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# Comparative Study of Effect of PAC and GAC on Removal of COD Contributing Component of Sugar Industry waste water

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## Abstract

*Rapid globalization leads us on the way to industrialization. Pollution of water by organic and inorganic chemicals is of serious environmental concern. Sugar industry is one of the biggest consumer of water, and can also introduce serious pollutant to the environment. Chemical as well as biological treatments to these waste waters are in practice since long. Powdered Activated Carbon (PAC) prepared from wood and nutshell charcoal with specific surface area of 5602.352 $cm^2$/gm and particle size 44 μ m and Granular Activated Carbon (GAC) prepared from wood and nutshell charcoal with specific surface area of 10.50 $cm^2$/gm and particle size 1.08 mm are used as adsorbents to the combined waste water of Sugar mill at room temperature. The different dosage of PAC and GAC is kept in contact for 24 hours and analyzed before and after treatment. The results of COD removal follow the Freundlich and Langmuir adsorption isotherm. Among PAC and GAC -PAC removes 62.26% of COD at the dose of 20gm/L further at the dose of 30 gm/L its 66.04%, whereas GAC removes 62.26% of COD at the dose of 20gm/L and found exhausted for higher dosages. It proves that rates of adsorption increases with the reduction in particle size.*

**Key words:** Adsorption isotherm, adsorption intensity (1/n), adsorption energy (b x $10^3$), adsorption capacity (K, $\Theta_0$).

## Introduction

Water is one of the essential enablers of life on earth. But pure water is not available to a large fraction of the population of the planet. While availability is an issue, contamination is another major concern which threatens the survival of many[1]. The Sugar Industry in India is one of the oldest and largest industries in the country. These mills require large volume of water of high purity and generate equally large volume of waste water which is highly complex and polluted[2]. In the present study, it was aimed to carry out experiments using Powdered Activated Carbon and Granular Activated Carbon for the removal of organic contaminants especially COD contributing components from the combined waste water of Sugar Industry, which is situated in South Gujarat region of India.

A number of conventional treatment technologies have been considered for treatment of wastewater contaminated with organic substances. Among them, adsorption process is found to be the most effective and economical method. It has aroused considerable interest during recent years. Current research has focused on modified or innovative approach that more adequately address the removal of organic pollutants. Commercial activated carbon is regarded as the most effective material for controlling the organic load[3,4]. Adsorption of various substances onto carbon surface is an exceedingly complex process to solve the water quality problem has been discussed thoroughly by Faust and Aly[5]. A novel Freundlich type multi-components adsorption isotherm was employed successfully to describe the adsorption of organic pollutants on activated carbon from the multi-component aqueous solution[6]. The presentation of the solute adsorbed per unit weight/ volume of the adsorbent as the function of the equilibrium concentration in the bulk solution at constant temperature is termed as the 'Adsorption Isotherm'[7]. Some investigators found that particular organic suspended solids could interfere with the adsorption process, both in term of adsorption capacity and adsorption rate; however the effect of the solids diminished as the size of the adsorbent increased[8]. Rates of adsorption increases with the reduction in particle size and it is inversely proportional to the square of the carbon particle diameter[9]. The effect of particle size and shape of adsorbent on adsorption is measured by computerized image analyzer[10].

Various activated carbons were tested for its better quality and quantity of different particle size for a comparison of its adsorption characteristics. GAC shows advantages in its application because it exhibits high porosity, large surface and is easy to reactivate after its exhaustion and is easy to handle. Adsorption is a surface phenomenon. Physical adsorption takes place at lower temperatures, while chemical adsorption is effective with localized high energy producing ionic bonds. So, isotherm tests are carried out for the effective dose of carbon by batch experiments. Column tests are conducted to find out operating capacity of the carbon with optimum flow rate and bed depth. Reactivation of spent carbon from waste water effluent is performed by thermal regeneration process or electro chemical process. Activated carbon is widely used and adsorption models have been effectively applied for partial or complete removal of various substances[11]. Removal of COD contributing components, TOC and cadmium of the waste water by adsorption process on fly ash and PAC was well studied. The data follow Freundlich and Langmuir type behavior. The reaction rate was evaluated for different time intervals and at different pH[12]. The adsorption characteristics of three types of activated carbon for 17β-estradiol were studied by long term experiments to assess the time which is necessary to reach equilibrium between the solid and the liquid phase. The equilibrium concentrations were calculated to be at 49–81% of the initial concentration in the concentration range between 1 and 100 ng/l, with 0.51 ng/l for a 1 ng/l and between 5.9 and 14.6 ng/l for 100 ng/l initial concentration[13].

Some researchers studied on adsorption characteristics of Congo red on coal-based mesoporous activated carbon. They found the monolayer adsorption capacity of ACs was found to increase with increasing both the mesoporous volume and the mesoporous contribution to their porous texture. They derived a correlation between the Langmuir adsorption capacity and the degree of mesopore development. The higher the both mesopore volume and mesopore contribution to the total pore volume, the higher is the adsorption capacity of the activated carbon with respect to Congo red dye[14]. The adsorption characteristics of Congo red dye differs for PAC and GAC. The result by using Taguchi method may be summarized as i. From S/N ratio of PAC the influencing parameters are in the order temperature > pH >Contact time. ii. Similarly for GAC the influencing parameters are in order pH >contact time>temperature. The optimum condition for adsorption CR dye on PAC is found to be pH of 2, temperature $50^0$C, contact time 300 minutes. But for GAC, Taguchi method optimizes the adsorption at pH-2, temperature $60^0$C and time-300 minutes[15].

An experimental investigation carried out on a pilot scale membrane bioreactor (MBR) with the addition of powdered activated carbon (PAC) to analyze improvements in effluent quality and in the filtration process. The results refer COD removal stability appeared to increase as PAC concentration increased. No effects were observed on the nitrification processes. The filtration process was evaluated in terms of sludge filterability, fouling rate and fouling reversibility. The fouling rate decreased with an increasing PAC concentration and showed complete reversibility both in presence and in absence of PAC[16]. In the Fenton oxidation reaction at pH 3 when PAC was added to the pre-treated supernatant, the COD removal efficiency was 80.22 %. When UF membrane separation was performed using this specimen, COD removal efficiency is 98.48 %. When the membrane separation was undertaken without PAC the COD removal efficiency is 94.93 %. Thus, the COD removal efficiency was higher when PAC was added compared with that observed when PAC was not added.[17]

One of the drinking waterworks supply was studied by bench-scale experiments examining effects of different particle size of granular activated carbon (GAC), pH, dosage, and temperature, and by forming the Freundlich adsorption model. The adsorption performance could be significantly improved by using small-size GAC, low pH, low temperature, and an increase in the dosage of GAC[18]. The study of two algal odorants dimethyl trisulfide and β-cyclocitral adsorption on granular activated carbon (GAC) was investigated. It was concluded that among the four isotherm models (Langmuir, Freundlich, Temkin, and Dubinin–Radushkevich), Freundlich isotherm showed the best fitting with the equilibrium data in terms of the coefficient of determination ($R^2$) and Chi-square ($\chi^2$)[19]. Volatile Organic Compounds (VOCs) such as methanol, ethanol, methyl ethyl keton, benzene, n-propanol, toluene, and o-xylene were adsorbed in a laboratory-scale packed-bed adsorber using granular activated carbon (GAC) at 101.3 kPa. The adsorption isotherm, and adsorbed amount and adsorption heat of VOCs were obtained using the breakthrough curve: the former for comparison with the conventional isotherm models, the latter for correlation with the physical properties of VOCs[20].

Chromium is mainly used in the leather and wood industries can be removed from waste by various processes like adsorption. The Cr(VI) adsorption by commercial granular activated carbon (GAC) as adsorbent from diluted solutions, and batch systems with controlled pH, In addition- effect of pH on the Cr(VI) adsorption, adsorption equilibrium, and kinetic were studied under experimental conditions (pH = 6, MA = 6g, for 90min.). On the GAC surface, carboxylic groups were found to be in higher concentrations (MAS= 0.43 m mol/g CAG), which increase the Cr(VI) adsorption, principally in acidic pH values[21].

The performance of the granular activated carbon (GAC) fixed bed adsorption; the continuous photo-catalysis systems and a combination of the two were studied to evaluate their capabilities in removing the herbicide of metsulfuron-methyl (MM) from waste water. Removal of MM via adsorption using GAC fixed beds of 5, 10 and 15 cm depths (operated at meter per hour) achieved a removal of 35, 55 and 65% of MM respectively[22]. The performances of GAC adsorption and GAC bioadsorption in terms of dissolved organic carbon (DOC) removal were investigated with synthetic biologically treated sewage effluent (BTSE), synthetic primary treated sewage effluent (PTSE), real BTSE and real PTSE. The main aims of the study are to verify and compare the efficiency of DOC removal by GAC (adsorption) and acclimatized GAC (bioadsorption). The results indicated that the performance of bioadsorption was significantly better than that of adsorption in all cases, showing the practical use of biological granular activated carbon (BGAC) in filtration process. The most significance was observed at a real PTSE with a GAC dose of 5 g/L, having 54% and 96% of DOC removal by adsorption and bioadsorption, respectively[23].

## Material and Method

Powdered Activated Carbon (PAC) with specific surface area 5602.352 $cm^2$/gm and particle size 44 μ m and Granular Activated Carbon (GAC) with specific surface area 10.50 $cm^2$/gm and particle size 1.08 mm were used as adsorbents for the treatment of sugar industry waste water. For present research work the adsorbent samples were prepared from wood and nutshell charcoal. The process of manufacturing of activated carbon included carbonization followed by activation. Where-in on pyrolysis of raw materials polynuclear aromatic system get resulted. The carbon formed would be further activated by burning it in atmosphere of $CO_2$, CO, $O_2$, $H_2O$ vapour, air or other selected gases at temperature between 300 to $1000^0$ C.

Wastewater samples were collected from the Khedut Sahakari Khand Udhyog Mandali Ltd, Bardoli. The pH and EC of the samples were measured on the site and the other parameters were analyzed in the lab according to the APHA. Samples were stored at temperature below $3^o$C to avoid any change in the physic-chemical characteristics.

The activated carbons were added to sugar industry waste water sample and the mixture was stirred well and was kept in contact until equilibrium state attain and that was 24 hours for this system. The known quantity (1 liter) of sample was treated with different amount of Powdered Activated Carbon and Granular Activated Carbon viz 1, 2, 5, 10, 15, 20, 30 gm/L stirred well and kept in contact for 24 hours at room temperature. Then the samples were filtered and analyzed especially for COD removal. The method for determination of COD practicable is dichromate reflux method followed from 'Standard methods for the water and waste water'[24]. The results for each dose are presented in table 1, 2 and figure 1, 2 and 3.

## Results and Discussion

Table 1 represents the data for Freundlich and Langmuir adsorption isotherms along with percent removal of COD exerting components of sugar industry waste water on to PAC. It can be observed that the percent removal of COD increases with increase in PAC concentration from 37.73% to 66.04%. The removal per unit weight is found to be decreased from 697.2 mg/gm to 40.67 mg/gm with increase in PAC dose. The logarithmic values of equilibrium concentration ($C_{eq}$) and removal per unit weight (x/m) were given in table which were used for the explanation of Freundlich adsorption isotherm model and plot whereas the inverse values needed for Langmuir isotherm model.

Table 2 represents the data for Freundlich and Langmuir adsorption isotherms along with percent removal of COD exerting components of sugar industry waste water on to GAC. It can be seen that the initial COD content of the waste water was 1847.58 mg/L which is reduced to 697.2 mg/L with 20gm/L of GAC and remains constant for higher dose. Percent removal of COD increases with increase in GAC concentration from 32.07% to 62.26% with 20 gm/L of GAC and remain constant for higher dose. The removal per unit weight is found to be decreased from 592.62 mg/gm to 38.346 mg/gm with increase in GAC dose. The logarithmic values of equilibrium concentration ($C_{eq}$) and removal per unit weight (x/m) were given in table which were used for the explanation of Freundlich adsorption isotherm model whereas the inverse values needed for Langmuir isotherm model.

Table 3 represents the comparison of effect of PAC and GAC on COD contributing components of the waste water from sugar industry clearly indicate that The maximum COD removal is found at 30gm/L of PAC concentration i.e. 66.04% whereas 62.27% for GAC.

**Table-1**
**Freundlich and Langmuir adsorption isotherm parameters for COD contributing components and percent removal of COD in presence of PAC**

| No | Adsorbent concentration m (gm/L) | Eq. Conc. $C_{eq}$ (mg/L) | Removal $x=C_0-C_{eq}$ (mg/L) | $q_e$= x/m (mg/gm) | Removal % | log $C_{eq}$ | log x/m | $1/C_{eq}x10^3$ | $1/q_ex10^2$ |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 0 | 1847.58 | ------- | ------ | ------- | 3.2666 | -------- | 0.5412 | ------- |
| 2 | 1 | 1150.38 | 697.2 | 697.2 | 37.73 | 3.0608 | 2.8434 | 0.8693 | 0.1434 |
| 3 | 2 | 1080.66 | 766.92 | 383.46 | 41.51 | 3.0337 | 2.5837 | 0.9254 | 0.2608 |
| 4 | 5 | 906.36 | 941.22 | 188.244 | 50.94 | 2.9573 | 2.2747 | 1.1033 | 0.5312 |
| 5 | 10 | 801.78 | 1045.8 | 104.58 | 56.6 | 2.9041 | 2.0194 | 1.2472 | 0.9562 |
| 6 | 15 | 766.92 | 1080.66 | 72.044 | 58.49 | 2.8848 | 1.8576 | 1.3039 | 1.3880 |
| 7 | 20 | 697.2 | 1150.38 | 57.519 | 62.26 | 2.8434 | 1.7598 | 1.4343 | 1.7386 |
| 8 | 30 | 627.48 | 1220.1 | 40.67 | 66.04 | 2.7976 | 1.6093 | 1.5937 | 2.4588 |

**Adsorbent:** Powdered Activated Carbon (PAC)
**Specific Surface Area:** 5802.352 $cm^2$/ gm
**Particle size**: 44 μ m
**Room temperature**: 26± $1^0$C
**Contact duration**: 24 Hours

**Table-2**
**Freundlich and Langmuir adsorption isotherm parameters for COD contributing components and percent removal of COD in presence of GAC**

| No | Adsorbent concentration m (gm/L) | Eq. Conc. $C_{eq}$ (mg/L) | Removal $x=C_0-C_{eq}$ (mg/L) | $q_e$= x/m (mg/gm) | Removal % | log $C_{eq}$ | log x/m | $1/C_{eq}x10^3$ | $1/q_ex10^2$ |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 0 | 1847.58 | ------- | ------ | ------- | 3.2666 | -------- | 0.5412 | ------- |
| 2 | 1 | 1254.96 | 592.62 | 592.64 | 32.08 | 3.0986 | 2.7728 | 0.7968 | 0.1687 |
| 3 | 2 | 1150.38 | 697.2 | 348.6 | 37.73 | 3.0608 | 2.5423 | 0.8693 | 0.2869 |
| 4 | 5 | 1080.66 | 766.92 | 153.384 | 41.51 | 3.0337 | 2.1858 | 0.9254 | 0.6520 |
| 5 | 10 | 836.64 | 1020.94 | 101.094 | 54.72 | 2.9225 | 2.0047 | 1.1953 | 0.9892 |
| 6 | 15 | 766.92 | 1080.66 | 72.044 | 58.49 | 2.8848 | 1.8576 | 1.3039 | 1.3880 |
| 7 | 20 | 697.2 | 1150.38 | 57.519 | 62.26 | 2.8434 | 1.7598 | 1.4343 | 1.7386 |
| 8 | 30 | 697.2 | 1150.38 | 38.346 | 62.26 | 2.8434 | 1.5837 | 1.4343 | 2.6078 |

**Adsorbent**: Granular Activated Carbon (GAC) **Room temperature**: 26± $1^0$C, **Specific Surface Area:** 10.50 $cm^2$/ gm **Contact duration**: 24 Hours, **Particle size**: 1.08 mm

**Table-3**
**Comparison of effect of PAC and GAC on COD contributing components of the waste water from sugar industry**

| No | Adsorbent concentration m (gm/L) of PAC and GAC | Eq. Conc. $C_{eq}$ (mg/L) | Removal % By PAC | Eq. Conc. $C_{eq}$ (mg/L) | Removal % By GAC |
|---|---|---|---|---|---|
| 1 | 0 | 1847.58 | ------- | 1847.58 | ------- |
| 2 | 1 | 1150.38 | 37.73 | 1254.96 | 32.08 |
| 3 | 2 | 1080.66 | 41.51 | 1150.38 | 37.73 |
| 4 | 5 | 906.36 | 50.94 | 1080.66 | 41.51 |
| 5 | 10 | 801.78 | 56.6 | 836.64 | 54.72 |
| 6 | 15 | 766.92 | 58.49 | 766.92 | 58.49 |
| 7 | 20 | 697.2 | 62.26 | 697.2 | 62.26 |
| 8 | 30 | 627.48 | 66.04 | 697.2 | 62.26 |

Figure 1 represents the plot of log $C_{eq}$ Vs log x/m (COD) for PAC and GAC. The straight line nature of the plot corresponds to slope 1/n and intercept K. 1/n is related to adsorption intensity whose value is 4.0445 for COD while intercept K on Y-axis related to adsorption capacity is found to be 1.3 for PAC. For GAC 1/n is related to adsorption intensity whose value is 5.3158 for COD while intercept K on Y-axis related to adsorption capacity is found to be 1.15

Figure 2 represents the plot of Langmuir parameters viz, $1/C_{eq}$ x $10^3$ and $1/q_e$ x $10^3$. The nature of the curve for COD onto PAC and GAC is linear however the intercept on X-axis related to adsorption energy (L/mg) i.e. b x $10^3$ is 0.84 L/mg (PAC) and 0.76 L/mg (GAC) for COD exerting components. These values can be used to calculate the adsorption capacity $\Theta_0$ i.e 282.84 (mg/gm) for PAC and 279.18 (mg/gm) for GAC.

Figure 3 represents the % removal of COD contributing components of the waste water from sugar industry in presence of PAC and GAC, clearly indicates that the maximum COD removal is found at 30gm/L of PAC concentration i.e. 66.04% whereas 62.27% for GAC.

The data presented in table 1 and 2 represent the Influence of different dose of PAC and GAC on COD contributing components. The removal of COD is explained on the basis of adsorption phenomenon and the extent can be co-related with increasing adsorption sites with increase in the dose. The other information are used to prove the adsorption isotherm-Freundlich and Langmuir adsorption isotherms for COD onto PAC and GAC which is used for the calculation of the Adsorption intensity, Adsorption energy and Adsorption capacity. The percent removal of COD seems to be increased with increase in dose of adsorbent for PAC. The plautue nature of the plot after 30gm/L for PAC and 20gm/L for GAC is due to the exhaustion of the adsorbent site. It can be explained on the basis of particle size. Rates of adsorption increases with the reduction in particle size and it is inversely proportional to the square of the carbon particle diameter. The logarithmic and inverse values of $C_{eq}$ and x/m are used for plot of isotherm.

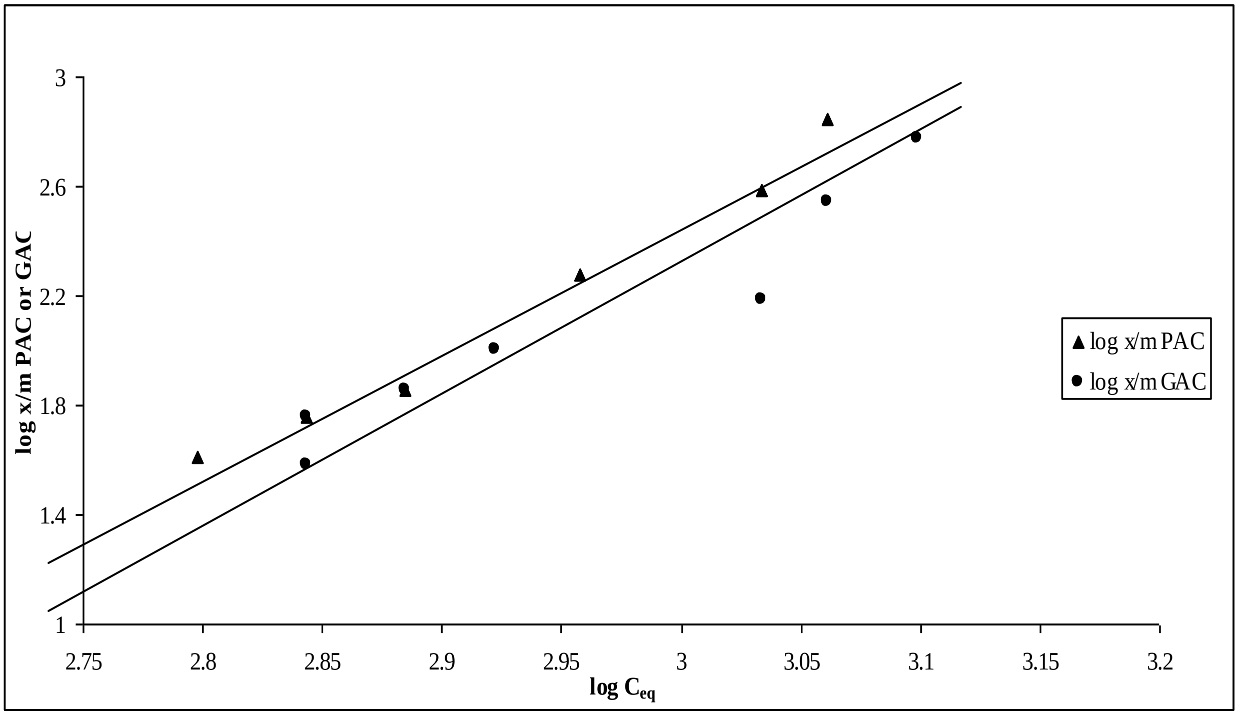


**Figure-1**
**Freundlich adsorption isotherm for COD contributing components onto PAC and GAC**

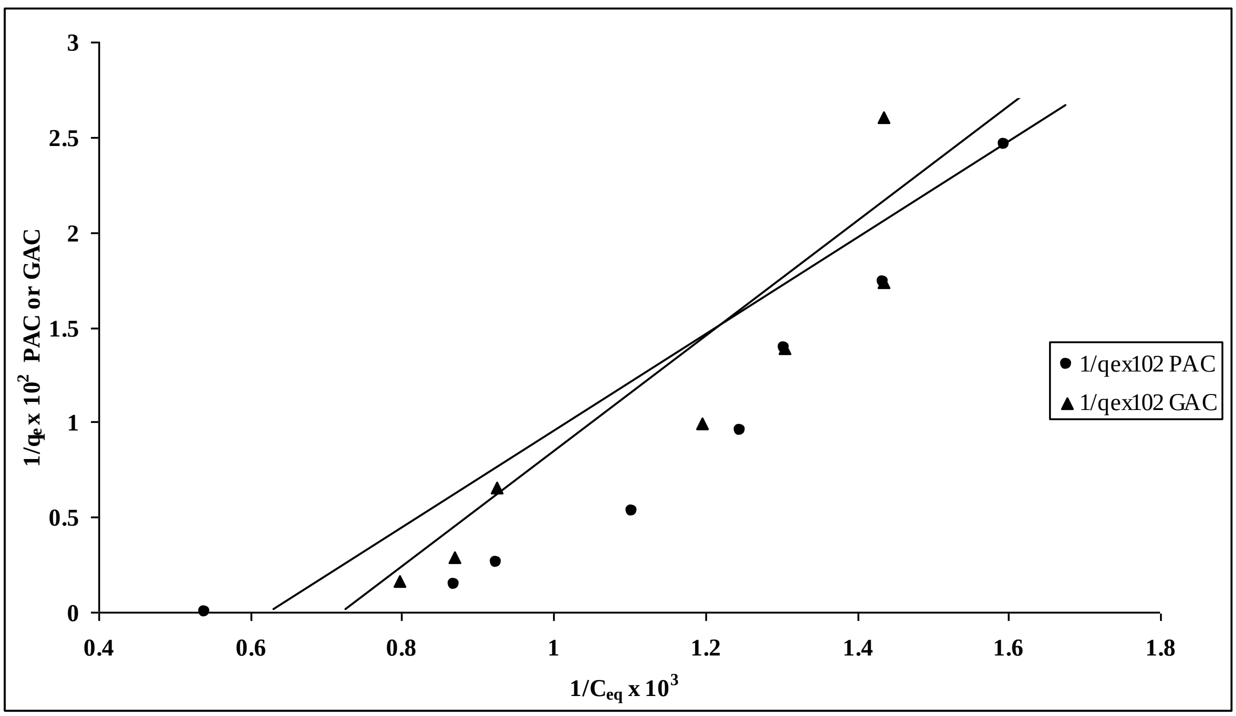


**Figure-2**
**Langmuir adsorption isotherm parameters for COD contributing components of COD in presence of PAC and GAC**

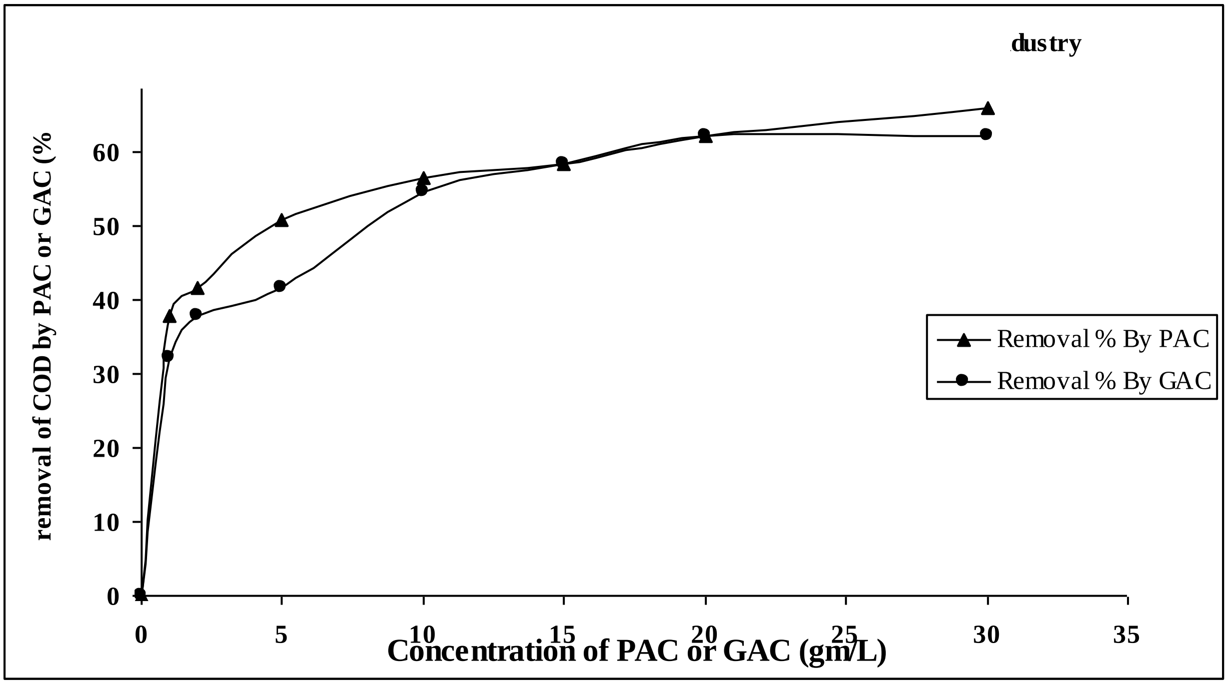


**Figure-3**
**Comparison of effect of PAC and GAC on % COD removal of sugar industry waste water**

The logarithmic value of equilibrium concentration and removal per unit weight gives the linear plot for COD by PAC and GAC confirm the applicability of Freundlich adsorption isotherm. It is the most widely used mathematical description of adsorption in aqueous systems. The equation is an empirical expression that covers the heterogeneity of the surface and exponential distribution of sites and their energies. With the purpose of linearization the equation is represented in logarithmic form as—

$$\log x/m = \log K + 1/n \log C_{eq}$$

The plot of log $C_{eq}$ versus log x/m gives straight line with a slope of 1/n and log K is the intercept of log x/m at log $C_{eq}$ = 0 which indicates that Freundlich adsorption isotherm model is applicable.

The same table shows the Langmuir adsorption isotherm for COD by PAC and GAC. Langmuir isotherm is a plot of the amount of impurity adsorbed by PAC against the amount of impurity that remains in solution. It is a preliminary test to check the efficiency of particular material.

These mode of action can be explained on the basis of Langmuir's model, i.e. 'Ideal localized monolayer model' according to which: i. The molecules are adsorbed at definite sites on the surface of the adsorbent. ii. Each site can accommodate only one molecule (monolayer). iii. The area of each site is a fixed quantity determine solely by the geometry of the surface. iv. The adsorption energy is the same at all the sites.

Such behavior on the basis of kinetic consideration, presuming that the adsorbed molecules cannot migrate across the surface of the interact with another neighboring molecules can be mathematically expressed as under

$$1/q_e = 1/\Theta_0 b \;\; x \;\; 1/C_{eq} + 1/\Theta_0$$

Where: $q_e$ = amount of solute adsorbed per unit weight of adsorbent(mg/gm), = x/m i.e. x is amount of adsorbate adsorbed (mg/L) m is weight of adsorbent (gm/L), $C_{eq}$ = equilibrium concentration of the solute (mg/L), $\Theta_0$ = Langmuir constant related to adsorption capacity (mg/gm), b = Langmuir constant related to adsorption energy (L/mg)

Plot of log $C_{eq}$ versus log x/m is a straight line in nature, presented in figure 1 suggests the applicability of this isotherm and indicate a monolayer coverage of the adsorbate on the outer surface of the adsorbent. The steep slope indicates high adsorptive intensity at high equilibrium concentration that rapidly diminished at lower equilibrium concentration covered by the isotherm. As Freundlich equation indicates the adsorptive capacity x/m is a function of the equilibrium concentration of the solute. Therefore, higher capacity is obtained at higher equilibrium concentrations.

Figure 2 represents the plot of Langmuir adsorption isotherm for COD contributing components onto PAC and GAC. The straight line nature of the plot confirms the applicability of the Langmuir model and also the monolayer coverage. The Langmuir constant $\Theta_0$ in mg/gm related to adsorption capacity indicate availability of more surface active region onto adsorbent site and b x $10^3$ L/mg related to adsorption energy in terms of x/m is a characteristic of the system.

Figure 3 represents the % removal of COD content of sugar industry waste water. The percent removal of COD seems to be increased with increase in dose of adsorbent for PAC. The plautue nature of the plot after 30gm/L for PAC and 20gm/L for GAC is due to the exhaustion of the adsorbent site. It can be explained on the basis of particle size. Rates of adsorption increases with the reduction in particle size and it is inversely proportional to the square of the carbon particle diameter.

## Conclusion

This study leads us to the conclusion that the final combined waste water of Sugar manufacturing unit is highly polluted having higher COD value. Due to some practical limitation only COD parameter is emphasized in this paper when the final combined waste water of Sugar mill is treated with finely divided low cost material PAC and Granular Activated Carbon (GAC) at room temperature for 24 hours of contact duration the following results are achieved. i. The maximum COD removal is found at 30gm/L of PAC concentration i.e. 66.04% whereas 62.27% for GAC. ii. Comparison of PAC and GAC for removal of COD proves that Adsorption is a surface phenomenon. Rates of adsorption increases with the reduction in particle size and it is inversely proportional to the square of the carbon particle diameter. iii. At room temperature PAC and GAC work as adsorbents and follow Freundlich and Langmuir isotherm models. The results give straight line which confirms the applicability of isotherm. i. The Freundlich constant K an intercept on X axis is related to adsorption capacity is found to be 1.3 while the slope 1/n is related to adsorption intensity is found to be 4.0445, ii. The straight line of the Langmuir plot gives intercept on Y axis called b x $10^3$ L/mg i.e. adsorption energy is 0.84 and the calculated adsorption capacity $\Theta_0$ mg/gm is 282.85 iii. The Freundlich constant K an intercept on X axis is related to adsorption capacity is found to be 1.15 while the slope 1/n is related to adsorption intensity is found to be 5.3146 for GAC iv. The straight line of the Langmuir plot gives intercept on Y axis called b x $10^3$ L/mg i.e. adsorption energy is 0.76 and the calculated adsorption capacity $\Theta_0$ mg/gm is 279.18 for GAC.

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