

# IMPROVING OF EFFLUENT SEWAGE BY TREATMENT WITH LOCAL MATERIALS

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## **Abstract**

Natural materials that are available in large quantities or certain waste from agricultural operations may have the potential to be used as low cost adsorbents, as they represent solid waste, widely available and are environmentally friendly after use it. The aim of this research is to evaluate the applicability of using one of these locally available materials (sawdust) to improve the sewage effluent quality.

The sawdust, low cost locally available material and solid waste, was tested as biosorbent for the removal of biochemical oxygen demand (BOD<sub>5</sub>) from effluent sewage. Adsorption of BOD<sub>5</sub> onto this low-cost adsorbent was studied by continuous mode system. The effects of initial BOD<sub>5</sub> concentration, flow rate, bed height, particle size, activated carbon/sawdust ratios and pH on adsorption capacity were studied. The experiments showed that the adsorption through the sawdust instead of commercial activated carbon was found to be very effective for the removal of BOD<sub>5</sub>, where the maximum removal percentage reached 86.23%.

## **1. Introduction**

Fresh water is already a limiting resource in many parts of the world. In the next century, it will become even more limiting due to increased population, urbanization, and climate change, thus leading to water crisis and serious consequences on the environment. This limitation is caused not just by increased demand for water, but also by pollution in freshwater ecosystems. Iraq is one of the countries that is suffering from water crisis and is threatened by it more and more during the next years. Pollution decreases the supply of usable water and increases the cost of purifying it, however, liquid and solid wastes produced by human settlements and industrial activities pollute most of the watercourses throughout the world.

Wastewater is essentially the water supply of the community after it has been used in a variety of applications. From the standpoint of sources of generation, wastewater may be defined as a combination of the liquid (or water) carrying wastes removed from residences, institutions, and commercial and industrial establishments, together with such groundwater, surface water, and storm water as may be present (**Metcalf and Eddy, 2003**).

Untreated wastewater generally contains high levels of organic material, numerous pathogenic microorganisms, as well as nutrients and toxic compounds. It thus entails environmental and health hazards and, consequently, must immediately be conveyed away from its generation sources and treated appropriately before final disposal. The ultimate goal of wastewater management is the protection of the environment in a manner commensurate with public health and socio-economic concerns.

The most widely used parameter of organic pollution applied to both wastewater and surface water is the 5-day BOD ( $BOD_5$ ). This determination involves the measurement of the dissolved oxygen used by microorganisms in the biochemical oxidation of organic matter. BOD test results are now used to (1) determine the approximate quantity of oxygen that will be required to biologically stabilize the organic matter present, (2) determine the size of waste-treatment facilities, (3) measure the efficiency of some treatment processes, and (4) determine compliance with wastewater discharge permits (**Metcalf and Eddy, 2003**).

In fact, the BOD values indicate the amount of biodegradable organic material (carbonaceous demand) and the oxygen used to oxidize inorganic material such as sulfides and ferrous iron. It also may measure the oxygen used to oxidize reduced forms of nitrogen (nitrogenous demand) unless their oxidation is prevented by an inhibitor. The BOD test has its widest application in measuring waste loading to treatment plants and in evaluating the BOD removal efficiency of such treatment systems (**Quevauviller et al., 2006**).

Biochemical Oxygen Demand or BOD as it is commonly abbreviated, is one of the most important and useful parameters (measured characteristics) that indicates the organic strength of a wastewater. BOD measurement permits an estimate of the waste strength in terms of the amount of dissolved oxygen required to break down the wastewater. The BOD test is one of the most basic tests used in the wastewater field. It is essentially a measure of the biological and chemical component of the waste in terms of the dissolved oxygen needed by the natural aerobic biological systems in the wastewater to break down the waste under defined conditions.

Sawdust is an abundant by-product composed of fine particles of wood. It is considered as an agricultural waste and a by-product of manufacturing industries and can easily be understood to be more of a hazard, especially in terms of its flammability and is used either as cooking fuel or as packing material. It is easily available in the countryside at zero or negligible price. Sawdust contains various organic compounds. It is composed of three major constituents of a wood cell (lignin, cellulose and hemicellulose), cellulose 40-50%, lignin 23-33% in softwoods; 16-25% in hardwoods, and hemicelluloses 15-25% (**Shaban et al., 2009, Batzias and Sidiras, 2007**). Sawdust has proven to be a promising effective material for the removal of dyes from wastewaters. Moreover, it is actually an efficient adsorbent that is effective to many types of pollutants, such as dyes, oil, salts, heavy metals (**Velizar et al., 2009**) and others. **Baral et al., (2006)** conducted studies on adsorption of hexavalent chromium by varying various parameters. The adsorption capacity was found to be pH dependant. Sawdust was found to be very effective and reached equilibrium in 3h (adsorbate concentration 30mg/l). Salts treatment of sawdust by (calcium chloride, zinc chloride, magnesium chloride and sodium chloride) enhanced the adsorption properties of the original material and since sawdust is an industrial waste/byproduct and the salts used can be recovered as spent liquids from various chemical operations, this process of adsorbent upgrading/modification might be considered to take place within an 'Industrial Ecology' framework (**Batzias and Sidiras, 2007**).

**Velizar et al., (2009)** presented the results of the batch and column adsorption of copper and some associated ions by employing linden and poplar sawdust as a low-cost adsorbent.

## **2. Experimental procedure and methods**

### **2.1 Source of Wastewater**

In the present study, the wastewater used was unchlorinated secondary effluent taken from Al-Maamera Wastewater Treatment Plant every day over the period of the experimental run. The desired amount of effluent was to be taken daily and fed into the column. Detailed analysis of raw effluent was conducted immediately to determine its physio-chemical properties.

### **2.2. Preparation of the adsorbent**

The raw sawdust used was collected from a local furniture manufacturing industry and sieved in the size ranges (0.15-0.4) and (0.85-1.15) mm. After collection and sieving, the sawdust was washed with distilled water to remove muddy materials and impurities and then dried in an electrical oven at 100-110 °C, the sawdust was then immersed in 2N NaOH aqueous solution for 8 h (Meena et al., 2004). It was observed that a dark-red solution was generated during this treatment, which indicated the removal of lignin from the adsorbent material. Thereafter, it was washed several times with distilled water to remove the lignin content and excess of NaOH. The sawdust was repeatedly washed with distilled water till no red coloration was observed. After washing, sawdust dried in an oven at 100-110 °C for 6 h (Bhattacharya et al., 2006). It was then immersed in 0.2N H<sub>2</sub>SO<sub>4</sub> for 8 h to remove the traces of alkalinity and other impurities. The acid-treated sawdust was again thoroughly washed with distilled water to remove the excess of H<sub>2</sub>SO<sub>4</sub> and other coloring materials till the wash-found water was colorless. After the wash water became colorless, the treated sawdust adsorbent material was dried at 100-110 °C and stored in a dessicator for use as an adsorbent.

Sawdust was treated with sodium hydroxide solution to increase the adsorption property of the adsorbent. Carboxylate ligands are believed to be responsible for metal binding by the biomasses. This means that metal binding can be enhanced by increasing the number of carboxylate ligands in the biomass. Cellulose, hemicellulose, and lignin, which are major constituents of most plant tissues, contain methyl esters, which do not bind metal ions significantly. However, these methyl esters can be modified to carboxylate ligands by treating the biomass with a base such as sodium hydroxide, thereby increasing the metal-binding ability of the biomass (Rehman et al., 2006).

Treatment of biosorbents with NaOH solution positively affected adsorption capacity because they can be considered as chemically activated, that is chemical activation is a single step process and is held in presence of dehydrating reagents such as KOH, K<sub>2</sub>CO<sub>3</sub>, NaOH, ZnCl<sub>2</sub> and H<sub>3</sub>PO<sub>4</sub> which influence pyrolytic decomposition and inhibit tar formation, the carbon yield obtained is higher and the temperature used in chemical activation is lower than that of physical activation (Deng et al., 2009).

### **2.3 Column studies**

Continuous flow adsorption studies were conducted in a glass column made of Pyrex glass tube of (0.8m) height and (0.05m) inner diameter. At the bottom of the column, a stainless sieve was attached followed by a layer of glass beads. A known quantity of the prepared sawdust was packed in the column to yield the desired bed heights of the adsorbent, and then an upper retaining sieve was inserted on top of the bed and firmly secured in place by layer of glass beads in order to provide a uniform flow of the solution through the column. The samples of solutions at the outlet of the

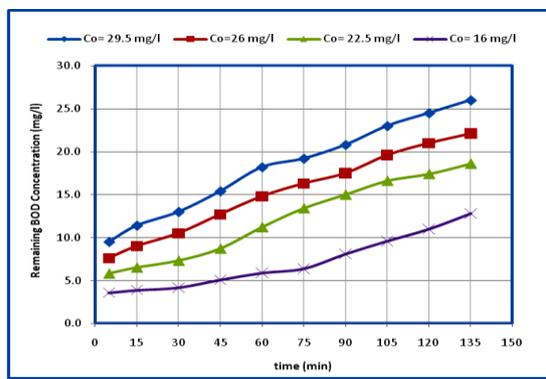
column were collected at regular time intervals and the concentrations were measured. All the experiments were carried out at room temperature ( $25 \pm 1^\circ\text{C}$ ).

### 3. Results and discussion

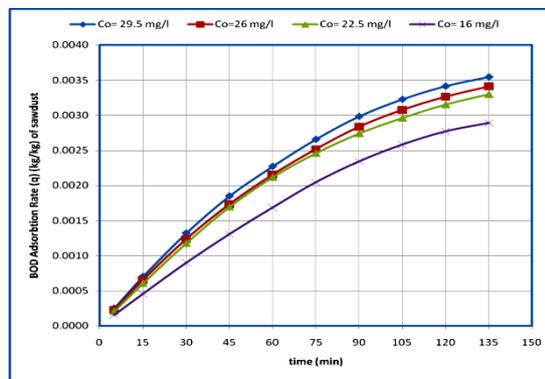
#### 3.1 The Effect of the Influent Concentration

Four experiments were carried out with different influent  $\text{BOD}_5$  concentrations of (29.5, 26, 22.5, and 16) mg/l respectively. The other initial conditions (temperature of  $25^\circ\text{C}$ , pH of 7.5, flow rate of  $3.33 \times 10^{-6} \text{ m}^3/\text{sec}$ , particle size of 0.15-0.4 mm, bed height of 0.2 m and  $\text{AC}/\text{SD}=0$ ) were kept constant.

The breakthrough curves for the above experiments were plotted in Fig.(1) by plotting the remained  $\text{BOD}_5$  concentration against time. This figure showed that the break point was reached fastly as the influent concentration increased.



**Figure (1):** breakthrough curves of isothermal adsorption for different  $\text{BOD}_5$  influent initial concentrations.

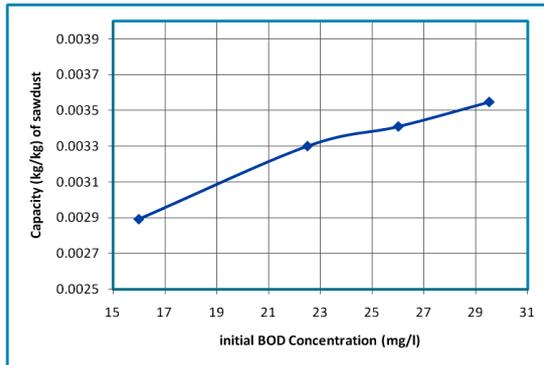


**Figure (2):** Adsorption rate for different  $\text{BOD}_5$  influent initial concentrations.

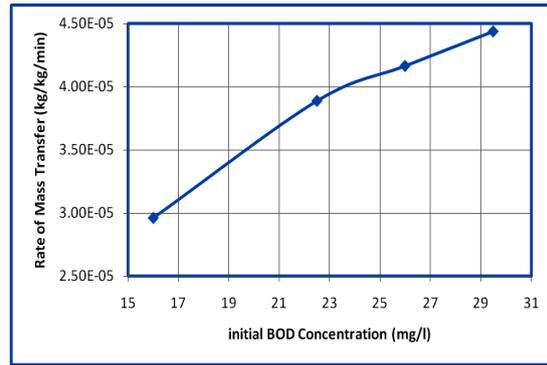
To show the adsorption rate with different influent concentrations of  $\text{BOD}_5$ , the solute adsorbed (the uptake) versus time were plotted in Fig.(2). This figure shows that the total quantity of solute removed from solution (adsorption rate) at any period of time increased with increasing influent  $\text{BOD}_5$  concentration which would be anticipated with the basis of the increased driving force for mass transfer with increased concentration of solute in solution. A linear portion exists for each curve at the early period of experiment. This means that only external mass transfer resistance (film diffusion) is rate determining step because there is no concentration gradients within the particle. With increasing influent concentration, the linear segment of the curves extends over a shorter period of time.

This observation coincides with the consideration that film diffusion controls as the rate limiting step until surface area becomes essentially saturated. Deviation from linearity occurs because of the increasing influence of interparticle transport on the overall rate of mass transfer as the experiments progress.

The effect of the influent  $\text{BOD}_5$  concentration on the adsorption capacity of the sawdust was shown in Fig. (3), by plotting the adsorption capacity (maximum value of each curve in Fig.(2)) versus the different influent  $\text{BOD}_5$  concentrations.



**Figure (3):** effect of the influent initial BOD<sub>5</sub> concentrations on the capacity of the sawdust.

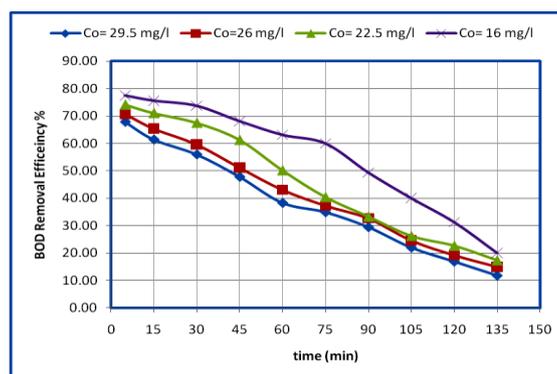


**Figure (4):** effect of the influent initial BOD<sub>5</sub> concentrations on the rate of mass transfer.

This figure showed that the capacity increased as the influent BOD<sub>5</sub> concentration increased. This was due the fact that as the influent concentration increases the concentration difference will increase too (i.e., the difference between the BOD<sub>5</sub> concentration in the bulk solution and the corresponding concentration on the surface of the adsorbent particles).

The rate of mass transfer for the initial stages of adsorption were obtained from the slopes of the linear portions of the curves in Fig. (2). The relation between mass transfer rate and BOD<sub>5</sub> concentration was represented in Fig.(4). This figure showed that the mass transfer rate increased with increasing influent BOD<sub>5</sub> concentration.

The removal yield showed a decreasing trend as the initial MB concentration was increased as shown in Fig. (5). At lower concentrations, all BOD<sub>5</sub> present in the adsorption medium could interact with the binding sites so higher adsorption yields were obtained. At higher concentrations, lower adsorption yields were observed because of the saturation of the adsorption sites.

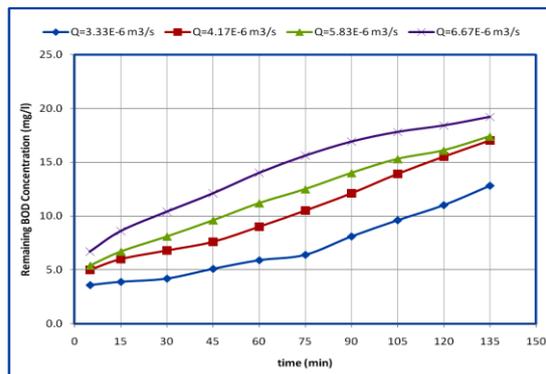


**Figure (5):** effect of the influent initial BOD<sub>5</sub> concentrations on the adsorption removal efficiency of sawdust.

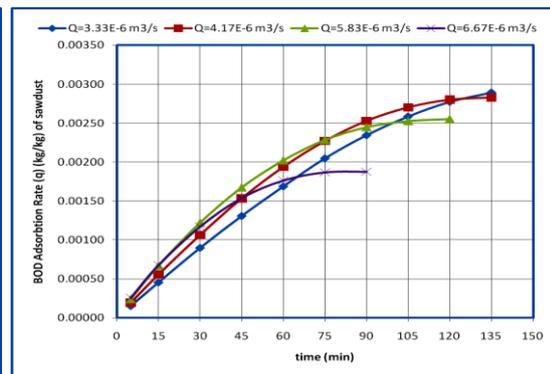
### 3.2 The Effect of Flow Rate

The effect of the flow rate on the adsorption of BOD<sub>5</sub> using sawdust was investigated by varying the flow rate ( $3.33 \times 10^{-6}$ ,  $4.17 \times 10^{-6}$ ,  $5.83 \times 10^{-6}$ , and  $6.67 \times 10^{-6}$ ) m<sup>3</sup>/sec, with constant bed height of 0.2m, initial BOD<sub>5</sub> concentration of (16-17)mg/l, pH solution of 7.5, particle size of (0.15-0.4)mm, temperature of 25 °C , and activated carbon/sawdust (AC/SD=0) .

The break through curves for the above experiments were plotted in Fig.(6). It can be seen from this figure that as the flow rate of wastewater increased, the break through point time of the curve decreased, and from these results the wastewater flow rate of ( $3.33 \times 10^{-6}$  m<sup>3</sup>/sec) was adopted for other experiments conditions.



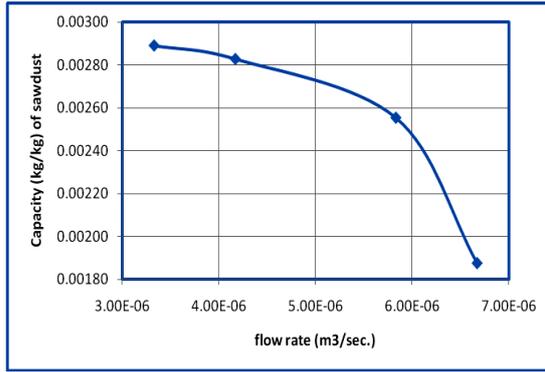
**Figure (6)** breakthrough curves of isothermal adsorption for different influent flow rates.



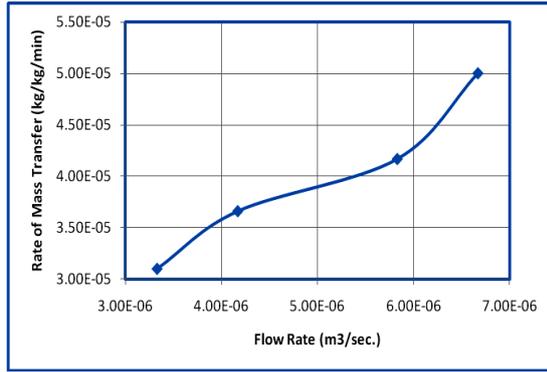
**Figure (7):** Adsorption rate for different influent flow rates.

Examining Fig. (7), the adsorption rate versus time. It can be seen that there is a marked increase in the rate of adsorption and the capacity of the sawdust with decreasing flow rate. Also it can be observed from the same figure that the lower flow rate the longer the linear portion of the curve, indicating that film diffusion remains rate limiting for longer periods. Increasing flow rate in this region causes reduction to the surface film, thereby, decreasing resistance to mass transfer. So deviation from linearity occurs because of the increasing influence of intraparticle transport on the overall rate of mass transfer when the external surface area becomes essentially saturated with solution.

The adsorption capacity (accumulative adsorption rate) of sawdust decreased as the flow rate of wastewater increased, as shown in figure (8). This was due to, a good available contact time which will affect the amount of the capacity, that for low flow rate the wastewater molecules would have a sufficient contact time to occupy the space within the particles.



**Figure (8):** effect of the influent flow rates on the capacity of the sawdust.



**Figure (9):** effect of the influent flow rates on the rate of mass transfer of BOD<sub>5</sub>.

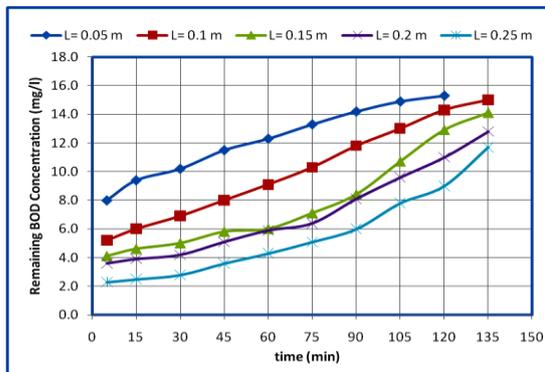
Figure (9) represented the rate of mass transfer for the initial stages of adsorption for different flow rates. This figure showed that the mass transfer rate increased as the flow rate of wastewater increased. Increased flow rate in this region may be expected to give a compression or reduction of the film thickness. Thereby decreasing resistance to mass transfer and increasing the mass transfer rate.

### 3.3 The effect of Bed Height.

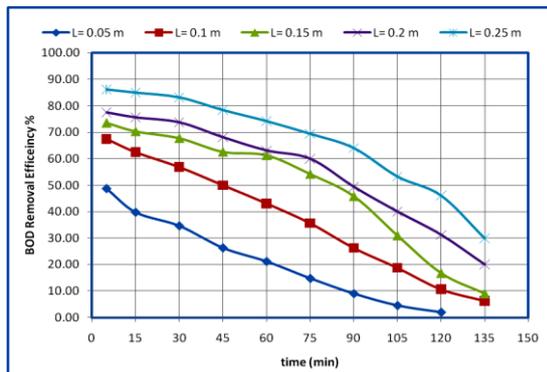
Five experiments, were carried out with different sawdust bed heights (0.05, 0.1, 0.15, 0.2, and 0.25) m. The other initial conditions were kept constant (particle size = 0.15-0.4 mm, BOD<sub>5</sub> initial concentration of (15.5-16.7) mg/l, flow rate of  $3.33 \times 10^{-6}$  m<sup>3</sup>/sec, pH of 7.5, temperature of 25°C, and activated carbon/sawdust ratio (AC/SD=0).

The breakthrough curves for the above experiments were plotted, (remaining BOD<sub>5</sub> against time for a given bed height) as shown in Fig. (10). Examining Fig. (10), it can be seen that the break point value increased as the bed height increased.

The effect of different bed heights on the removal efficiency of sawdust beds and the adsorption rates where shown in figures (11) and (12).



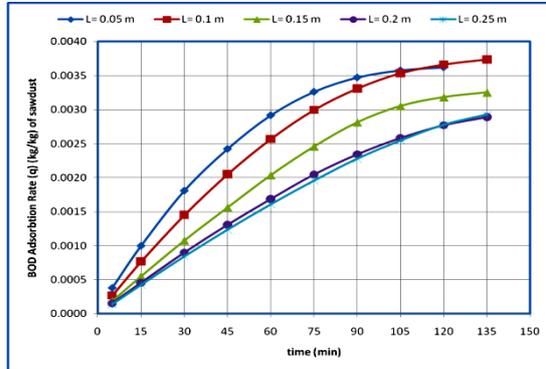
**Figure (10):** breakthrough curves of isothermal adsorption for different packed heights



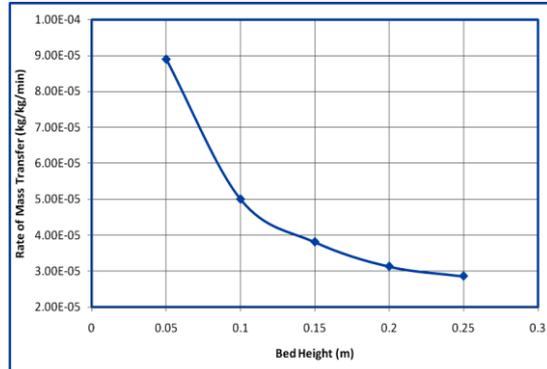
**Figure (11):** Adsorption removal efficiency for different packed heights.

The curves in fig.(11) showed that the removal efficiency increased with increasing bed height, and that at the end of the experimental time the removal efficiencies of the bed heights (0.05, 0.1, 0.15, 0.2, and 0.25)m were (1.92% at 120 min, 6.25%, 9.03%, 20%, and 29.94% at 135 min) respectively. The experiments

conducted with beds of 0.2 and 0.25 m could be continue for more time which leads to increase the adsorption rates as can be seen from fig.(12). The increasing of bed height will provide an extra surface area for the adsorption process to carry on. This means the service life of the sawdust bed increased with the increasing of the sawdust bed depth at constant flow rate of wastewater and constant linear velocity but the contact time will increase at bed depth increasing.



**Figure (12):** Adsorption rate for different packed heights.



**Figure (13):** effect of the bed heights on the rate of mass transfer of BOD<sub>5</sub>.

The increasing of the bed height will increase the capacity, because additional spaces will be available for the wastewater molecules to be adsorbed on these unoccupied areas, furthermore, increasing the bed height will give a sufficient contact time for these molecules to be adsorbed on the sawdust surface.

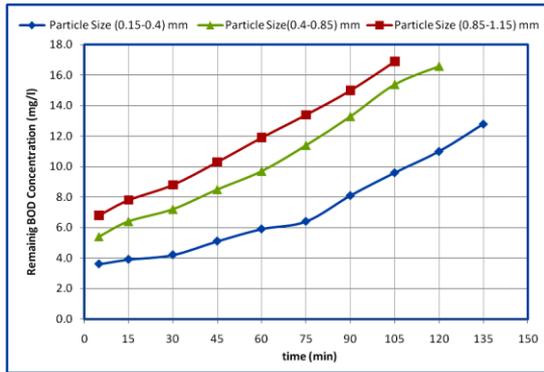
Figure (13) showed that the mass transfer rate decreased with increasing sawdust bed, this indicate that for deeper beds lead to the increasing in the required time to reach the saturation of adsorbent bed.

### 3.4 The Effect of Particle Size

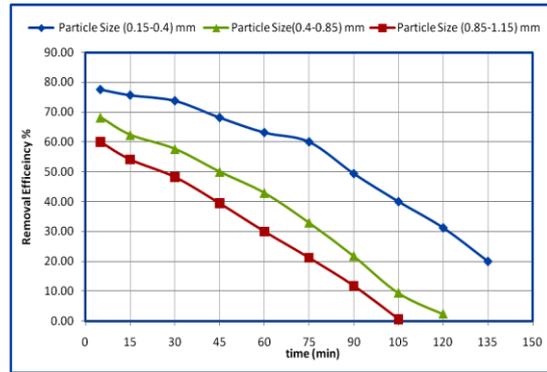
Adsorption is a surface phenomenon, as such, the extent of adsorption is proportional to specific surface area, which can be defined as that portion of surface area that is available for adsorption. Thus the amount of adsorption accomplished per unit weight of a solid adsorbent is greater the more finely divided and the more porous the solid (Weber, 1972).

Three experiments, were carried out at different particle size ranges (0.15-0.4, 0.4-0.85 and 0.85-1.15) mm. The other initial conditions were kept constant (bed height of 0.2 m, BOD<sub>5</sub> initial concentration of (16-17) mg/l, flow rate of  $3.33 \cdot 10^{-6}$  m<sup>3</sup>/sec, pH of 7.5, temperature of 25 °C, activated carbon/sawdust ratio (AC/SD=0).

The break through curves for the above experiments were plotted (effluent BOD<sub>5</sub> concentrations versus time) as shown in figure (14). This figure showed that the required time for reaching the break through point increased when particle size decreased. This due to the fact that when the particle size decreases, the surface area will be available for adsorption increased for a given adsorbent weight (i.e. providing more space for the adsorbate molecules to occupy the new surface area and therefore it will increase the time of saturation).



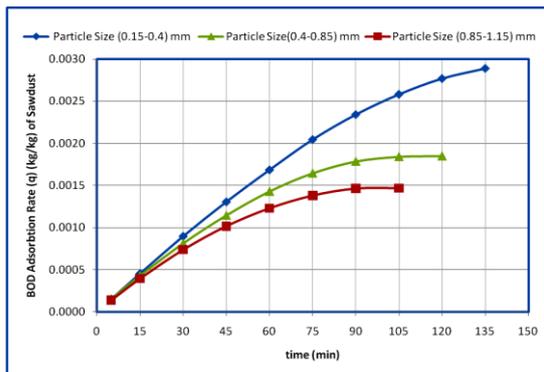
**Figure (14)** Breakthrough curves of isothermal adsorption for different particle sizes.



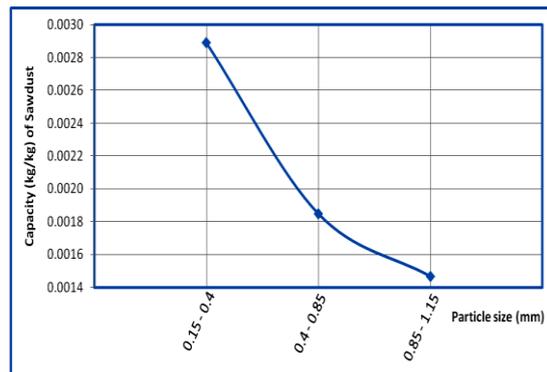
**Figure (15)** the effect of the particle size on the BOD<sub>5</sub> removal efficiency.

Figure (15) shows the effect of particle size in the removal efficiency of BOD<sub>5</sub>. It can be seen that the removal efficiency increases as the particle size decreases.

The adsorption rate was plotted versus time for a given particle size and was shown in Fig. (16). It can be seen from the figure that the adsorption rate increased for smaller particle size in comparison with the larger particle size.

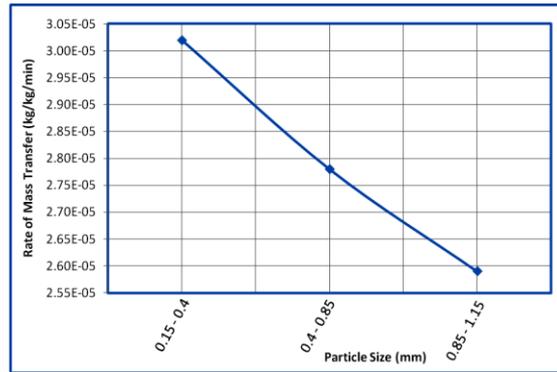


**Figure (16)** Adsorption rate for different particle sizes.



**Figure (17)** effect of particle sizes on the capacity of sawdust

The effect of the particle size on the capacity of sawdust was shown in Fig. (17), it can be seen that increasing the particle size will decrease the adsorption capacity of the sawdust. This was attributed to the fact that for smaller particles of the adsorbent the micropores are believed to be more readily accessible by the wastewater molecules and the transport is mainly due to film diffusion which is more effective than intraparticle diffusion. As the particle size increases, the transport due to intraparticle diffusion will be more dominant. Since it is slow and not very effective process, therefore the adsorption capacity will decrease when particle size increases. As same as the rate of mass transfer will increase when the particle size decreases as shown in Fig. (18).



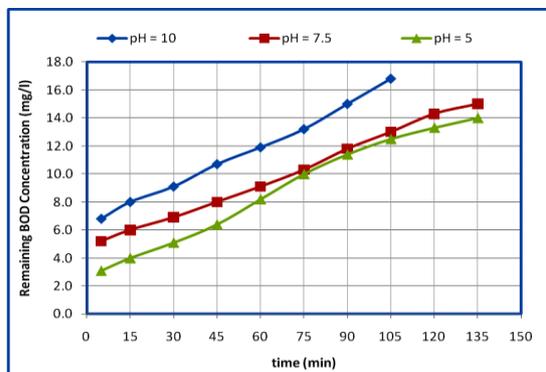
**Figure (18)** effect of the particle sizes on the rate of mass transfer of BOD<sub>5</sub>.

This mass transfer was due to diffusion transfer (film diffusion) which is faster than the diffusion due to intraparticle transport that happens after the diffusion process.

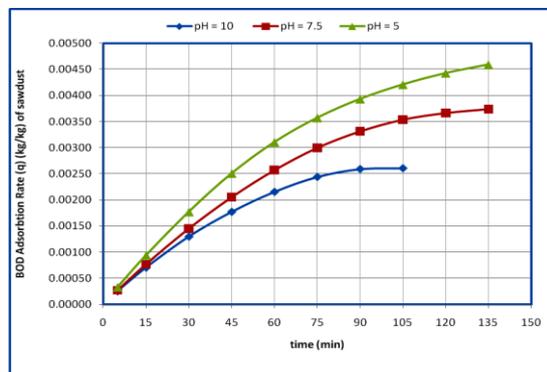
### 3.5 The Effect the pH of the Solution

Three experiments, were carried out with different solution pH; 5, 7.5, and 10. The other initial conditions were kept constant, BOD<sub>5</sub> concentration of (16-17) mg/l, bed height of 0.1 m, flow rate of  $3.33 \times 10^{-6} \text{ m}^3/\text{sec}$ , particle size of (0.15-0.4)mm, temperature of 25 °C and activated carbon/sawdust (AC/SD=0).

The breakthrough curves for the above experiments were plotted, Fig. (19), this figure showed that the breakpoint increased as the pH value decreased.



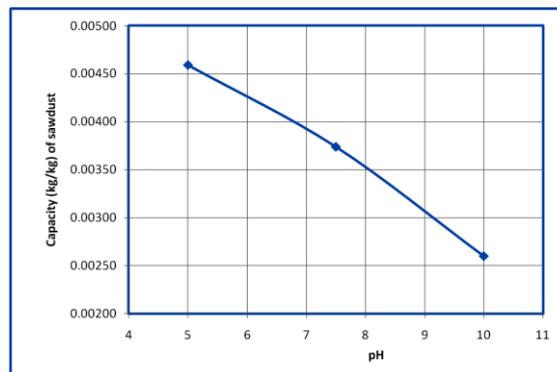
**Figure (19):** breakthrough curves of isothermal adsorption for different pH of the influent.



**Figure (20):** Adsorption rates of different influent pH.

The solute adsorbed (the uptake) versus time were also plotted in Fig. (20). From Fig. (20) it can be seen that the total amount of solute removed from solution at any period of time increased with decreasing pH. This is due the fact that the cell walls of sawdust mainly consist of cellulose and lignin, and many hydroxyl groups, such as tannins or other phenolic compounds. Adsorbents containing high levels of cellulose acquired negative surface charge on contact with water (Ferrero, 2007, Asadi et al., 2008). As pH of the system decreased, the number of negatively charged adsorbent sites decreased and the number of positively charged surface sites increased. This will also increasing the driving force which increases the accumulation of the pollutants on the sawdust surface.

Figure (21) illustrated the effect of solution pH on the capacity of sawdust. It can be seen that the adsorption capacity of sawdust increases with decreasing solution pH. The reason for the better adsorption capacity observed at low pH may be attributed to the larger number of H<sup>+</sup> ions present, which in turn neutralize the negatively charged adsorbent surface, thereby reducing hindrance to the diffusion of organics at higher pH. At higher pH, the capacity of the adsorbent decreased. The reduction in adsorption may be possible due to the abundance of OH<sup>-</sup> ions, causing increased hindrance to diffusion of organics contributing to BOD<sub>5</sub> ions. **Aluyor and Badmus, 2008**, in their study, observed similar effect on COD removal from industrial wastewater. Also the mass transfer rates for initial stages of adsorption increases with decreasing solution pH.



**Figure (21):** effect of the pH of the influent on the capacity of the sawdust.

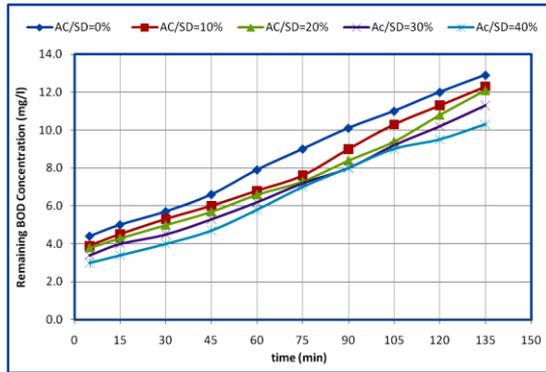
### 3.6 The Effect of Different Activated Carbon/Sawdust Ratios

The effect of different commercial activated carbon /sawdust height ratios were investigated for BOD<sub>5</sub> adsorption onto sawdust by adding different height ratios of (1mm particle size) activated carbon to the sawdust bed which was of (0.15-0.4mm particle size).

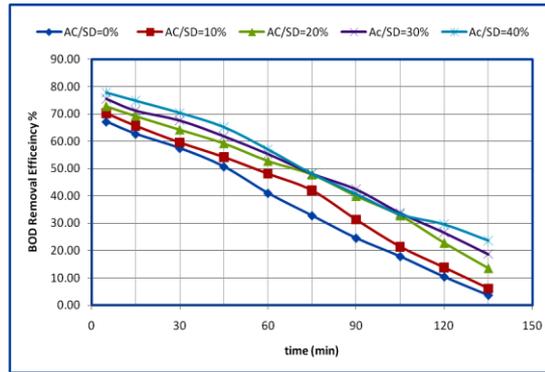
Five experiments were conducted using different height ratios of (0%, 10%, 20%, 30%, and 40%). All experiments were carried out at constant conditions, flow rate of  $3.33 \times 10^{-6}$  m<sup>3</sup>/sec, initial BOD<sub>5</sub> concentration of (13.1-14) mg/l, sawdust bed height of 0.1m, temperature of 25 °C and solution pH of 7.5.

The experimental breakthrough curves are presented in Fig.(22), and the BOD<sub>5</sub> removal efficiency versus time were plotted in Fig. (23).

It can be seen from figures (22) and (23) that adding activated carbon to the sawdust bed increased the amount of solute removed from solution at any period of time and hence increasing the removal efficiency.



**Figure (22)** breakthrough curves of isothermal adsorption for different AC/SD ratios.



**Figure (23)** the effect of the AC/SD ratios on the BOD<sub>5</sub> removal efficiency.

In fact, the commercial activated carbon has higher surface area, adsorption capacity and higher porosity than any other adsorbent. Therefore, increasing the activated carbon ratio caused an increase in the available adsorption sites and the total adsorption capacity of the bed, and this expected to increase in the adsorption capacity.

#### 4. Conclusions

In this research, the ability of using local adsorbent of sawdust was studied, and the following points can be concluded based on the results discussed earlier.

From BOD<sub>5</sub> results, the capacity of sawdust increased with increasing of bed height, influent concentration, activated carbon/sawdust ratio and also increased with decreasing flow rate, particle size and pH. Much larger bed volumes are required when using the proposed adsorbent in continuous column operations due to higher adsorbent requirement when compared to GAC. The rate of mass transfer for initial stage of adsorption increased with increasing influent concentration, flow rate and also increased with decreasing particle size, pH and bed height.

#### 5. References

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