

## EFFECTIVENESS & ECONOMY OF SAWDUST WOOD ADSORBENTS IN REMOVING ANIONIC DYES OF AQUEOUS SOLUTIONS

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### ABSTRACT

Adsorption of methyl orange (MO) from the aqueous solution on a low-cost adsorbent made of sawdust wood (SW) is studied experimentally. In the current research, a comparative study has been conducted between the natural sawdust wood (SW) and the activated sawdust wood (ASW). Conditions of experimental study were included the followings: 10 ppm of methyl-orange, pH=6 and a dosage in the range of 0.5 - 2 gm for different sizes of sawdust. During the study, Fourier-transform infrared spectroscopy-(FTIR) as well as (UV/V) is spectroscopy techniques were used. Results obtained indicate that the removal efficiency of methyl orange at  $25 \pm 3$  C exceeded 38.7%. A good fit for equilibrium data was obtained as per Langmuir model ( $R^2 > 0.99$ ). Maximum adsorption capacity of MO on ASW is 1.8 mg g<sup>-1</sup>. Results analysis of the activated sawdust waste show more a potential effectiveness as cheap adsorbent for the dyes removal from industrial wastewater.

**Keywords:** Industrial Water; Dyes Treatment; Adsorbent Low-Cost Sawdust

### 1. INTRODUCTION

Amongst the many shapes of global pollution found today, water pollution rates as the main one. It had been estimated by the World Bank that industrial water pollution caters approximately for 20% of world water pollution. Many researchers' attention had been attracted to this manifestation. The main sources of water pollution include: Industrial activities such as dye industries, coal, textiles, food, plastics, paper, pharmaceutical and petrochemical industries. Agricultural activities too contribute through the followings: Pesticides spread in farming and forestry in general, aquaculture and veterinary drugs, municipal wastewater as well as other global and environmental changes [Silva et al., 2016, Machado et al., 2014, Zhou. et al., 2015]. Even if having degraded concentrations, massive threats lie within these environmental contamination sources by the toxic effects they acquire, teratogenicity, carcinogenicity and mutagenicity [Yagub et al., 2014, Alayati et al. 2016, Atout et al., 2017]. Dyes of many kinds are employed in different fields of industry to add colours to products such as cosmetics, plastics, rubber and textiles, among many other products. The wastewater of such products has a considerable harmful content of synthetic dyes which are disposed of in river streams and into the environment in general with no effective treatment.

Due to their high stability, dyes demand temperatures as high as in excess of 200 °C to decompose [Machado et al., 2014, Pereira et al., 2010, Rafatuallh et al., 2009]. This makes dyes very hazardous and capable of inflicting sever harms. Dyes are also resistant to heat, biological degra-

ation and light by their molecular structure. Most conventional methods had been applied to get rid

of organic dyes in waste water treatment. These methods are either physiochemical or biological. The first type includes oxidation and filtration destruction via UV/Ozone treatment, flocculation, ion membrane exchange separation, floating and electrocoagulation, while the second type takes the form of anaerobic treatment [Carvalho et al., 2015, Gozmen et al., 2009, El-Ashtoukhy et al., 2015, Nawaz et al., 2014, Sathian et al., 2014]. Anyway, whenever the wastewater streams are ample, such treatments demand great costs. Adsorption is one of the dye removal methods that had acquired increasing consideration by virtue of its potential effectiveness and its additional advantages like being reusable, high selectivity at molecular level, economy as per energy demands, flexibility in dealing with many chemicals, immunity towards toxic constituents and ease of handling [Yagub et al., 2014, Tapia et al., 2016]. Adsorption is a wastewater treatment method of physical-chemical mechanism wherein the dissolved molecules get attached to an adsorbent surface by chemical and physical properties. There are a variety of interactions available to perform according to dyes' origins and nature of adsorbents. These interactions include van der Waals forces & electrostatic interactions [Ebrahimi et al., 2013, Attallah et al., 2013]. Provided that a proper choice of adsorbent is made, dyes and pollutants of mineral, organic and/or anionic nature can be efficiently removed from both industrial and waste water via the process of adsorption. Some of the suggested adsorbents are: jackfruit peel [Foo et al., 2012],

orange peel [Azouaou et al. 2010], coffee grounds [Ardejani et al., 2007], pine bark [Argun et al., 2009], corn silk [Ahmad et al., 2012] and rice husk [El-Shafey et al., 2010]. These inexpensive agricultural substances are either available in abundance or are some certain waste products from agricultural and/or industrial activities. Therefore, it is necessary to prepare an easily available, low-cost, equally effective alternative as an adsorbent intended for wastewater treatment. Since it is produced as a solid waste in large quantities at sawmills, sawdust wood is a potential candidate here. It primarily contains cellulose and lignin. Recently, many studies reflected interest in the use of sawdust to remove heavy metals ions from aqueous solution via adsorption. Due to considerably recognized findings obtained so far, researchers have been encouraged in this matter [Perieira et al., 2010, Rafatiualh et al. 2009, Arigun et al., 2009, Ahmad et al., 2012, ElShaifey et al., 2010, Djilali et al., 2016]. In the current research, the sawdust wood is recognized as being environment-friendly, cost-effective, and locally available. Sawdust wood has the feature of an adsorbent material that can remove dyes from methyl orange solution; that is why it is widely used in leather processing industries. Effects of different parameters such the adsorption dose, contact time, and particle size were studied in this work.

**1.1 Adsorption Isotherms:** It is for clarifying the interaction of adsorbate with adsorbents as well as further estimating the capacity of adsorbents that the absorption isotherms are used. In the literature; there are different isotherm models with Freundlich and Langmuir models receiving the top priority amongst all. Langmuir isotherm theory assumes monolayer coverage of adsorbates over a homogeneous adsorbents surface [Ardejani et al., 2007, Argun et al., 2009]. A rudimentary assumption is that adsorption occurs at specific homogeneous sites within the adsorbent and that no further adsorption can happen there once a dye molecule occupies that site. The Langmuir adsorption isotherm had offered a successful model for clarifying the process of removing dyes from aqueous solutions relying on adsorption [El-Shafey et al., 2010]. Eq. (1) can represent that.

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (1)$$

where  $q_e$  refers to the amount of solute at equilibrium ( $\text{mg g}^{-1}$ ),  $C_e$  is the solute concentration ( $\text{mg L}^{-1}$ ) at equilibrium,  $K_L$  and  $q_m$  refer to Langmuir constants related to the energy of adsorption and adsorption capacity ( $\text{mg g}^{-1}$ ), respectively [Boudrahem et al., 2015, Aydin et al., 2009]. The

equation can be rearranged to yield a linearized form:

$$\frac{1}{q_e} = \frac{1}{q_m K_L} \frac{1}{C_e} + \frac{1}{q_m} \quad (2)$$

Plotting the reciprocal of the concentration against the reciprocal of adsorption capacity results a straight line having a slope value of  $1/q_e$  and an intercept value. The main features of Langmuir isotherm may be depicted in terms of a constant that is dimensionless and which is called the Separation Factor  $R_L$ ; the value which can be calculated through this equation:

$$R_L = \frac{1}{1 + K_L C_o} \quad (3)$$

where  $K_L$  ( $\text{L/mg}$ ) represents the Langmuir constant and  $C_o$  ( $\text{mg/L}$ ) represents initial concentration relating to the adsorbate. The shape of the isotherm is indicated by the value of  $R_L$  which is considered: Linear when  $R_L = 1$ ; Favorable when ( $0 < R_L < 1$ ) or irreversible ( $R_L = 0$ ) and is considered unfavorable whenever  $R_L > 1$  [El-Geundi et al., 2012]. The Freundlich isotherm model explains the interaction between adsorbate molecules and adsorbents with multilayer adsorption on heterogeneous surfaces [Ahmad et al., 2012]. The Freundlich equation written below is an empirical equation for the adsorption isotherm:

$$q_e = K_F C_e^{1/n} \quad (4)$$

The following linearized equation is deduced through taking logarithms of the two sides of the equation:

$$\log(q_e) = \frac{1}{n} \log C_e + \log K_F \quad (5)$$

There is a relation of relevance between  $K_F$  of the Freundlich expression and the adsorbate's adsorbent capacity;  $1/n$  depends upon the adsorption strength, simultaneously indicating affinity between the adsorbate and the adsorbent. When  $1/n$  is less than 1 the process of adsorption is chemical for sure. Alternately; the process is physical more than any whenever  $1/n$  is more than 1. The value of  $1/n$  approaches zero more and more whenever the surface is more of a heterogeneous one [Ahmad et al., 2012].

## 2. Experimental Procedures

**2.1 Adsorbent and characterization:** The natural sawdust wood used in the present study was collected from Iraqi local carpentry workshops. The collected sawdust wood (SW) was washed with distilled water many times so as to remove water-soluble materials and dirt particles. The washing step was sustained until no color was

observed in the wash water. The eroded materials were then thoroughly dried in an oven at a temperature range of 80-110 °C. The dried sawdust was sieved after grinding; using (Besmak sieves from 2.36mm to 300µm). It was dried afterwards in an air circulating oven and had been stored in a desiccator until it was time to be used. The cleaned sawdust wood was carried to a furnace (Tianjin Taisite Instrument CO., LTD) to be burned completely at 300°C and it was cooled after that. The activated sawdust wood (ASW) used in this research had been washed using distilled water to remove water soluble materials that might be present in the sawdust prior to the adsorption study. It was then sieved to (300-2054) µm size and was used without any further treatment.

Figure 1a,b shows the sawdust before and after burning.

**2.2 Adsorbate and analytical measurements:** All materials and reagents used in this research are of an analytical grade; distilled water was used for preparing the synthetic dye solution. The anionic dye of MO was supplied by laboratory of the university. Figure 2 provides the chemical structure of MO. NaOH (98% purity) and HCl (37% purity) were purchased from India. The concentration of methyl orange in the aqueous solution had been determined at maximum absorption wavelength (464.5 nm) while using a UV–spectra meter (UV-1800 Shimadzu, Japan) spectrophotometer connected to a PC. The pH measurements were made using pH meter (Model 2906, Jenway Ltd, UK).

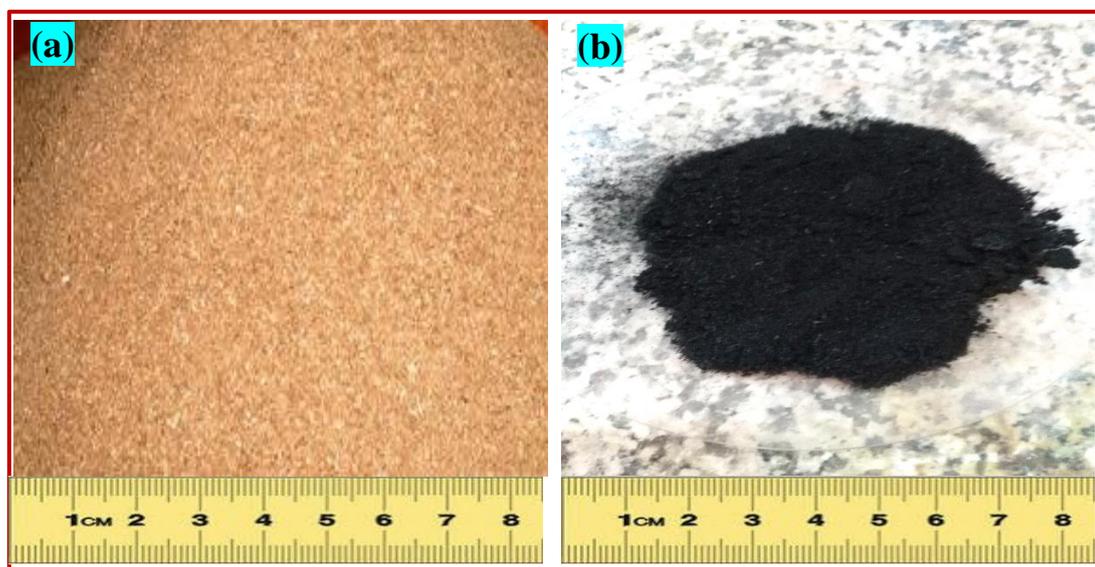


Figure 1: The sawdust waste (a) before burn (b) after burn.

**2.3 Batch Adsorption Procedure:** A series of stoppered reagent bottles were used to implement experiments for batch adsorption. 0.5 g of an adsorbent had been weighed prior to introduction into the reagent bottles that contained various concentrations of MO.

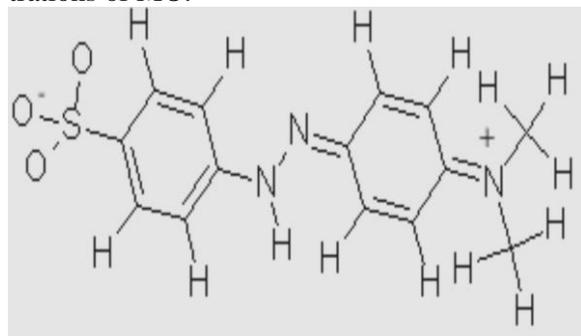


Figure 2: Discription of the structure of methyl orange.

An aqueous solution of 250 ml volume had been used within the research. For the sake of

reaching the desired figure of pH, a selection of either HCL or NaOH has been made. Next to that, a magnetic stirrer was employed to shake the bottles at room temperature ( $25 \pm 3$  °C) for a specified time to obtain a state of equilibrium. The solutions were filtered next to that and a UV–VIS Spectrophotometer was used to assess the final MO concentrations.

The initial concentration (10 mg/L), contact time (30–120 min), solution pH is 6 and adsorbent concentration (0.5–2 g) had been studied. The amount of MO at equilibrium ( $q_e$ ) was calculated from the mass balance equation given as below [Liu et al., 2017]:

$$q_e = \frac{V(C_0 - C_e)}{M} \quad (6)$$

where  $q_e$  (mg/g) is the amount of methyl orange per mass unit of watermelon adsorbents at certain time  $t$ ,  $C_e$  and  $C_0$  (mg/L) are as mentioned

above with (at time  $t$ ) and initial concentrations of methyl orange, respectively,  $M$  is the mass of watermelon adsorbent (mg) and  $V$  is the volume of the solution (mL). The percent MO removal via sawdust wood (Adsorption %) had been assessed for each equilibration using the expression presented as:

$$\text{Adsorption \%} = (C_0 - C_t)/C_0 \times 100 \quad (7)$$

### 3. RESULTS AND DISCUSSION

**3.1 FTIR Analysis:** FTIR spectrum in the range of 500-4000  $\text{cm}^{-1}$  is shown in Fig.3. An FT-IR apparatus type shimadzu (4000-400 $\text{cm}^{-1}$ ) was used to identify the functional groups and structural details in the SW that might be involved in the adsorption processes. FTIR analysis was carried out to identify the functional groups in the sawdust wood that might be involved in the adsorption process.

Figure 3 shows the FTIR analysis pertaining to SW before the adsorption process for varied sizes. The sawdust's complex structure is manifested via

the peaks given in Figure 3a for 2054  $\mu\text{m}$ . -OH or hydroxyl groups are referred to by the significant peak around 3383.26  $\text{cm}^{-1}$  while the presence of C-H bond is regarded as the cause for the bond at 2933.83  $\text{cm}^{-1}$ . Additionally, both peaks of 1660.77  $\text{cm}^{-1}$  and 1732.13 refer to the carbonyl group -C=O. On the other hand, the aromatic rings are represented by the 1510.31  $\text{cm}^{-1}$  whereas 1371.43 and 1427.37  $\text{cm}^{-1}$  are associated with the -CH<sub>3</sub> and C-O in phenols, respectively. Peaks indicating the complex structure of sawdust are shown in Figure 3b for 300  $\mu\text{m}$  sawdust. The hydroxyl groups -OH are represented by the significant peaks around 3363.67  $\text{cm}^{-1}$  while the presence of C-H bond is considered as the cause for the bond at 2918.4  $\text{cm}^{-1}$ . Alternately, carbonyl group -C=O are referred to by the peaks of 1734.06 and 1658.84  $\text{cm}^{-1}$ . Aromatic rings are represented by 1548.89  $\text{cm}^{-1}$  whereas both 1373.43 and 1425.44  $\text{cm}^{-1}$  are associated with the -CH<sub>3</sub> and C-O in phenols, respectively.

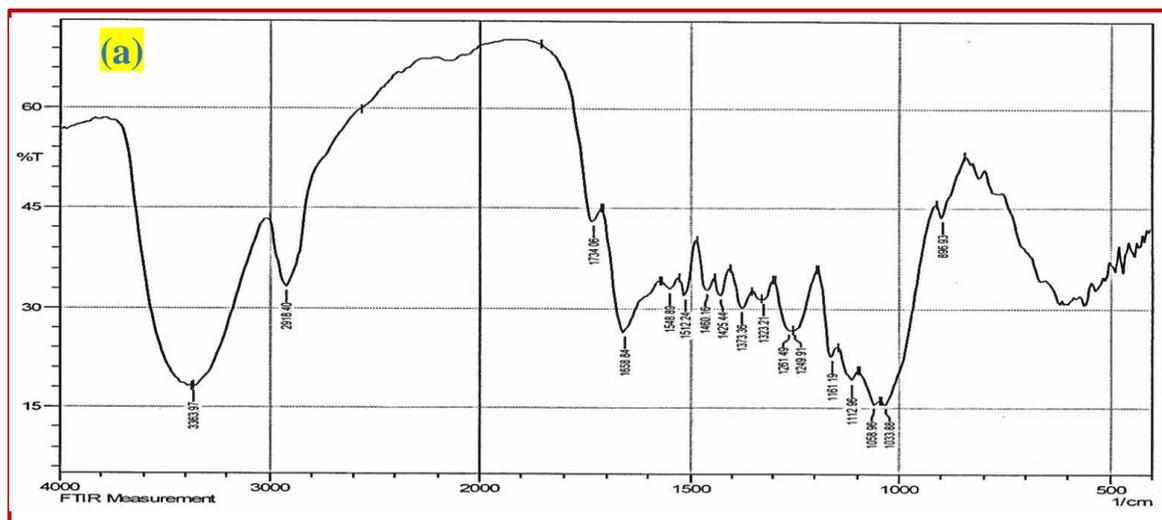


Figure 3: (a) FTIR analysis of natural sawdust waste before adsorption for 2.054 mm.

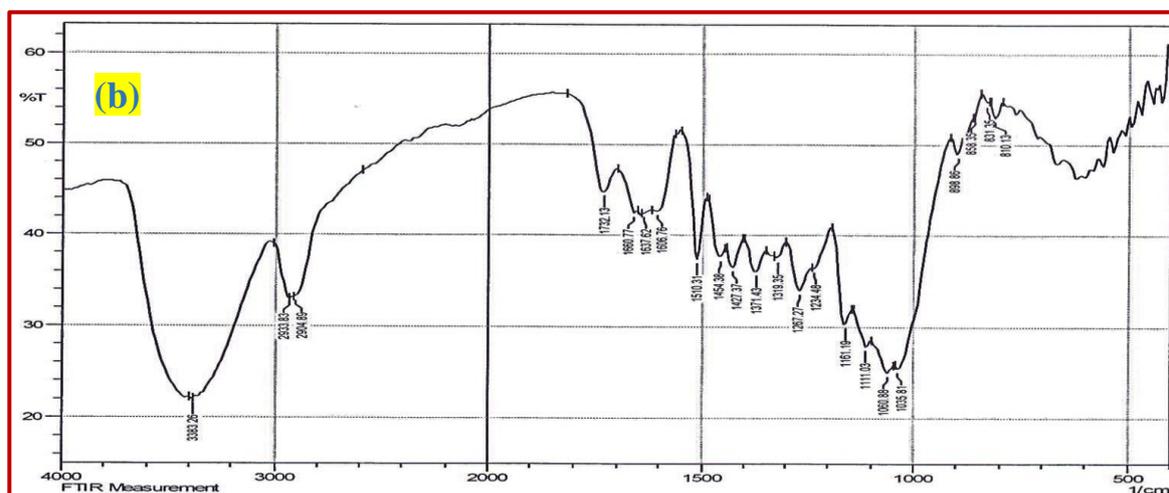


Figure 3: (b) FTIR analysis of natural sawdust waste before adsorption for 300  $\mu\text{m}$ .

### 3.2 Effect of contact time on adsorption process:

In order to assess equilibrium time, the research carried out adsorption for MO onto both sawdust wood-(SW) and activated sawdust wood-(ASW) adsorbents maintaining optimal concentration; the adsorption being function of contact time here. As evident from Figures 4 and 5, the adsorptions of MO for two adsorbents is slow in the first 30 min of contact time. Right after that, it took 90 min for the equilibrium time to be reached in a gradual manner as per SW adsorbent while ASW adsorbent took 120 min to reach equilibrium. That is to say; an increase in percentage MO removal

of 8% (from 30% to 38%) had been achieved through increasing contact time from 30 min to 120 min for SW adsorbent, whereas for ASW adsorbent the achieved increase was 10% (from 50% to 60%). As evident from Figure 5, the percentage removal of ASW is greater than it for SW adsorbent. The rapid percentage removal observed at the lower time is ascribed to availability of empty reactive site on the surface of adsorbents, whereas at higher time, the changes of percentage removal drops by both slow pore diffusion as well as adsorbents' saturation [El-Shafey et al., 2010, Agarwal et al., 2016].

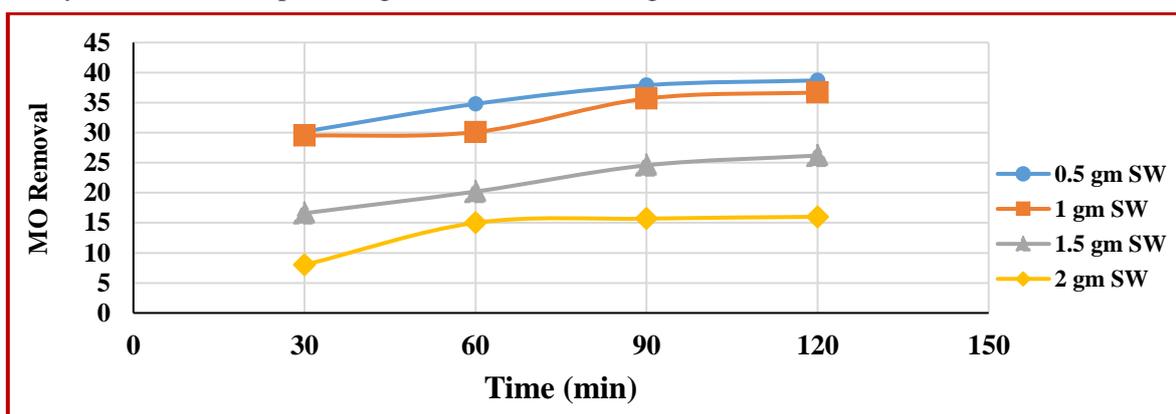


Figure 4: Effect of contact time on MO removal ( $C_0=10$  mg /L, pH 6.0, agitation speed: 300 rpm, temperature:  $27\pm 3$ oC, 2.054 mm).

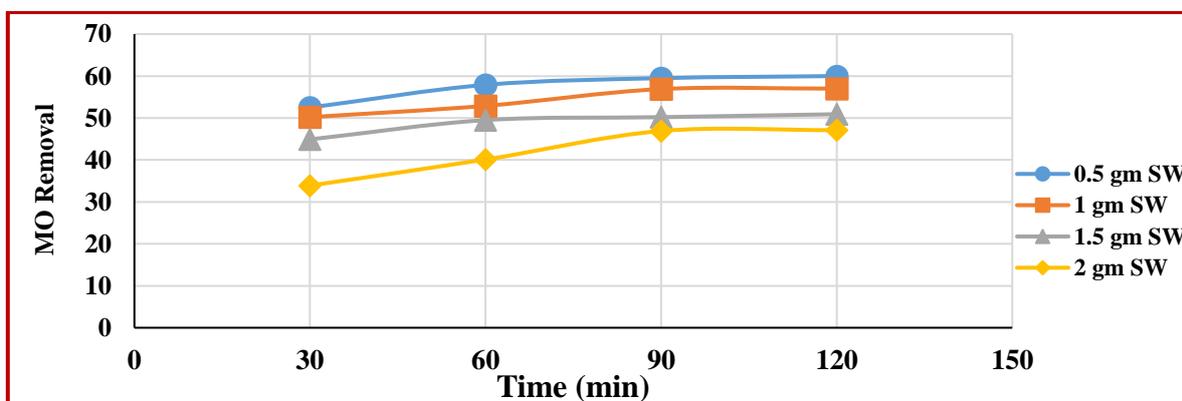


Figure 5: Effect of contact time on MO removal  $C_0=10$  mg /L, pH 6.0, agitation speed: 300 rpm, temperature:  $27\pm 3$ °C, 2.054 mm.

### 3.3 Effect of Adsorbent Dose on Adsorption:

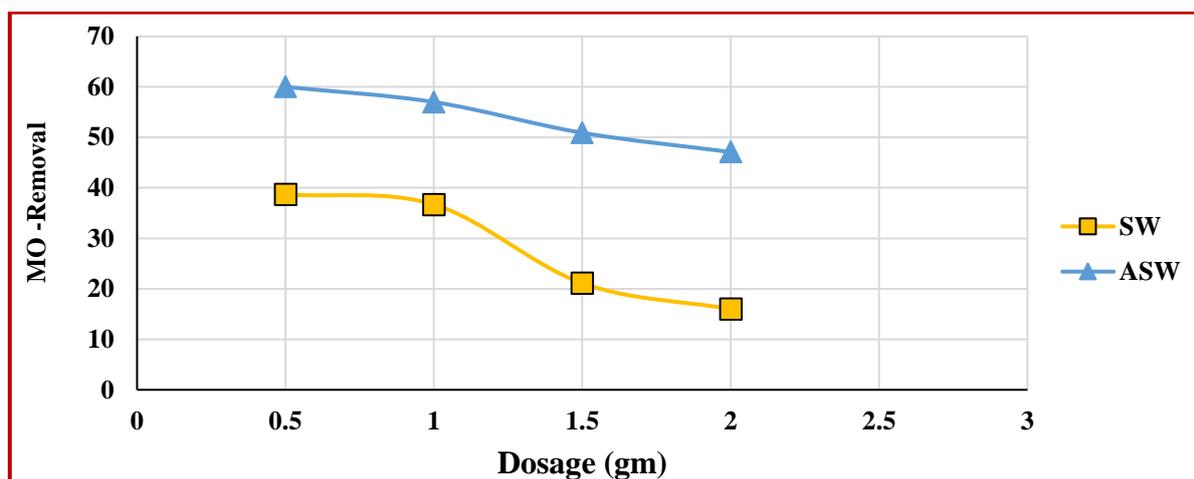
The mass of sorbent was varied in the range of 0.5–2 g for the removal of MO from aqueous solution by SW and ASW. Adsorbent dose is an important parameter because it determines the capacity of an adsorbent for a given initial concentration of adsorbate. The effect of adsorbent dose was studied with MO dye removal keeping all the experimental conditions constant. The adsorption of MO by SW and ASW at different adsorbent doses for 10 mg/L of methyl orange concentration at pH 6 was studied. The results given in figure 6a show that as the adsorbent' mass increases from 0.5 to 1.5 gm, the percent MO adsorption increase from 16 to 38.7% for sawdust and from 47.1 to 60

% for activated sawdust as shown in the same figure. However, any increase in dose of adsorbent causes a counter decrease in the amounts of dye adsorbed per unit mass of adsorbent. A sorption process causing unsaturation of the adsorption would result due to increase in adsorbent dose at constant dye volume and concentration. In cases where concentration ratios of SW to solute are considerably high; a fast-superficial adsorption onto the adsorbent surface is there such that it produces a lower concentration of solute within the solution than for lower adsorbent to solute concentration ratio. The reason here is due to only a certain quantity of dye being adsorbable by a fixed mass of SW. Thus, the greater the adsorbent dos-

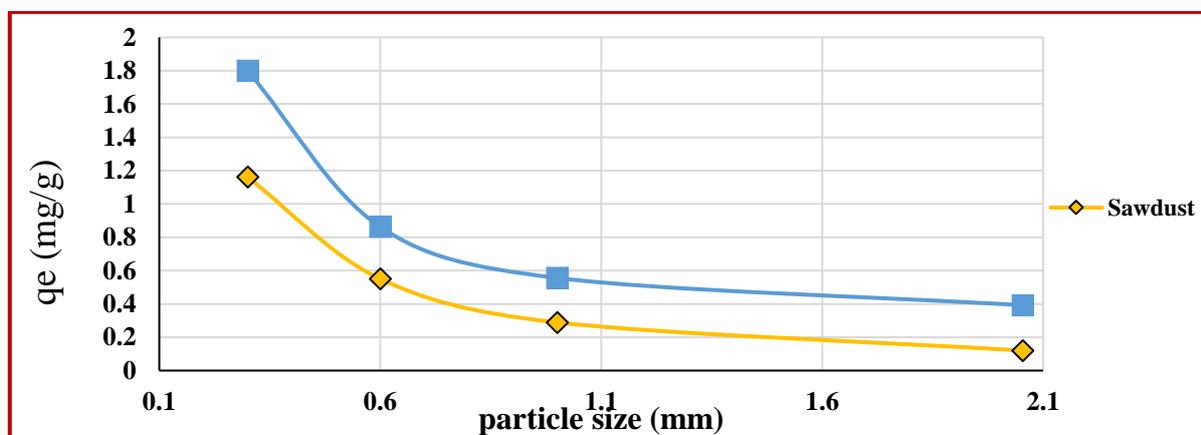
age is, the greater gets the volume of effluent which can be purified by a fixed mass of adsorbent.

The drop in the quantity pertaining to MO adsorbent found for the increase in mass of adsorbent is attributed to the gradient of concentration between the solute concentration and the (on the surface of the adsorbent) or to the split in the flux. Therefore, the adsorbed quantity of dye by the adsorbent's unit weight is made to drop for an increased

dosage of adsorbent. This causes adsorption capacity to drop down as a result of increasing the dosage of adsorbent; this is clearly depicted in Fig 6b. Moreover, the decrease in adsorption can be ascribed to aggregation or overlapping of adsorption sites which leads to limiting total sorbent surface area that is there for dye molecules as well as resulting into increasing length of diffusion path. However, the adsorption capacity showed a decreasing trend with increasing adsorbent dosage.



**Figure 6a:** Effect of dosage on MO removal on SW ( $C_0 = 10$  mg/L, pH 6.0, agitation speed: 300 rpm, temperature:  $25 \pm 3^\circ\text{C}$ ,  $2054 \mu\text{m}$ , and 120 min).



**Figure 6b:** Effect of contact time on adsorption capacity of MO dye (pH 6, 120 min contact time  $C_0 = 10$  ppm).

**3.4 Effect of Particle Size:** For the sake of carrying out batch adsorption experiments, different particle sizes of an adsorbent ( $300 \mu\text{m}$  to  $2054 \mu\text{m}$ ) had been used at an initial concentration of  $10 \text{ mg/L}$  pH 6.0. Decreasing the particle size led to increasing MO removal. It is evident from Figure 7 that minimum particle size reflected greater adsorption in comparison with larger sizes. The surface area for adsorption is increased due to adsorbents having small size. This inverse proportionality between particle size and adsorption capacity

advocates the idea that the dye did not penetrate the particles. Rather instead, the dye adsorbed on the outer surface preferentially which is probably because of steric hindrance of large dye molecules. Efficiency of MO removal increased gradually with the increase in adsorbent particle size reaching its maximum at particle size of  $0.3 \text{ mm}$  (49%) and (75%) for SW and ASW respectively. These observations agree with that obtained by [Abdelwahab et al., 2017].

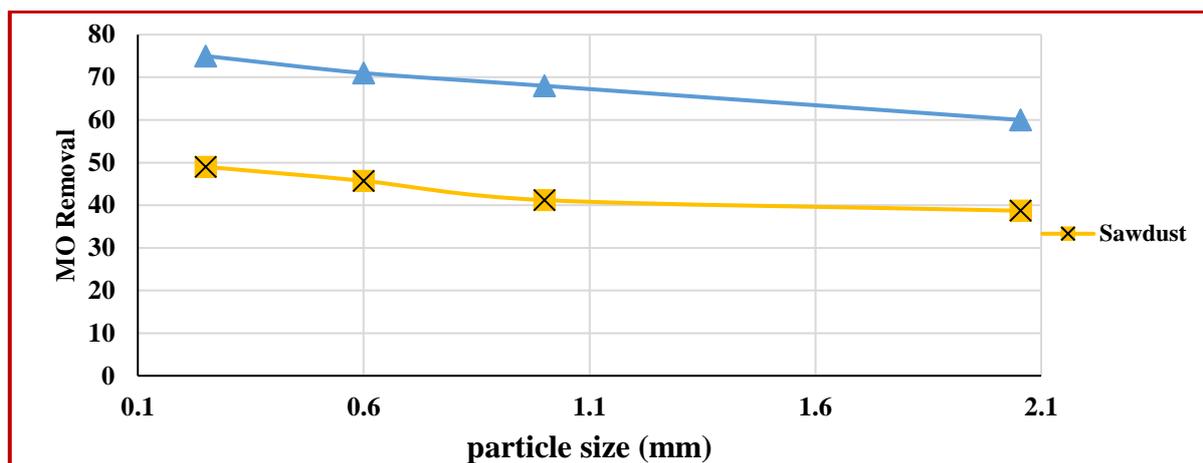


Figure 7 Effect of adsorbent particle size on MO removal ( $C_0$ : 10 mg /L, pH 6.0, agitation speed: 300 rpm, temperature:  $27 \pm 3^\circ\text{C}$ , 120 min, 0.5gm dosage).

**3.5 Adsorption Isotherms:** The Freundlich and Langmuir isotherm equations were applied with adsorption data pertaining to SW and ASW employed in both equations. The Langmuir maximum adsorption capacities for ASW and SW had been found as 1.8 and 1.16 mg/g, respectively. The maximum dye adsorption capacity pertaining to ASW adsorbent samples had been found to be

around 150% higher than that pertaining to MO samples. Apparently, the Langmuir model best fits the experimental results over the experimental range with good coefficients of correlation ( $R^2 > 0.96$ ). This result resembles that of [Mohammed et al., 2016]. Figures 8 and 9 show the Langmuir model for sawdust and activated sawdust wood.

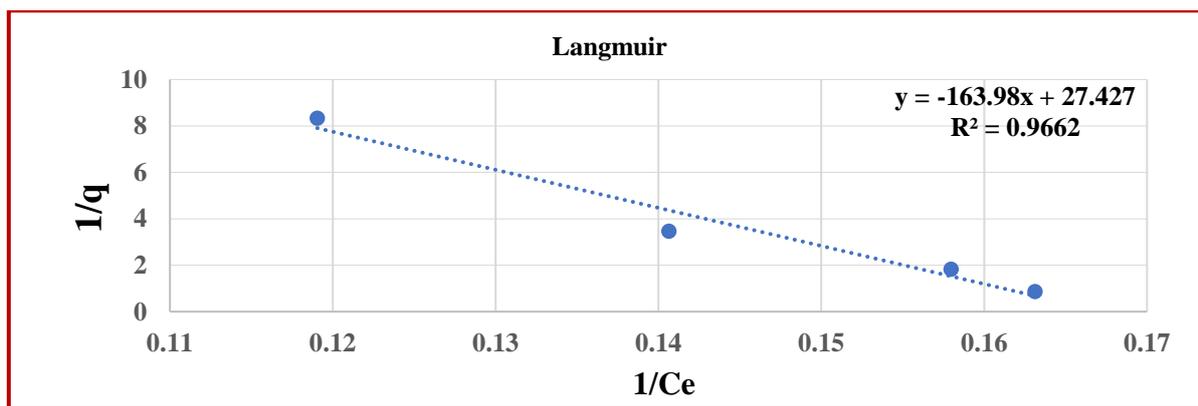


Figure 8: Langmuir isotherme plot for adsorption of MO on SW sample.

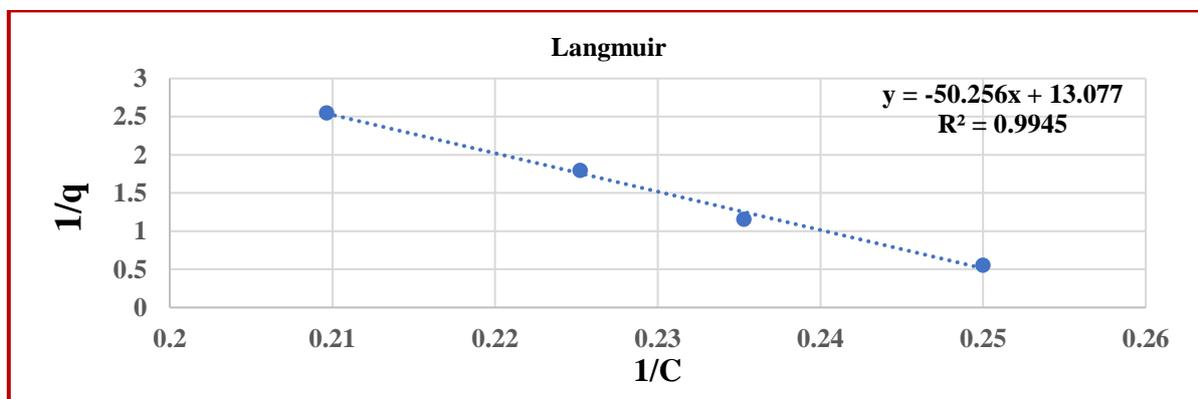


Figure 9: Langmuir isotherm plot for adsorption of MO on ASW sample

Table 1 gives the values of linear correlation  $R^2$ ,  $q_m$  and  $K_L$ .

**Table 1:** Isotherm parameters for MO adsorbed onto ASW and SW samples obtained by using Langmuir model

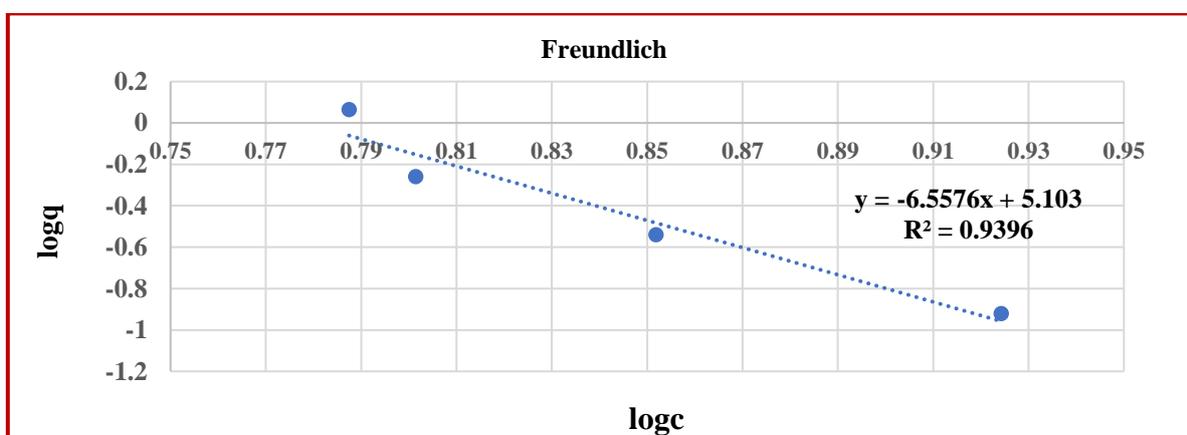
SAMPLE	QM	KL	R <sup>2</sup>
ASW	0.08	0.96	0.995
SW	0.0365	1.01	0.966

1/n and KF are Freundlich constants which are both related to adsorption intensity of the sorbent and adsorption capacity, respectively. The plot of logCe versus log q in Figure 10 for SW and Figure 11 for ASW is employed to evaluate the slope 1/n and the intercept KF. The values of n, and KF as well as a linear correlation R<sup>2</sup> with Freundlich isotherms are all given in Table 2.

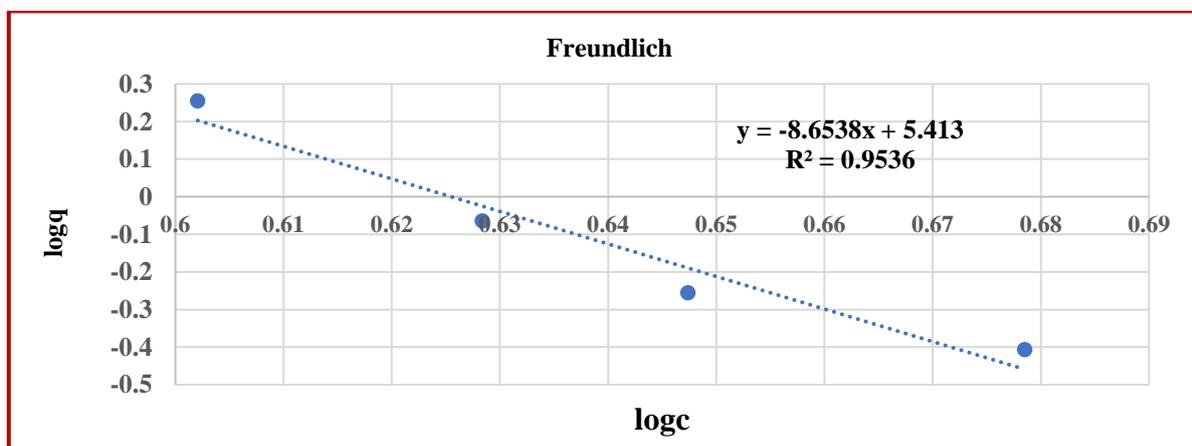
The best fit exhibited R<sup>2</sup> = 0.934 for SW and R<sup>2</sup> = 0.95 for ASW. The value of n was -0.15 for the SW sample and 0.16 for the ASW sample. Because of that and speaking about the whole concentration range of the researched red dye; the adsorption property of ASW proved to excel over that of SW. The KF value of the Freundlich equation (Table 2) indicated that ASW excelled over SW in matters of adsorption capacity.

**Table 2:** Isotherm parameters for MO adsorbed onto ASW and SW samples obtained by using Freundlich model.

SAMPLE	N	K <sub>F</sub>	R <sup>2</sup>
ASW	0.11	5.413	0.95
SW	0.15	5.103	0.94



**Figure 10:** Freundlich isotherm plot for adsorption of MO on SW sample.



**Figure 11:** Freundlich isotherm plot for adsorption of MO dye on ASW sample.

#### 4. Conclusions

The research proposed an economic and efficient procedure to remove dyes within treatment of wastewater using adsorption on a cheap material that occurs naturally. Adsorption is a successful option to remove anionic dyes due to being easy in handling and due to adaptability to varying conditions of wastewater. The values of removal efficiency were found to be around the figure of 60%. This depends upon solution comp-

osition, type of adsorbent and other adsorption conditions. Sawdust proved to be the best adsorbent for removal of dyes from aqueous solutions under certain physicochemical conditions. The adsorbents studied here are very effective in removing dyes from industrial wastewater. There is a direct proportionality between adsorbent dose and the percent MO removal while the correlation between size of adsorbent material and percent MO

removal reflects an inverse proportionality. The maximum adsorption can be obtained by using particle size of 0.3  $\mu\text{m}$  in neutral media (pH 6).

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