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Effect of SiO_2/Chitosan Composition Variation on Functional Groups in Hydrophobic Cellulose Paper for Water/Oil Separation

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Abstract: This research aims to investigate the effect of varying SiO_2-Chitosan compositions on the functional groups of cellulose paper. Which exhibits hydrophobic properties, which can be used for water and oil separation purposes. The composite layer consists of a silica-chitosan solution coated onto cellulose paper via the dip-coating method. Varying compositions of SiO2-chitosan investigated include (0.6g:0.3g), and (0.3g:0.6g). Characterization by FTIR revealed the presence of the main functional groups, i.e., Si-O-Si, C-H, C-O, N-O, N-H, C-N, and O-H, with a range of 938-3784 cm^{-1} strain. The identification of these functional groups confirms to the incorporation of the SiO2-chitosan compound on the surface of the cellulose paper. This functionalization introduces a hydrophobic layer, as evidenced by the characteristic FTIR spectra of the SiO_2-chitosan compound. The results of this study confirm that the modification of SiO2-Kitosan composition significantly affects on the chemical properties and physical stability of the composite coating. This result explains the great potential of cellulose-based materials in water and oil separation technology. In addition, these results enable the development of sustainable materials with great efficiency in fluid separation applications. When cellulose is modified with silica, it enhances mechanical strength, thermal stability, and selectivity of the membrane, making it suitable for fluid separation applications, such as filtration.

Keywords: Cellulose Paper; SiO_2-Chitosan; Functional Groups; Hydrophobic; FTIR (Fourier Transform Infrared Spectroscopy).

1. Introduction

The problem of water pollution caused by oily wastewater effluents of wastewater is a growing concern at present. Such effluents, generated through several types of activity including industries,

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residential, and transportation, can have adverse effects on the environment and human health, even in the long term. Besides harming the ecosystem, such wastes can also impair the purity of water, which is life-supporting [1]. The rapid development of industry and urbanization has led to an increasing generation of oily waste. "Meanwhile, the demand for clean water is rising due to rapid population growth and economic development, especially in regions with limited water availability [2]"..Existing water sources are becoming increasingly scarce and are further threatened by untreated oily effluents Such effluents normally produce an oil layer over the surface of the water, and in most cases, such a layer will hinder the availability of oxygen in the water [3]. Consequently, Consequently, aquatic life will suffer, and the aquatic ecosystem will be destroyed. Hence, several effective alternatives must be developed in overcoming such a problem, such as improvements in technology for easier separation of oil and water [4].

One efficient and effective method to separate water and oil is by using cellulose-paper-based membranes. Cellulose paper is a sheet composed of cellulose fibers, a naturally occurring compound that is insoluble in water [5]. However, pure cellulose paper is not effective, particularly due to its poor selectivity in separating oil in aqueous environments. To enhance its performance, cellulose paper must be modified to make its surface hydrophobic, i.e., water-repellent yet oil-absorbent [6]. The modification process typically involves the use of supplementary materials in the form of nanoparticles that form a coating [7]. One of the most common nanoparticle pairs used for effective oil-water separation is silica and chitosan [8].

Silica, a substance long utilized for many purposes, including as a coating, in its hydrophobic form has a low-energy surface, meaning its particles do not easily interact with water molecules. Therefore, silica tends to repel water and maintain a dry surface [9]. In contrast, chitosan is naturally hydrophilic, but when combined with silica and modified, it can effectively make the cellulose paper surface hydrophobic [10]. Additionally, chitosan has functional groups that strongly adsorb oils, particularly those with high molecular weights [11]. Thus, silica and chitosan form a highly effective coating for separating water and oil. When combined with silica and chitosan nanoparticles, modified cellulose paper can repel water while absorbing oil with high efficiency [12]. This represents a highly effective and efficient solution for cleaning oily water using cellulose-paper-based materials, offering a sustainable and innovative approach [13].

The success of SiO_2-chitosan bonding can be measured by analyzing the functional groups present in a sample. These functional groups reflect the molecular interactions between SiO_2 and chitosan [14]. These groups can be analyzed through sample characterization using an FTIR spectrometer [15]. An FTIR spectrometer is a device used to analyze molecular interactions between various molecules in a sample through its infrared spectrum. In this study, the focus is on evaluating the impact of composition variation in SiO_2-chitosan [16]. Prior studies have not sufficiently discussed the impact of composition variation in SiO_2-chitosan on functional groups. For this reason, this study evaluates the impact of composition variation in SiO_2-chitosan with compositions of (0.6 g:0.3 g) and (0.3 g:0.6 g). This variation can be observed in the FTIR spectra, as they show changes in peak intensity and position corresponding to variations in the proportions of SiO_2 and chitosan used.

2. Materials and Method

This research involves several key steps to prepare a SiO_2-chitosan composite solution. First, silica is extracted from rice husk ash, which is a key material in this study. After extracting silica, a SiO_2-chitosan composite solution is prepared with composition variations of (0.3 g:0.6 g) and (0.6 g:0.3 g). Next, a cellulose paper coating is applied using the dip-coating technique. During the dip-coating process, materials with specific characteristics are prepared. After coating, the cellulose paper is dried in an oven at 100°C for 15 minutes. Drying is performed to remove excess water, ensuring the prepared materials are more stable. To identify the functional groups in the prepared composite solution, FTIR characterization testing is conducted. The characterization testing aims to determine the molecular structure of the prepared materials. The materials and tools used in this study include Fourier Transform Infrared (FTIR), analytical scales, furnaces, dropper pipettes, glass funnels, spatulas, watch glasses, magnetic stirrers, and measuring cups. Using materials such as silica, chitosan, stearic acid, ethanol, hexane, cellulose paper, and distilled water, two variations of SiO_2-chitosan compositions were successfully prepared, which are worth exploring further.

2.1. Silica extraction of rice husk ash.

Rice husk was washed to remove impurities and then dried in a 110°C oven for 24 hours. Next, 40 g of rice husk was weighed and placed in a furnace at 700°C for 6 hours to obtain ash. The rice husk ash was ground with a mortar and sifted through a 200-mesh sieve. The ash was weighed and mixed with 250 ml of aquades solution, then stirred at 200°C for 1 hour. The goal is that the rice husk ash is completely clean from impurities and does not affect the resulting silica. Rice husk ash was dissolved in distilled water as shown in Figure 1.

Figure 1. Rice husk ash stirring process with distilled water

Next, the solution was filtered using filter paper and dried in an oven at 110°C. The dried ash was weighed, and 125 ml of NaOH solution was added, followed by stirring at 80°C for 1 hour. The solution was filtered, and HCl was added to adjust the pH to 7, producing white silica gel. The silica gel was allowed to stand for 48 hours, washed with aquades five times, filtered, and dried in an oven at 110°C for 4 hours to obtain xerogel. The xerogel was finely ground and sifted through a 100-mesh sieve.

2.2. Manufacture of SiO_2-Citosan composite solution

Mix 30 grams of stearic acid with 100 ml of ethanol solution. Stir the mixture at 80°C for 15 minutes until fully dissolved. Add 0.3 grams of chitosan to the solution and stir until

dissolved, which takes approximately 10 minutes. Repeat the same process for 0.6 grams of chitosan. Prepare a new solution by mixing 10 grams of stearic acid with 50 ml of hexane. Add 0.6 grams of silica to the solution and stir until fully dissolved. Repeat the same process for 0.3 grams of silica. This process is carried out as shown in figure 2.

Figure 2. Process of mixing silica into solution

2.3. Cellulose Paper Coating SiO_2-Chitosan Composite Solution

After preparing the SiO_2-chitosan composite solution, the next step involves coating cellulose paper with the solution using the dip-coating technique. The coating process can be seen in Figure 3.

Figure 3. The process of coating cellulose paper

This process is carried out by dipping the cellulose paper into the SiO_2-chitosan composite solution, then removing it and allowing a thin layer to form, which will evaporate, while excess solution drips off. Subsequently, the coated cellulose paper is heated in an oven at 100°C for 15 minutes. The cellulose paper is dyed three times for each sample.

2.4. Characterization of FTIR

Samples that have undergone the drying/sintering process were characterized using FTIR to identify compounds present on the coated cellulose paper. Fourier Transform Infrared (FTIR) is a widely used analytical method in chemical research to understand the molecular structure of organic and inorganic compounds. FTIR allows the observation of the infrared absorption spectrum of the sample, enabling the identification of chemical bonds within the compound [17].

3. Results and Discussion

The FTIR characterization aims to identify the functional groups in cellulose paper coated with SiO_2-chitosan with composition variations of (0.6 g:0.3 g) and (0.3 g:0.6 g). FTIR (Fourier Transform Infrared Spectroscopy) is a spectroscopic technique used to analyze chemicals based on the absorption spectra of infrared radiation by molecules. In this study, FTIR was used to determine the functional groups of cellulose paper coated with a SiO_2-chitosan layer with composition variations of (0.6 g:0.3 g) and (0.3 g:0.6 g). Characterization with these composition variations was conducted to analyze changes in the functional groups present. For example, in the first variation (0.6 g:0.3 g), an increased presence of functional groups indicates stronger interactions between the cellulose paper and the SiO_2-chitosan layer. In contrast, in the variation (0.3 g:0.6 g), changes in the intensity of functional groups suggest structural variations in the material.

3.1 The Results of the Characterization of FTIR SiO_2 - Chitosan with a Variation of (0.6g- 0.3g)

The following form of graph of the results of the characterization of FTIR, which can be seen in the picture below:

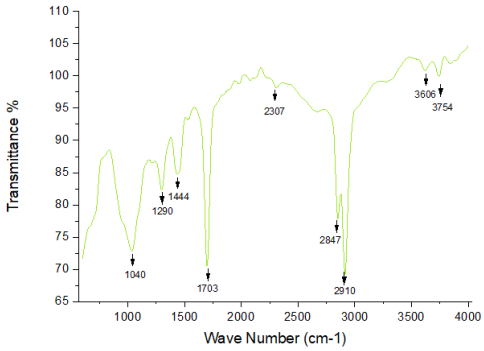

Figure 4. FTIR results of variations (0.6g: 0.3g)

Figure 4 shows the FTIR characterization graph of SiO_2-chitosan composite layers with composition variations of 0.6 g:0.3 g. From the graph, several peaks can be observed, representing the functional groups of the composite layer. These include peaks at 1040 cm^{-1}, 1290 cm^{-1}, 1444 cm^{-1}, 1703 cm^{-1}, 2307 cm^{-1}, 2847 cm^{-1}, 2910 cm^{-1}, and 3606 cm^{-1}. For instance, the peak at 1040 cm^{-1} represents the Si-O-Si bonds of the SiO_2 layer, and the peak at 1703 cm^{-1} corresponds to the C=O bond of chitosan. Additionally, the peak at 2307 cm^{-1} can be attributed to the NH_2 bond of chitosan, which is involved in interactions with SiO_2. Based on the results of FTIR, the results of the functional groups of SiO_2 - Chitosan with a Variation of (0.6g- 0.3g) can be seen in table 1.

Table 1 Functional Groups of SiO_2 - Chitosan with a Variation of (0.6g- 0.3g)

Peak Number	Wavenumber (cm^{-1})	Transmittance (%)
1	3754	99,91
2	3606	100,88
3	2910	69,23
4	2847	77,81
5	2307	98,17
6	1703	70,49
7	1444	84,74
8	1290	82,29
9	1040	72,85

Based on the composition variation of 0.6 g:0.3 g, the peak at 3754 cm^{-1} corresponds to the O-H bond, a characteristic of water or alcohol groups. The peak at 3606 cm^{-1} indicates the presence of hydrogen-bonded O-H, a characteristic of alcohol, phenol, or adsorbed water. The peak at 2910 cm^{-1} corresponds to C-H bonds. The peak at 2847 cm^{-1} represents additional aliphatic C-H bonds, commonly associated with $–CH_2$ and $–CH_3$ groups. The peak at 2307 cm^{-1} corresponds to C-N bonds. The peak at 1703 cm^{-1} indicates the presence of C=O bonds. The peak at 1444 cm^{-1} represents C-H bonds in methylene and methyl groups. The peak at 1290 cm^{-1} corresponds to C-N bonds. Finally, the peak at 1040 cm^{-1} indicates the presence of Si-O-Si bonds, a characteristic of silica and chitosan structures modified with silica. These functional groups create a hydrophilic-oleophobic balance, allowing water to pass through while repelling oil, making the membrane efficient for applications like wastewater treatment, oil spill cleanup, and industrial filtration.

3.2 The Results of the Characterization of FTIR SiO_2 - Chitosan with a Variation of (0.3g- 0.6g)

The following form of graph of the results of the characterization of FTIR, which can be seen in the picture below:

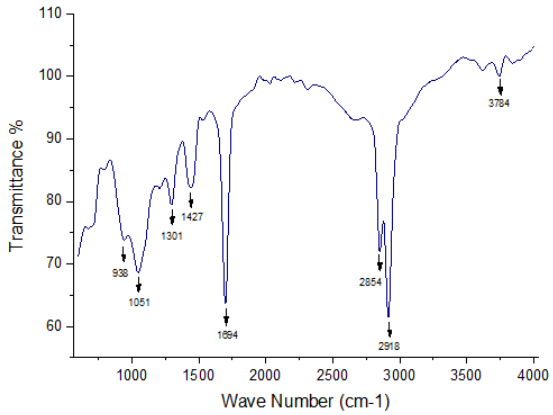

Figure 5. FTIR results of variations (0.3g: 0.6g)

Figure 5 shows the FTIR characterization graph of SiO_2-chitosan composite layers with composition variations of 0.3 g:0.6 g. From the graph, several functional group peaks can be observed in the SiO_2-chitosan composite layer. The deposition results reveal several peaks at specific wavenumbers, such as 938 cm^{-1}, 1051 cm^{-1}, 1301 cm^{-1}, 1427 cm^{-1}, 1694 cm^{-1}, 2854 cm^{-1}, 2918 cm^{-1}, and 3784 cm^{-1}. The peak at 938 cm^{-1} indicates the presence of C-O-Si bonds, while the peak at 1051 cm^{-1} indicates the presence of Si-O-Si bonds. The peak at 1301 cm^{-1} indicates the presence of C-O bonds from the chitosan group. Additionally, the peaks at 2854 cm^{-1} and 2918 cm^{-1} indicate C-H bonds associated with specific chemical structures. Based on the results of FTIR, the results of the functional groups of SiO_2 - Chitosan with a Variation of (0.3g- 0.6g) can be seen in Table 2.

Table 2. Functional Groups of SiO2 - Chitosan with a Variation of (0.3g- 0.6g)

Peak Number	Wavenumber (cm-1)	Transmittance (%)
1	3784	100,03
2	2918	61,44
3	2854	71,94
4	1694	63,67
5	1427	82,26
6	1301	70,49
7	1051	68,58
8	938	73,77

Based on the composition variation of 0.3 g:0.6 g, the peak at 3784 cm^{-1} reflects the presence of an O-H bond. The O-H group likely originates from silica (particularly from Si-OH, a silanol group) or adsorbed water on the composite surface. The peaks at 2918 cm^{-1} and 2854 cm^{-1} reveal the presence of C-H bonds. These vibrations arise from the organic structure of chitosan, specifically from methylene (CH_2-) and methyl (CH_3) groups. The presence of these groups confirms the incorporation of chitosan in the composite synthesis, and it is essential to maintain properties such as biocompatibility and antimicrobial activity. The peak at 1694 cm^{-1} indicates the presence of a C=O bond, suggesting the modification of chitosan, such as through cross-linking with silica (via silanol groups). The peak at 1427 cm^{-1} reveals the presence of C-H bonds. The peak at 1301 cm^{-1} indicates the presence of C-N bonds in the chitosan structure. This confirms chitosan's potential to enhance the composite's adsorption properties, particularly for charged molecules such as metal ions. The peak at 1051 cm^{-1} reveals the presence of Si-O-Si and C-O bonds. This vibration is characteristic of the siloxane group (Si-O-Si) in the silica framework. This confirms the successful fabrication of silica and the potential for interactions between silica and chitosan, which enhance the composite's mechanical properties. The peak at 938 cm^{-1} corresponds to the silanol (Si-OH) group on the silica surface. Silanol groups form hydrogen bonds with chitosan-bound amide and hydroxyl groups, promoting interactions between components and enhancing adsorption capacity and surface properties. The graphical form of the FTIR data from the two samples can be seen in Figure 6 below.

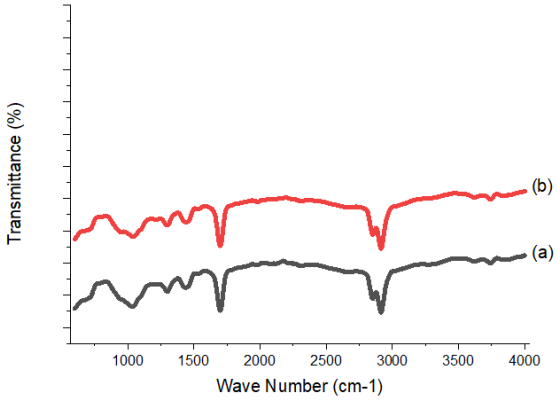

Figure 6. FTIR graph of the two samples.

Figure 6 shows the graphical form of the FTIR test data results, where the x-axis shows the wave number, while the y-axis shows the transmission. The black graph shows the data results from the composition variation of 0.6g: 0.3g, the red graph shows the data results from the composition variation of 0.3g : 0,6g. The FTIR peaks confirm interactions between alkyl groups (-CH_3, -CH_2) and hydroxyl groups (-OH) derived from chitosan and silica. For example, when chitosan-derived hydroxyl groups interact with silica-derived alkyl groups, the formation of a bond enhances the material's hydrophobic properties. Additionally, the C=O peak indicates the potential formation of carboxylic acid and ester bonds, which influence the efficiency of water-oil separation. For instance, ester bonds can enhance the material's ability to repel water and attract oil, thereby improving separation efficiency. The Si-O-Si peak confirms the role of silica in the cellulose matrix, enhancing the material's physicochemical properties for separation applications. For example, when silica is added to the cellulose matrix, the material's structure is strengthened, improving its resistance to environmental impact.

Variations in silica and chitosan composition in both samples allow for the prediction of changes in functional groups responsible for hydrophobic properties and water-oil separation capabilities. Consequently, these variations lead to changes in the chemical and physical properties of the material, affecting the efficiency of the separation process. Silica facilitates chemical interactions, while chitosan contributes properties that reduce water affinity. In addition, silica enhances the material's overall hardness due to its crystalline nature, and chitosan provides protection against microorganism contamination due to its antibacterial properties. When combined in a separation material, silica and chitosan create a mixture that optimizes water-oil separation efficiency.

Based on a dual plot of two composition variations, the FTIR spectrum shows variations in peak intensity and position, which indicate interactions between silica, chitosan, and cellulose. Compositional variations affect the presence of functional groups, thereby influencing the hydrophilic and hydrophobic properties of the composite material. For instance, when silica is added to the mixture, significant variations in the FTIR spectrum are observed, indicating interactions between silica and other components. This demonstrates that silica plays a significant role in shaping the properties of the composite material. Additionally, the presence of chitosan and cellulose also contributes to influencing the material's properties. For example, the addition of chitosan can make the material more hydrophilic, while cellulose contributes to its hydrophobic

nature. Therefore, variations in FTIR peak intensity and position can be interpreted as reflecting the complex interactions between the three components. Compositional variations in the composite material can significantly affect the efficiency of water-oil separation [18].

4. Conclusion

The first mixture (0.6 g silica: 0.3 g chitosan) can be optimized in terms of efficiency in terms of water and oil separation, with a strong interaction between chitosan and silica appearing at 1040 cm^{-1} (Si-O) and high silica proportion. Silica will have a high value of adsorption for water and oil, and in such proportion, silica can play a dominant role in characterization for separation. The mixture (0.3 g silica: 0.6 g of chitosan) consists in a high proportion of hydroxyl groups (O-H) with a role of enhancing contact with water, but less strong interaction between silica and chitosan, with a role in enhancing efficiency in oil separation.

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