EFFECT OF MAGNETIC OF ADDITION $Co_{0.5}Ni_{0.5}Fe_2O_4$ NANOPARTICLES ON THE MICROSTRUCTURE AND ELECTRIC TRANSPORT PROPERTIES OF YBCO SUPERCONDUCTOR

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for the award of the degree of

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JANUARY 2017

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I dedicate my disquisition work to my family and many friends. A special feeling of gratitude to my parents whose words of encouragement and push for tenacity ring in my ears. My family that have never left my side.and a very special thanks to my friends, without them none of my success would be possible, and along with all hard working and respected lecturers.

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ABSTRACT

In this research, the effect of small amount of $Co_{0.5}Ni_{0.5}Fe_2O_4$ magnetic nanoparticles were investigated to improve the microstructure and electric transport properties of YBCO superconductor. Pure $YBa_2Cu_3O_7$ was synthesized by solid state reaction method and underwent several characterization which were Meissner effect, phase confirmation of YBCO composite was done X-ray Diffractometer (XRD). Then,microstructural analysis is performed using Scanning Electron Microscope (SEM) also critical temperature, T_c was measured by using four point probe method. For Meissner effect, the sample with 0.04 wt% of $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles showed the longest time levitating on the permanent magnet with 20.87 seconds which can be proven that the sample have higher T_c compared to other. Next, the XRD pattern indicated that all the samples were in single orthorhombic crystal structure. The internal microstructure of YBCO showed porosity in grain and size particle decreased when the small amount of nanoparticles was added. Lastly, the T_c cannot be identified through four point probe method due to the result were significantly different with temperature of liquid nitrogen.

ABSTRAK

Dalam kajian ini, kesan sedikit nanozarah magnet $Co_{0.5}Ni_{0.5}Fe_2O_4$ telah disiasat untuk meningkatkan mikrostruktur dan pengangkutan elektrik sifat YBCO superkonduktor. $YBa_2Cu_3O_7$ Pure disintesiskan oleh kaedah tindak balas keadaan pepejal dan mengalami beberapa pencirian yang kesan Meissner, pengesahan fasa YBCO komposit dilakukan X-ray Diffractometer (XRD), analisis mikrostruktur dilakukan dengan menggunakan Mikroskop Imbasan Elektron (SEM). Selain itu, suhu kritikal, T_c diukur dengan menggunakan kaedah penduga empat titik. Untuk kesan Meissner, sampel dengan 0.04 wt% daripada nanopartikel $Co_{0.5}Ni_{0.5}Fe_2O_4$ menunjukkan masa yang paling lama terapung pada magnet kekal dengan 20.87 saat. Ini membuktikan bahawa sampel dengan 0.04 wt% daripada nanopartikel $Co_{0.5}Ni_{0.5}Fe_2O_4$ mempunyai lebih tinggi T_c berbanding dengan yang lain. Seterusnya, corak XRD menunjukkan bahawa semua sampel adalah dalam struktur kristal otorombik tunggal. Mikrostruktur dalaman YBCO menunjukkan keliangan dalam bijirin dan saiz zarah berkurangan apabila jumlah nanopartikel yang kecil ditambah. Akhir sekali, suhu kritikal, T_c tidak dapat dikenal pasti melalui kaedah penduga empat titik kerana hasilnya adalah jauh berbeza dengan suhu cecair nitrogen.

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LIST OF SYMBOLS

T_c	-	critical temperature
J_c	-	critical current density
H_c	-	critical field
H	-	applied field
B	-	magnetic field
R	-	resistivity
s	-	second
wt%	-	weight percentage
λ	-	wavelength
2θ	-	Bragg angle
K	-	kelvin
^oC	-	degree celcius
Å	-	angstrom (10^{-10})
d	-	interplanar spacing in Bragg's law

LIST OF ABBREVIATIONS

XRD	-	X-Ray Diffractometer
SEM	-	Scanning Electron Microscope
$YBa_2Cu_3O_7$	-	yttrium barium copper oxide
Y_2O_3	-	yttrium oxide
$BaCO_3$	-	barium carbonate
CuO	-	copper (II) oxide
$Co_{0.5}Ni_{0.5}Fe_2O_4$	-	Cobalt nickel ferric oxide

CHAPTER 1

INTRODUCTION

1.1 BACKGROUND OF THE PROBLEM

Superconductivity is study about materials that offers no electrical resistance that is zero resistivity, $R = 0$ and expels magnetic fields,or show perfect diamagnetism ($B_{\text{Inside}}=0$) when the material is cooled to adequate temperature (normally in liquid helium temperature range). In 1911, K. Onnes observed the behaviour of superconductivity of mercury in liquid helium and he noticed that the resistance disappeared below critical temperature $T_c \sim 4$ K. Critical temperature of several materials such as lead (Pb), aluminum (Al) and some alloys have been discovered. High temperature of an oxide superconductor, LaBaCuO recorded at 77 K was found by Georg Bednorz and Alex Muler (1986) and it strongly depends on their structure.

Generally, superconductors are divided into two types; Type-I and Type-II. In Type-I, the superconductors must be kept at below critical temperature, T_c to ensure the magnetic susceptibility stay at negative one (- 1) and based on Figure 1.1, the critical field, H_c higher than applied field, H. But the superconductivity can be destroyed if the applied magnetic field is stronger than critical magnetic field, $H > H_c$, the magnetic field are able to penetrate into superconductor causes an extinguish of the superconducting state and it no longer be superconductor (perfect conductor) even though the temperature below critical temperature, T_c and this phenomena show Type-I superconductor perfectly obeys Meissner effect. Meissner effect describe the properties of superconductor where superconducting materials creates current which oppose the magnetic field inside the superconductor. Walther Meissner and Robert Ochsenfeld found Meissner effect in superconductors which is also known as perfect diamagnetism ($B_{\text{inside}} = 0$).

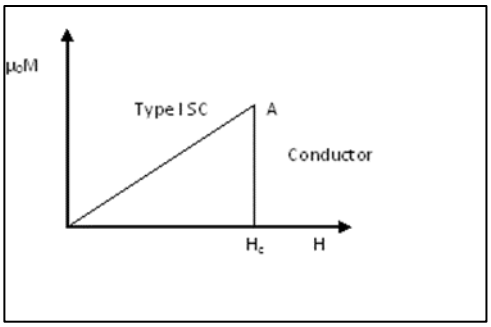


Figure 1.1: The magnetization versus applied magnetic field for Type-I superconductor

Besides, Type-II superconductor can behave like Type-I superconductor (Meissner phase) as shown in Figure 1.2. $H < H_{c1}$ the magnetic field destroyed magnetic flux and magnetization inside superconductor become zero. Nonetheless, at one point the applied magnetic field, H reaches the critical magnetic field, H_{c1} the magnetic fluxes enter uniformly and slowly start loses the superconductivity. When H_{c1} passed upper critical magnetic field, H_{c2}, the superconductivity behavior completely disappears. This is called 'mixed state' ($H_{c1} < H < H_{c2}$) in which quantized vortices flux penetrates the material without demolishing superconductivity. The addition of nanoparticles or impurities can be increased the current density, J_c that act as flux pinning center.

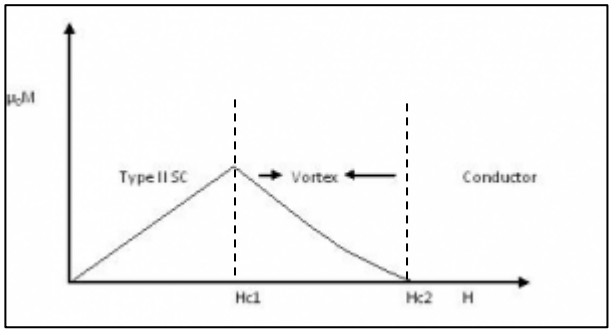


Figure 1.2: Magnetization versus applied magnetic field for type II superconductor.

1.2 PROBLEM STATEMENT

Copper oxide based superconductor ($YBa_2Cu_3O_7$) at the normal state can be applied at electronic device but we cannot use in application because the critical current density, *Jc* is too low. Regarding this issue, the addition of magnetic nanoparticles, $Co_{0.5}Ni_{0.5}Fe_2O_4$ into $YBa_2Cu_3O_7$ is needed to increase the critical temperature to be used for various type of application. Furthermore, due to low critical current density J_c in Type-II superconductor, the vortex motion different amount of magnetic nanoparticles might change critical current density, J_c, critical temperature, T_c, microstructure and Meissner effect of $YBa_2Cu_3O_7$ superconductor compared to electrical properties in pure YBCO.

In the other hand, in order to measure the electrical properties of superconductor, four point probe must be developed in FIST laboratory in Universiti Malaysia Pahang. Four-point probe technique is easy and fast method to measure the direct current (DC) resistance in superconductor.

1.3 OBJECTIVES OF RESEARCH

i. To study the change in the microstructure of $YBa_2Cu_3O_7$ with the addition of 0.00 wt%, 0.01 wt%, 0.02 wt%, 0.03 wt% and 0.04 wt% magnetic $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles.
ii. To study the electrical transport properties of YBCO with the addition of 0.00 wt%, 0.01 wt%, 0.02 wt%, 0.03 wt% and 0.04 wt% magnetic $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles.
iii. To develop a laboratory scale method in determining the electrical properties of YBCO at liquid nitrogen liquid.

1.4 Scope of the Study

In this study, the effect of magnetic of addition $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles on the microstructure and electric transport properties of YBCO superconductor has been investigated. $YBa_2Cu_3O_7$ superconductor is used as a sample and the $Co_{0.5}Ni_{0.5}Fe_2O_4$ is used as magnetic nanoparticles. It involves three major processes which are hand grinding, sintering and pelletization. The samples were added with magnetic nanoparticles with different weight percentage which were 0.01 to 0.04 wt% and in the end of the product are pellets of $YBa_2Cu_3O_7$superconductor. The Meissner effect is analyzed and showed the levitation of superconductor. The $YBa_2Cu_3O_7$ superconductor is measured by using four-point probe technique as a fast and easy method to find electrical properties and critical temperature. Furthermore, in order to characterize the microstructure and phase formation of $YBa_2Cu_3O_7$, it has been studied by using Scanning Electron Microscope (SEM) and X-Ray Diffraction (XRD) method respectively.

CHAPTER 2

LITERATURE REVIEW

2.1 HISTORY 0F $YBa_2Cu_3O_7$ SUPERCONDUCTOR

Superconductivity is the unique phenomena that occur in a certain material like YBCO. The basic properties of superconductor are zero resistivity and perfect diamagnetism ($B = 0$). In 1987, Wu and a team discovered that Type II superconductor which is YBCO that possesses high critical temperature T_c around (~ 92 K) over the boiling point of liquid nitrogen (77 K). So, the lattice parameter, the valance ratio and the sample treatment play an important role to achieve superconductor above 77 K. (Wu et al, 1987)

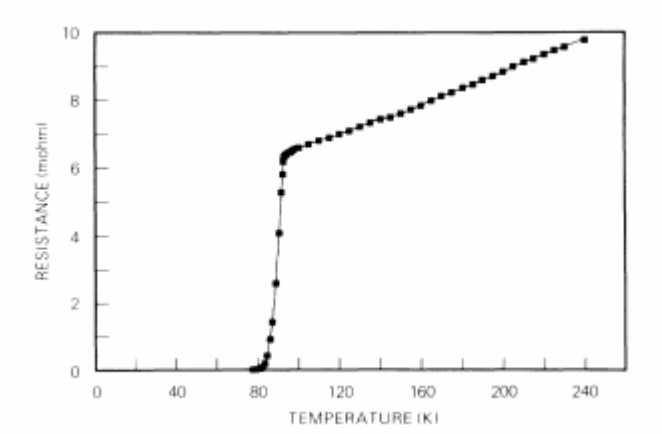


Figure 2.1: The effect of magnetic field on resistance .

Source:Wu et al, 1987

2.2 STRUCTURE OF $YBa_2Cu_3O_7$ SUPERCONDUCTOR

The structure of $YBa_2Cu_3O_7$ as shown in Figure 2.2 plays an important role for understanding of superconducting material. Basically, structure of YBCO superconductor has been determined as a deformed, oxygen deficient multi-layered perovskite superconductor. The $YBa_2Cu_3O_{7-x}$ where $0 \leq x \leq 1$ as shown in Figure 2.2 has orthorhombic symmetry; the structure consists of copper-oxygen planes (CuO_2) with yttrium (Y) and barium (Ba) atoms. The presences of oxygen atoms are essential for superconductivity. The cupper-oxygen chains are important for superconductivity and give orthorhombic structure to $YBa_2Cu_3O_7$.

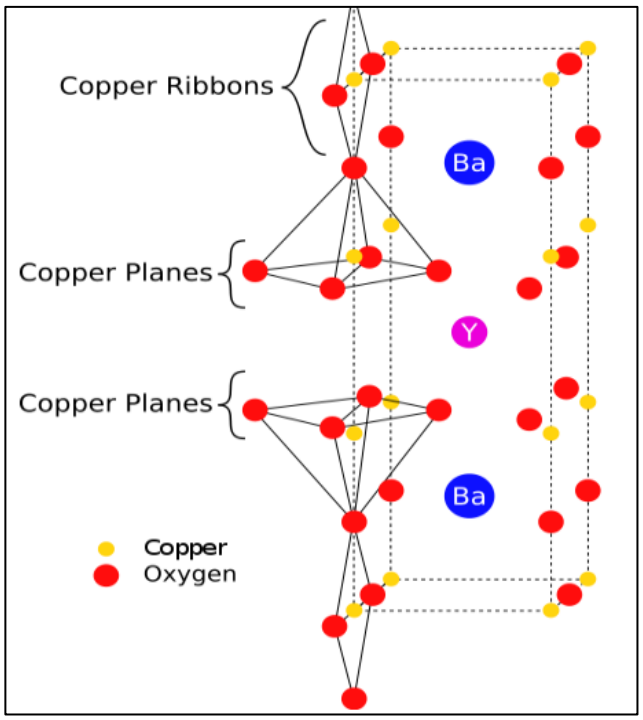


Figure 2.2: Structure of YBCO

Source: Dutta (2014)

2.3 EFFECTS OF MAGNETIC NANOPARTICLES ADDITION INTO HIGH TEMPERATURE SUPERCONDUCTOR

According to previous research about addition of nanoparticles in BSCCO, Hafiz et al. (2011) has reported the addition of nano-sized $Co_{0.5}Ni_{0.5}Fe_2O_4$ by using co-precipitation method, there was slightly significant changes to the zero resistance temperature ($T_{c\ zero}$) for low $Co_{0.5}Ni_{0.5}Fe_2O_4$ at 102 *K* compared to non-added sample at 101 *K* where can be attributed to an enhancement of the high-*T*c phase. However, with the increase in $Co_{0.5}Ni_{0.5}Fe_2O_4$ content showed a significant decrease in $T_{c\ zero}$. Besides, the addition of nano-sized $Co_{0.5}Ni_{0.5}Fe_2O_4$ in sample showed a higher critical current density, J_c but based on observation, the highest J_c was recorded in the x= 0.01% sample with degradation with increasing temperature as a consequence of thermally activated flux creep. According to researcher, the enhancement due to the small $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles addition enhances the transport critical current density in $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10}$ (Bi-2223) superconductor.

In addition, Abd-Shukor et al. (2014) has investigated the effect of addition nano-sized, where the onset transition temperature ($T_{c\ onset}$) showed no significant changes for low Co_3O_4 ($x \leq 0.02$wt%). Nevertheless, the T_c decreased monotonically with increase in Co_3O_4 content indicating a pair-break like mechanism for x= 0.03 to x= 0.12wt% showed a sudden broadening of the superconducting-transition width was also observed for $x \geq 0.11$wt%. Furthermore, in this research showed homogeneous distribution of Co_3O_4 throughout the sample was observed under scanning electron micrographs (SEM).

CHAPTER 3

RESEARCH METHODOLOGY

3.1 INTRODUCTION

In this research, $YBa_2Cu_3O_7$ superconductor were synthesized by the conventional solid-state reaction technique. The major steps involve are hand grinding, sintering and pelletization. The sample of YBCO are combination of yttrium oxide (Y_2O_3), barium carbonate ($BaCO_3$), and copper (II) oxide (CuO). Precursors YBCO were first prepared using appropriate stoichiometric ratios of high purity Y_2O_3 (99.9 %), $BaCO_3$ (99.9 %), and CuO (99.9 %) according to the chemical formula below: -

$$½\ Y_2O_3 + 2\ BaCO_3 + 3CuO \rightarrow YBa_2Cu_3O_7 \qquad (3.1)$$

From this equation, the weight of each raw material was calculated as in appendix and the process will use 4.54 g (Y_2O_3), 15.87 g ($BaCO_3$), and 9.59 g (CuO). All the element were grinded thoroughly and sintered in a furnace at 900˚C for 24 hours. For this case, sintering was repeated twice with intermediate grinding. After that, the magnetic nanoparticles $Co_{0.5}Ni_{0.5}Fe_2O_4$ were added to the precursor powder of YBCO with varied from x = 0.01 to 0.04 wt% of the total mass of the sample. The mixed powder was then grinded and pressed into pellets and sintered at 900˚C for 24 hours. After the pallet cooled down at room temperature, the YBCO pallet will be testing with several processes to characterize the YBCO superconductor.

The Meissner effect of $YBa_2Cu_3O_7$ were observed before proceed other characterizations. The structure and phase identification of the sintered samples were examined by powder XRD using a Rigaku MiniFlex II with CuK_α radiation. Scanning Electron Microscope (SEM) were used to characterize the microstructure of $YBa_2Cu_3O_7$. The critical temperature, T_c, resistivity and current density were measured by using Four- Point Probe.

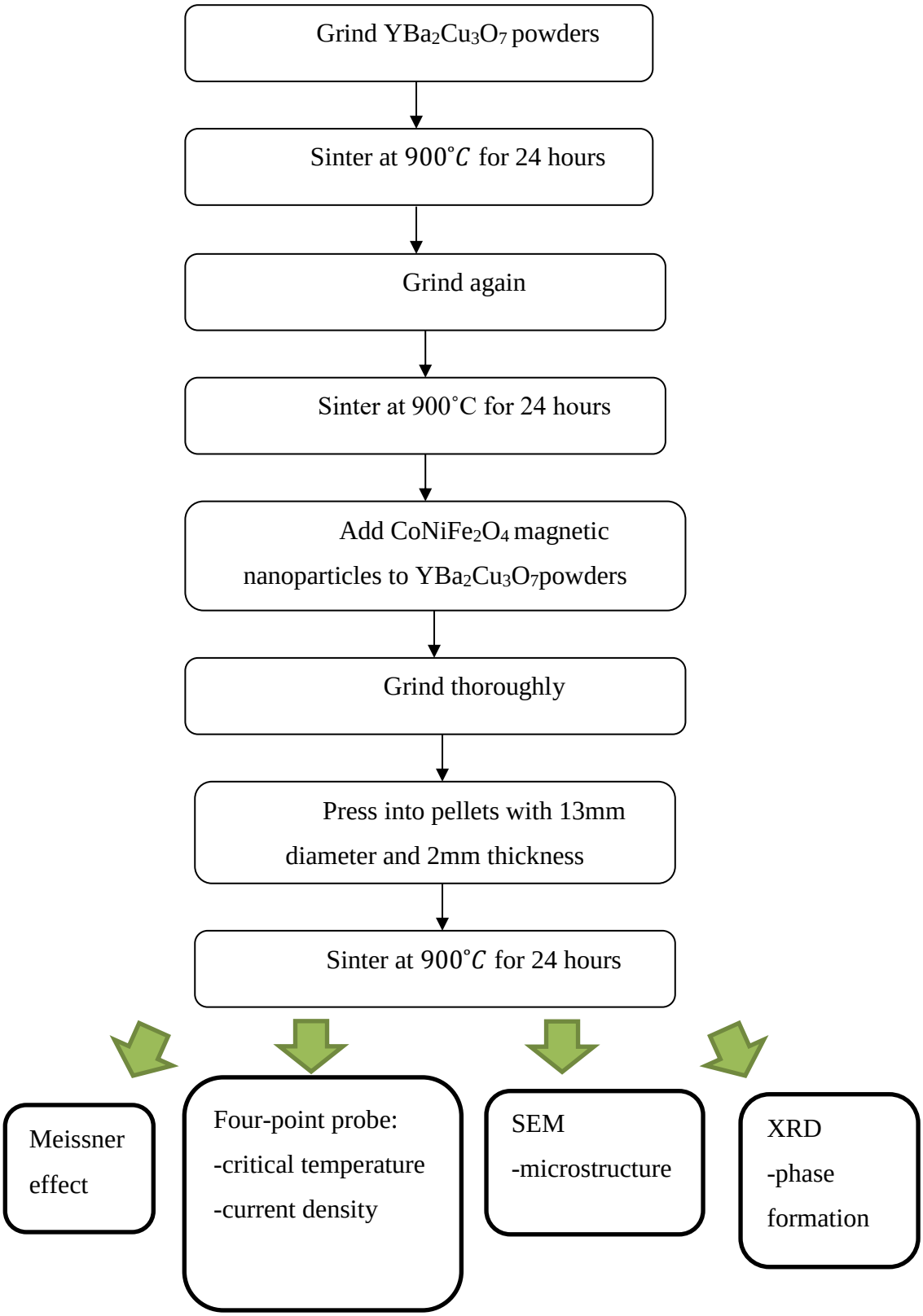


Figure 3.1: Flowchart of samples preparations and characterizations.

3.2 MATERIALS AND APPARATUS

All the chemical, materials, apparatus and machine that were used to prepared and characterized $YBa_2Cu_3O_7$ superconductor with 0.01 wt%, 0.02 wt%, 0.03 wt% and 0.04 wt% addition were provided as shown table below.

Table 3.1: Chemical and Materials

Yttrium oxide, Y_2O_3	7.565g
Barium carbonate, $BaCO_3$	26.445g
Copper oxide, CuO	15.990g
Cobalt nickel ferric oxide, $Co_{0.5}Ni_{0.5}Fe_2O_4$	1g
Methanol, CH_3OH	2L
Liquid nitrogen, N_2	1L
Distilled water	-

Table 3.2: Sample Preparation

Electronic balance	Vial (20ml)
Hydraulic press	Spatula
Evacuable Pellet Dies	Crucible and cover
Furnace	Aluminum foil
	Mortar and pestle
	Petri dish
	Plastic wash bottle
	Beaker (50ml & 100ml)
	Tissue paper
	Micrometer screw gauge

Table 3.3: Sample Characterization

X-Ray Diffraction (Rigaku miniFlex II)	Thermocouple (t type)
Scanning Electron Microscope (EVO 50)	Cable/ wire
Four-point probe	Heat shrink sleeve
	Banana plugs
	Soldering iron & stand
	Solder
	Millimeter
	Silver paint
	Neodymium magnet (N35)

3.3 METHOD OF PREPARATION

3.3.1 SOLID STATE METHOD

i. Grinding Method

In solid state chemistry, a mortar and pestle is often used to prepare reactants for a solid-state synthesis (the ceramic method). Mortar and pestle is used to crush, grind and mix solid substances. Nowadays, there are various tools used for grinding in laboratory such as grain mills (automatic) for large amount of sample. The kind of mortar and pestle used in the preparation of a solid-state material is very important. If the sample being ground is harder than the material comprising the mortar, significant contamination of the sample can be expected. For this reason, porcelain (fired clay) is not typically used for solid state synthesis. In addition, porcelain is a porous material and can be difficult to clean. Instead porcelain, Agate (a form of quartz) is usually the best choice of material in term of easy to handle even though the price is expensive.

The $YBa_2Cu_3O_7$ sample which are in powder form are weighed using electronic balance based on chemical equation which are 4.54 g (Y_2O_3), 15.87 g ($BaCO_3$), and 9.59 g (CuO). All the element mixed together by using mortar and pestle for two hour to get the gray powder of YBCO. Grinding is continued until the mixture is homogeneous and intermediate grinding is needed where grind more than one time. $Co_{0.5}Ni_{0.5}Fe_2O_4$ powder was added to immaculate superconducting YBCO powder in various wt%. The differ composites were ground independently for 2 hour. The different wt% of $Co_{0.5}Ni_{0.5}Fe_2O_4$ added to YBCO are x= 0.01,0.02, 0.03, 0.04 wt%.

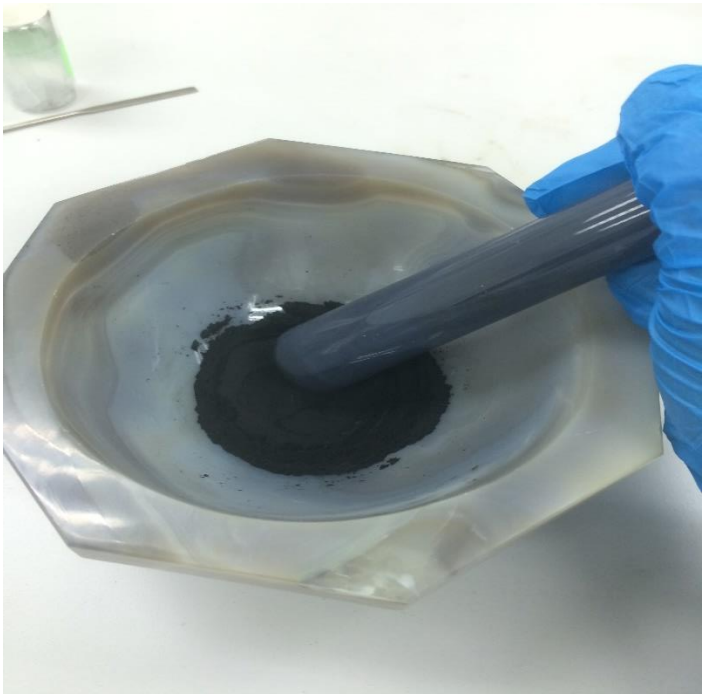


Figure 3.2: Hand grinding using mortar and pestle

ii. Sintering Method

Sintering is thermal treatment of fine-grained material at a temperature below the melting point of the main constituent, for the purpose of increasing its grain size and strength by bonding together the particles. The atomic diffusion takes place and the welded areas formed during compaction grow until eventually may be lost completely. Re-crystallization and grain growth may follow, and the pores tend to become rounded and the total porosity, as a percentage of the whole volume, tends to decrease. However, sintering can proceed only locally, for example at contact point of grains, without any appreciable change in the average overall density of a powder compact.

For this research, the mixture powder of $YBa_2Cu_3O_7$ is placed in the crucible and then sintered for 24 hours with temperature 900 ˚C in Nabertherm furnace. In this case, $YBa_2Cu_3O_7$ powder required second sintered at the same time and temperature with intermediate grinding before powder is added with magnetic nanoparticles and third sintering after pelletization and permitted to cool gradually inside the furnace to room temperature.

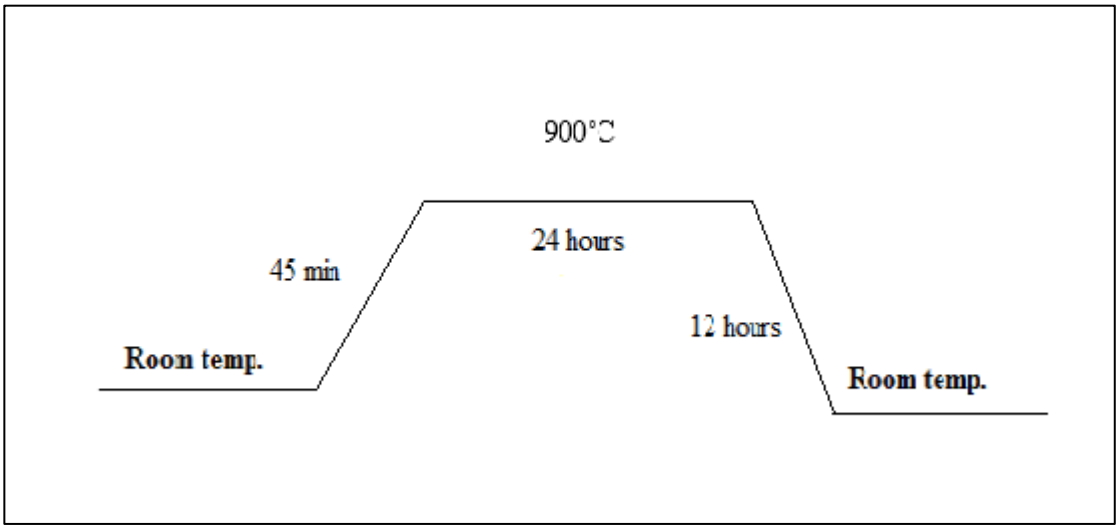


Figure 3.3: Heating profile for sintering method

3.3.2 Pelletization method

Pelletization is a technique that converts fine powders or granules of spherical beads or pellet. Pelletization leads to a refinement in the flowability, mixing properties and appearance thereby avoiding generation of immoderate dust and reducing segregation, eliminating undesirable properties also improving the chemical and physical properties of fine powder. (Supriya et al. 2012)

All composition of sintered powders are weighed 1.5 g to make four sample each of wt% element and bulk sample for SEM and four point probe characterization. All the weighed powders are pelletized using hand pellet press with pellet press dies. The diameter and thickness of mold or die is 13 mm and 3 mm respectively. Hydraulic press was used to pressed the sample with applied pressure is 8 tons and left for 30 seconds before taken out. Next, the pellet was sintered for 24 hours and used for characterization.

3.4 MATERIAL CHARACTERIZATION

3.4.1 Meissner effect

All the precursor powder of YBCO with addition $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles varied from x = 0.01 to 0.04 wt% were immersed in liquid nitrogen about 2 minutes. The sample were put on the Neodymium magnet (N35) and the time of sample levitating were recorded by using stopwatch. The method was repeated three times and the average reading are taken to get accurate result.

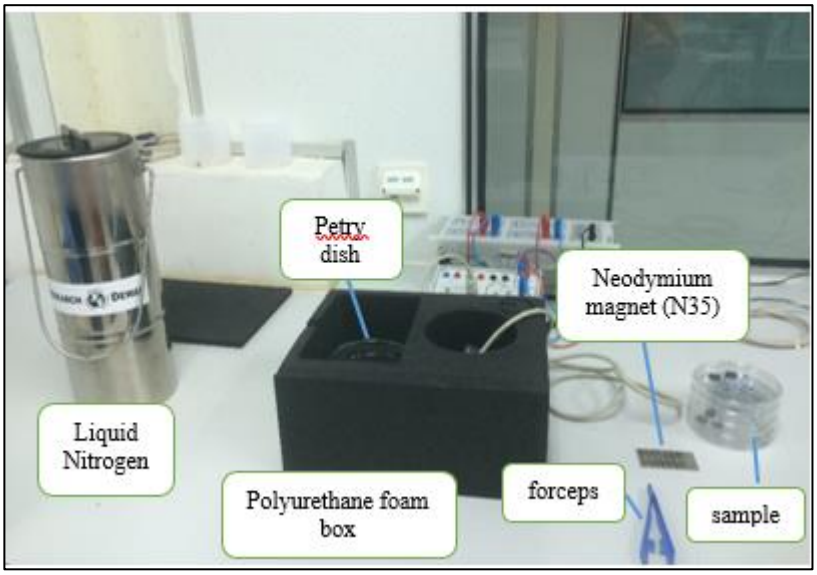


Figure 3.4: The equipment for Meissner effect testing.

3.4.2 X-Ray Diffractometer (XRD)

X-ray diffraction depends on constructive interference of monochromatic X-rays and a crystalline sample. These X-ray are created by a cathode beam tube, separated to deliver monochromatic radiation, collimated to concentrate, and directed toward the sample. The interaction of the incident ray with the sample produces constructive interference and a diffracted beam when conditions fulfill Bragg's Law.

$$n\lambda = 2d \sin \theta \tag{3.2}$$

This law relates the wavelength of electromagnetic radiation to the diffraction angle and the lattice spacing in a crystalline sample. These diffracted X-rays are then detected, processed and calculated. By filtering the specimen through a range of 2θ angles, all possible diffraction directions of the lattice should be attained because of the random orientation of the powdered material. Transformation of the diffraction peaks to *d*-spacing from the result, the crystal structure can be calculated by using formula below:

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2} \quad (3.3)$$

Where *h, k, l* are miller indices and *a, b, c* are unit cell length or parameter.

Figure 3.5: X-Ray Diffractometer (XRD) machine.

X-ray diffraction was conducted using a Rigaku Miniflex X-ray diffractometer. The sintered powder will be testing to analyze phase formation of $YBa_2Cu_3O_7$ superconductor, $Co_{0.5}Ni_{0.5}Fe_2O_4$ magnetic nanoparticles and CuO with scan range angle from 20˚ to 80˚. The X- Ray of Kα Cu radiation filter by Ni has a wavelength, (λ =1.54Å). The samples were scanned at a rate of 2.0˚/min while sampling at every 0.02˚. the XRD will the running the sample and display the result through desktop within one hour.

3.4.3 Scanning Electron Microscopic (SEM)

Scanning Electron Microscope (SEM) provides details surface information by tracing a sample in a raster pattern with an electron beam was used in analysis $YBa_2Cu_3O_7$ superconductor. High energy electron beam is used to excite the specimen and the signals are collected and analyzed so that an image can be constructed. Primary electrons are focused and deflected by electronic lens to produce a narrow scan beam that bombards the specimen in high vacuum condition. The image magnification can reach until ~10000X.

Figure 3.6 shows the Scanning Electron Microscope (SEM) that used for analysis microstructure of $YBa_2Cu_3O_7$ superconductor. This Evo 50 SEM provide the image magnification reach to 5000X resolution. Firstly, the sample must be coated with a thin layer of conducting material which is palladium. Sputter coater machine (Figure 3.7) is used to coated the $YBa_2Cu_3O_7$ superconductor with 40 mA current flow in vacuum state, $1.32^{-0.02}$ Pa for 70 s. All the sample must be coated because to prevent charging of a sample with an electron beam and to increase signal to noise ratio.

Figure 3.6: Scanning Electron Microscope (SEM) machine.

Figure 3.7: Sputter coater machine.

3.4.4 Four Point Probe Method

A four-point probe is a simple apparatus for measuring the resistivity of superconductor samples. By passing a current through two outer probes and measuring the voltage through the inner probes allows the measurement of the substrate resistivity. The four-point device was set up to conduct this experiment. It is consisted of two constant current wire connected at the ends of $YBa_2Cu_3O_7$ superconductor, two voltage wire are connected to voltmeter as shown in Figure 3.8.

Thermocouple is a sensor that consist of two wire legs made from different metals. Thermocouple consist various type such as type K, type J and type T. Type T thermocouple are made copper and constantan metal was used in this experiment. The Type T is a very stable thermocouple and is often used in extremely low temperature applications such as cryogenics or ultra low freezers.

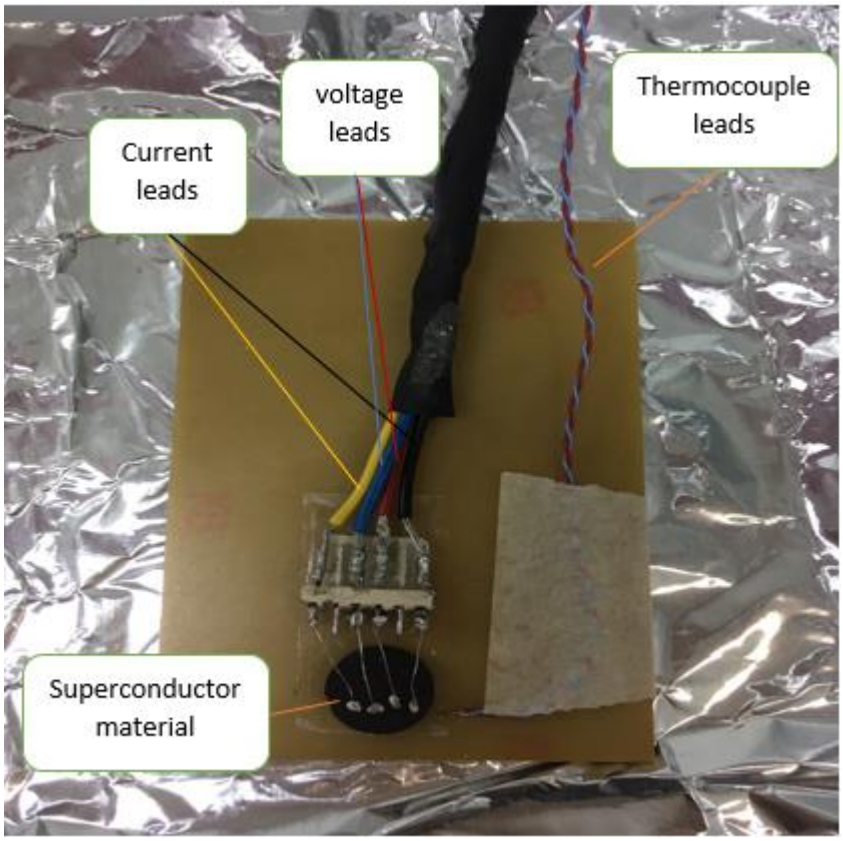


Figure 3.8: Four point probe device.

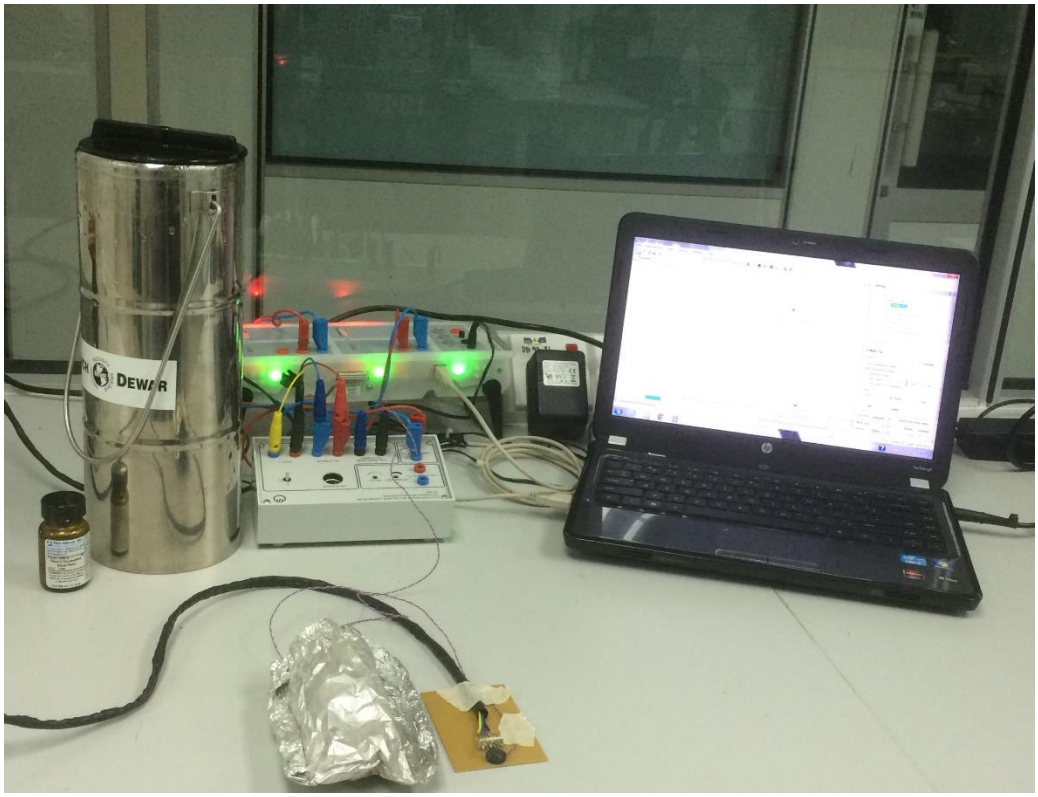


Figure 3.9: The equipment for Four point probe testing.

CHAPTER 4

RESULT AND DISCUSSION

4.1 CHARACTERIZATION OF YBa2Cu3O7 SUPERCONDUCTOR

4.1.1 Meissner Effect

The Figure 4.1 shown the $YBa_2Cu_3O_7$ superconductor with addition of 0.01 wt%, 0.02 wt%, 0.03 wt% and 0.04 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles were levitated on the permanent magnet. During Meissner effect testing, sample with 0.04 wt% took the longest time to levitate on the permanent magnet which is 20.87 seconds followed by 0.03 wt%, 0.02 wt% and 0.01 wt%. the average reading is shown in Table 4.1.

Table 4.1: The time of samples levitating above the permanent magnet.

Sample	Time (seconds)			
	1st reading	2nd reading	3rd reading	Average reading
0.01 wt%	15.58	17.26	16.33	16.39
0.02 wt%	19.70	16.90	19.12	18.57
0.03 wt%	19.88	19.50	20.46	19.95
0.04 wt%	18.55	24.90	19.16	20.87

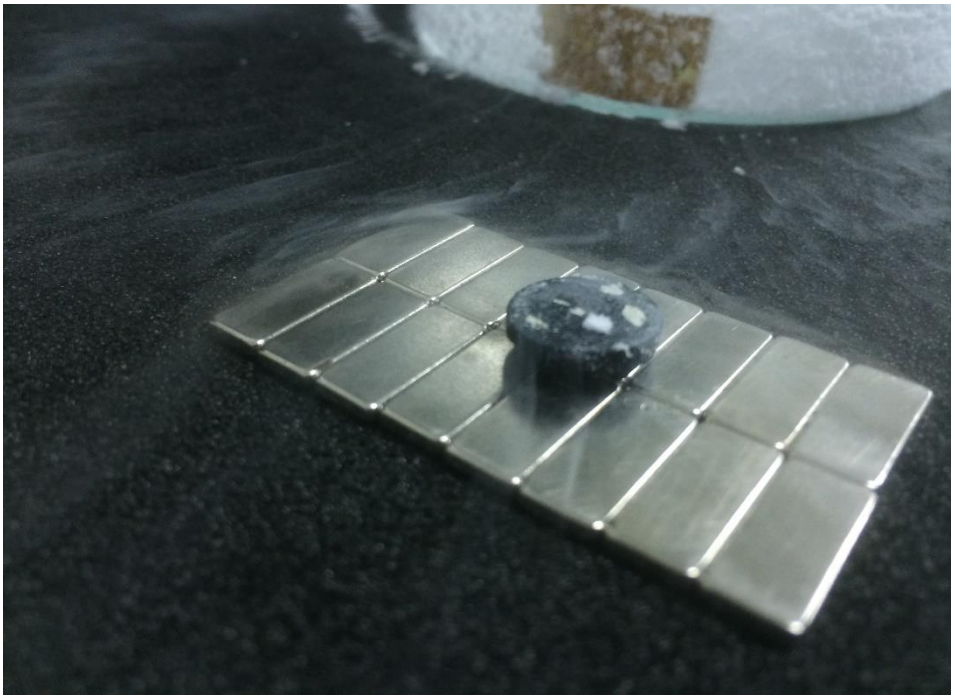


Figure 4.1: The YBCO superconductor levitate due to Meissner effect.

When superconductor at a temperature below its critical temperature, *Tc*, the magnetic field will not allow to enter freely. The superconductor will levitate until it reaches its *Tc*. The sample 0.04 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticle showed the longest levitation time with average 20.87 second while the shortest levitation time goes to sample with 0.01 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticle. Hence, the higher the levitation times, the higher the *Tc*. The addition of nanoparticles give the effect of YBCO superconductor to increase the *Tc* with appropriate weight percentage.

4.1.2 Structural Analysis by XRD

The XRD powder patterns of YBCO and YBCO+ xCoNiFeO (x = 0.01 wt%, 0.02 wt%, 0.03 wt%, 0.04 wt%) are shown in Figure 4.2 and 4.3 respectively. The XRD pattern indicate that all the samples are in single orthorhombic crystal structure with space group Pmmm (symmetry class for orthorhombic). For pure YBCO which is non-added sample, shown the highest peak at (013) with angle 32.77˚ act as reference sample.

Figure 4.3 shows the combination four graph of YBCO+ $xCo_{0.5}Ni_{0.5}Fe_2O_4$ (x = 0.01wt%, 0.02 wt%, 0.03 wt%, 0.04 wt%). The graph slightly shift as the angle increase compared to pure YBCO, which were mostly the highest peak at (013) with angle 33.09˚. Some $Co_{0.5}Ni_{0.5}Fe_2O_4$ peaks are also seen in the pattern. It is also observed that the structure has not changed even after addition of $Co_{0.5}Ni_{0.5}Fe_2O_4$. The symmetry has also not changed after addition. It concludes that the addition of $Co_{0.5}Ni_{0.5}Fe_2O_4$ to pure YBCO has no significant effect on the structure and symmetry of the composites. The lattice parameters were calculated and shown in Table 4.2.

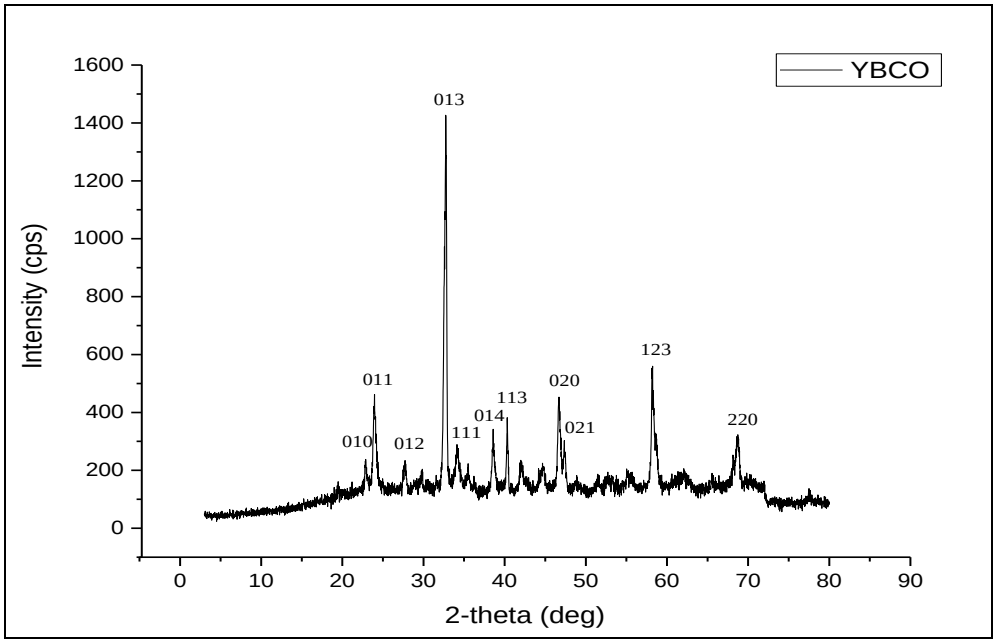


Figure 4.2: XRD pattern of pure $YBa_2Cu_3O_7$ superconductor.

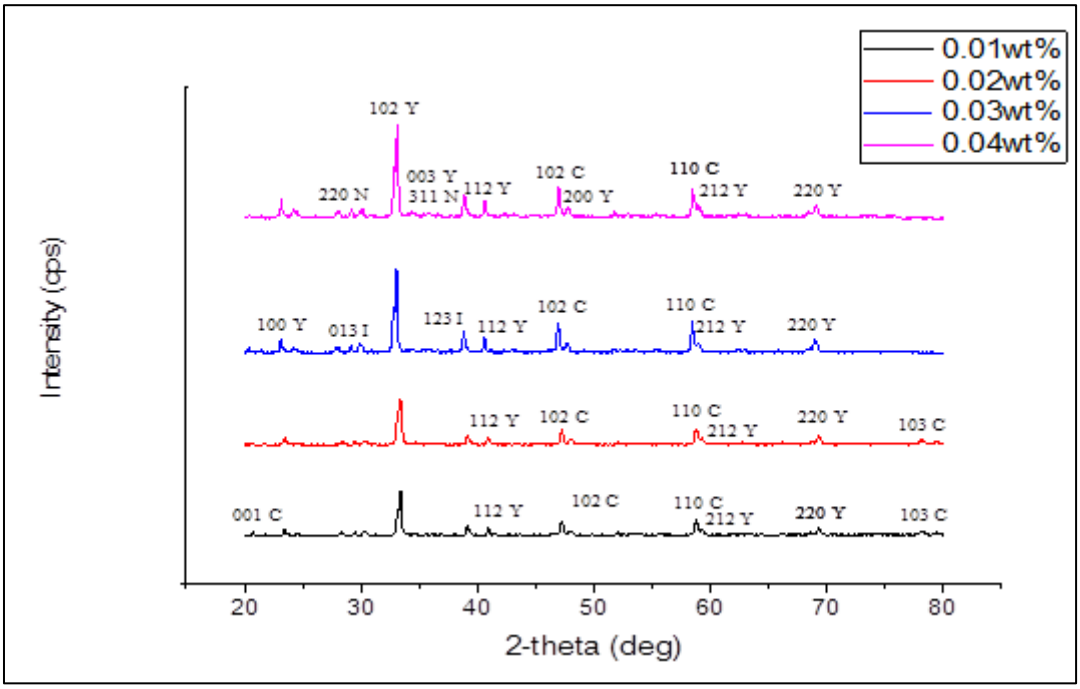


Figure 4.3: XRD pattern of $YBa_2Cu_3O_7$ with different addition of magnetic nanoparticles $Co_{0.5}Ni_{0.5}Fe_2O_4$. In the graph, Y, C, I indicate $YBa_2Cu_3O_7$, CuO and $Co_{0.5}Ni_{0.5}Fe_2O_4$ respectively.

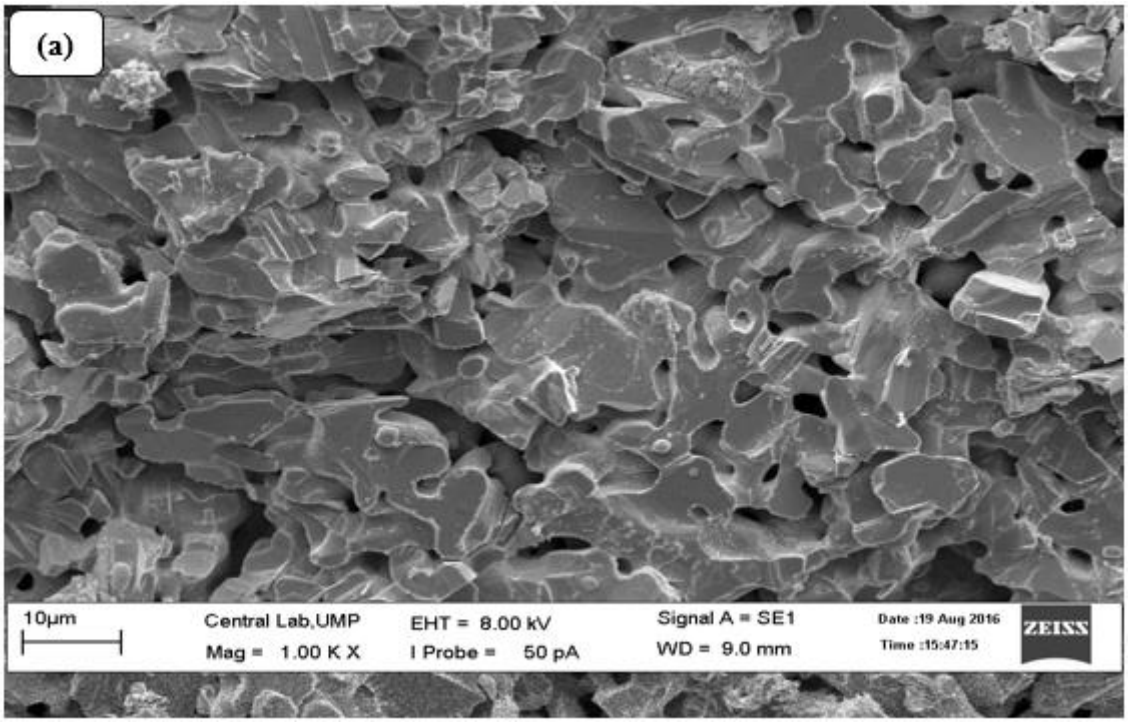
Table 4.2: Lattice parameters calculated from XRD graphs.

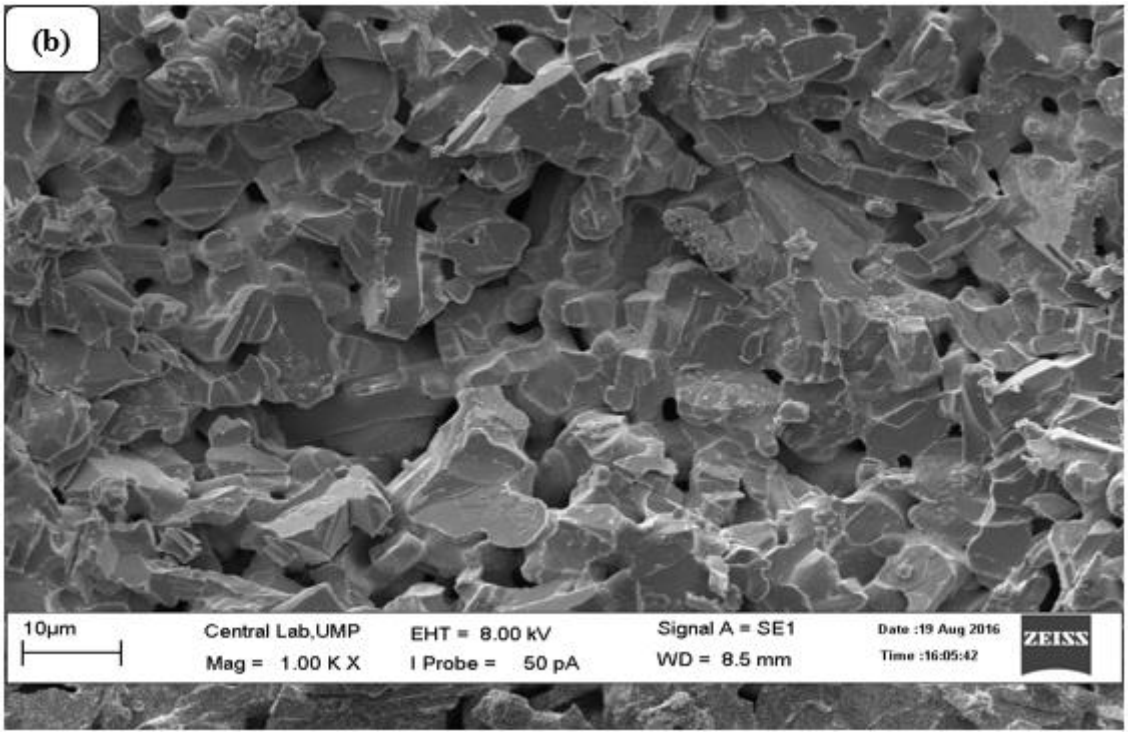
CoNiFeO (wt.%)	a (Å)	b (Å)	c (Å)
YBCO	3.4058	3.8890	11.7175
0.01	3.8510	3.8732	7.6310
0.02	3.8710	3.8997	7.5180
0.03	3.8486	3.8656	7.4824
0.04	3.7980	3.8649	7.5701

Refinement of the X-ray diffraction data shows that the *a* and *b* parameters of non-added sample and added sample are very close. The lattice parameters of the non-added sample were *a* =3.4058 Å, *b* = 3.889Å and *c* = 11.7175Å. However, all of the addition $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticle samples showed different c parameter with non-added samples. The XRD result also shows the unknown element that maybe could be effect the crystal structure and lattice parameter. This is maybe due to impurities in sample during grinded and pelletized the samples.

4.1.3 Scanning Electron Microscopy (SEM) Analysis

Figure 4.4 (a) – (d) shows the SEM micrographs for $YBa_2Cu_3O_7$ superconductor samples with magnetic nanoparticles $Co_{0.5}Ni_{0.5}Fe_2O_4$ has large plate grains randomly oriented in all direction with size varying from 1 to 10μm. Grains are very closely packed to each other. With the addition of $Co_{0.5}Ni_{0.5}Fe_2O_4$ the grain size decreases to a large extent with the increase of pores and voids in the sample. The grains become rounder in shape and increase in porosity also surface area as shown in Figure 4.4 (d) with 0.04 wt% of nanoparticles.




(b)
10µm
Central Lab,UMP
Mag = 1.00 K X
EHT = 8.00 kV
I Probe = 50 pA
Signal A = SE1
WD = 8.5 mm
Date :19 Aug 2016
Time :16:05:42
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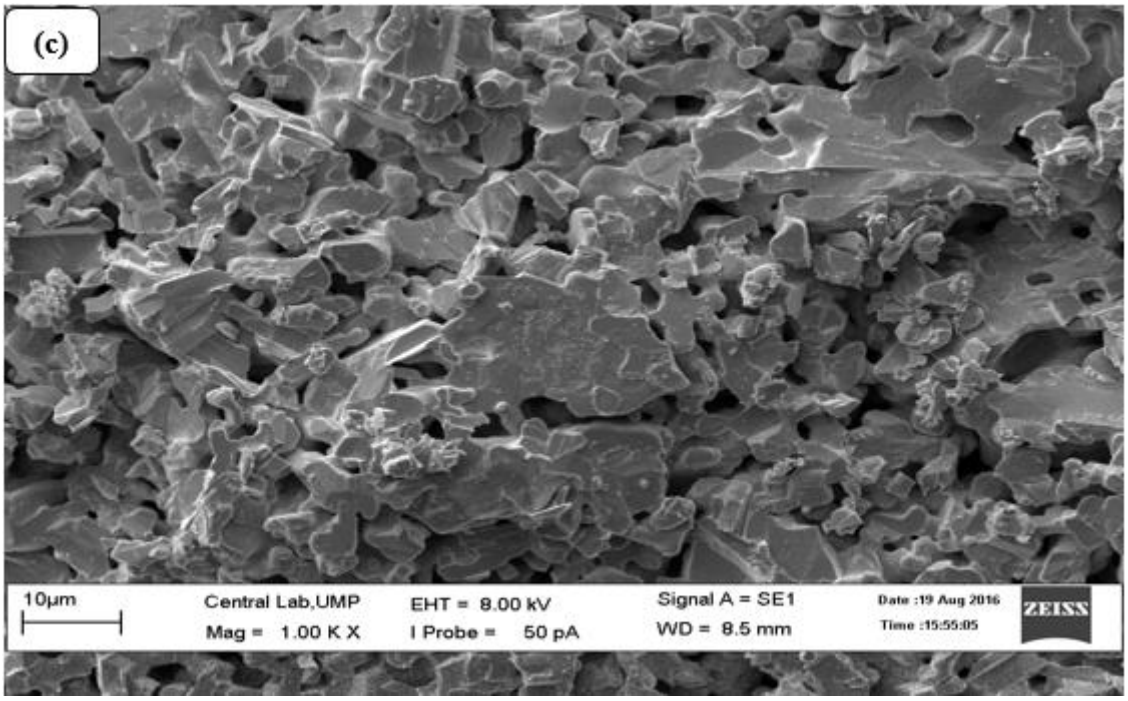
(c)
10µm
Central Lab,UMP
Mag = 1.00 K X
EHT = 8.00 kV
I Probe = 50 pA
Signal A = SE1
WD = 8.5 mm
Date :19 Aug 2016
Time :15:55:05
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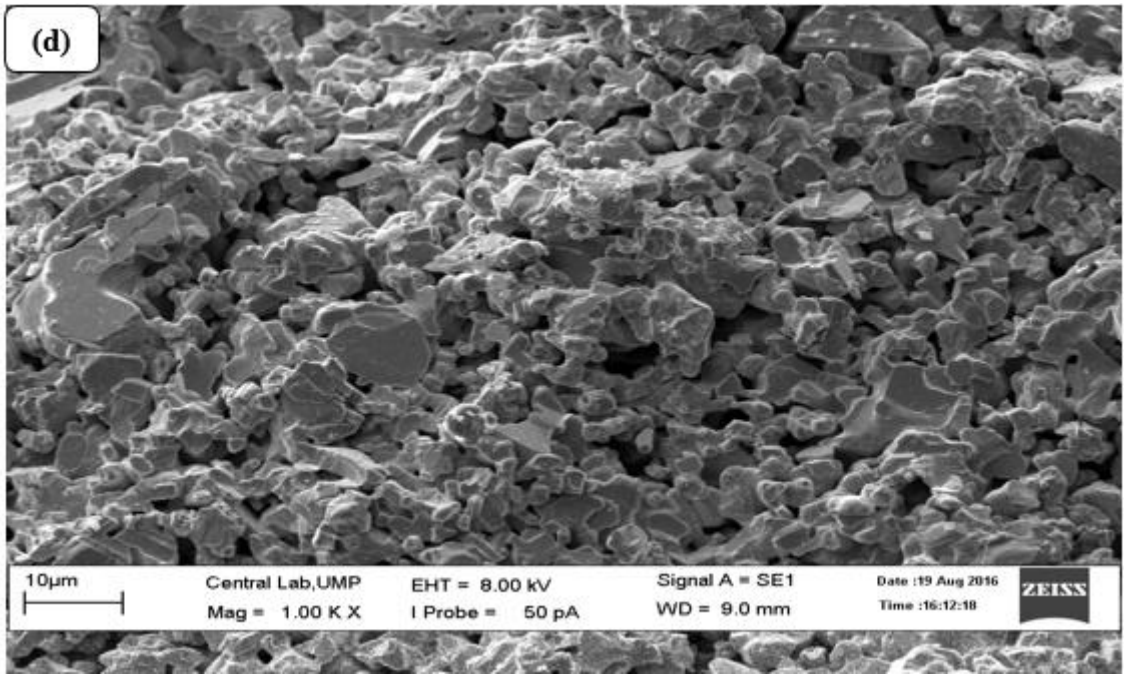

Figure 4.4 (a) - (d): Internal microstructure of sample with magnetic nanoparticles addition with magnification of 1000X, (a) x = 0.01wt%, (b) x = 0.02wt%, (c) x = 0.03wt% and (d) x = 0.04wt%

During sintering process with temperature exceed 850˚C, the grain of YBCO superconductor will develop and formed strong grains which is plate- like grain. Besides, the addition of nanoparticles showed the effect on the microstructure of YBCO superconductor. Based on the Figure 4.4 (a) and (b), the grain of sample YBCO with addition 0.01 wt% and 0.02 wt% of $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticle showed a little porosity with dominant plate-like grain while Figure 4.4 (c), the grain reduced in size with 0.03% nanoparticles and (d) obviously showed the porosity in microstructure and particle size decrease. When porosity in YBCO material increase, it may affect the electrical properties where increase flux pinning leads to increased critical current density, J_c. (Roth et al. 1990)

4.1.4 Four Point Probe Method Analysis

Measurement of the resistivity dependence of temperature for different samples with various amounts of $Co_{0.5}Ni_{0.5}Fe_2O_4$ is shown in Figure 4.5 – 4.7. From the graph below, it does not show proper reading of temperature dropping. Based on the voltage reading from Cassey lab2 software, it measured the potential drop for sample with 0.02wt%, 0.03wt% and 0.04wt% of nanoparticle. From potential drop reading, the critical temperature, T_c is calculated but the value T_c is too big and cannot be considered as T_c value. For sample with 0.01wt% nanoparticle, it does not show any reading. This is because the sample was cracked due to humidity of atmosphere and maybe destroyed the superconductivity.

In addition, the transition temperature is consistent maybe due to the equipment problem or some error during carried out this testing. The temperature cannot reach the liquid nitrogen temperature which is 77.2 K and it is limited to 280K.

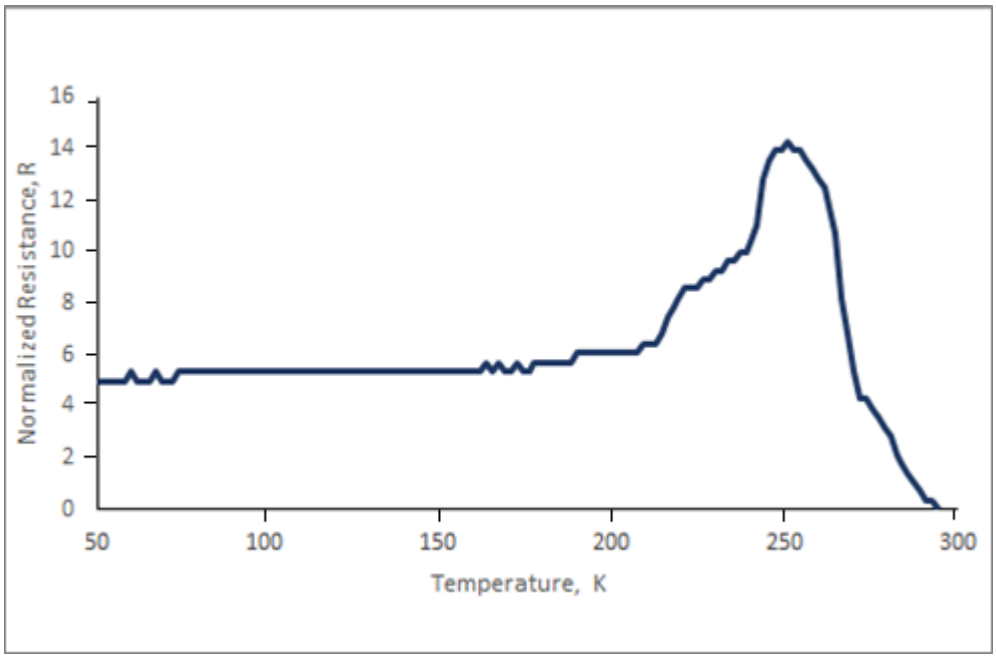

Figure 4.5: Normalized resistance versus temperature for $YBa_2Cu_3O_7$ superconductor with addition of 0.02 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$.

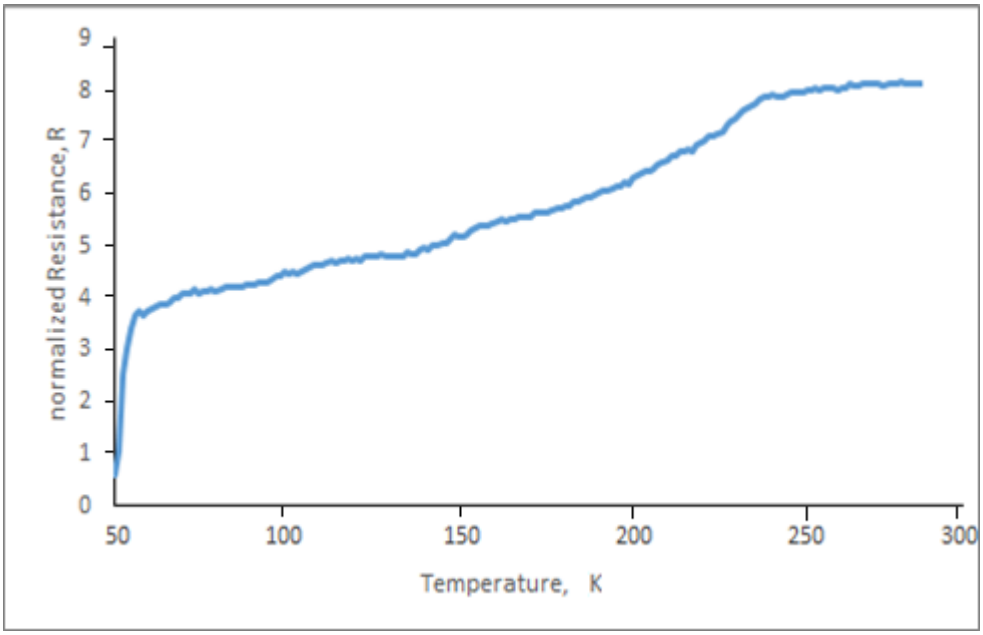

Figure 4.6: Normalized resistance versus temperature for $YBa_2Cu_3O_7$ superconductor with addition of 0.03 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$

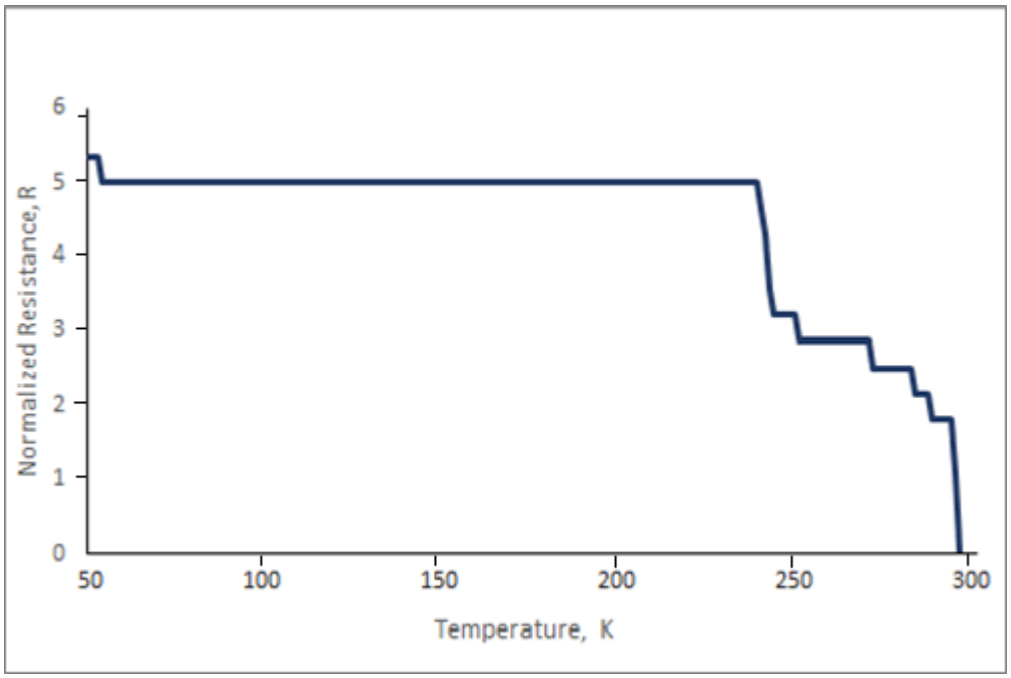

Figure 4.7: Normalized resistance versus temperature for $YBa_2Cu_3O_7$ superconductor with addition of 0.04 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$

There are some precaution steps need to more attention. during sample preparation, such as the grinding and pelletizing process, make sure that all equipment used in clean condition. After sintered the sample, color of YBCO superconductor sample is black. If there are green color on the sample surface, grind again because the green color shows uneven grinding of CuO element with others element. In terms of sample storage, superconductor sample should be stored in an airtight container or with some drying agent like silica gel and store the container in a dry place.

CHAPTER 5

CONCLUSION AND RECOMMENDATION

5.1 CONCLUSION

In the nut shell, this project focus on effect in microstructure and electrical properties magnetic of $YBa_2Cu_3O_7$ superconductor with addition of 0.01wt%, 0.02 wt%, 0.03 wt% and 0.04 wt% $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles were studied. Thus, there are many conclusions that can be conclude.

$YBa_2Cu_3O_7$ superconductor was synthesized using solid state reaction method. The effect of addition of $Co_{0.5}Ni_{0.5}Fe_2O_4$ to YBCO was studied by various characterization techniques. Firstly, Meissner effect analysis show the sample with 0.04wt% of $Co_{0.5}Ni_{0.5}Fe_2O_4$ nanoparticles levitate the longest time on the permanent magnet compared to 0.03wt%, 0.02wt% and 0.01wt%.

Next, the phase of YBCO was confirmed by XRD analysis where the highest peak was mainly dominant correspond to YBCO phase with orthorhombic structure. The addition of $Co_{0.5}Ni_{0.5}Fe_2O$ nanoparticle to pure YBCO does not show any changes in crystal structure, thus, there are no significant effect on the structure and symmetry of the composites. The lattice parameter for all added sample showed differ value of unit cell which were c = 7.000 Å while the pure YBCO is c = 11.000 Å. This is maybe due to impurities element in sample during sample preparation.

For SEM characterization, the microstructure of YBCO with addition nanoparticle show some changes in term of particle size and porosity in grain. The porosity grain is obviously seen in sample with 0.04wt% nanoparticles. Lastly, for four-point probe characterization, the T_c value cannot be identified due to the T_c of this experiment is limited to 280K.

5.2 RECOMMENDATION

There are a lot of ways to improve this project. Firstly, in order to get the excellent result, a long duration time is needed so that we can repeat the experiment many times. Next, for four point probe technique, it is involve with dealing electrical instrument. So, the connection wire in four point probe device need to check frequently by using multimeter to avoid short circuit. In addition, the instrument in FIST laboratory must be do regularly maintenance to ensure no technical error occurs during experiment. In addition, the addition of nanoparticles is recommended by larger different of weight percentage. Last but not less, the effectiveness of four point probe that developed in laboratory scale still under observation. To get a good result, the design of four point probe device should be in proper technique.

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APPENDICES

APPENDIX 1

Calculation of YBCO weight;

Chemical equation:-

½ Y_2O_3 + 2 $BaCO_3$ + 3CuO + ½O_2→ $YBa_2Cu_3O_7$

Molar mass of,

Y= 88.90585 g/mol

Ba= 137.327 g/mol

C= 12.0107 g/mol

O= 15.9994 g/mol

Cu= 63.546 g/mol

- ½ Y_2O_3

  [(2×88.90585 g/mol) + (3×15.9994 g/mol)]

  = 112.9050 g/mol

- 2 $BaCO_3$

  2[137.327 g/mol + 12.0107 g/mol + (3×15.9994 g/mol)]

  = 394.6718 g/mol

- 3CuO

  3[63.546g/mol + 15.9994 g/mol]

  = 238.6362 g/mol

b) Total molar mass

112.9050 g/mol + 394.6718 g/mol + 238.6362 g/mol

=746.2130 g/mol

c) Mass of YBCO

i. ½ Y_2O_3

(112.9050 g/mol÷746.2130 g/mol) × 30 g

= 4.54 g

ii. 2 $BaCO_3$

(394.6718 g/mol ÷ 746.2130 g/mol) × 30 g

= 15.87 g

iii. 3CuO

(238.6362 g/mol ÷ 746.2130 g/mol) × 30 g

= 9.59 g

Total product need for this experiment are: -

4.54 g + 15.87 g + 9.59 g = **30g**

APPENDIX 2

Crystal structure of all composition YBCO superconductor is calculated by using Orthorhombic structure formula: -

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}$$

For pure YBCO, lattice phase (010), the value of d is 3.889

$$\frac{1}{3.851^2} = 0 + \frac{1^2}{\mathrm{b}^2} + 0$$

b^2 = 15.1243

b = 3.889Å

For lattice phase (011), the value of d is 3.719

$$\frac{1}{3.719^2} = 0 + \frac{1^2}{3.889^2} + \frac{1^2}{0^2}$$

c^2 = 137.3000

c = 11.7175Å

For lattice phase (111), the value of d is 2.627

$$\frac{1}{2.627^2} = \frac{1^2}{\mathrm{a}^2} + \frac{1^2}{3.889^2} + \frac{4^2}{11.7175^2}$$

a^2 = 11.6186

a = 3.4086 Å

a) 0.01 wt% of $CoNiFe_2O$

For lattice phase (100), the value of d is 3.851

$$\frac{1}{3.851^2} = \frac{1^2}{\mathrm{a}^2} + 0 + 0$$

a^2 = 14.8302

a = 3.851Å

For lattice phase (110), the value of d is 2.7309

$$\frac{1}{2.7309^2} = \frac{1^2}{3.851^2} + \frac{1^2}{b^2} + 0$$

b^2 = 15.0020

b = 3.873Å

For lattice phase (114), the value of d is 1.5640

$$\frac{1}{1.5640^2} = \frac{1^2}{3.851^2} + \frac{1^2}{3.873^2} + \frac{4^2}{c^2}$$

c^2 = 58.241

c = 7.631 Å

b) 0.02 wt% of $CoNiFe_2O$

For lattice phase (003), the value of d is 2.506

$$\frac{1}{2.506^2} = 0 + 0 + \frac{3^2}{c^2}$$

c^2 = 56.5203

c = 7.518Å

For lattice phase (200), the value of d is 1.9355

$$\frac{1}{1.9355^2} = \frac{2^2}{a^2} + 0 + 0$$

a^2 = 14.9846

a = 3.871Å

For lattice phase (112), the value of d is 2.2181

$$\frac{1}{2.2181^2} = \frac{1^2}{3.871^2} + \frac{1^2}{b^2} + \frac{2^2}{7.518^2}$$

b^2 = 15.2084

b = 3.8997 Å

c) 0.03 wt% of $CoNiFe_2O$

For lattice phase (200), the value of d is 1.9243

$$\frac{1}{1.9243^2} = \frac{2^2}{a^2} + 0 + 0$$

a^2 = 14.8117

a = 3.8486 Å

For lattice phase (102), the value of d is 2.6826

$$\frac{1}{2.6826^2} = \frac{1^2}{a^2} + 0 + \frac{2^2}{c^2}$$

c^2 = 55.9868

c = 7.4824Å

For lattice phase (112), the value of d is 2.2039

$$\frac{1}{2.2039^2} = \frac{1^2}{3.8486^2} + \frac{1^2}{b^2} + \frac{2^2}{7.4824^2}$$

b^2 = 14.9430

b = 3.8656 Å

a) 0.03 wt% of $CoNiFe_2O$

For lattice phase (100), the value of d is 3.798

$$\frac{1}{3.798^2} = \frac{1^2}{a^2} + 0 + 0$$

a^2 = 14.4248

a = 3.798 Å

For lattice phase (102), the value of d is 2.6810

$$\frac{1}{2.6810^2} = \frac{1^2}{3.798^2} + 0 + \frac{2^2}{c^2}$$

c^2 = 57.3063

c = 7.5701 Å

For lattice phase (112), the value of d is 2.2029

$$\frac{1}{2.2039^2} = \frac{1^2}{3.798^{\,2}} + \frac{1^2}{b^2} + \frac{2^2}{7.5701^{\,2}}$$

b^2 = 14.9381

b = 3.8649 Å

APPENDIX 3

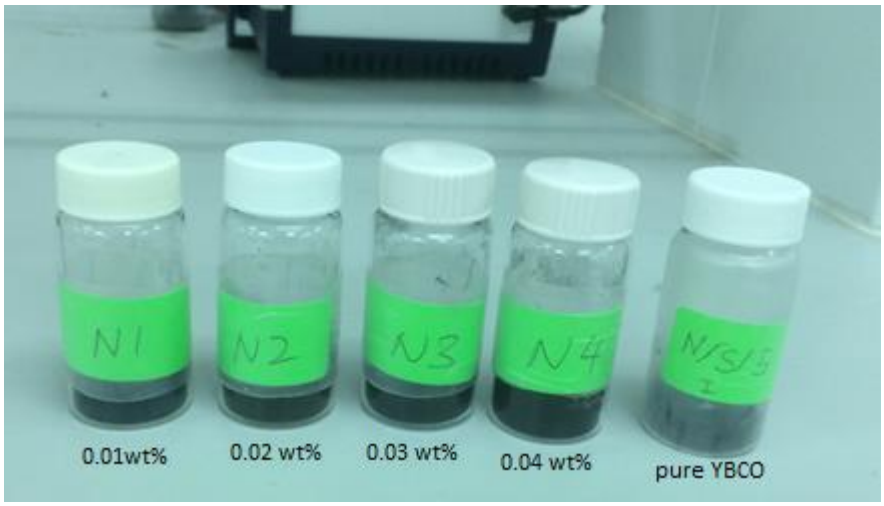

Figure 1: The samples with different composition nanoparticles $Co_{0.5}Ni_{0.5}Fe_2O_4$ are put into different vials

Figure 2: Magnetic nanoparticle

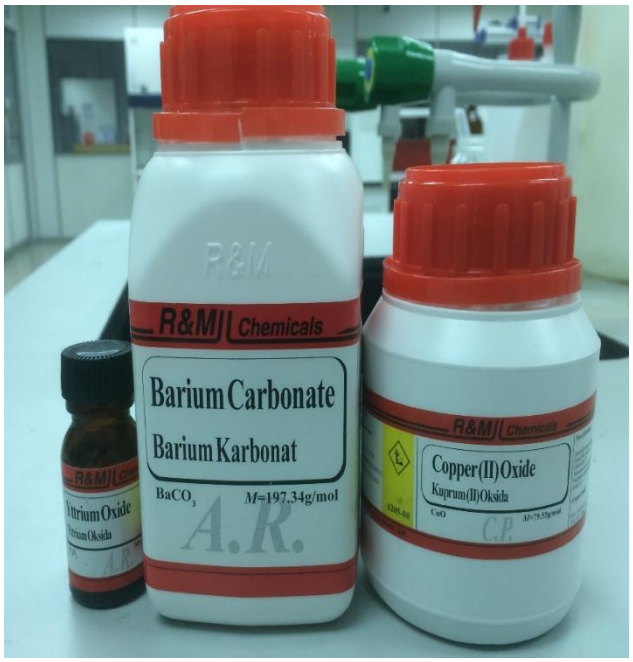

Figure 3: The chemical used to produce YBCO superconductor

Figure 4: Hydraulic Press

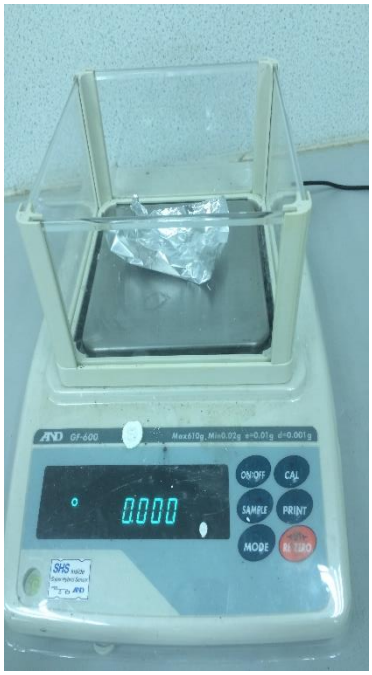

Figure 5: Electronic Balance

Figure 6: the box furnance used for sintering