# COMPARISON OF THE PROPERTIES OF MAGNETIC PEANUT HULLS (RED AND WHITE) BY USING ORGANIC DYE

Latt Latt Aung<sup>1</sup>, Ohn Ohn Soe<sup>2</sup>

#### Abstract

This research work deals with the prepared magnetic peanut hulls used for investigation of colour removal efficiency. Peanut hulls (red and white) and prepared magnetic peanut hulls (red and white) were characterized by using EDXRF, FT IR and SEM techniques. The composition of the magnetic peanut hulls which was determined by EDXRF, showed that calcium and iron were the main component in the peanut hulls. FT IR spectra showed the presence of ionizable groups such as carboxyl, carbonyl and hydroxyl in both magnetic peanut hull (red and white) samples. SEM micrographs indicated the porous nature of the surface. Therefore dyes can enter the pores of the samples. In this research removal percent of methylene blue(MB, a cationic dye) by prepared magnetic peanut hulls (red and white) were studied in terms of contact times, concentrations, dosages, pH, stirring The highest removal efficiency of MB by speeds and temperatures. magnetic peanut hulls red (97.55 %) and white (96.86 %) were found at 120 min of contact times. The optimum concentration of MB was 20 ppm for the colour removal by both sample red (97.55 %) and white (96.86 %). The optimum dosages of both samples were observed to be 0.2 g. The optimum pH value for removal efficiencies of both samples were observed as pH 7. The optimum stirring speed and temperature were observed to be 200 rpm and 30 °C for both red and white magnetic peanut hulls samples. Magnetic peanut hulls (red) showed higher colour removal efficiencies than white sample.

Keywords: ferrofluid, peanut hulls, magnetic peanut hulls, methylene blue, colour removal efficiency

#### Introduction

Water quality control standards and regulations against hazardous pollutants have become stricter in many countries. Dyes are widely used in the textile, food, cosmetics, pharmaceutical, tanneries, electroplating factories and host other industries (Sayan, 2006). The methods of colour removal from industrial effluents include biological treatment, chemical coagulation

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followed by sedimentation, floatation, adsorption, oxidation and photocatalytic discolouration (Ozkan-Yucel and Gokcay, 2010). Among these methods, sorption processes appear to be preferable techniques. Adsorption has been proven to be an excellent method for removing dyes from aqueous solutions because of its significant advantages.

Materials whose physical properties can be varied by application of external magnetic fields belong to a specific class of smart materials. In many cases magnetically responsive composite materials can be formed by modification of originally diamagnetic materials by magnetic nanoparticles, present in different types of magnetic fluids (ferrofluids). Such composite materials have already found many important applications in various areas of biosciences, biotechnology, medicine and environmental technology (Safarik and Safarikova, 2002). The application of magnetite in the field of wastewater treatment is becoming an interesting area of research. Nanoparticles exhibit good adsorption efficiency especially due to higher surface area and greater active sites for interaction with metallic species and can easily be synthesized several researches have used it as an adsorbent (Hritcu et al., 2009). World annual production of shelled peanuts was 42 million tones in 2014. Since peanuts are grown as a plant, they are a renewable resource (Brown et al., 2000). As a by-product, the hulls are easy and inexpensive to obtain because industries are in need of only the peanut as the main ingredient in their products. Peanut hulls are very porous and have low solubility. The majority of their chemical composition is fiber (60 -67 %) including cellulose and lignin. The peanut hull is a complex material consisted of polyphenol such as catechol, pyrogallic acid and m-trihydroxybenzene, mineral, lipid, and cellulose, etc (Kargi and Cikla 2006).

Magnetically responsive peanut hulls materials, prepared by modification of diamagnetic materials by magnetic fluids (ferrofluids). Ferrofluid modified biological waste (peanut hulls) has been successfully used for the separation and removal of water soluble organic dyes and thus this low cost adsorbent could be potentially used for wastewater treatment. In addition to traditional inorganic or synthetic organic materials, recently also materials of biological origin, such as microbial and algae cells, or lignocellulose particles, have been transferred into magnetically responsive nanobiocomposite materials after magnetic fluid treatment. Such materials could be used for the separation and removal of both organic and inorganic xenobiotics or for the construction of magnetically responsive, whole cells biocatalysts. Modification of agricultural by-product could enhance their natural adsorption capacity or add another additional value to the by-product (Taha and Magraby, 2014).

The present work is concerned with the synthesis of magnetic peanut hull as a biosorbent for removal of dye.

#### **Materials and Methods**

#### Materials

In this research, Peanut hulls were collected from broker house at Ba Yint Naung, Yangon Region. All chemicals used were of analytical reagent grade.

#### **Preparation of Magnetic Peanut Hulls**

The raw peanut grains samples (red and white) were roasted with sand bath to obtain roasted peanut hulls. The roasted peanut hulls were washed with tap water to remove dirt and other impurities and then spraied with distilled water. The washed roasted peanut hulls were dried in an oven at 80 °C and then crushed in milling machine to form a powder. The powder was sieved with 600 µm mesh and stored in the dried plastic bottle.

First, 2.1 g of iron(II) sulphate and 3.1 g of iron(III) chloride were placed into a 250 mL beaker containing 80 mL distilled water. Then, the solution was heated to 80 °C, with stirring for 30 min. After that, 25 mL of 25 % (v/v) ammonium hydroxide solution was slowly added into the beaker containing solution mixture. The black precipitate was obtained. To prepare magnetic peanut hulls, 10 g each of peanut hulls powder (red and white) were suspended in the ferrofluid solution at 80 °C. The suspension was mixed on a stirrer for 30 min. The magnetically modified peanut hulls particles were then left to cool to room temperature and washed with distilled water and then dried until complete dryness and stored in the dried plastic bottle (Taha and Magraby, 2015).

# Characterization of the Peanut Hulls (Red and White) and the Prepared Magnetically Modified Peanut Hulls (Red and White) by EDXRF, FT IR and SEM

Peanut hulls (Red and White) and prepared magnetically modified peanut hulls (Red and White) were characterized by EDXRF, FT IR and SEM (Taha and Maghraby, 2014).

## **Colour Removal Efficiency of Magnetic Peanut Hulls**

In this work, methylene blue model solution was used for investigation the colour removal efficiency of unroasted peanut hulls, roasted peanut hulls, ferro powder and magnetic peanut hulls. A 0.2 g adsorbent was added into 100 mL of 20 mg/L concentration of methylene blue solution and initial pH value of dye solutions. This solution was taken in 250 mL conical flask, stirring speed 200 rpm and thermostatically controlled at 30 °C for 120 min on rotary shaker. The solutions was then filtered and measured by UV-visible 1240 spectrophotometer. All samples were studied at same procedure (Taha and Maghraby, 2015).

# **Colour Removal Efficiency of the Prepared Magnetic Peanut Hulls (Red and White) by Using Methylene Blue Model Solution**

In this research, the prepared magnetic peanut hulls were used for investigation of colour removal efficiency. The stock solution of MB (1000 mg/L) was prepared in distilled water. All working solutions were prepared by diluting the stock solution with distilled water to the required concentration. Adsorption experiments were carried out in a rotary shaker at different speed (50-250 rpm) and ambient temperature (20-40 °C), using 250 ml conical flasks containing 100 ml different concentrations of dye solutions solutions were previous adjusted with 0.1 M HCl or NaOH using pH meter. Different doses of sorbent (0.025, 0.05, 0.1, 0.2 and 0.3 g) were added to each flask. After shaking the flasks for predetermined time intervals (20-180 min), the samples were withdrawn from the flasks and the dye solutions were separated from the sorbent by filtration then centrifugation. Dye concentrations in the supernatant solutions were estimated measuring absorbance at maximum wavelengths of dyes with a UV-visible (1240) spectrophotometer (Shimadzu, Japan) (Taha

and Maghraby, 2015). The removal percent was calculated by the following equation

 $R \% = A_0 - A_e / A_0 x 100$ 

Where  $A_0$  and  $A_e$  (mg/L) are the liquid-phase concentrations of dye at initial and at any time t, respectively.

## **Results and Discussion**

## **Preparation of Magnetically Modified Peanut Hulls**

In this research, roasted peanut hulls powder and ferrofluid solution were mixed at 80 °C and stirred for 30 minutes. Figure 1 shows photograph of magnetically modified peanut hulls (red and white) and Figure 2 shows photograph of test for magnetic properties of magnetic peanut hulls.



Figure 1: The photograph of prepared magnetic peanut hulls red and white



Figure 2: Test for Magnetic Properties of Magnetic Peanut Hulls

# Composition and Physical Properties of Peanut Hulls and Magnetic Peanut Hulls

The composition of the magnetic peanut hulls which was determined by EDXRF. Table 1 and Figure 3 show the EDXRF spectra. As can be seen, iron and calcium were the main component in the peanut hulls and magnetic peanut hulls. In this result, magnetic peanut hulls was contained more iron content than peanut hulls.

The FT-IR spectra of peanut hulls and magnetic peanut hulls biomass were studied in the range of 400–4000 cm<sup>-1</sup> using (FTIR-8400 S- Shimadzu, Japan). Table 2 and Figure 4, 5, 6 and 7 show the FT IR spectra and FT IR spectral data of peanut hulls (red and white) and the prepared magnetically modified peanut hulls (red and white) samples. The results of the FT-IR spectra of raw biosorbent revealed the presence of peak in the region of (3000-2850) cm<sup>-1</sup> which is due to the C-H stretching and indicates the presence of -CH and CH groups in the structure of peanut hull biomass. The band at (1820-1670) cm<sup>-1</sup> allocates the C=O stretching vibrations. Presence of broad bands in the region of (3600-3200) cm<sup>-1</sup> indicates the presence of O-H group (carboxylic acids, phenols and alcohols) on the surface of biosorbent as in cellulose, pectin and lignin. The presence of peaks in the region of (1680-1620) cm<sup>-1</sup> might be due to presence of C= C bonds. The peak at (1410-1310)  $cm^{-1}$  was caused by the CH<sub>2</sub> bending. The peak at (1350-1260)  $cm^{-1}$  is indicative of the OH in plane bending cellulose. Analysis of the FT IR spectral showed the presence of ionizable groups (carbonyl, carboxyl and hydroxyl) able to interact with protons, metal or positive dye ions these functional groups may be the major biosorption sites for methylene blue removal.

The scanning electron micrograph of peanut hulls were illustrated the porous nature of the surface morphology. As shown in Figure 8 (a and b) the peanut hulls material has more fiber and more active sites, while in Fig. 8 (c and d) it showed the peanut hulls is completely covered with iron oxide, and all the iron oxide particles are aggregated to form a spherical and cage-like structure. The iron oxide particles exhibit magnetic behavior, creating more negative charges. The presence of the new particles on the peanut surface not only to change material to be magnetic but also it may increase the sorption process due to increase the surface area with small particles added. Therefore dyes can enter the porous of the samples.

### **Colour Removal Efficiency of Magnetic Peanut Hulls**

In this research, colour removal efficiency of unroasted peanut hulls, roasted peanut hulls, ferro powder and magnetic peanut hulls were studied by using methylene blue model solution. Table 3 and figure 9 show the removal percent of all samples. According to these data, the highest colour removal percent of the magnetic peanut hulls is (97.58 %). Therefore colour removal efficiencies of magnetic peanut hulls samples is the best.

## Colour Removal Efficiency Effect of contact times

In the present work, colour removal efficiency of magnetic peanut hull (red and white) samples were studied using methylene blue. Table 4 shows the relationship between contact times and removal percent of methylene blue by magnetic peanut hulls red and white. Figures 10 show plot of methylene blue removal percent as a function of contact time by magnetic peanut hull (red and white) samples. The removal efficiency was found to increase for both adsorbents with increasing contact time from 20 minutes to 180 minutes. The highest colour removal efficiency of methylene blue by magnetic peanut hulls red (97.55 %) and white (96.86 %) were found at 120 minutes of contact times.

## **Effect of concentration**

In the experiment, colour removal efficiency of magnetic peanut hull (red and white) samples were studied using methylene blue. Table 5 shows the relationship between concentrations and removal percent of methylene blue by magnetic peanut hulls red and white. Figures 11 show plot of methylene blue removal percent as a function of concentration by magnetic peanut hull (red and white) samples. Removal of dye increased with increase initial concentration, further the adsorption was rapid in the early stages and then gradually increased and became almost constant after the equilibrium point. The optimum concentration of methylene blue was 20 ppm for the colour removal by both samples red (97.55 %) and white (96.86 %) respectively.

#### Effect of dosage

In this study, effects of dosages of the prepared magnetic peanut hull (red and white) samples were studied by using the dosages such as 0.025, 0.05, 0.1, 0.2 and 0.3 g. Table 6 shows the relationship between dosages of prepared magnetic peanut hulls red and white with removal percent of methylene blue. The percentages of dyes remove increased as the adsorbent dose was increased over the range 0.025–0.3 mg. Figures 12 show the effects of adsorbent dosage. So, the magnetic peanut hulls biomass of 0.2 g was chosen for subsequent experiments. Maximum biosorption capacity of magnetic peanut hulls was achieved by using 0.2 g biosorbent dosage.

## Effect of pH

In the present work, colour removal efficiency of magnetic peanut hull (red and white) samples were studied using methylene blue. Table 7 mentioned the relationship between pH and removal percent of methylene blue by magnetic peanut hulls red and white. Figures 13 show plot of methylene blue removal percent as a function of pH 4, 6, 7, 8 and 10 on magnetic peanut hulls red and white samples. The highest removal efficiency of methylene blue by magnetic peanut hulls red (97.55 %) and white (96.86%) were found at pH 7. Therefore optimum pH value for colour removal efficiencies of both samples were pH 7.

#### Effect of stirring speed

In this experiment, colour removal efficiency of magnetic peanut hull (red and white) samples were studied using methylene blue. Table 8 shows the relationship between stirring speed and removal percent of methylene blue by magnetic peanut hulls red and white. It is normally found that with higher stirring speed, there is faster removal and higher removal at equilibrium. However, the results obtained in this work, was showed the effect of stirring speed on the colour removal in the range of stirring speed studied, 50, 100, 150, 200 and 250 rpm in Figures 14. The optimum stirring speed was observed to be 200 rpm for both red and white magnetic peanut hulls samples.

#### **Effect of temperature**

In the present work, colour removal efficiency of magnetic peanut hull (red and white) samples were studied using methylene blue. Table 9 shows the relationship between temperature and removal percent of methylene blue on magnetic peanut hulls (red and white). The results regarding the effect of temperature on the biosorption potential of peanut hulls biomass are shown in Figures 15. Experiments were performed at five temperatures within the range of 20-40 °C. The results clearly showed that biosorption of methylene blue dye onto peanut hulls biomass is endothermic process. The increase in temperature results increase in biosorption of dye from aqueous solution. This increase may indicate the increased tendency of MB due to an increase in its kinetic energy with increasing temperature, and this situation leads to more adsorption on the surfaces of peanut hulls. Maximum dye removal was achieved at 30 °C. In this result, 30 °C is the most suitable temperature with 120 min contact time, 20 ppm concentration, 0.2 g dosage, pH 7 and 200 rpm stirring speed.

 

 Table 1: Relative Abundance of Elemental Compositions of Peanut Hulls and Magnetic Peanut Hulls (Red and White)

	% Composition								
Sample	Ca	Fe	S	Ti	Mn	Cu	Zn	Sr	Rb
Peanut hulls (Red)	30.556	15.958	4.449	1.223	1.019	0.762	0.41	0.397	0.212
Peanut hulls(White)	42.317	12.334	-	-	0.777	0.684	0.451	0.790	-
Magnetic Peanut hulls(red)	0.390	99.125	-	-	0.485	-	-	-	-
Magnetic Peanut hulls (white)	1.009	99.47	-	-	0.465	-	-	0.056	-



Figure 3: EDXRF spectrum of (a) peanut hulls (red) (b) peanut hulls (white) (c) magnetic peanut hulls (red) and (d) magnetic peanut hulls (white)

No.	0	bserved V	alues (ci	<b>m</b> <sup>-1</sup> )	Literature value (cm <sup>-1</sup> )	Interpretation
	P(Red)	P(White)	MP(R)	MP(W)		
1	3441	3449	3362	3426	3600-3200	$v_{OH}$ O-H stretching
2	2945	2926	2899	2924	3000-2850	$v_{CH}$ C-H stretching
3	1735	1743	1735	1743	1820-1670	$v_{C=0}$ C= O stretching
4	1641	1641	1639	1629	1680-1620	$v_{C=C}$ C=C stretching
5	1512	1514	1510	-	1600-1400	Aromatic CH <sub>2</sub> stretching
6	1419	1425	1404	1408	1410-1310	$\delta_{CH}$ CH bending
7	1263	1263	1259	1265	1350-1260	$\delta_{OH}$ O-H in plane bending
8	1143	1155	-	1107	1260-1000	v <sub>C-C-O</sub> C-C-O asymmetric stretching
9	895	-	-	-	1000-675	$\delta_{CH}$ C-H bending
10	607	611	-	-	720-590	<sup>υ</sup> <sub>Fe-O</sub> Fe-O group (asymmetric)

Table 2: Assignment of FT IR Spectral Data of Peanut Hulls and<br/>Magnetic Peanut Hulls, (Red and White )



Figure 4: FT IR spectrum of peanut hulls (red)



Figure 5: FT IR spectrum of peanut hulls (white)



Figure 6: FT IR spectrum of the prepared magnetic peanut hulls (red)



Figure 7: FT IR spectrum of the prepared magnetic peanut hulls (white)









**Figure 8:** SEM images of (a) peanut hulls (red) (b) peanut hulls (white) (c) magnetic peanut hulls (red) (d) magnetic peanut hulls (white)

Table 3: Colour Removal Efficiency of Magnetic Peanut Hulls

No.	Sample	Removal (%) ± STD	
1	Unroasted Peanut Hulls	$60.23 \pm 0.05$	
2	Roasted Peanut Hulls	$62.85 \pm 0.09$	
3	Ferro Powder	$70.91 \pm 0.02$	
4	Magnetic Peanut Hulls	97.58 <u>+</u> 0.03	







Figure 10: Photographs of colour removal efficiency of all samples with model solution

Table 4	: Removal	Percent	of N	Methylene	Blue	by	the	Magnetic	Peanut
	Hulls (Re	d and W	nite)	as a Funct	tion of	f Co	ntac	t Times	

	Contact	Magnetic Peanut Hulls	Magnetic Peanut Hulls
No.	Time (min)	(Red)	(White)
	1  mile (mm) -	Removal % ± STD	Removal % ± STD
1	20	$60.35 \pm 0.03$	$55.54 \pm 0.11$
2	40	$77.66 \pm 0.09$	$75.88 \pm 0.11$
3	60	$90.36 \pm 0.06$	$77.05 \pm 0.03$
4	80	$90.73 \pm 0.03$	$78.59 \pm 0.03$
5	100	$96.99 \pm 0.01$	$90.86 \pm 0.03$
6	120	$97.55 \pm 0.06$	$96.86 \pm  0.07$
7	140	$97.49 \pm 0.01$	$96.82 \pm 0.01$
8	160	$96.94 \pm 0.07$	$96.82 \pm 0.01$
9	180	$96.88 \pm 0.03$	$96.77 \pm 0.02$



Figure 11:Removal percent of methylene blue model solution by magnetic peanut hulls (red and white) as a function of contact times

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No.	Concentration	Magnetic Peanut Hulls (Red)	s Magnetic Peanut Hulls (White)
	(ppm)	<b>Removal % ± STD</b>	<b>Removal % ± STD</b>
1	5	$81.58 \pm 0.2$	$22  79.05 \pm 0.11$
2	10	$86.12 \pm 0.1$	$0  82.17 \pm 0.08$
3	15	$89.89 \pm 0.1$	1 $83.28 \pm 0.05$
4	20	$97.55 \pm 0.0$	$96.86 \pm 0.06$
5	25	$75.50 \pm 0.0$	$71.50 \pm 0.07$
Exp	erimental condition		
Wei	ght of Peanut Hulls	= 0.1 g Temperature	$= 30 \ ^{\circ}\mathrm{C}$
Stir	ring rate	= 200 rpm Contact time	= 120 min

Table 5: Removal Percent of Methylene Blue by the Magnetic PeanutHulls (Red and White) as a Function of Concentrations



Concentration (ppm)

**Figure 12:** Removal percent of methylene blue model solution by the magnetic peanut hulls (red and white) as a function of concentration

 Table 6: Removal Percent of Methylene Blue by the Magnetic Peanut

 Hulls (Red and White) as a Function of Dosages



Figure 13: Removal percent of methylene blue model solution by the magnetic peanut hulls (red and white) as a function of dosages

NT.	TT	Magnetic Peanut	Hulls (Red)	) Magnetic Peanut Hulls (White)				
No. pr		Removal %	± STD	Removal % ± STD				
1	4	66.23	± 0.75	$60.86 \pm 0.02$				
2	6	77.00	± 0.02	$70.92~\pm~0.05$				
3	7	97.55	± 0.01	$96.86 \pm 0.03$				
4	8	91.81	± 0.02	$88.94 ~\pm~ 0.05$				
5	10	87.98	± 0.02	$81.08~\pm~0.06$				
Exper	rimenta	l condition						
Conce Stirrir Weigl	entration ng rate ht of do	$\begin{array}{ll} \text{pn of dye} &= 20 \text{ ppm} \\ &= 200 \text{ rpm} \\ \text{psage} &= 0.2 \text{ g} \end{array}$	Tempera Contact	ature $= 30 \ ^{\circ}\text{C}$ time $= 120 \ \text{min}$				
	100	Г						
	95	-		$\sim$				
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	pH							

Table 7: Removal Percent of Methylene Blue by the Magnetic PeanutHulls (Red and White) as a Function of pH

Figure 14: Removal percent of methylene blue model solution by the magnetic peanut hulls (red and white) as a function of pH

Table 8: Remova	l Percent	of Methy	lene Blue	by the	Magnetic	Peanut
Hulls (F	led and W	'hite) as a I	Function of	f Stirring	g Speeds	

No.	Stirring	Magnetic Peanut Hulls (Red)	Magnetic Peanut Hulls (White)
	Speed (rpm)	<b>Removal % ± STD</b>	<b>Removal % ± STD</b>
1	50	$87.52 ~\pm~ 0.15$	$85.20~\pm~0.12$
2	100	$93.18~\pm~0.05$	$92.66~\pm~0.05$
3	150	$93.37~\pm~0.06$	$93.30~\pm~0.71$
4	200	$97.55~\pm~0.03$	$96.86~\pm~0.10$
5	250	$95.21~\pm~0.06$	$94.06~\pm~0.06$

Experimental condition	l		
Concentration of dye	= 20 ppm	Temperature	= 30 °C
pH	= 7	Contact time	= 120 min
Weight of dosage	= 0.2  g		



Figure 15: Removal percent of methylene blue model solution by magnetic peanut hulls (red and white) as a function of stirring speeds



 Table 9: Removal Percent of Methylene blue by Magnetic Peanut Hulls

 (Red and White) as a Function of Temperatures

Figure 16: Removal percent of methylene blue model solution by magnetic peanut hulls (red and white) as a function of temperatures

#### Conclusion

In this research, the prepared magnetic peanut hulls (red and white) were used as materials to remove the dyes from wastewaters. Removal of methylene blue by the prepared magnetic peanut hulls (red and white) were studied using model solutions of methylene blue.

In this work, the magnetic peanut hulls biomaterial was prepared by treatment with ferrofluid to remove colour. Characterization of the prepared magnetic peanut hulls was carried out by EDXRF, FT IR and SEM The composition of the magnetic peanut hulls which was techniques. determined by EDXRF, showed that calcium and iron were the main component in the magnetic peanut hulls. FT IR spectra showed the presence of ionizable groups such as carboxyl, carbonyl and hydroxyl in both sample magnetic peanut hulls (red and white). SEM micrographs indicated the porous nature of the surface. Therefore dyes can enter the porous of the The adsorption efficiencies of methylene blue solution on the samples. magnetic peanut hulls (red and white) were investigated as a function of the contact times, concentrations, dosage of samples, pH, stirring speeds and temperatures. The highest removal efficiency of MB by magnetic peanut hulls red (97.55 %) and white (96.86 %) were found at 120 min of contact times. The optimum concentration of MB was 20 ppm for the colour removal by both sample red (97.55 %) and white (96.86 %). The optimum dosages of both samples were observed to be 0.2 g. The optimum pH value for removal efficiencies of both samples were observed as pH 7. The optimum stirring speed and temperature were observed to be 200 rpm and 30 °C for both red and white magnetic peanut hulls samples. Magnetic peanut hulls (red) showed higher colour removal efficiencies than white sample.

Peanut hulls are readily available and relatively cheaper than synthetic resins. Therefore magnetically modified peanut hulls can be used to reduce the impacts of industrial water pollution on the aquatic environment.

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