Preparation and Characterization of Fe/SiO_2 Composite

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Abstract. Fe/SiO_2 soft magnetic composites have gained considerable attention in the field of materials science due to their unique magnetic properties, high electrical resistivity, and thermal stability. These materials are particularly useful in various applications, including transformers, inductors, and electromagnetic devices. Silica is an oxidizing compound that is widely used in industry. Silica can be obtained from various sources including agricultural waste, in this case bamboo leaves. This research aims to prepare composite consisting of magnetic materials Fe and silica by ball milling technique. Silica is extracted from bamboo leaf gathering from local farm, then burning in electrical furnace 600°C for 4h. Bamboo leaf ash then was leached using hydrochloric acid (HCl) extraction of 5M, while Fe powder is purchased from chemical stores. The silica and Fe powder grind in ball milling reactor of 240 rpm for 24h. The Fe mass fraction (x) varied from 0.1 to 0.9. The samples were characterized using X-ray fluorescence (XRF), X-ray diffraction (XRD), Fourier transform infrared (FTIR) spectroscopy, Scanning electron microscopy-Energy Dispersive X-ray Spectroscopy (SEM-EDS) and vibrating sample magnetometer (VSM) techniques, I-V characteristic for electrical resistivity. Experimental result confirmed that $Fe\text{-}SiO_2$ composite prepared by ball milling show particle crystalline Fe dispersed in matrix amorphous SiO_2, however the shape and size of Fe particle are irregular. XRD result shows the appearance of silica at $2\theta = 21^{\circ}$ the type quartz and α-Fe around $2\theta = 44.68^{\circ}$ as well as FTIR spectra demonstrate the presence Si-O-Si bond at wavelength of 1099 cm^{-1} and Fe-O bond at wavelength of 470 cm^{-1}. Electrical resistivity reveals the dcreaase with increasing Fe fraction and The presence percolation around x = 0.7. VSM measurement present superparamagnetic behavior with magnetic saturation (M_s) is proportional to the increasing mass fraction of Fe, from 28 emu/g for x = 0.1 to 170 emu/g for x = 0.9.

Keywords: Fe/SiO_2 composite, ball milling, structural properties, magnetic properties, functional groups.

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1 Introduction

Nanoparticle materials offer many advantages due to their unique size and physicochemical properties. Due to the wide application of magnetic nanoparticles in biotechnology, biomedicine, materials science, engineering, and the environment, attention is focused on the synthesis of various types of synthesis methods [1]. Magnetic nanoparticles exhibit amazing phenomena such as high irreversibility fields, high saturation fields, superparamagnetism, anisotropy [2]. Magnetic nanoparticles with negligible coercivity and remanent fields are needed for biomedical applications, so superparamagnetic nanoparticles are highly desirable for these applications [3]. The disadvantages of metal nanoparticles are low particle stability and easy aggregation [4]. One solution is to immerse in silica. In addition, silica nanoparticles can easily bind to various ligands or antibodies by modifying the surface of SiO_2 [5].

In recent decades, research on magnetic composite materials has received great attention due to their potential applications in various fields, including information technology, biomedical, energy, and the environment. Magnetic nanoparticles, in particular, offer unique properties that allow for a wide range of applications, such as medical imaging, disease therapy, water purification, and biomolecule separation [5].

Magnetic nanoparticles dispersed in porous matrices have attracted the attention of researchers in the last two decades because of their wide range of applications, including as data storage materials, gas sensors, magnetooptics. Various commonly used magnetic materials are α-Fe and iron oxide [2, 6], Ni [7], FeSiCr [8, 9]. Some of the commonly used insulator materials are SiO_2 [2, 6, 7, 8, 9], Al_2O_3 [10] and TiO_2 [11] because they have high resistivity and good thermal stability. Anas et.al., [12] studied the tunneling giant magnetoresistance of coevaporated Fe-SiO thin films.

The methods used for the synthesis of magnetic nanoparticles are various such as mechanical milling [2, 13], ion implantation [14], co-precipitation, microemulsion technique, sonochemistry process, sol–gel method, flow injection, electrochemical production, supercritical fluid techniques, thermal decomposition, hydrothermal routes, microwave techniques, spray pyrolysis, laser pyrolysis, flame spray pyrolysis, gas-phase synthesis, arc discharge, and oxidation methods [15]. The method has drawbacks, such as complexity in controlling size distribution, high cost and contamination with the chemicals materials used.

Therefore, ball milling method is the cheaper tools and fewer parameters to control [2]. In this study, the effect of Fe mass fraction on the size-dependent crystal structure and magnetic properties of the ball milled Fe-SiO_2 composite was investigated. The particle size was tuned by adjusting the Fe mass fraction. The results of ball milling are then characterized by various methods such as X-ray Fluorescence (XRF), X-ray Diffraction (XRD), Fourier Transform Infrared (FTIR), Vibrating Sample Magnetometer (VSM) and Scanning Electron Microscope (SEM). In addition, the interaction between Fe and O were analyzed.

2 Methodology

2.1 Raw Materials

The raw bamboo leaf (BL) was collected from local farm in Konda, South Konawe Regency – Indonesia. Initially, the collected BL is washing with distilled water to remove adhering contaminations present in them and then it dried by exposing it to sunlight for 24h. Fe powder of 10 m and hydrochloric acid (37%) was purchased from CV. Intraco Makassar. Another material, such as distilled water, alcohol, filter paper, was purchased from the local chemical store in Kendari.

2.2 Bamboo Leaf Ash

The sample preparation process is carried out by taking foliage samples, washing, cutting bamboo leaves with small sizes, drying under the sunlight, for ashing bamboo leaves is used electrical furnace for 4 hours at temperature of 600 °C, grinding using mortar and sieving of 100 mesh.

2.3 Silica Extraction from Bamboo Leaf Ash

The extraction silica from bamboo leaf ash (BLA) is done by mixing 20 gr of bamboo leaf ash with 240 mL HCl (5M), which was stirred using a hot plate magnetic stirrer at a temperature of 110°C for 5 hours at a speed of 240 rpm. The leaching results obtained are filtered using whattman 41 filter paper to obtain a precipitate, and then the precipitate is rinsed by distilled water until pH neutral, finally dried in an oven with a temperature of 110°C for 5 hours.

2.4 Fe/SiO_2 Composite Preparation

Nanocrystalline Fe particles in a silica matrix, with Fe mass fraction varying between x = 0.1 and 0.9, were prepared by mechanical ball milling. The starting Fe and silica powders were milled using Cr-steel balls, inside jars sealed under an ambient atmosphere, for 6 h. Then, the milled Fe and silica were mixtured and grinded in ball milling for 24h. The grinding speed was held constant of 200 rpm.

2.5 Sample Characterization

Fe/SiO_2 composites that have taken the form of physical powders are characterized using XRF, XRD, FTIR and VSM to determine the chemical composition, crystal structure, functional groups and magnetic properties of each sample respectively.

The resulting samples were characterized by X-Ray Fluorescence (XRF), Fourier Transform Infrared (FTIR), X-ray diffraction (XRD) and scanning electron microscopy (SEM) equipped by energy-dispersive x-ray spectrometer (EDS). Magnetic measurements were carried by VSM, with a maximum applied field of 1 T to determine the chemical composition, crystal structure, functional groups and magnetic properties of each sample respectively. In addition, the electrical resistivity was performed.

3 Results and Discussion

3.1 Chemical Composition in Fe/SiO_2 Composite

The chemical composition in Fe/SiO_2 composite is determined by XRF. Table 1 shows the percentage of chemical content based on the Fe mass fraction. XRF detected that the iron oxide compound that appeared was Fe_2O_3. The interaction between iron and oxygen is biased during the milling process but is not expected to be large. To see more about this interaction, it can be seen from the results of characterization with XRD and FTIR. Table 1 is also reveal that the total Fe and silica in the composite is from 99.33% to 99.10% with a nominal Fe mass, the rest consists of TiO_2, MnO, Al_2O_3, MgO, CaO, N_2O and K_2O compounds. These compounds are thought to be derived from silica extracted from the leaves of the Petung bamboo (*Dendrocalamus asper*).

Table 1. Percentage of chemical compound content for various Fe mass fraction

No	Compounds	Compounds Percent in each of Fe/SiO_2 sample (%)				
		0.1	0.3	0.5	0.7	0.9
1	Fe_2O_3	12,04	21,93	31,47	44,47	73,65
2	SiO_2	87,29	77,28	67,72	54,67	25,45
3	TiO_2	0,3994	0,5793	0,6375	0,7310	0,8011
4	MnO	0,1337	0,0992	0,0675	0,0520	0,0361
5	Al_2O_3	0,0114	0,0100	0,0125	0,0157	0,0282
6	MgO	0,0059	0,0064	0,0068	0,0080	0,0119
7	CaO	0,0672	0,0569	0,0497	0,0296	0,0080
8	Na_2O	0,0016	0,0019	0,0026	0,0035	0,0063
9	K_2O	0,0440	0,0367	0,0283	0,0189	0,0054

Figure 1 shows the content of Fe and SiO_2 in Fe/SiO_2 composites for variations in the Fe mass fraction. An increase of Fe in the composite indicates an increase in the percentage of Fe, however this increase is not linear, it tends to be exponential. Meanwhile, the relationship between Fe increase and SiO_2 reduction is linear with the coeffience of determination (R^2) of 1.

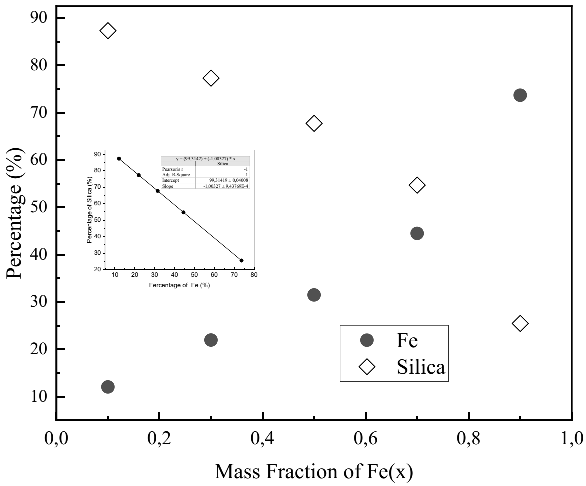

Fig. 1. Percentage of silica and Fe against variation of Fe mass fraction

3.2 Structural Properties of Fe/SiO_2 Composite

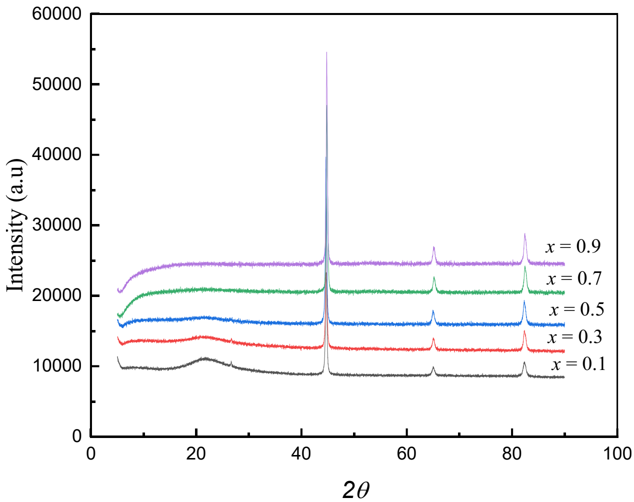

Fig. 2. XRD pattern of sample with variation of Fe:SiO_2 ratio (Fe above is higher)

The structural properties of Fe/SiO_2 composite is analyzed by XRD diffraction. X-ray diffraction patterns of various $Fe:SiO_2$ ratios are shown in Figure 2. The XRD pattern is dominated by 3 peaks situated at $2\theta = 44{,}69°$; $65{,}65°$ and $82{,}32°$ with main peak at $2\theta = 44{,}60°$ which is identified as α-Fe. These peaks are the characteristic peaks corresponding to the crystallographic planes (101), (200) and (211) of a crystalline structure of Fe, respectively. The data agree well with the results of Lopez et.al.[2] and Luo, et.al. [16]. A broad peak was observed at $2\theta = {\sim}17$ - $29°$ corresponding to the formation of amorphous silica [17, 18] appearing just for the sample $x = 0{,}10 - x = 0{,}30$.

3.3 Functional Groups of Fe/SiO_2 Composite

Figure 3 shows the FTIR spectra of prepared Fe/SiO_2 with different mass fraction of Fe. FTIR can be used to study the surface chemical structure of Fe/SiO_2 and the bonding interactions between Fe and SiO_2. Specific bands corresponding to Si-O-Si stretching and Fe-O stretching can provide insights into the chemical environment of the composite. The spectrum exhibits a strong broad-absorption band centered around 1090 and 800 cm-1 which are characteristics of silica. In all spectra, the absorption bands around 3400 cm−1 correspond to O – H stretching mode. The spectra of Fe/SiO_2 shows the absorption bands near 470 cm−1 which are assigned to Fe–O stretching mode. The interaction Fe – Si – O which usually appears at wave numbers of 1050 cm-1 [19, 20] was not detected in this study.

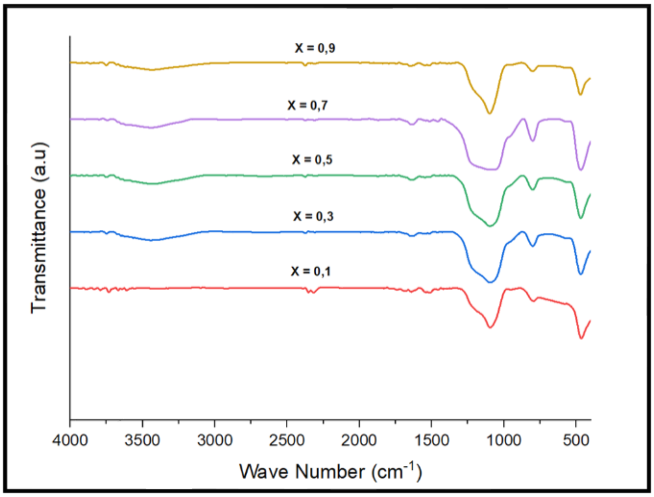

Fig. 3. FTIR spectra of Fe/SiO_2 for the different mass fraction of Fe

3.4 Electrical Resistivity of Fe/SiO_2 Composite

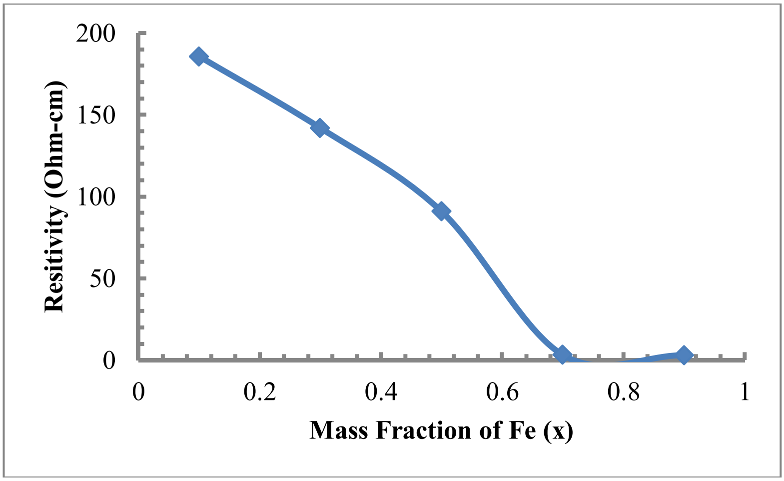

Fig. 4. Electrical resistivity of Fe/SiO_2 composite for the different mass fraction of Fe

The electrical resistivity of $Fe\text{-}SiO_2$ composite decrease with the increasing of the mass fraction of Fe (x) as shown in Figure 4. The electrical resistivity is the range 186 Ω-cm to 3 Ω-cm. The data is very close to the result of Honda, et.al. [21]. We can see the discontinuous decrease at around $x = 0.7$, at which the mechanism of the electrical resistivity is changed. This region is known as percolation zone.

3.5 Magnetic Properties of Fe/SiO_2 Composite

The M-H curve of sample with the selected mass fraction of Fe is shown by Figure 5. The sample shows the absence of the remanence magnetization (Mr) and coersive field (Hc) indicating that the sample has superparamagnetic (SP) behavior. The higher the Fe content, the greater the saturation magnetization (Ms). This is in accordance with Ma, et al. [22] where magnetization increases with an increase in the volume fraction of Fe particles.

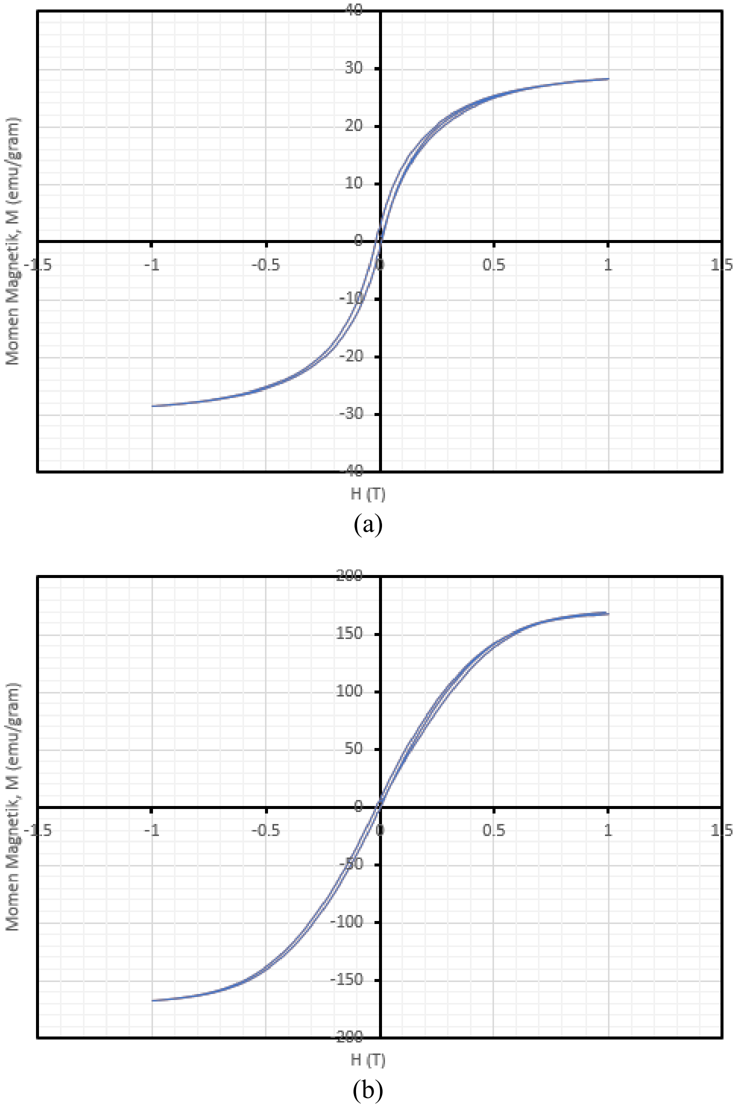

Fig. 5. M-H curve of sample with the mass fraction of Fe; (a) 0,1 (b) 0,9

4 Conclusion

Fe-SiO_2 composite has been prepared by ball milling with the variation mass fraction of Fe. The sample shows particle crystalline Fe dispersed in the amorphous SiO_2 matrix however the shape and size of Fe particle are irregular. There are a weak interaction between Fe and O in the sample. The Fe-SiO_2 composite present superparamagnetic behavior.

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