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Removal of Nonsteroidal Anti-Inflammatory Drugs (NSAIDs) from Water Using Empty Fruit Bunch (EFB) Based Bio-sorbent

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Abstract. Naproxen has been found in a variety of water sources, including drinking water and groundwater. The effectiveness of wastewater treatment plants and other modern pharmaceutical removal procedures are investigated, with a particular focus on adsorption. There are several significant findings in this report that the quantity of oil palm wastes particularly oil palm EFB has created disposal problems. Utilizing EFB as a pollutant adsorbent in wastewater is a viable option. The aim of this research is to determine the adsorption capacity and the efficiency of EFB in removal of NSAIDs from water. As a result, EFB must be used to mitigate agricultural waste and water pollution. The highest percentage naproxen sodium removal shows at concentration of 100 ppm after 24 hours in 2.5 g of EFB, which was 98.66%. For 5.0 g of EFB, the highest percentage removal recorded in 1000 ppm concentration of naproxen sodium solution after 24 hours was 45.74 %. Aspects for EFB to achieve maximum adsorption capacity and efficiency are smaller EFB and contact time in 24 hours. In conclusion, EFB can be utilized as a cheap raw material to be a bio-sorbent.

1. Introduction

NSAIDs are a kind of medicines that has analgesic (pain-relieving) and antipyretic (fever-lowering) effects, similarly as anti-inflammatory effects in higher doses. However, the persistence of NSAIDs within the atmosphere has raised serious questions about their possible danger to terrestrial and marine ecosystems [1-8]. Analgesics and nonsteroidal anti-inflammatory medications are one of the most common anthropogenic groups of xenobiotics found in soil, sediments, surface, ground, and even drinking water.

Furthermore, conventional wastewater treatment plants (WWTPs) are not fully equipped to remove these micro pollutants at this time. Most drugs that make it to the environment are unaltered or only slightly metabolized, such as conjugated with charged molecules [9-10]. There is no one approach utilized in sewage treatment for micro pollutants usage due to the enormously diverse structure of micro pollutants, their unsaturated and saturated character, the presence of numerous functional groups such as halogen, distinct side chains, and linear or branching structure [11].

The use of EFB to produce valuable bio-chemicals is used as a cost-effective and long-term waste management solution in the palm oil industry. The EFB which is a residual product after extracting oil from fresh fruit bunches (FFB), is one of the numerous leftovers from the palm oil industry [11]. Converting EFB into value-added adsorbents solves the disposal problem while still replacing traditional adsorbents. Because of its variety, low cost, and high content of lignin, cellulose, and hemicellulose, EFB has gained popularity as an adsorbent. Physical and chemical alterations to EFB will turn it into high-adsorption-capacity value-added adsorbents [12]. In this study, EFB is tested for its potential to act as bio-sorbent in removal of Naproxen from the water matrix.

2. Material and Methods

2.1. Materials

Palm oil empty fruit bunch used in this experiment to test the adsorptivity rate of empty fruit bunch. To test the adsorption of EFB, Naproxen sodium solutions were prepared in Environmental Laboratory at Faculty of Earth Science in Universiti Malaysia Kelantan Campus Jeli. EFB was collected from Felda Kemahang, Tanah Merah, Kelantan. Naproxen sodium 275 mg was bought at Fedrowell Pharmacy, Jeli, Kelantan.

2.2. Preparation of Empty Fruit Bunch (EFB) as Bio-sorbent

Empty fruit bunch samples shown in Figure 1 was collected from Felda Kemahang, Tanah Merah, Kelantan. The EFB is placed in the oven at 75°C for 2 hours. After EFB completely dried, it was cut into little pieces of approximately 2-3 cm. The purpose of this procedure was to lower the moisture content of the EFB. No chemical treatments were used to guarantee that the biomass was used is a cost-effective and low-cost manner [12].

Figure 1. Raw EFB

2.3. Preparation of Naproxen stock solutions

Naproxen sodium is a white to creamy, crystalline solid, freely soluble in water at neutral pH. Sunprox as naproxen sodium tablets is available as light blue and oval-shaped tablets containing 275 mg of naproxen sodium. About 50 tablets were crushed in powder form using pastel and mortal. The powdered tablets were weighed 20 g on a weighing scale. The naproxen sodium was then placed in a beaker, and 3 ml of methanol was added. More methanol was added until the naproxen sodium dissolved. 30 ml of methanol were added until all the powder was dissolved.

Later, the methanol solution of naproxen was then air dried to evaporate the methanol. Three days were taken until the solution was dried entirely, leaving a white powder on the bottom of the beaker. Demineralised distilled water was added until it reached 1 litre and shook on an orbital shaker at 150 rpm for three days until it dissolved completely.

A working stock solution was created from which four known concentration solutions (standard solutions) were created. A calibration curve of the solution must be made to estimate the concentration of the naproxen in the solution. Absorbance for each point was measured, and the absorbance vs concentration graph is shown in Figure 2. The regression line is Y= 6E-05x, and it was generated using the least squares approach from four points. The correlation coefficient between absorbance and concentration is 0.986.

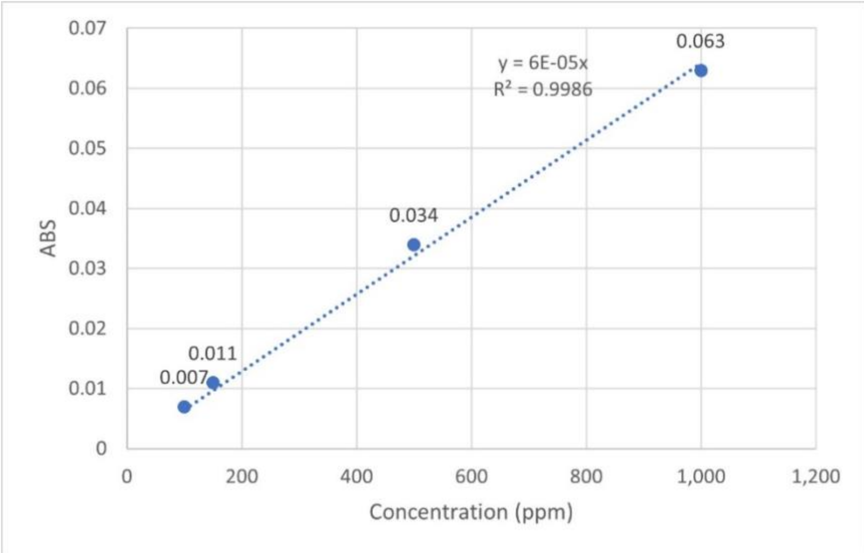

Figure 2. Calibration curve of Naproxen sodium solution

2.4. Absorption Experiment

5 different compound concentrations (0 ppm, 100 ppm, 150 ppm, 500 ppm, 1000 ppm) with 3 different EFB weight (0 as blank, 2.5 g, 5.0 g) was used. Different weight of the EFB are added to a beaker with 50 ml of working solution at different concentrations. The beaker was sealed with aluminium foil and set aside for 60 minutes. Then, using a syringe membrane filter, aqueous samples are extracted from the solutions, and the 1 ml of concentrations are evaluated using a Hach Dr6000 UV-Vis spectrophotometer. UV-Vis Spectroscopy was used to evaluate changes in concentration readings of the solutions a 0-hour, first hour and after 24 hours. All the tests were carried out at room temperature and were performed three times for each concentration, with the average computed and graphed. The solution temperature was maintained at 30°C. The initial and final concentration of the compound was measured.

2.5. Adsorption capacity and removal efficiency

The adsorption procedure was carried out for 24 hours at room temperature to achieve absorption equilibrium. After 24 hours, the mixture was filtered to measure the concentration of naproxen sodium solution. All the adsorption studies were carried out in triplicate. The Equation 1 for calculating equilibrium adsorption capacity, qe, (mg/g) was used where; C_i is initial diclofenac concentration (mg/L), C_e is equilibrium diclofenac concentration (mg/L), V is a volume of the diclofenac solution (L) and W is the weight of the adsorbent that was used (mg).

$$\text{qe}=\frac{(C_i - C_e)\,V}{W} \qquad (1)$$

The adsorbance of each solution was measured at the wavelength of peak absorption, using the same container for each. Then, the unknown concentration was calculated by substituting adsorbance in regression line, Y= 6E-05x. The steps were repeated for all the adsorbance readings of concentration of naproxen sodium solution in EFB at 0-hour, 1 hour and 24 hours. The original and final concentrations of naproxen were measured, and the percentage of naproxen removal was determined using the Equation 2, where C_i represented as the initial concentration of the sample (mg/L) and C_e as the final concentration of the sample (mg/L).

$$\text{Removal percentage of Naproxen (\%)} = \frac{c_i - c_e}{c_i} \times 100\% \qquad (2)$$

3. Results and Discussion

3.1 Effect of adsorbent dosage

Figure 3 shows a graph bar of removal efficiency of Naproxen sodium using 2.5 g of EFB against concentration of naproxen sodium solution, ppm at 1 hour and 24 hours. Concentration of naproxen sodium solution at 100 ppm achieved the highest removal efficiency of naproxen sodium from water (98.66 %). 500 ppm of naproxen sodium solution have the lowest removal efficiency from water which is 58.29 %.

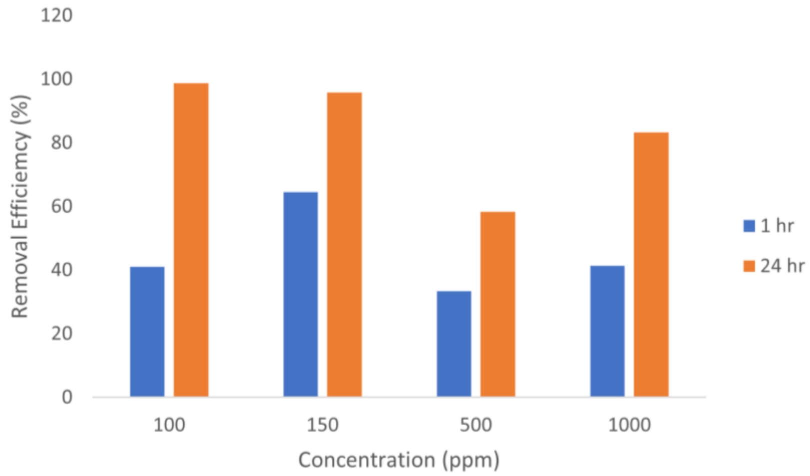

Figure 3. Graph of removal efficiency of Naproxen sodium, % using 2.5 g of EFB against concentration of Naproxen sodium solution, ppm at 1 hour and 24 hours

Moreover, Figure 4 shows a graph bar of removal efficiency of Naproxen sodium solution using 5.0 g of EFB against time. Concentration of Naproxen sodium solution at 1000 ppm achieved the highest removal efficiency of naproxen sodium from water (45.74 %) in 24 hours. 500 ppm of naproxen sodium solution have the lowest removal efficiency from water which is 20.44 % in 24 hours.

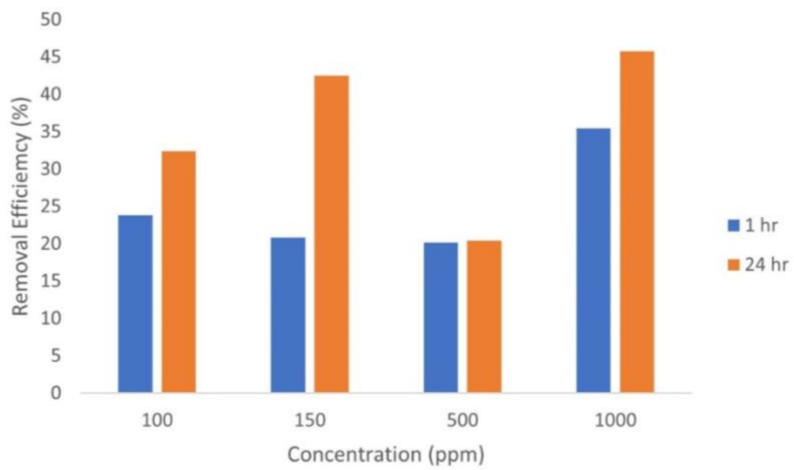

Figure 4. Graph of removal efficiency of naproxen sodium, % using 5.0 g of EFB against concentration of naproxen sodium solution, ppm at 1 hour and 24 hours

The EFB fiber surface has functional groups, such as hydroxyl groups, that increase its hydrophilic property, reducing its affinity to bind to hydrophobic molecules and thus enhancing the adsorption process. Because lignin and disrupting the crystalline structure of the cellulose are sealed, the accessibility of the pores and surface of the cellulose is low without altering the structure of the cellulose. Pollutant adsorption capacities of lignocellulosic materials are remarkable. During the adsorption test, cellulose is one of the components in the EFB fiber that has a hydrophilic property that causes water uptake and capillary action. The capillary action that happens in the microscopic crevices inside the fiber composite allows water to naturally transfer, resulting in a moisture content increase. The substantial intake of water composite can be explained by the high amount of cellulose in EFB fiber [12].

3.2 Effect of Concentration

According to Figure 5 and 6, the graph of equilibrium of Naproxen sodium against concentration of Naproxen sodium solution, ppm, using 2.5 g and 5.0 g of EFB in 1 hour and 24 hours. In Figure 5, at 150 ppm in 1 hour with 2.5 g of EFB recorded the highest equilibrium adsorption which was 25.4 % while at 500 ppm, recorded the lowest equilibrium adsorption which was 17.9 %. The line of 5.0 g of EFB recorded the lowest equilibrium adsorption at 100 ppm (10.28 %) meanwhile the highest equilibrium adsorption recorded at 1000 ppm which was 18.95 %.

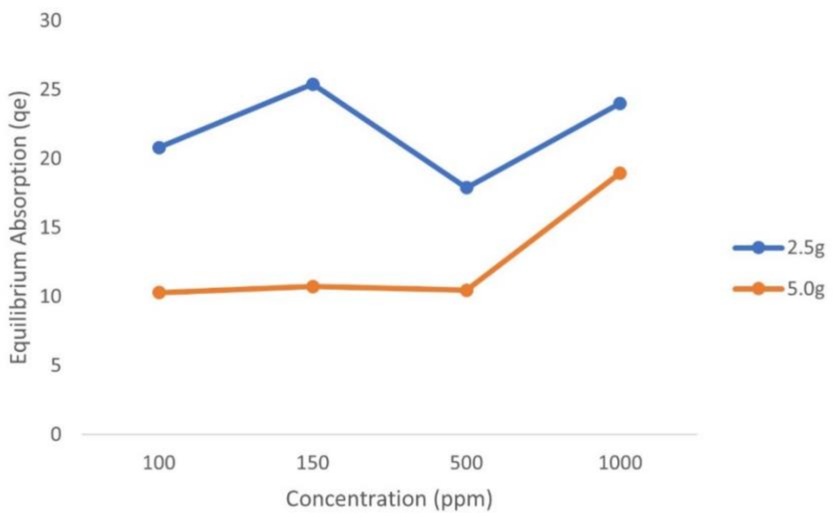

Figure 5. Graph of equilibrium absorption of naproxen sodium solution, qe in 1 hour against concentration of naproxen sodium solution, ppm, using 2.5 g and 5.0 g of EFB

Furthermore, in Figure 6, the highest equilibrium adsorption recorded on 2.5 g of EFB in 24 hours is at 100 ppm as 50.12 % meanwhile the lowest recorded is at 500 ppm as 31.22 %. The highest equilibrium adsorption recorded on 5.0 g of EFB in 24 hours is at 1000 ppm as 24.45 % meanwhile the lowest recorded is at 500 ppm as 10.62 %. However, as the fiber approaches moisture equilibrium, the weight increases the composites, resulting in minor differ in the percentage of water absorption. The water will first fill up the empty spaces quickly through capillary action until the voids become limited, which is largely reliant on the voids present in the composite itself. The number of voids in a composite that allows water to absorb can have an impact on its deterioration and interfacial bond [3].

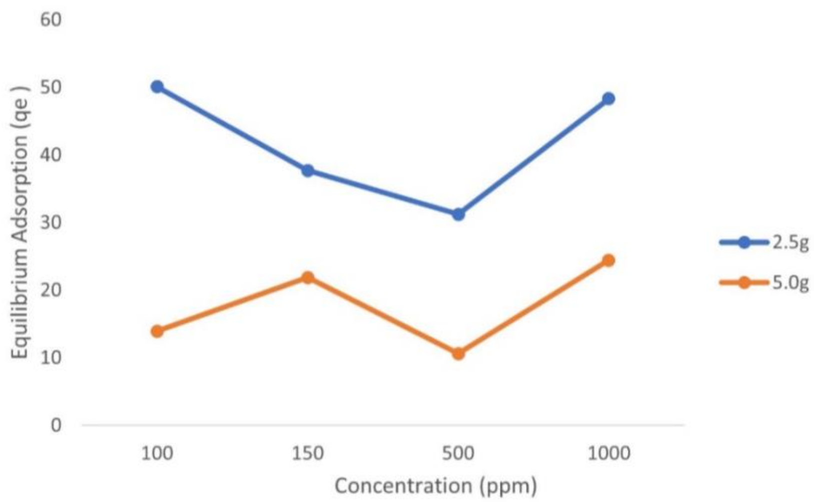

Figure 6. Graph of equilibrium absorption of naproxen sodium, qe in 24 hours against concentration of naproxen sodium solution, ppm, using 2.5 g and 5.0 g of EFB

Even though the concentration increases, the volume remains constant. As of it, the 5.0 g of EFB does not entirely immerse in the solution. The contact time with salute concentration was low. The structure cannot be compressed as EFB was untreated. Raw EFB has a solid texture with no ripples due to a covering of plant wax, whereas treated samples have a rough surface with varying degrees of wrinkles and grooves [11,12]. It has a straw-like structure and most likely cannot be compressed as it is void. Then, the concentration of naproxen sodium solution is unstable. A low concentration of naproxen sodium was more stable than the concentrated solution.

EFB cellulose has a low sorption capacity due to the presence of many hydroxyl groups on its surface. The availability of hydroxyl surface functional groups of EFB cellulose is thought to resist naproxen sodium molecules in the absorption media, limiting absorption uptake [11,12]. Furthermore, the water adsorption behaviour of the fibreboard is affected by the fibre's tendency to absorb water due to the formation of hydroxyl groups. Through the creation of hydrogen bonds, the hydroxyl group will adsorb moisture or water [5]. Thus, the process of permeation would be higher in larger EFB meanwhile the capillary action would be higher in the smaller EFB. This is because smaller EFBs have high accessibility to fill up the empty spaces in the core. Since there was less exposure to the core in larger EFB, the solution will diffuse inside the EFB through the permeation process.

Amin et al. [3] discussed that water could migrate into interior hydrophilic zones and, in some instances, permeate the solid. To attain this high absorption capacity, adsorbates do not accumulate on the pore surfaces. Permeation happens instead, transporting adsorbates into the solid structure, under the pore surfaces, to adsorption sites that are not on the pore surfaces. Because of their capacity to swell in water and create additional space for adsorbates, EFB offers excellent promise as adsorbents. In naproxen sodium adsorption, several attractive H-bonding interactions between adsorbate functional groups and those on the adsorbent surface play an essential role.

4. Conclusion

This study proves the potentials of EFB waste to be utilized as low-cost adsorbents for naproxen sodium removal. The absorption experiments showed that the adsorption contact time and adsorbent dosage had a great influence on the adsorption performance of naproxen sodium concentration by EFB. The extent of this removability to other type of contaminant of emerging concern still unknown and require an additional testing.

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