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RESEARCH PAPER

Bentonite-chitosan bionanocomposite for adsorption of used lubricant oil

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Abstract: The growth of the transportation industry has led to the expansion of the automotive sector. Automotive business activities have the potential to cause several environmental problems, one of which is environmental pollution caused by used lubricating oil waste. One alternative effort to process lubricating oil waste is the adsorption method. This research aims to determine the ability or effectiveness of bentonite-chitosan composites in adsorbing waste lubricating oil. This research began by synthesizing Bentonite-Chitosan composites. The successfully synthesized material was then characterized using X-ray diffraction (XRD) and Fourier Transform Infra Red (FTIR) instruments. After that, it will be applied to used lubricating oil waste with variations in time and mass variations. The ability of the Bentonite-Chitosan composite to adsorb waste lubricating oil will be seen through the viscosity test results. The viscosity test results show a significant influence on the viscosity of the used lubricating oil. Meanwhile, the results of viscosity testing for mass variations show that the more bentonite-chitosan composite used, the greater the viscosity increase value. The highest viscosity value was obtained in the bentonite-chitosan composite of 15 g with a time of 5 hours at 7,75 cSt.

Keywords: Bentonite, chitosan, adsorption, lubricating oil, viscosity

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INTRODUCTION

The growth of the transportation industry has driven an increase in automotive businesses. The number of registered motor vehicles in Riau Province has reached 4,216,014 units [1], and this figure is expected to continue rising along with population growth. This trend encourages the expansion of automotive businesses in motor vehicle maintenance and repair services. These automotive activities can cause various environmental issues, including pollution [2]. The disposal of used lubricants contributes to the degradation of soil, water, and ecosystems. Emissions of metals such as calcium, magnesium, iron particles, and zinc from the combustion of mineral oils used as lubricants raise concerns about environmental degradation. Furthermore, the current and prospects of mineral oil usage as vehicle engine lubricants are predicted to increase [3]. The higher the consumption of lubricants, the greater the volume of used lubricant waste generated [4].

Given these issues, there is a need for methods to process the oil used for lubricating. One alternative approach for waste treatment is the adsorption process. This method is considered promising due to its simplicity and relatively low cost. A natural material that can be used as an economical adsorbent is bentonite. Bentonite is an abundant natural material in Indonesia and is inexpensive. It is a unique mineral due to its swelling properties, cation exchange capacity, and intercalation ability [5]. Its significant swelling ability allows bentonite to adsorb various metal ions. Because of its swelling properties and large surface area, bentonite can be used as an adsorbent. [6].

The photodegradation process is more widely used in industry because it is more economical and does not cause toxic side effects, one of which is adsorbents containing natural polymers such as chitosan [7]. Chitosan is widely used as an adsorbent because it has amine and hydroxyl groups, which cause chitosan to have high chemical reactivity, which plays a role in ion exchange. Chitosan is a biopolymer that is effectively used as a heavy metal adsorbent because of its non-toxic properties, high mechanical strength, biocompatibility, biodegradability, and functionality [8].

In this study, we want to synthesize bentonite-chitosan to degrade used lubricating oil. Chitosan is widely used as an adsorbent because chitosan has amine and hydroxyl groups, which cause chitosan to have high chemical reactivity and act as an ion exchanger [9]. Bentonite functions as an absorber of dirt in used lubricating oil because bentonite has a specific crystal structure [10]. This research is expected to produce Bentonite-Chitosan material that can reduce the impact of environmental pollution caused by used lubricating oil waste, and this study also hopes that the used lubricating oil can be reused.

MATERIALS AND METHODS

The equipment used in this research is chemical glassware supporting analysis, analytical balance, oven, magnetic stirrer, hot plate, X-ray diffraction (XRD), Fourier Transform Infra-Red (FTIR), and Personal Protective Equipment (PPE). The materials used in this research were activated bentonite, chitosan, distilled water, glacial acetic acid, sulfuric acid, and lubricating oil.

Synthesis of Bentonite-Chitosan

The chitosan solution was prepared by dissolving 2 g of chitosan powder in 200 mL of 5% (v/v) acetic acid and stirring at room temperature for 4 hours. In addition, bentonite suspension was prepared by dispersing 8 g of bentonite powder in 600 mL of distilled water and stirring for 30 minutes. The chitosan solution was then slowly added to the bentonite suspension at a temperature of 60˚C and stirred for 24 hours. The final composite was washed with distilled water until it reached a neutral pH. The product obtained was dried at 70˚C to achieve modified bentonite powder [7].

CHARACTERIZATION

X-ray diffraction (XRD)

X-ray diffraction is used to determine the structure, crystallinity, and purity level. XRD is an analysis used to identify crystallite material in a material by utilizing X-ray electromagnetic wave radiation [8]. XRD analysis was carried out to determine changes in the structure of bentonite before and after modification with chitosan.

Fourier Transform Infra-Red (FTIR)

FTIR is an analytical techniquethat measuresg the vibrations of molecules excited by infrared radiation in a certain wavelength range. The most widely used region for IR analysis is the 4000-400 cm^{-1} range because all molecules have their characteristic absorbance and main molecular vibrations in this region [8]. FTIR analysis was carried out to determine the functional groups in the formed bentonite-chitosan composite.

Preparation of Used Lubricating Oil

The prepared 800 mL sample of used lubricant was put into a beaker glass, then stirred at a speed of 400 rpm and 160 mL of H_2SO_4 (p.a) was added. The mixture was stirred continuously for 3 hours until a homogeneous solution was obtained, then left for 24 hours until 2 layers were formed and the oil layer was taken for adsorption [9].

Adsorption of Used Lubricating Oil

Determination of Optimum Adsorption Time

A total of 3 beakers, each filled with 100 mL samples of used lubricating oil. 5 g of Bentonite-Chitosan composite was added to each beaker. Stirred using a magnetic stirrer (400 rpm) with varying times of 1, 3 and 5 hours at a temperature of 70°C. Then, at each stage, a settling time of 24 hours is given [9]. After that, a parameter test was carried out on the viscosity of the lubricating oil to determine the optimum adsorption time.

Determination of Optimum Mass

A total of 3 beakers, each filled with 100 mL samples of used lubricating oil. 5, 10, and 15 g of Bentonite-Chitosan composite were added to each beaker and stirred using a magnetic stirrer (400 rpm) for (optimum adsorption time) at a temperature of 70°C. Then, at each stage, a settling time of 24 hours is given [9]. After that, a parameter test was carried out on the viscosity of the lubricating oil to determine the optimum mass.

Viscosity Test

Viscosity is the most important parameter in lubricants because the viscosity value will affect the lubricating power [15]. Viscosity can be expressed as resistance to fluid flow, which is the friction between fluid molecules and each other. The measuring instrument used to determine the viscosity of a liquid is a viscometer. The viscometer used in this research was an Ostwald viscometer [10].

RESULTS AND DISCUSSION

X-ray diffraction (XRD)

Results Analysis using XRD was carried out to determine changes in the structure of bentonite before and after modification with chitosan. The success of the modification process is determined from the results of XRD analysis by comparing the basal spacing (Å) of the bentonite and bentonite-chitosan diffractograms. The results of characterization using XRD can be seen in Figure 1. Each clay mineral has distinctive diffraction peaks because the diffraction angle 2θ is related to the crystal lattice of the mineral being analyzed. The typical characteristic of bentonite minerals in XRD characterization is characterized by the appearance of spectrum peaks in the corner areas (2θ = 19°; 20-23°; 26°; and 39°), which are diffraction planes of basal spacing [11].

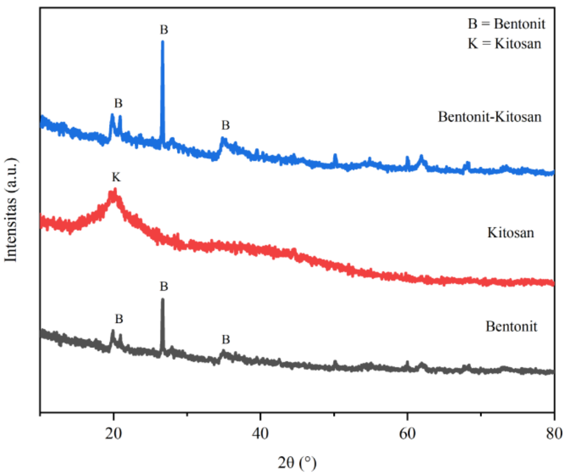

Figure 1. Diffractogram of Bentonite, Chitosan, and Bentonite-Chitosan Composite

Based on the images of the XRD patterns of bentonite and bentonite-chitosan, it can be seen that the interaction process of chitosan and bentonite does not cause structural changes in bentonite, which is characterized by the 2θ peak, which still shows the typical peak of bentonite. The bentonite diffractogram in this study shows typical peaks of the bentonite mineral, namely at 2θ = 19.91°, 20.93°, 26.70° and 35.07° with basal spacing of 4.4538 Å, 4.2403 Å, 3.3355 Å and 2.5562 Å, respectively. This result is supported by research [12], which shows the main peaks in the area 2θ = 19.96°, 35.08° and 61.92° with basal spacing of 4.44 Å, 2.55 Å and 1.49 Å, respectively, which is characteristic of bentonite minerals.

After modification with chitosan, the 2θ peaks experienced a shift in the interplane distance to 19.85°, 20.89°, 26.67° and 35.04° with basal spacing of 4.4671 Å, 4.2471 Å, 3.3396 Å and 2.5586 Å, respectively. In the chitosan diffractogram, the appearance of a peak at 2θ = 20.22° indicates the presence of a chitosan diffraction pattern. Based on research [11], the X-ray diffraction pattern of chitosan shows a diffraction peak pattern at the 2θ position of around 10 and 20. Based on this data, it can be concluded that the sample used is chitosan. Although the chitosan diffraction peak is not visible in the bentonite-chitosan diffraction pattern, changes in the 2θ value can indicate a structural change in bentonite to bentonite-chitosan. It could be due to chitosan sticking to the bentonite surface or vice versa, even in small quantities.

Fourier Transform Infrared Results (FTIR)

The FTIR Characterization process is carried out to determine the functional groups present in each material. The functional groups contained in the material absorb energy with certain wave numbers in the infrared region. FTIR analysis was carried out to identify the presence of functional groups originating from chitosan contained in the bentonite structure. The FTIR spectrum can be seen in Figure 2 and Table 1. The FTIR spectrum for bentonite shows the characteristic absorption band for bentonite at 3623.26; 1633.78; 1419.84; 997.32; 790.30; cm^{-1}. The FTIR spectrum of bentonite shows an absorption band similar to the results of research [13], which showed an absorption band at 3636.93, 1637.5, 1443.18, 998.06, 783.71, cm^{-1}; this spectrum is similar to standard bentonite.

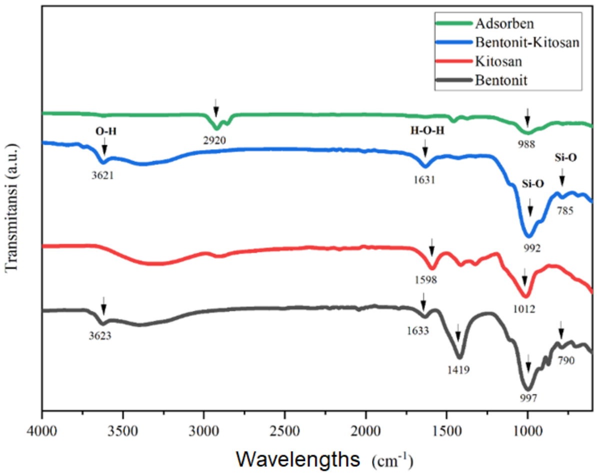

Figure 2. FTIR Spectra of Bentonite, Chitosan, Bentonite-Chitosan Composite and Bentonite-Chitosan Adsorbent After Contact with Used Lubricating Oil

Table 1. FTIR Analysis Results

Wave number (cm-1)		Functional group
Bentonite	**Bentonite-Chitosan**	
3623,26	3621,19	O-H
1633,78	1631,55	H-O-H
997,32	992,15	Si-O
790,30	785,96	Si-O

Absorption in the 3100-3700 cm-1 area is formed due to the stretching vibration of O-H and is strengthened by absorption in the wave number 1600-1700 cm^{-1}, which is the bending vibration of H-O-H. The absorption bands that appear at wave numbers 3100-3700 cm^{-1} and 1600-1700 cm^{-1} come from H_2O molecules bound through hydrogen bonds in bentonite [12]. The Si-O stretching vibration absorption in bentonite appears at a wave number of 1419.84 cm^{-1}; this result is supported by research [13] at a wave number of 1443.18 cm^{-1}, which is the Si-O stretching vibration in bentonite. Absorption at wave number 790.30 cm^{-1} shows Si-O stretching vibrations; this result is supported by research [9]. Typical absorption bands will appear at wave number 3620.1 cm-1 -OH group; 1636.4 cm-1 H-O-H; 794.6 cm^{-1} Si-O.

Research shows that the typical absorption peaks in bentonite, chitosan, and bentonite-chitosan include peaks at wave numbers 3100-3700 and 1600-1700 cm^{-1} [11]. A shift in some absorptions shows the interaction between chitosan and bentonite. Bentonite has O-H stretching vibrations and H-O-H bending vibrations of water molecules at wave numbers 3623.26 and 1633.78cm^{-1}. The O-H group is not much different in the bentonite-chitosan composite, namely 3621.19 and 1631.55 $cm^{-1.}$

In the spectrum of the bentonite-chitosan adsorbent that has been contacted with used lubricating oil, it can be seen that changes occur, namely the disappearance of absorption peaks at certain wavelengths. The disappearance of the absorption peak and changes in the absorption band pattern have proven that the impurities contained in the used lubricating oil have been bound to the adsorbent. It shows that the bentonite-chitosan composite succeeded in adsorbing impurities contained in used lubricating oil.

Adsorption Results of Used Lubricating Oil

Lubricating oil that has been used for a long time will experience changes in chemical composition or composition; apart from that, it will also experience changes in physical and mechanical properties. This is caused by the influence of pressure and temperature during use and dirt that enters the lubricating oil itself. Sulfuric acid has the property of working to reduce the surface tension of liquids, so it can be used to remove several contaminants contained in used lubricating oil [9]. This preparation stage makes the adsorption stage easier because it reduces contaminants. Preparation of used lubricating oil is carried out by adding H_2SO_4 (Sulfuric Acid) to form two layers. The top layer will be separated, and the adsorption process will continue. In the lubricant adsorption process using bentonite-chitosan composite, a cation exchange process occurs in the bentonite-chitosan composite so that metal and non-metal impurities that have not been dissolved in the preparation process can be adsorbed by the bentonite-chitosan composite. The adsorption process was carried out by varying the adsorption time and mass of the bentonite-chitosan composite. The ability or effectiveness of the Bentonite-Chitosan composite in adsorbing waste lubricating oil will be seen through the viscosity test results.

Viscosity test

Lubricating oil is widely used to protect the engine from friction with other components, which can cause damage or wear. When used, lubricating oil is contaminated with various impurities, causing a decrease in the effectiveness and quality of the lubricating oil over a certain period. The level of wear on the engine occurs more easily with thinner lubricants. Viscosity is the most important parameter in lubricants because the viscosity value will affect the lubricating power. One way to measure lubricating oil's viscosity is using an Ostwald viscometer. Ostwald viscometer is a tool used to determine the viscosity of a liquid. This tool is made of U-shaped glass and can hold a certain amount of liquid. This can be determined by measuring the time required for a liquid to flow in a capillary tube from point a to point b [14]. The viscosity of lubricating oil is determined by comparing it with samples with a certain viscosity. In this study, the comparison lubricating oil used new lubricating oil.

Optimum Adsorption Time

This study's time variations were 1, 3, and 5 hours. Time variations were carried out to determine the optimum adsorption conditions for the bentonite-chitosan composite to adsorb impurities in used lubricating oil. The viscosity test results for time variations can be seen in Figure 3. From the viscosity test results, time does not have much effect on the viscosity of used lubricating oil. There was no significant change between variations in adsorption time. The viscosity data obtained at 1, 3, and 5 hours were 3.37 cSt, 3.37 cSt, and 4.0 cSt, respectively. The highest viscosity will be taken to determine the optimum adsorption mass, namely at 5 hours.

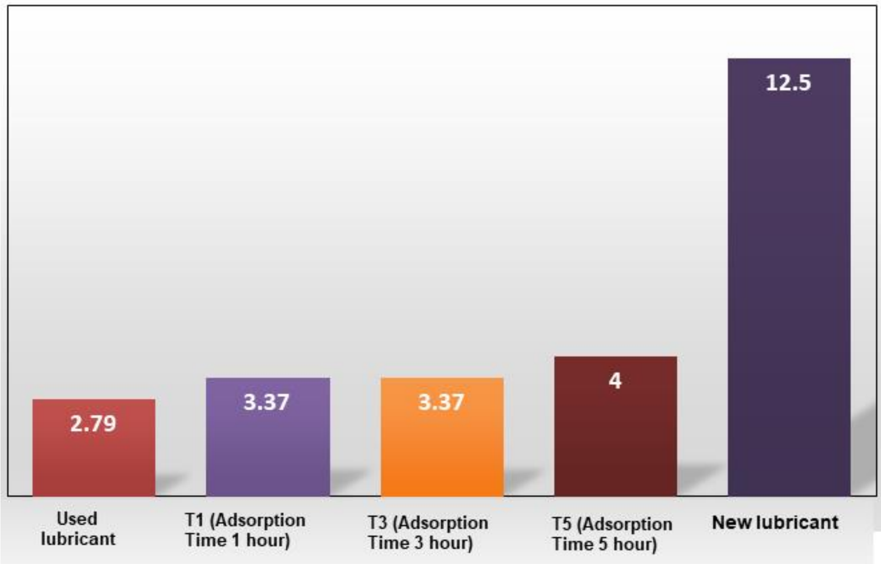

Figure 3. Time Variation Viscosity Test Results

Optimum Mass Determination Results

In this study, the mass variations used were 5, 10, and 15 g. The viscosity test results for mass variations can be seen in Figure 4. From the viscosity test results, it can be seen that there was a greater increase in viscosity compared to the time variation. The viscosity data obtained for masses of 5, 10, and 15 g were 4.0 cSt, 4.87 cSt, and 7.75 cSt, respectively. From the results of viscosity testing for mass variations, it can be seen that the more bentonite-chitosan composite used, the greater the viscosity increase value. The highest viscosity value was obtained in the bentonite-chitosan composite of 15 g with a time of 5 hours at 7.75 cSt.

Even though it is not yet close to the new lubricating oil standard, there has been a fairly large increase in viscosity. This indicates that the high value of contaminants that can be absorbed in this mixture variation increases the viscosity of the used lubricating oil samples. The increase in viscosity of lubricating oil over time shows that the adsorption time of lubricating oil is not significant in increasing the viscosity of the fluid being adsorbed. This shows that the mass of the adsorbent has more influence on the adsorption of lubricating oil than the adsorption time. Other factors such as adsorption temperature, lubricating oil and surface chemical properties of the material can also influence the extent to which lubricating oil adsorption affects viscosity. Other parameter tests are needed for further research, such as pour point, flash point, and colour measurements of used lubricating oil.

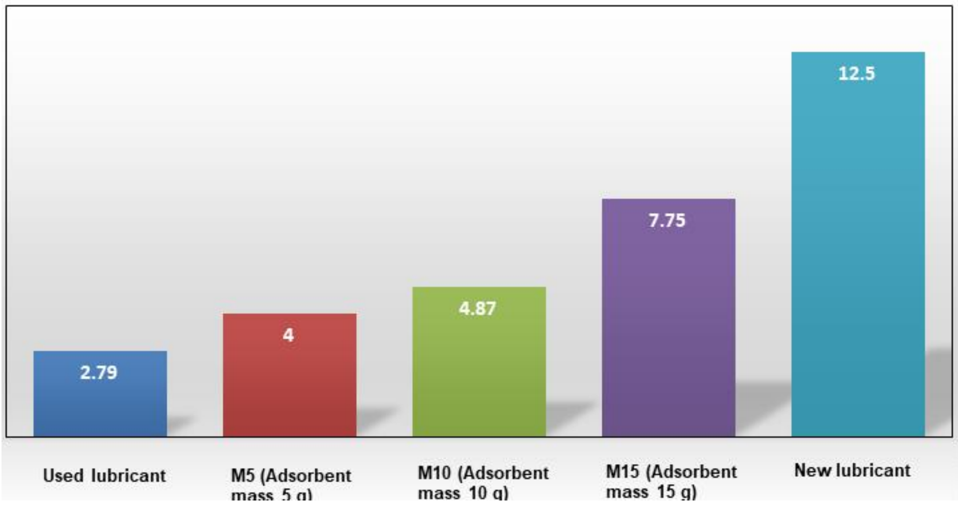

Figure 4. Mass Variation Viscosity Test Results

CONCLUSION

The conclusion from the research that has been carried out is that the results of the Bentonite-Chitosan composite synthesis were 7.0284 g with a per cent yield of 70.22%. From the X-ray diffraction (XRD) analysis results, it can be seen that the interaction process of chitosan and bentonite does not cause structural changes. Although the chitosan diffraction peak is not visible in the bentonite-chitosan diffraction pattern, changes in the 2θ value can indicate a structural change in bentonite to bentonite-chitosan. It could be due to chitosan sticking to the bentonite surface or vice versa, even in small quantities. From the Fourier Transform Infra-Red (FTIR) analysis results, a shift in some absorptions shows the interaction between chitosan and bentonite. In the spectrum of the bentonite-chitosan adsorbent that has been contacted with used lubricating oil, it can be seen that changes occur, namely the disappearance of absorption peaks at certain wavelengths. The disappearance of the absorption peak and changes in the absorption band pattern have proven that the impurities contained in the used lubricating oil have been bound to the adsorbent. It shows that the bentonite-chitosan composite succeeded in adsorbing impurities contained in used lubricating oil. The viscosity test results show that time does not have much influence on the viscosity of used lubricating oil. The results of viscosity testing for mass variations show that the more bentonite-chitosan composite used, the greater the viscosity increase value. The highest viscosity value was obtained in the bentonite-chitosan composite of 15 g with a time of 5 hours at 7.75 cSt.

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