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Analysis and Characterization of Fe_3O_4/Silica Composite from Rice Husk Ash

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ABSTRACT
In this investigation, Fe_3O_4/Silica composites were synthesized from rice husk ash utilizing the coprecipitation technique, aiming to elucidate the influence of varied heating temperatures on the surface morphology and elemental composition of the composites. Comprehensive characterizations were conducted employing Fourier Transform Infrared (FTIR) Spectroscopy, X-ray Diffraction (XRD), and Scanning Electron Microscopy (SEM). These analyses unveiled a heterogeneous distribution of Fe_3O_4 nanoparticles and affirmed the amorphous characteristic of the silica constituent, with the XRD results prominently displaying a broad peak at approximately $2\theta = 38°$, signifying the amorphous nature. Despite the alterations in heating temperatures, SEM observations indicated a negligible effect on the nanoparticles' surface morphology, whereas notable variations were discerned in their elemental composition. The outcomes of this study provide insightful contributions to the understanding of the structural properties of Fe_3O_4/Silica composites, suggesting avenues for refining synthesis methodologies for enhanced environmental and technological utilization.

Keywords: Adsorber, Fe_3O_4/Silica, Rice Husk

ABSTRAK
Pada penelitian ini, komposit Fe_3O_4/Silika disintesis dari abu sekam padi dengan menggunakan teknik kopresipitasi, yang bertujuan untuk mengetahui pengaruh suhu pemanasan yang bervariasi terhadap morfologi permukaan dan komposisi unsur komposit. Karakterisasi komprehensif dilakukan dengan menggunakan Spektroskopi Fourier Transform Infrared (FTIR), Difraksi Sinar-X (XRD), dan Scanning Electron Microscopy (SEM). Analisis ini mengungkap distribusi nanopartikel Fe_3O_4 yang heterogen dan menegaskan karakteristik amorf dari konstituen silika, dengan hasil XRD yang secara mencolok menampilkan puncak yang luas pada sekitar $2\theta = 38°$, yang menandakan sifat amorf. Terlepas dari perubahan suhu pemanasan, pengamatan SEM menunjukkan efek yang dapat diabaikan pada morfologi permukaan nanopartikel, sedangkan variasi penting terlihat pada komposisi unsurnya. Hasil dari penelitian ini memberikan kontribusi yang mendalam terhadap pemahaman tentang sifat struktural komposit Fe_3O_4/Silika, menunjukkan jalan untuk menyempurnakan metodologi sintesis untuk meningkatkan pemanfaatan lingkungan dan teknologi.

Kata kunci: Adsorber, Fe_3O_4/Silika, Sekam Padi

1. Introduction

The current state of affairs in every aspect of life, such as the rapid increase in population and the growth of industries, has impacted the depletion of clean water supplies [1]. The rising level of pollution due to wastewater containing organic and inorganic compounds from human and industrial activities has garnered significant concern over the last decade [2]. Numerous strategies, including the use of membranes, ion exchange, extraction, and precipitation, have been employed to overcome this problem [3]–[5]. However, adsorption remains a particularly promising method due to its ease of operation and high effectiveness [6], [7]. Industrial wastewater discharged into water bodies often contains organic pathogens and inorganic contaminants, which are extremely harmful to the environment [8].

Several metal oxide adsorbents can be applied to treat wastewater, including magnetite (Fe_3O_4), titanium dioxide (TiO_2), zinc oxide (ZnO), alumina oxide (Al_2O_3), magnesium oxide (MgO), and zirconium oxide (ZrO_2). Among these, magnetite nanoparticles (Fe_3O_4) are particularly intriguing due to their magnetic properties, high surface area, optical and catalytic properties, and strong adsorption bonds to organic pollutants [9]. One of the most promising coating materials for Fe_3O_4 is silica (SiO_2). Silica protects Fe_3O_4 nanoparticles from oxidation and agglomeration across different pH ranges and enhances their chemical stability and adsorption of hard metal ions. These functions make it suitable for magnetic separators in microbiology and detoxifying biological fluids. Additionally, silica's good hydrophilicity, stability, and biocompatibility enhance the performance of Fe_3O_4 nanoparticles in attaching organic molecules to the surface of nanoparticles through covalent bonds. Alizadeh et al. (2020) synthesized Fe_3O_4/SiO_2 for use as an adsorbent in the removal of methylene blue dye [10]. SiO_2 based on natural materials is particularly attractive for study due to the abundance of these materials. For instance, rice husk, a plentiful waste product in Indonesia, contains about 87% - 97% silica by dry weight after complete combustion [11].

Indonesia is often referred to as an agricultural country because a significant portion of its population is employed in the agriculture sector. In this sector, rice stands out as one of the primary products. According to data from the Central Statistics Agency (BPS), Indonesia is projected to produce 55.67 million tons of dry-milled grain (GKG) rice in 2022. Nevertheless, the by-products of rice processing, especially rice husks, have not been utilized optimally. To maximize the use of rice husks, purification technology is required to obtain high-purity silica material. One technique employed to obtain high-purity SiO_2 is coprecipitation. This chemical fabrication technique causes a solute to precipitate, forming the desired product. Compared to other conventional methods, this technique offers several advantages, including the production of high-purity products, a simple deposition process, and a relatively quick processing time. This makes it possible to produce powders with controlled crystal sizes using this method [12].

This study aimed to synthesize a Fe_3O_4/SiO_2 composite, where SiO_2 was produced from rice husk ash waste. The synthesis process employed the coprecipitation technique with varied heating temperatures, and the changes in surface morphology were then observed using an SEM-EDX instrument.

2. Methods

2.1. Synthesis of SiO_2 from Rice Husk

To synthesize SiO_2, 500 grams of rice husks were first washed and soaked in distilled water for 24 hours. They were then dried in an oven. Subsequently, the dried rice husks were heated to 700°C in a furnace to produce ash. The resulting rice husk ash was then crushed. For activation, 50 mL of distilled water and ZnCl were added to the ash. The mixture was placed in a closed container and dried in the oven.

2.2. Synthesis of Fe_3O_4/SiO_2

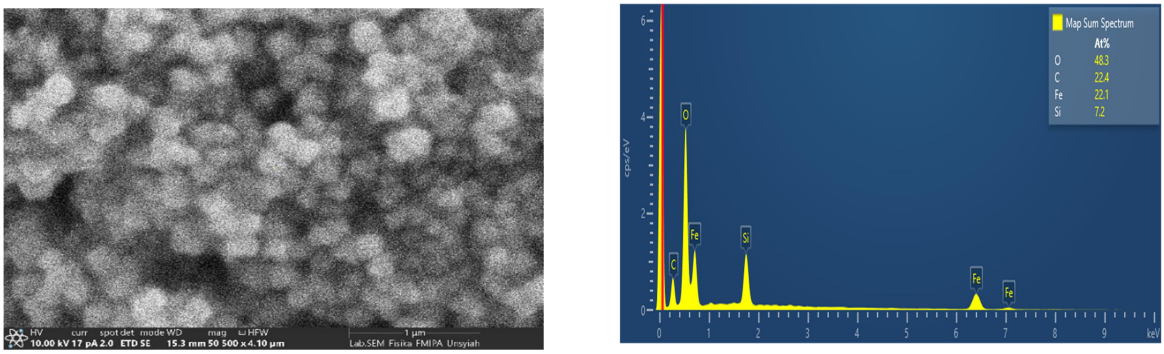
The synthesis of Fe_3O_4/SiO_2 began with mixing SiO_2 and Fe_3O_4 in ethanol. This was done by stirring the mixture at 500 rpm using a magnetic stirrer. Each mixture was then treated with concentrated NH_4OH and stirred in a fume cupboard using a magnetic stirrer. Subsequently, the solution was removed from the fume cupboard and washed with distilled water. The precipitate was washed with ethanol at 500 rpm until the pH reached 7. Afterward, the precipitate was dried in an oven. The dried samples were crushed for 1 hour to form a powder. Finally, the powder sample underwent characterization using XRD, SEM-EDS, and FTIR.

3. Result and Discussion

3.1. SEM- EDX Analysis of Fe_3O_4/SiO_2 Composites

The SEM images reveal that the distribution of Fe_3O_4 nanoparticles is uneven, with many clusters exhibiting the morphology of the Fe_3O_4 particles. Figure 3 displays the SEM-EDX results of the Fe_3O_4/SiO_2 composites derived from rice husk ash at various heating temperatures. At 700°C, the sample's homogeneous surface shows elemental compositions of O = 48.3%, C = 22.4%, Fe = 22.1%, and Si = 7.2%. This suggests

the possibility of interparticle agglomeration among Fe_3O_4 nanoparticles to incomplete sample preparation or non-uniform grinding in the furnace [13], [14]. The particles are round, with an 82.84 – 110.9 nm size range. However, the surface morphology images reveal gaps and distances between particles. Overall, while the heating temperatures did not significantly impact the surface morphology of the composites, they did affect the elemental compositions and their respective percentages.



(a) (b)

Figure 1. Morphology images and chemical compositions of Fe_3O_4/SiO_2 from rice husk ash at (a) T = 700°C with 50000x magnification.

3.2. XRD Analysis of Fe_3O_4/SiO_2 Composites

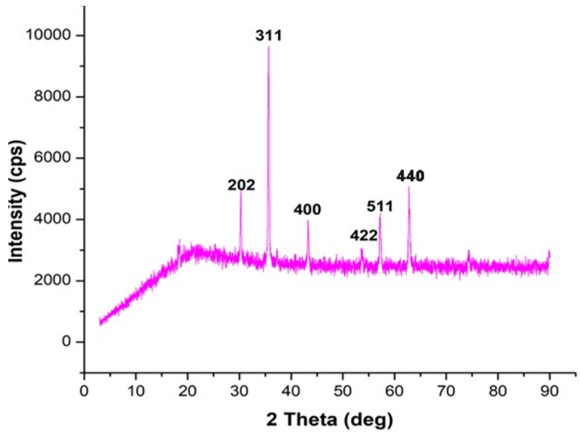


Figure 2. Image of Fe_3O_4/SiO_2 structure from rice husk ash at T = 700°C.

Table 1. Peak graphic of XRD in sample.

No.	2-theta (deg)	hkl	Height (cps)	FWHM (deg)	Int. I (cps deg)	Int. W (deg)	Asym. factor
1	30.275(17)	202	1632(117)	0.190(16)	415(18)	0.25(3)	2.2(10)
2	35.608(7)	311	5399(212)	0.186(7)	1433(20)	0.265(14)	1.1(2)
3	43.279(14)	400	1225(101)	0.15(2)	296(14)	0.24(3)	1.8(9)
4	53.589(9)	422	602(71)	0.14(3)	148(15)	0.25(5)	0.5(4)
5	57.149(17)	511	1393(108)	0.22(2)	404(20)	0.29(4)	0.51(19)
6	62.774(11)	440	2052(131)	0.211(15)	591(20)	0.29(3)	1.0(2)

The XRD results of the Fe_3O_4/SiO_2 composite from rice husk material, heated at 700°C, indicate that the silica ash obtained from well-burned rice husks is either in the initial treatment stage or amorphous. As illustrated in Figure 2, the XRD pattern shows peaks at 2θ = 30.275°, 35.608°, 43.279°, 53.589°, 57.149°, and 62.774°, which correspond to the (202), (311), (400), (422), (511), and (440) reflections, respectively. This is evidenced by the sloping peak at around 2θ = 38°, characteristic of amorphous silica. The XRD

testing determined the crystal structure, the highest peak, and the phase formation of the Fe_3O_4/SiO_2 sample. The XRD patterns of Fe_3O_4, Fe_3O_4-SiO_2, and Fe_3O_4-SiO_2-NH_2 indicate that the crystalline structure of the modified nanoparticles remained unchanged during the process. This suggests that the physical properties of the magnetite particles were not affected by the conjugation and surface modification of the Fe_3O_4 nanoparticles. Shao et al. (2012) observed that the SiO_2 peak is located at a 2θ angle of 24°, indicating the presence of amorphous silica. The diffraction peak of Fe_3O_4 particles increases with the addition of SiO_2 [15]. According to JCPDS No. 190629, the Bragg peaks are indexed as magnetite. The Miller Indices (hkl) are (220), (311), (400), (422), (511), and (440), corresponding to the SiO_2-Fe_3O_4 peaks. The average crystal size, calculated using the Scherrer equation, is approximately 60 nm [16].

3.3. FTIR Analysis of Fe_3O_4/SiO_2 Composites

Figure 3 presents the FTIR results of the Fe_3O_4/SiO_2 composite synthesized from rice husk material, heated at 700°C. The FTIR spectra (Figure 3) also reveal the amorphous nature of the resulting silica ash. The characteristic fingerprint of amorphous silica is evident in three wave number intervals: 500-750 cm^{-1}, 1200-1250 cm^{-1}, and 3300-3500 cm^{-1}. These wave numbers correspond to the stretching vibrations of asymmetric and symmetric Si-O-Si bonds.

The FTIR test results were conducted to identify the functional groups present in the tested samples. FTIR spectroscopy is a powerful technique that identifies specific functional groups in a material based on their absorption patterns in the infrared region. The wave absorption patterns observed in the FTIR results provide information about the types of chemical bonds and functional groups present in the Fe_3O_4/SiO_2 composite.

The specific peaks in the absorption spectrum correspond to vibrations associated with different chemical bonds. The Fourier Transform Infrared (FTIR) spectrometry spectra of the Fe_3O_4/SiO_2 SNPs revealed certain absorption peaks that suggested the presence of Si-O-Si, O-Si-O, Fe-O, and Fe-O-Si bonds [17]. The FTIR analysis of the Fe_3O_4/SiO_2 composite at a heating temperature of 700°C confirms the amorphous nature of the resulting silica and highlights specific wave numbers associated with Si-O-Si vibrations. This information contributes to a comprehensive understanding of the structural and chemical characteristics of the synthesized composite material. In other words, these vibrations indicate the presence of silicon-oxygen networks in the amorphous silica structure. The FTIR test results for the Fe_3O_4/SiO_2 composite provide insights into the functional groups formed, as evidenced by the wave absorption patterns observed in the tested samples [18], [19].

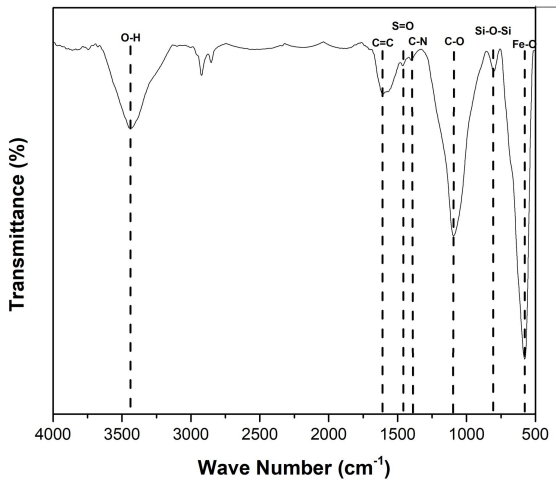


Figure 3. Analysis of Fe_3O_4/SiO_2 clusters from rice husk ash at T = 700°C.

4. Conclusion

In this comprehensive study, Fourier Transform Infrared (FTIR) Spectroscopy, Scanning Electron Microscopy (SEM), and X-ray Diffraction (XRD) analyses were meticulously employed to explore the impact of varying heating temperatures on the morphological and elemental properties of Fe_3O_4/Silica composites synthesized from rice husk ash. SEM assessments revealed an uneven distribution of Fe_3O_4 nanoparticles and highlighted the minimal influence of thermal treatment on surface morphology. Simultaneously, alterations in elemental composition demonstrated a considerable dependence on the thermal conditions applied, indicating the sensitivity of the composite's chemical properties to heating

temperatures. The FTIR spectroscopy data provided invaluable insights into the chemical bonding and functional groups present in the composites, revealing characteristic absorption bands associated with Si-O-Si and Fe-O vibrations, which further substantiate the formation and integration of Fe_3O_4 and silica within the composite structure. The XRD analysis was particularly revealing, identifying the amorphous nature of silica through a broad diffraction peak at approximately $2\theta = 38°$ and elucidating the crystalline structure of Fe_3O_4, thus offering profound insights into the composite's phase composition and structural integrity. These combined observations from SEM, FTIR, and XRD analyses significantly advance our understanding of the structural and chemical dynamics of Fe_3O_4/Silica composites. This research underscores the critical role of synthesis parameters in defining the composites' morphological, elemental, and bonding characteristics, laying a foundational basis for optimizing these materials for specific applications in environmental and technological fields where precise material properties are paramount for enhanced operational efficiency.

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