractive metallurgy, gem designing, coal industry, building industry, agribusiness, drug store and waste treatment. The science base for mechanochemistry, notwithstanding, gives off an impression of being restricted, particularly the investigation of energy. Henceforth, it does not have an arrangement of methodical and appropriate hypothesis to depict the mechanosynthesis preparing. (Wang Ming, 2010).

Mechanosynthesis amalgamation and ignition blend have a place with various research regions, however there are bunches of likenesses between them, particularly the way that the structure development of resultant in both procedures all falls behind the compound change. Thus, it is plausible that the basic macrokinetics hypothesis can be utilized to explore the mechanosynthesis response prepare. The hypothesis and research techniques for auxiliary macrokinetics were firstly advanced to apply in mechanosynthesis look into territories by the creators, expecting to enhance the dynamic hypothesis of mechanosynthesis response. Taking for instance in this work, the mechanosynthesis blend of $ZnFe_2O_4$ by high-vitality ball processing the blend of Fe_2O_3 and ZnO powders, and the auxiliary macrokinetics were considered.

2.4.2 Sol – gel Method

In order to add activated carbon with the copper zinc ferrite nanoparticle, appropriate amount of carbon and copper zinc nanoparticles were undergo sol-gel method so that both of the powder were mixed homogenously via sol-gel route with absolute ethanol as the fuel. (Toksha, et al., 2008) in his research discussion stated that the analysis of X-ray diffraction pattern of the calcined powder synthesized using this route shows that the final product is $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with the expected inverse spinel structure. The size of the particles was determined by Scherrer formula using most incense (311) peak. The particle size appears to increase almost linearly with annealing time, most likely due to the fact that longer annealing time enhances the coalescence process resulting in an increase in the particle size. Thus, it appears that particle size may be controlled by

varying either of the two parameters i.e. annealing temperature and time (Toksha et al., 2008).

According to (Klein, 1994), there are many advantages in sol-gel route : can create thin bond-coating to give astounding bond between the metallic substrate and the top, producing thick coating providing good corrosion protection, will easily produce materials into complex geometries in a gel state, can create high immaculateness items in light of the fact that the organo-metallic antecedent of the sought clay oxides can be blended, broken down in a predetermined dissolvable and hydrolysed into a sol, and in this way a gel, the structure can be exceptionally controllable, possible to have low temperature sintering capability, usually at 200 – 600 °C and is a simple, economic yet efficient method to produce high quality nanoparticles.

CHAPTER 3

MATERIALS AND METHODS

3.1 Introduction

This research begin with synthesizing of CuO-Zn nanoparticles by mechanosynthesis reaction method where (X = 0.2). 99.9 % pure AR grade Iron (III) Oxide (Fe_2O_3) with molar weight of 159.69 g /mol purchased from Sigma Aldrich, AR grade Zinc Oxides (ZnO) purchased from R&M Chemicals and Copper (II) Oxide (CuO) with molar weight 79.55 g /mol from R&M Chemicals were used as starting materials to prepare copper zinc ferrite nanoparticles ($Cu_xZn_{1-x}Fe_2O_4$) where X = 0.2. The powders of copper oxide, zinc oxide and iron oxide were dried in oven at 150 ℃ in air for 12 hours prior to use.

3.2 Research Methodology

Basically, the experimental research work consists of five (5) phases as illustrated in Figure 3.1.

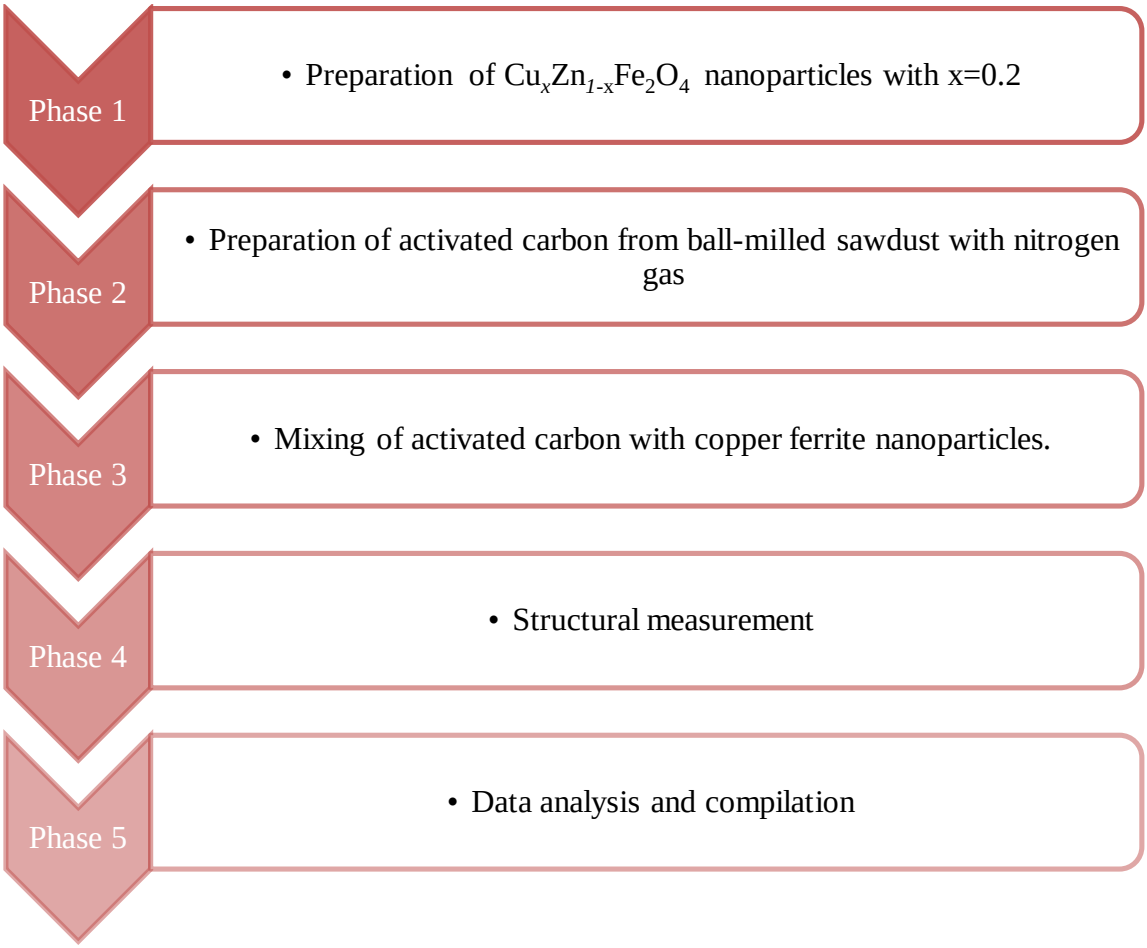


Figure 3.1: Flow chart of the general overview methodology in this research

3.3 Material Synthesis Methods

3.3.1 Preparation of Copper Zinc Ferrite Nanoparticles

As the most prior step in this phase, the powders of copper oxide, zinc oxide and iron oxide were dried in oven at 150 °C in air for 12 hours prior to use. The 90 grams of the mixture then were mechanosynthesisly milled for 8 hours at 600 rpm with ball to powder ratio of 10:1. The as-milled copper zinc ferrite nanoparticles were then calcined at 750 °C for an hour. Copper zinc ferrite nanoparticles then added with carbon as dopping with respective percentage of weight. Then, all the samples were then hydrothermally sol-gelled to obtain a homogenous mixture of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and

activated carbon as the dopant. Ethanol were used as fuel and 4 mixture of the sample with varied mass of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and the activated carbon with respective percentage weight were stirred with 30 ml of absolute ethanol heated 100 ℃ for 4 hours to form a sol. The brown gel then heated with 120 ℃ for an hour for water removal. Then, the brown gel were calcined at 800 ℃ for 3 hours until all the gel were burnt completely and a brown loose powder of copper zinc ferrite with activated carbon as dopant was obtained. The flowchart of preparing desired copper zinc ferrite nanopowder is shown in Figure 3.3.

Figure 3.2: Copper zinc ferrite produced after calcined

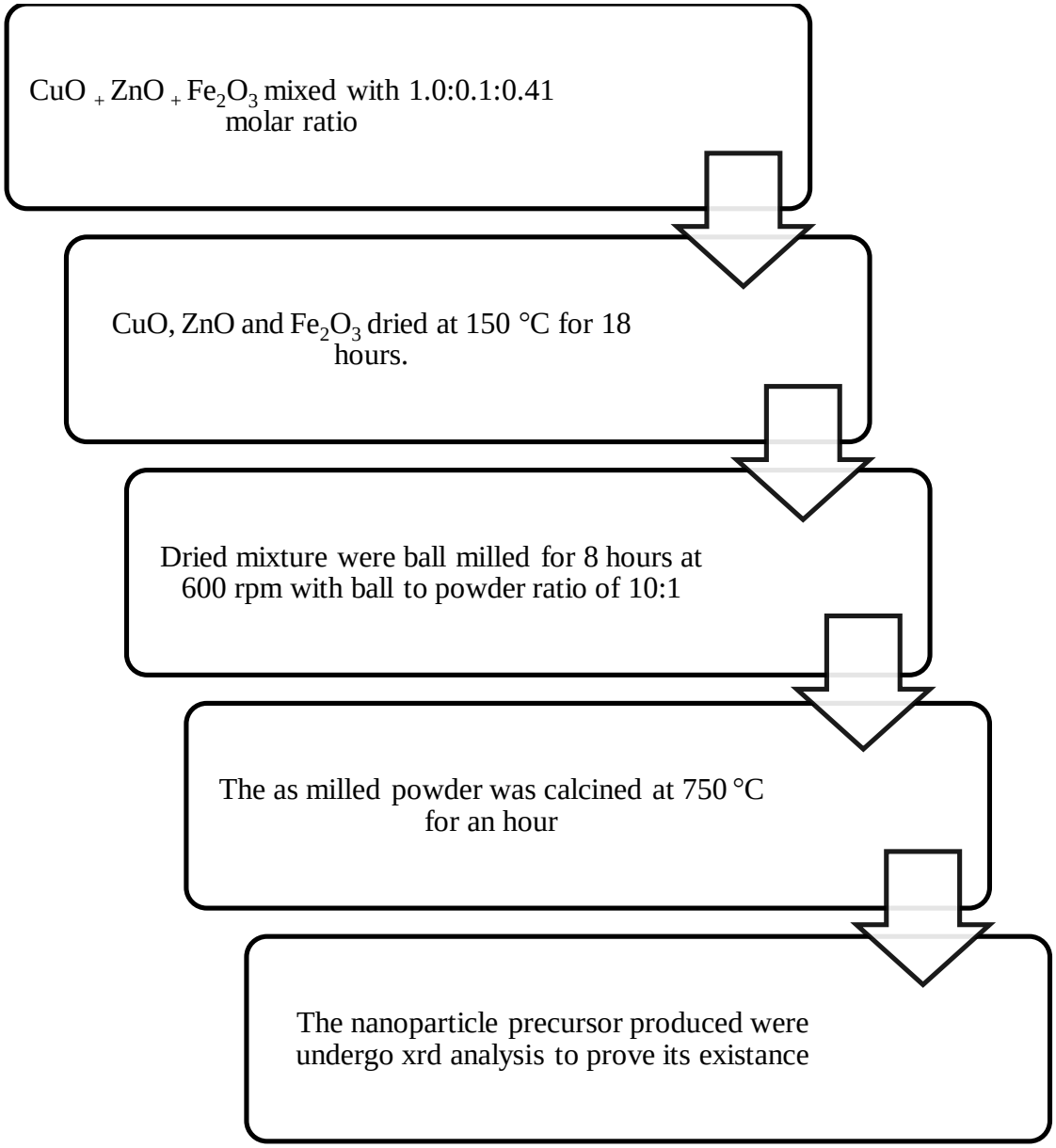


Figure 3.3: Flowchart of the preparation of copper zinc ferrite nanoparticles

3.3.2 Prolysis of Activated Carbon from Ball-milled Sawdust with Nitrogen Gas

Activated carbon were produced by following pyrolysis method. Referring to (Salehi et al., 2009), a 10 g sample of dried sawdust was placed in the tube furnace as the reactor, and nitrogen gas was used as the sweeping gas with flow rates of 1, 2, 4, and 6 L/min. The experiments were carried out to a final temperature of 500 ℃, at a heating rate of 1000 °C /min and stayed at that temperature for 20 minutes or until no further

critical arrival of white vapour gas was watched. Therefore, in this research the activated carbon was produced similarly with (Salehi et al., 2009) with slight different route. The sawdust were taken from nearest sawmill and then were ball-milled for 2 hours at 600 rpm producing fine and smooth sawdust. The sawdust then were burned at 600 °C for 3 hours in a tube furnace with the presence of nitrogen gas. A black porous activated carbon then produced. The process called pyrolysis. Pyrolysis is basically a process of thermal decomposition of organic matter which in this case the sawdust, with inert atmospheric conditions and can also in a limited supply of air. With that, it can release the volatiles and the formation of char (Sinha et al., 2000). This method was repeated until the desired amount of carbon achieved.

Figure 3.4: Activated carbon from sawdust

3.3.3 Mixing of Activated Carbon with Copper Ferrite Nanoparticles

At this phase, copper zinc ferrite nanoparticles were added with activated carbon with four different weight percentage of copper zinc ferrite and activated carbon. All the designated sample were as in Table 4.1. Carbon and copper zinc ferrite nanoparticle were mixed homogenously by sol-gel method with the presence of absolute ethanol as fuel. After being weighted for the desired mass according the designated weight percentage, the powders that vigorously mix in a beaker on a hot plate with a medium magnetic stirrer

with 100 °C temperature for 2-3 hours until the mixture were mixed and slightly dry. The brownish to black mixture then were dried in the air of 120 °C temperature for an hour for the water removal. The dried gel than were calcined for 3 hours at 800 °C temperature. After being calcined, the fluffy brown powder were obtained. It is observed that as the weight percentage of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ decrease and the weight percentage of activated carbon, C, were increased, the powder's color were getting darker comparing to the unmixed $Cu_{0.2}Zn_{0.8}Fe_2O_4$ nanoparticles.

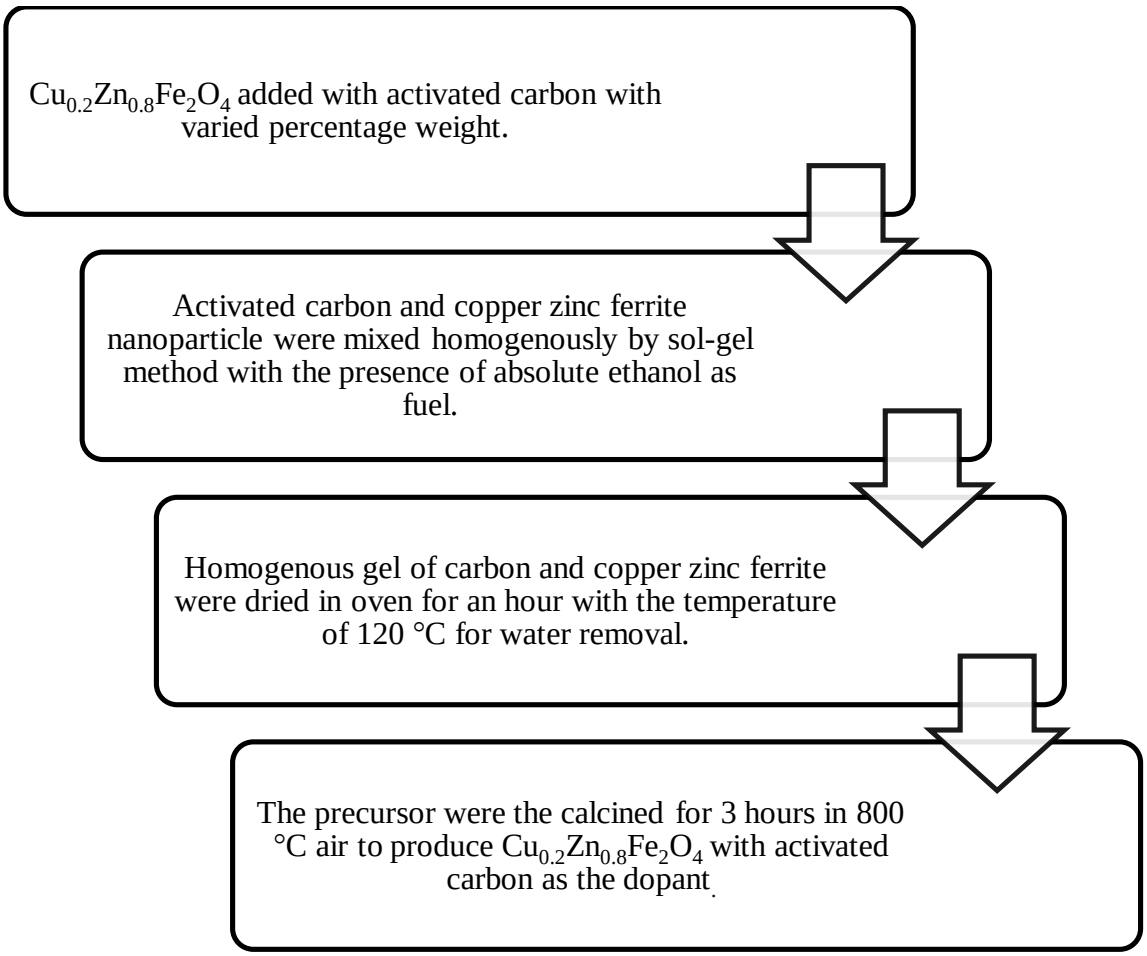


Figure 3.5: Flowchart of the mixing of activated carbon with copper zinc ferrite

The flowchart of the mixing of activated carbon with copper zinc ferrite was illustrated in Figure 3.5. The produced carbon doped copper zinc ferrite nanoparticles, Cu0.2Zn0.8Fe2O4, were the sent to XRD analysis to analysis the composition of the structure of carbon doped copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$.

3.4 Material Characterizations

Nowadays, with the advanced of technologies, instrument for the measurement and characterization of copper zinc ferrite nanoparticles, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant, can easily done for example XRD analysis, SEM analysis and FTIR study. All the instruments stated were used to measure the chemical, physical and structure of the $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant.

3.4.1 Fourier Transform Infrared Spectroscopy (FTIR) Study

In infrared spectroscopy, IR radiation is gone through a specimen and some of the infrared ray is absorbed by the specimen and some of the infrared is transmitted (passed through). The absorbed infrared were called and plotted as absorbance meanwhile the transmitted infrared were labelled as transmittance (Woods, 2015). The infrared spectra of all the samples were recorded in the range 4000-400 cm^{-1} in FTIR instrument (Figure 3.6) using KBr pellets (Zhao et al., 2010).

Figure 3.6: FTIR Spectroscopy, Perkin Elmer

Each of designated sample of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant were ground with KBr and compressed into thin pallet. The properties of chemical bonds formed were observed in the range 4000-400 cm^{-1} FTIR spectra.

3.4.2 X-Ray Powder Diffraction (XRD) Analysis

An X-ray diffraction pattern is plot of the density of x-ray scattered at different angles by a specimen. From the XRD pattern we can determine the crystalline phase in a mixture, measuring the quantitative phase of the crystalline phase in the mixture and also observed if any amorphous exist in the mixture. There are two kind of analysis in XRD analysis: qualitative analysis and quantitative analysis of XRD data. In qualitative analysis, the experimental data of XRD will be compared to the reference patterns to study the phase that are present. In quantitative analysis of XRD data, the position of the diffraction peak are used to calculate the unit cell dimensions (Woods, 2014). Therefore in this research, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant samples were undergo XRD analysis using Rigaku Mini Flex II instrument as shown in Figure 3.7.

Figure 3.7: XRD Analysis, Rigaku

3.4.3 Scanning Electron Microscopy (SEM)

SEM analysis is typically used for the study of metallurgy, biology, geology, and many more. High magnification image of sample can be observed in SEM analysis. Normally, SEM operates at high vacuum. The conventional principle of SEM is that a beam of electrons is generate by a favourable source, usually a tungsten filament or a field emission gun. Through a high voltage, the beam is accelerated and will pass through an apertures of a system and an electromagnetic lenses to create a thin beam of electrons. The beam than scans the surface of the sample. There are electrons emitted from the sample by the scanning beam and will be collected by a detector. All of the SEM image are in white and black resolution. In this paper, the sample of copper zinc ferrite nanoparticle, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and all the samples of copper zinc ferrite with activated carbon as the dopant, with the designated weight of percentage were magnified with 2500x magnification to observe the morphology of the sample.

Figure 3.8: Scanning Electron Microscopy, Quanto 450

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Introduction

In this research paper, the structural and chemical properties of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$, with activated carbon as the dopant were crucially observed and analysed. All of the significant properties and parameters that were analysed are FTIR study, XRD analysis, and SEM analysis.

4.2 X-Ray Diffraction Analysis

Figure 4.1 shows the XRD spectra of the copper zinc ferrite CuxZn1-xFe2O4 nanoparticles with X=0.2 precursor produced after calcined at 750°C. Peak (1 0 0), (1 0 4), (3 1 1), (1 1 0), (4 2 2), (5 1 1) and (4 4 0) shows polycrystalline hexagonal of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$. The peak of (1 0 0) and (1 1 0) were belongs to CuZn phase. Peak of (1 0 4), (3 1 1), (4 2 2) were correspond to magnetite proving that copper zinc ferrite were successfully produced which is also correspond to ferromagnetic: strongly attracted to magnetic fields. Therefore, it is proven that copper zinc ferrite does exhibit magnetic properties. Referring to (Jing Xu et al ., 2010), the synthesized Fe3O4 was transformed to α-Fe after annealed in furnace at 750 ℃ as confirmed by the XRD result.

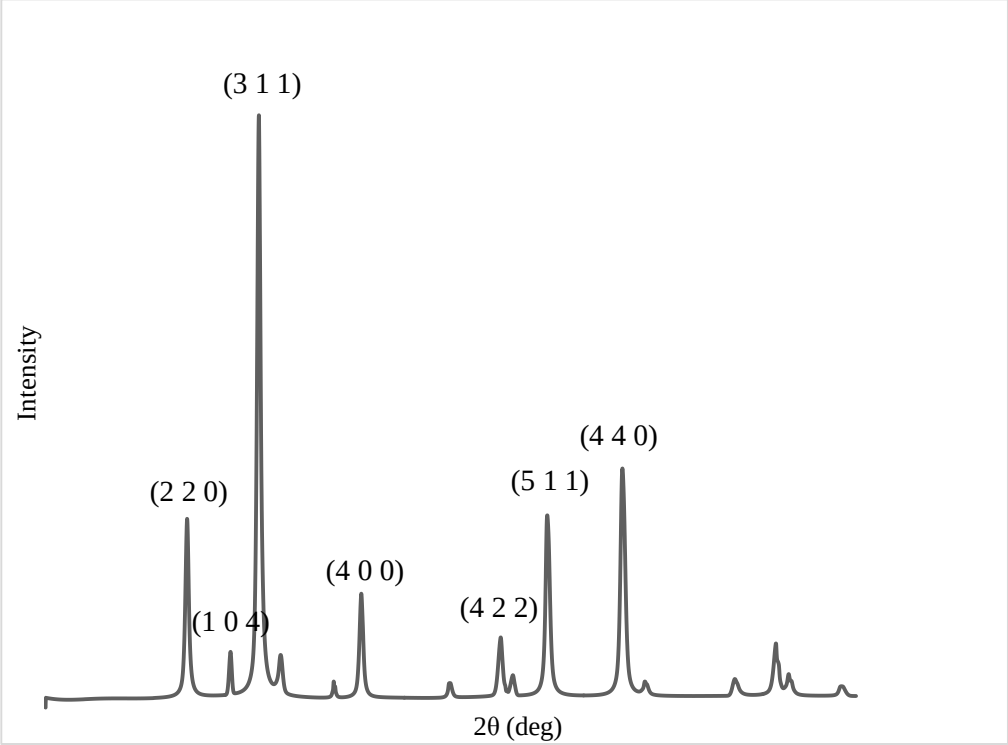


Figure 4.1: X-ray diffractograms of copper zinc ferrite $Cu_{0.2}Zn_{0.8}Fe_2O_4$ nanoparticles precursor produced after calcined at 750 degree

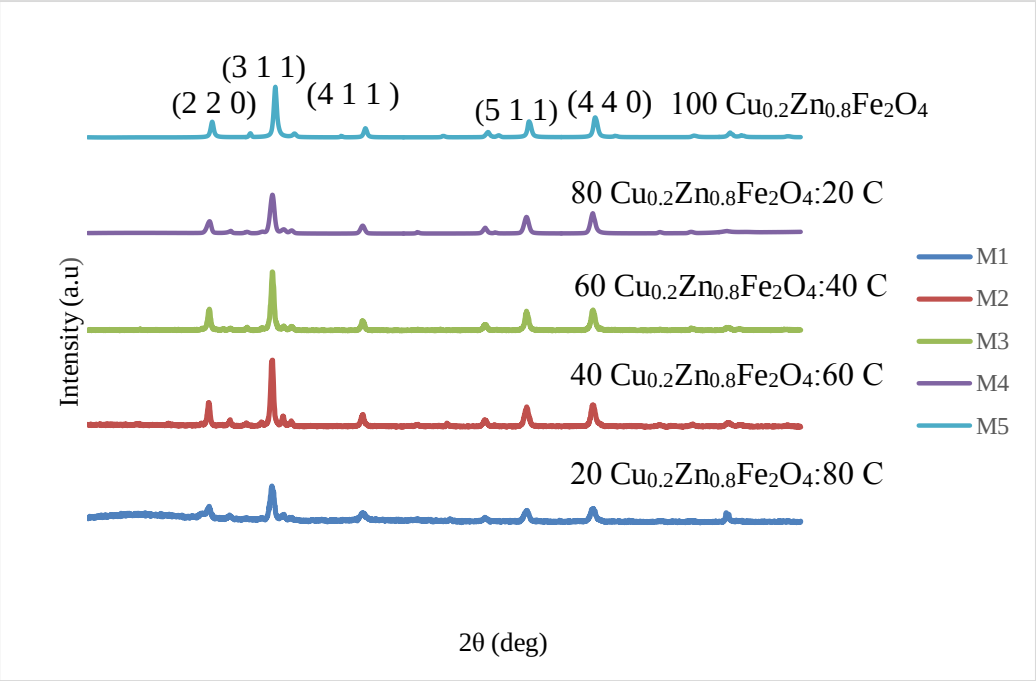


Figure 4.2: XRD Spectra of the copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant, with different designation on weight percentage of activated carbon on copper zinc ferrite powder, (Table 4.1).

Table 4.1

The designation of sample carbon copper zinc ferrite, carbon doped with $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with respective weight percentage.

DESIGNATION	**WEIGHT PERCENTAGE (%)** $Cu_{0.2}Zn_{0.8}Fe_2O_4$ **: C**
M1	20:80
M2	40:60
M3	60:40
M4	80:20
M5	100:0

Based from the analysis of Figure 4.2 the peak of (3 1 1) is the most sharp and high intensity of peak for all the weight percentages of activated carbon and $Cu_{0.2}Zn_{0.8}Fe_2O_4$. Peak (3 1 1) is correspond to magnetite of carbon copper zinc ferrite as it also correspond a strong attraction towards magnetic field (Chiolerio et al., 2016). As the plotted XRD patterns in Figure 4.2 is similar to the XRD pattern plotted in (Xu et al., 2007) research paper, about entitled preparation and magnetic properties of magnetite particles by sol-gel method, therefore can be concluded that as carbon were mixed with copper zinc ferrite, the magnetic properties was exhibit.

To have detailed characterization about the structure, the lattice constants a and c were calculate using the formula of (2):

$$\frac{1}{d^2{}_{hkl}} = \frac{4}{3}\frac{h^2 + hk + k^2}{a^2} + \frac{l^2}{c^2}$$

where (h k l) are the Miller indices of the respective crystalline planes, a = b and c stand for the lattice parameters of the hexagonal $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant. The structure and d_{hkl} is the distance between (h k l) planes. The standard value for hexagonal cell c/a = 1.633. From the data tabulated in Table 4.2, Table 4.3 and Table 4.4, the value of c/a are compatibly similar to the standard value for hexagonal cell.

Therefore can be also concluded that the $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and carbon doped $Cu_{0.2}Zn_{0.8}Fe_2O_4$ were matched perfectly for hexagonal cell.

Table 4.2
The calculated value of a, c and c/a at the peak (3 1 1)

Sample	a (Å)	c (Å)	c/a (Å)
M1	2.916	5.052	1.733
M2	2.851	4.938	1.732
M3	2.915	5.048	1.732
M4	2.851	4.938	1.732
M5	2.897	5.018	1.732

Table 4.3
The calculated value of a, c and c/a at the peak (5 1 1)

Sample	a (Å)	c (Å)	c/a (Å)
M1	1.867	3.232	1.173
M2	1.867	3.234	1.173
M3	1.868	3.236	1.732
M4	1.867	3.239	1.735
M5	1.861	3.223	1.732

Table 4.4
The calculated value of a, c and c/a at the peak (4 4 0)

Sample	a (Å)	c (Å)	c/a (Å)
M1	1.715	2.970	1.732
M2	1.715	2.970	1.732
M3	1.715	2.970	1.732
M4	1.715	2.970	1.732
M5	1.710	2.962	1.727

Hence, it can be observed that carbon does not disrupt magnetic properties copper zinc ferrite at all but in fact enhance the magnetic properties of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$. At the weight percentage of 40 %wt of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and 60 %wt of activated carbon, the peak of magnetite were the highest also can be concluded that it is the most ideal ratio of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and activated carbon, C that were obtained from sawdust of recycled sawdust. At the ratio of 40 %wt of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and 60 % wt of activated carbon the magnetite peak (3 1 1) is the highest

and in the nutshell also can be said that at that particular ratio, the magnetite peak of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ were enhanced.

4.3 Fourier Transform Infrared Spectroscopy (FTIR)

Fourier transform infrared spectroscopy (FTIR) spectra of Carbon copper zinc ferrite before and after adding activated carbon were measured using Pelkin Elmer Spectroscopy. FTIR study was to identify the chemical and structural changes occurred in copper zinc ferrite before and after adding of activated carbon also to identify the unknown compound. Each of designated sample of copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as dopant were ground with KBr and compressed into thin pallet. The properties of chemical bonds formed were observed in the range 4000-400 cm^{-1} FTIR spectra.

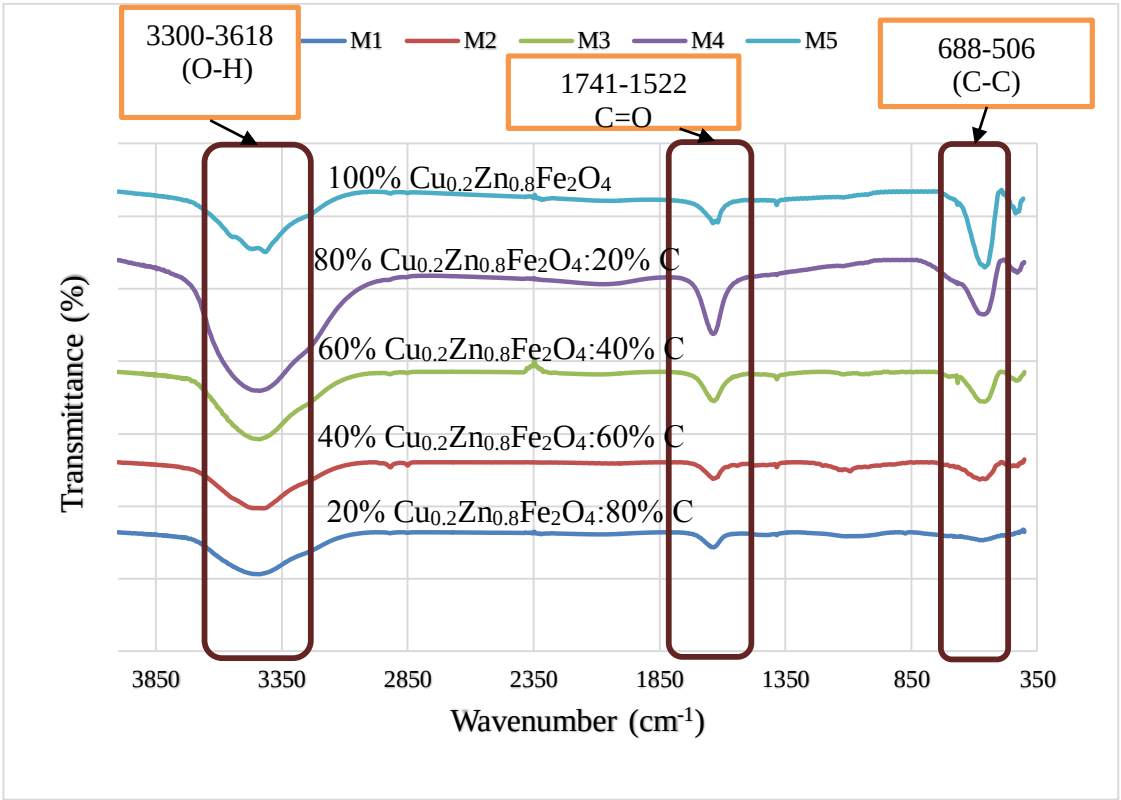


Figure 4.3: FTIR Spectra of activated carbon doped copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with different weight percentage of carbon and $Cu_{0.2}Zn_{0.8}Fe_2O_4$.

From results obtained in Figure 4.3, it is proven the presence of chemical and structural changes occurring in $Cu_{0.2}Zn_{0.8}Fe_2O_4$ as the percentage weight of carbon and copper zinc ferrite varied. According to Waldron, ferrites can be considered constantly reinforced crystal i.e. the molecules are attached to all closest neighbours by equal powers (ionic, covalent bonding or Van der Waals). FT-IR retention groups of solids are normally relegated to the vibration of particles in the crystal grid (Raut et al., 2014).

As observed from Figure 4.3, there is a strong transmittance of alcohol group the wavenumber of 3300-3618 cm^{-1}. It were strong and broad curve for all the samples of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant. As the weight percentage of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ decrease and the weight percentage of activated carbon increase, the transmittance peak of alcohol, (O-H) group at the wavenumber of 3300-3618 cm^{-1} become more broader which is less intense compared from the raw material of copper zinc ferrite nanoparticles (M5). From the graph, it were indicate that as the weight percentage of activated carbon increase, the intensity of the alcohol were decrease where M1 sample, (20%wt $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 80% C) has the lowest intensity of O-H group peak.

For the observation at the wavenumber of 1741-1522 cm^{-1} weak vibrations correspond to the C = O bond. The absorption band shows the strong stretching of C=O bond due to the huge changes in the dipole take place (Braihi, 2014). Similar to the O-H group, M1 sample, (20 %wt $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 80 %wt C) has the lowest intensity of C=O bond. M4 sample, (80 %wt C $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 20 %wt C), has the most intense peak of C=O.

On the other hand, at the wavenumber of 688-506 cm^{-1}, its exhibit the present of C-O stretching for all the samples. The carbon to carbon bond intensity however decrease as the weight percentage of the carbon increase.

4.4 Scanning Electron Microscopy (SEM) Analysis

The average size of the $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant were observed under Scanning Electron Microscope (SEM). The (SEM) micrograph of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant were referred to observe and confirmed the grain formation of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with activated carbon as the dopant. Figure below shows the (SEM) micrograph of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and $Cu_{0.2}Zn_{0.8}Fe_2O_4$ added activated carbon as the dopant at magnification of 2500x.

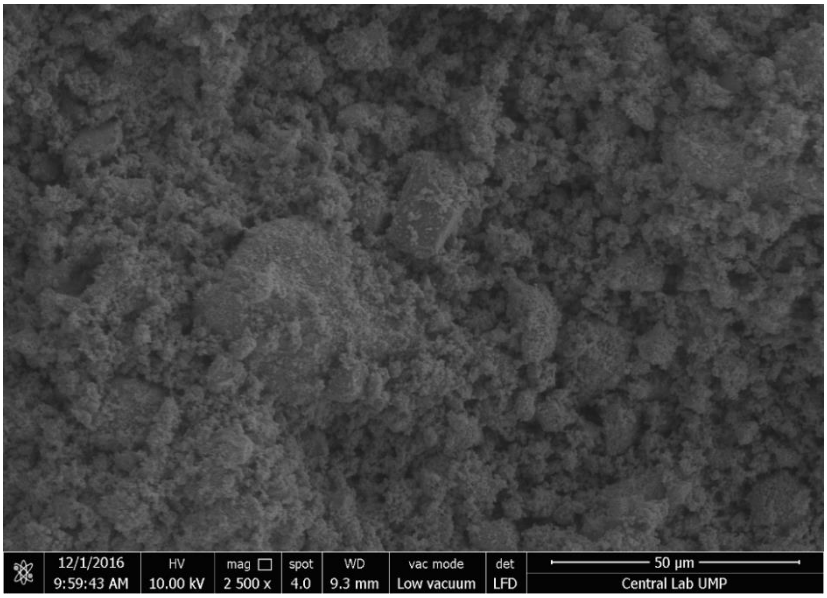


Figure 4.4: 100% $Cu_{0.2}Zn_{0.8}Fe_2O_4$

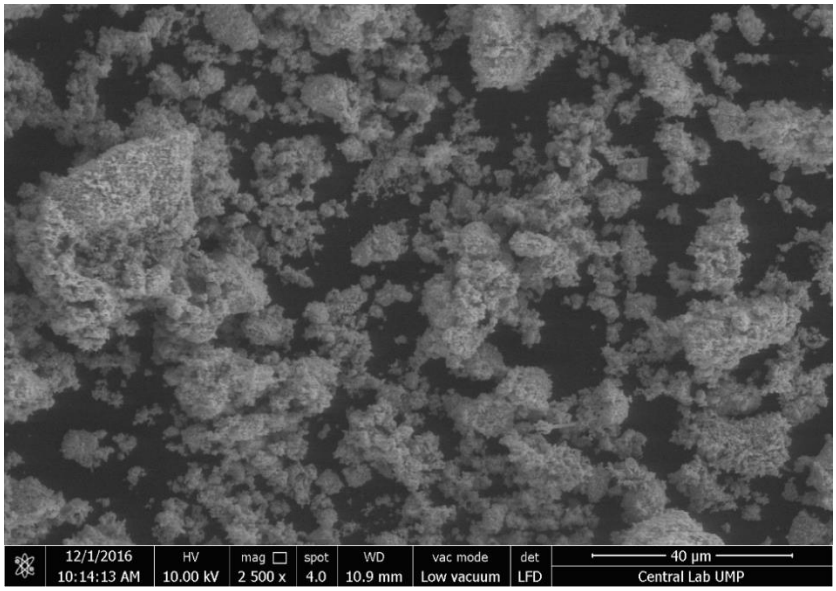


Figure 4.5: 80% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 20% C

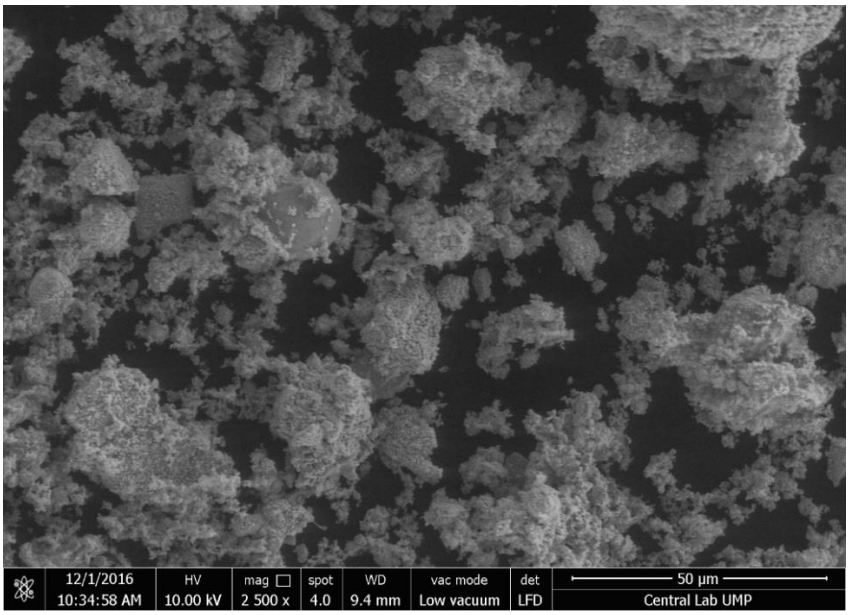


Figure 4.6: 60% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 40% C

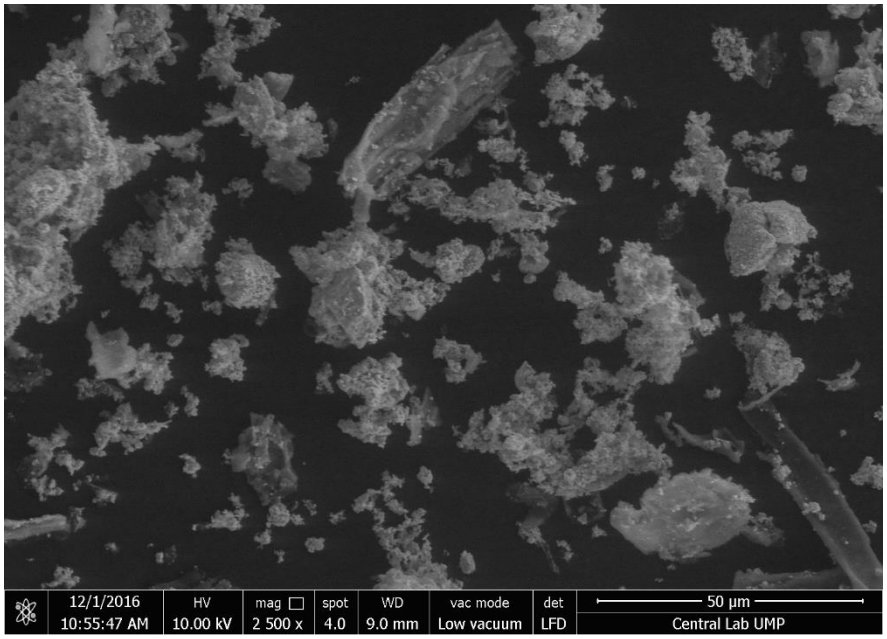


Figure 4.7: 40% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 60% C

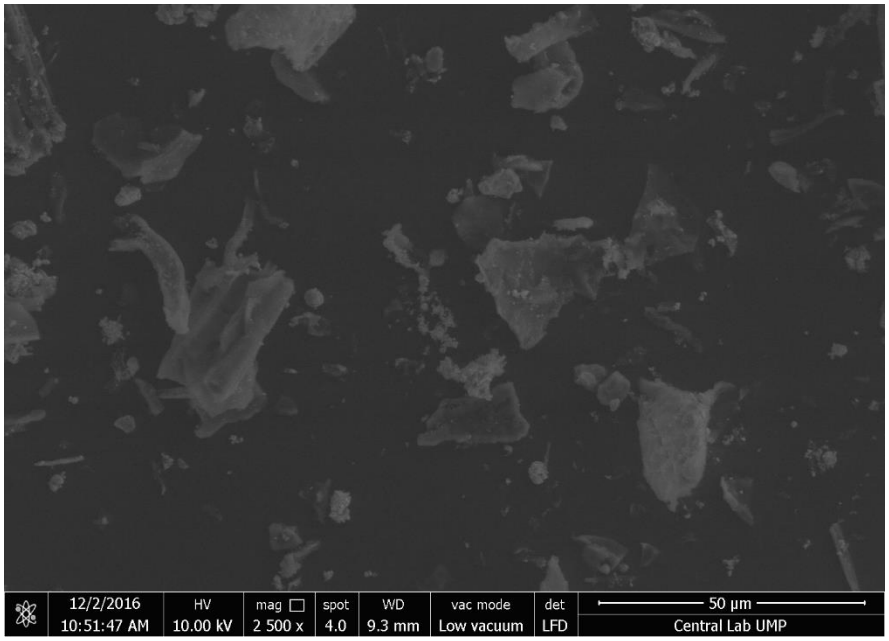


Figure 4.8: 20% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 80% C

Figure 4.5 is the SEM micrograph of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ after ball milled for 8 hours at 600 rpm with ball to powder ratio of 10:1 showing the homogeneity and uniformity of particle sizes of $Cu_{0.2}Zn_{0.8}Fe_2O_4$. From the figure 4.6 , as the carbon copper zinc ferrite nanoparticle added with activated carbon with weight percentage of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ and

carbon 80 % wt and 20% wt respectively that were produced by sol-gel method, the voids and pores are clearly observed. The particles were agglutinated together performing the void and space. This shows that there is reaction happened between carbon and copper zinc ferrite. Referring to the Figure 4.5, 4.6, 4.7 and 4.8, as the weight percentage of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ decrease and the percentage weight of carbon increased, the voids and space become much bigger and there are more agglutination of the particles.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

From the result obtained, it is proven that $Cu_xZn_{1-x}Fe_2O_4$ nanoparticles with x=0.2 successfully produced by mechanosynthesis method and the carbon copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with the activated carbon as the dopant were also successfully produced via sol-gel method. Therefore, we can also conclude that this route is not only easy and cheap to be set up, it also promised the effectiveness of the production of the samples.

Referring to the results obtained, activated carbon is proven does not interrupt the crystalline phase of copper zinc ferrite but do interrupt its structure as in the SEM analysis the structure of the crystal structure is changed: becoming more void and has lots of space as the percentage weight of carbon increase.

However, it can be seen the potential of carbon in magnetic nanoparticle application where not only it is easy to produced, lower the cost of the production, it also enhanced the magnetic structure of the $Cu_{0.2}Zn_{0.8}Fe_2O_4$. This were shown in XRD Data.

5.2 Recommendation

There are some recommendations in order to improve this research paper. More characterization method should be done such as vibrating sample magnetometer (VSM) analysis for magnetic properties measurement, impedance analyser, to study electrical properties/ dielectric properties at low frequency , Vector Network Analyser (VNA) which to study electrical/dielectric properties at high frequency and also field emission scanning electron microscope (FESEM) analysis for the structural properties and Vector Network Analyser which is to study electrical/dielectric properties at high frequency. Therefore more significant proof can be summarize regarding the effect of activated carbon to the metal oxides.

On the application of the copper zinc ferrite, $Cu_{0.2}Zn_{0.8}Fe_2O_4$ with the activated carbon as the dopant, further research need to be done in order to improve the voids and space of $Cu_{0.2}Zn_{0.8}Fe_2O_4$ after activated carbon was added.

REFERENCES

Braihi, A. J. (2014). Proposed Cross-Linking Model for Carboxymethyl Cellulose /StarchSuperabsorbent Polymer Blend. *International Journal of Materials Science and Applications*. ***3*(6):** 363.

Chiolerio, A., Sangermano, M., Vescovo, L., Allia, P., Tiberto, P., Coisson, M., … Marchegiani, G.. Photoinitiator-Free UV-Cured Acrylic Coatings Containing Magnetite Nanoparticles, **pp:** 2530–2535.

Godbole, B., Badera, N., Shrivastava, S. B., Jain, D., Chandra, L. S. S., & Ganesan, V. (2013). Synthesis, Structural, Electrical and Magnetic Studies of Ni- Ferrite Nanoparticles. *Physics Procedia*. ***49*:** 58–66.

Jing Xu, Haibin Yang, Wuyou Fu, Kai Du, Yongming Sui, Jiuju Chen, Yi Zeng, Minghui Li, G. Z. (2010). Preparation and Magnetic Properties of Magnetite Nanoparticles by Sol – Gel Method nanoparticles by sol – gel method.

Klein, L. C. (1994). A review of: " Sol-Gel Science - The Physics and Chemistry of Sol-Gel Processing " Edited by C. Jeffrey Brinker and George W. Scherer. *Materials and Manufacturing Processes*. ***9*(5):** 1007–1008.

Lasheras, X., Insausti, M., Gil de Muro, I., Garaio, E., Plazaola, F., Moros, M., … Lezama, L. (2016). Chemical Synthesis and Magnetic Properties of Monodisperse Nickel Ferrite Nanoparticles for Biomedical Applications. *The Journal of Physical Chemistry C*.

Masrour, R., Mounkachi, O., El Moussaoui, H., Hamedoun, M., Benyoussef, A., Hlil, E. K., … El Maalam, K. (2013). Physical proprieties of ferrites nanoparticles. *Journal of Superconductivity and Novel Magnetism*. ***26*(12):** 3443–3447.

Okuma, O., & Horikawa, T. (2015). Production of Activated Carbon With Large Specific Surface Area From a Sawdust Mixed With a Nitrogen Compound By Chemical Activation Using.

Raut, A. V., Barkule, R. S., Shengule, D. R., & Jadhav, K. M. (2014). Synthesis, structural investigation and magnetic properties of Zn^{2+} substituted cobalt ferrite nanoparticles prepared by the sol–gel auto-combustion technique. *Journal of Magnetism and Magnetic Materials*. **pp:** *358–359*, 87–92.

Salehi, E., Abedi, J., & Harding, T. (2009). Bio-oil from sawdust: Pyrolysis of sawdust in a fixed-bed system. *Energy and Fuels*. ***23*(7):** 3767–3772.

Sharma, H. B., Singh, N. G. B., Singh, S. B., & Devi, T. H. D. (2014). Synthesis and Characterization of Cobalt Ferrite (Cofe 2 O 4) Nanoparticles by Sol-Gel Autocombustion Method. **Issue: 3**.

Shinde, A. B. (2013). Structural and Electrical Properties of Cobalt Ferrite Nanoparticles. **(4):** 64–67.

Singh, R., Havlica, J., Hnatko, M., Šajgalík, P., & Alexander, C. (2016). Magnetic properties of

Co 1 − x Zn x Fe 2 O 4 spinel ferrite nanoparticles synthesized by starch - assisted sol – gel autocombustion method and its ball milling. **pp:** 1–33.

Sinha, S., Jhalani, A., Ravi, M. R., & Ray, A. (2000). Modelling of Pyrolysis in Wood : A Review. *SESI Journal*. **Vol.*10*:** 1–17.

Toksha, B. G., Shirsath, S. E., Patange, S. M., & Jadhav, K. M. (2008). Structural investigations and magnetic properties of cobalt ferrite nanoparticles prepared by sol – gel auto combustion method. **Vol.*147*:** 479–483.

Vatta, L. L., Sanderson, R. D., & Koch, K. R. (2006). Magnetic nanoparticles : Properties and potential applications. *Advanced Materials*. ***78*(9):** 1793–1801.

Viswanathan, B., Neel, P., & Varadarajan, T. (2009). Methods of activation and specific applications of carbon materials. *Methods of Activation and Specific Applications of Carbon Materials*. **Vol.160.**

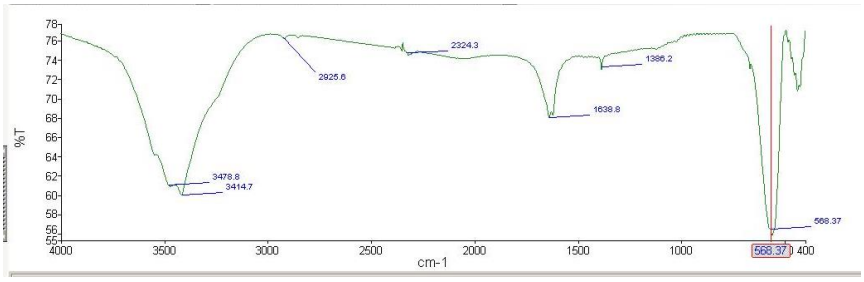
Woods, T.. Industrial Sample Analysis William Herschel.

Xu, J., Yang, H., Fu, W., Du, K., Sui, Y., Chen, J., Zou, G. (2007). Preparation and magnetic properties of magnetite nanoparticles by sol-gel method. *Journal of Magnetism and Magnetic Materials*. ***309*(2):** 307–311.

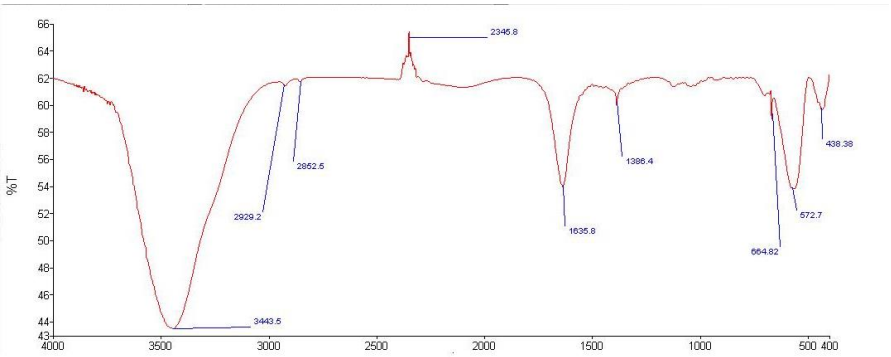
Zhao, Z. W., Ouyang, K., & Wang, M. (2010). Structural macrokinetics of synthesizing ZnFe2O4 by mechanical ball milling. *Transactions of Nonferrous Metals Society of China (English Edition)*. ***20*(6):** 1131–1135.

APPENDIX A
SAMPLE APPENDIX 1

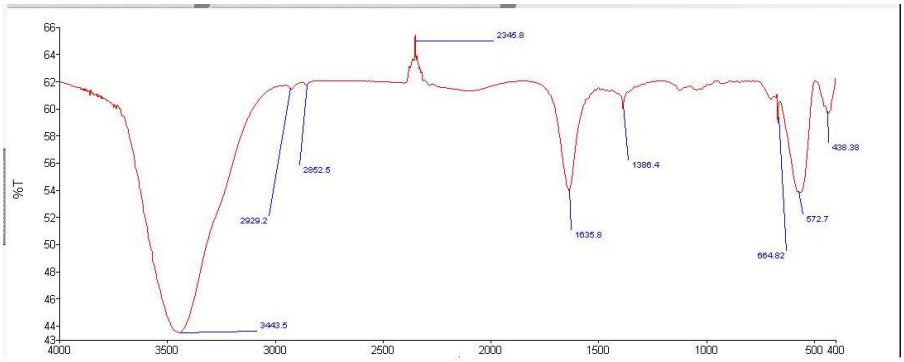
The raw data obtained from FTIR analysis to determine the chemical composition of carbon doped copper zinc ferrite nanoparticles.



100% $Cu_{0.2}Zn_{0.8}Fe_2O_4$

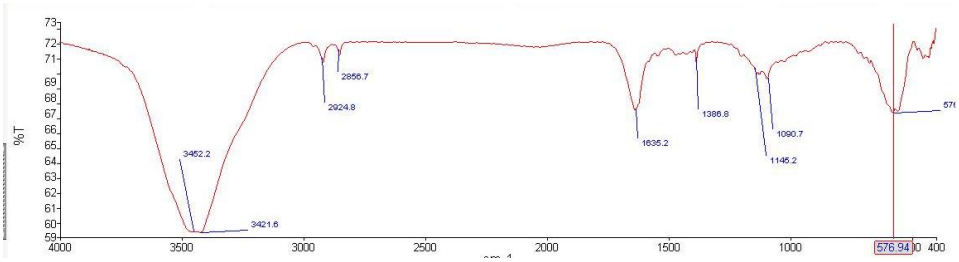


80% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 20% C

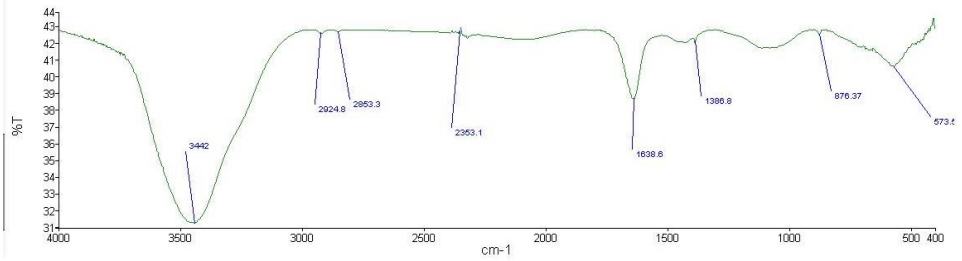


60% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 40% C

APPENDIX B
SAMPLE APPENDIX 2



40% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 60% C



20% $Cu_{0.2}Zn_{0.8}Fe_2O_4$: 80% C