

# Removal of phenol from dilute aqueous solutions in a multistage bubble column adsorber using activated carbon prepared from *Tamarindus indica* wood

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## Abstract

In the present investigation, a multistage bubble column adsorber is designed and fabricated to remove phenol from dilute aqueous solutions. The multistaging has been achieved by hydrodynamically induced continuous bubble generation, breakup due to rupture, coalescence, and regeneration. Activated carbon prepared by chemical activation of *Tamarindus indica* wood with zinc chloride has been used as the adsorbent for the removal of phenol. During the preparation of the activated carbon it was found that carbonization time and temperature are the two most important parameters that affect its quality. Different operating parameters that affect the performance of the multistage bubble column adsorber are the contact time, activated carbon loading, and superficial gas velocity. The optimum operating parameters are as follows: contact time (160 min); activated carbon loading (1 g/l); and superficial gas velocity (0.0263 m/s). At these optimum conditions the maximum phenol removal reported for initial phenol concentrations of 40, 60, 80, and 100 mg/l are 99, 97, 94, and 85%, respectively.

**Keywords:** Activated carbon; adsorption; adsorber; multistage bubble column; phenol removal

*JEPS* (2010), Vol. 4, pp. 1 – 7.

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## 1. Introduction

Water pollution due to the discharge of phenol and its derivatives in effluent from process industries has been a major cause of concern for environmental engineers in recent years. Phenol is one of the most harmful hazardous pollutants present in effluent from processing and manufacturing industries engaged in oil refining, coal tar processing, petrochemical production, coke manufacture, plastic industry, textile processing, leather processing, insecticides production, manufacture of dyes and dyeing, glass production, etc. [1-2]. Phenol, even in trace amounts, may cause nausea, vomiting, greenish or smoky colored urine, and even death from respiratory failure or cardiac arrests with long-term exposure [3-4]. The presence of chlorine can aggravate the situation as it can react with

phenols to produce mono-, di-, or trichlorophenols, which impart tastes and odors to waters. Chlorinated phenols have often been detected in treated wastewaters [5]. Fatal poisoning may also occur by adsorption of phenol by skin, if a large area of it is exposed. Increased awareness of its effect on the environment and the increasingly stringent environmental laws has created an urgent demand for the development of suitable equipment and processes for the complete removal of phenol from process wastewater.

Various treatment methods have been developed for the abatement of phenolic wastewaters [3-4]. The choice of treatment depends upon effluent characteristics such as concentration of phenol, pH, temperature, flow volume, biological oxygen demand, economics, and social factors like standards set by government agencies. A thorough

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review of the literature indicates that adsorption by activated carbon was found to be one of the most effective methods for the removal of phenol from wastewater [1, 4]. Activated carbons have the advantage of exhibiting a high adsorption capacity for organic pollutants due to their high specific surface area, adequate pore size distribution, and relatively high mechanical strength [6-7]. Activated carbons are also effective as catalysts and catalyst supports. However, the use of activated carbon is restricted due to the high cost of commercial activated carbons and problems associated with the regeneration of these carbons. Therefore, there is a need to search for low-cost potential adsorbents. Usually commercial activated carbons are prepared from coal, coconut shell, or bamboo. Recently, many researchers have shown that agricultural wastes and resources rich in lignocellulosic materials have emerged as a better choice for preparing activated carbons. There is a large number of studies regarding the preparation of activated carbons from agricultural wastes [8], nuts [2], nutshells [1, 9], fruit stones [10], bagasse [11], oil palm waste [12], and agricultural residues from sugarcane [13], rice [14], peanut [15, 16], sawdust [17], and canes from some easy-growing wood species [18]. These materials can be used as is for adsorption; however, activation by chemical or physical methods enhances their adsorptive capacity [2].

Traditionally, activated carbon-based adsorption has been carried out in packed beds, fluidized beds, and moving beds using granular and/or powdered activated carbons as adsorbents. Operational complexity and removal efficiency limitations have restricted the use of these types of equipment. There is a continuous search for more efficient equipment without additional mechanical complications. In recent years, bubble columns have drawn increased attention as suitable alternatives due to their various advantages over other gas-liquid (or gas-liquid-solid) contacting devices [19]. They are simple in construction and require little maintenance as there are no moving parts. They have high liquid phase content, excellent heat transfer properties, low initial cost, and offer a significant interfacial mass transfer area and good mixing properties at low energy consumption, as the gas phase serves the dual function of aeration and agitation [20]. Bubble columns can therefore prove to be a very important contacting system for the liquid-phase adsorption of organic pollutants using powdered activated carbons. However, due to various physical and equilibrium property limitations the maximum single stage efficiency of removal is limited to 70-80% [21]. To combat pollution, much higher efficiency of removal is necessary, and this can only be achieved if a multistage system can be developed. Multi-staging can be achieved either by using the required number of devices in a series or by manipulating the hydrodynamics of the device in a single column [22].

The present investigation is focused on two things –

development of a multistage bubble column adsorber, and preparation of an efficient activated carbon from the wood of the *Tamarindus indica* tree. The multistage bubble column has been designed to operate in three stages – the staging effect being achieved through hydro-dynamically induced continuous bubble generation, break-up through rupture, and regeneration. Creation of finer, small bubbles from initial large bubbles creates more area for mass transfer. Also, the continuous rupture and bursting of bubbles creates localized turbulence and recirculation, which helps in faster transfer of pollutants to the active sites of the solid adsorbents. This bubble column adsorber has been used to treat phenolic wastewater using the activated carbon prepared from the wood of the *Tamarindus indica* tree. The effect of different operating parameters on the efficiency of removal of phenol has been investigated.

## 2. Experimental

### 2.1. Preparation & characterization of activated carbon from *Tamarindus indica* wood

Branches of dried tamarind tree were collected locally and cut to small pieces nearly 1 inch × 1 inch. These precursors were washed with distilled water to remove the water-soluble substances on the wood surface and then dried under sunlight to remove the moisture. Activated carbon was prepared by the chemical activation method using zinc chloride as the activating agent. Activation was done by mixing 10 g of precursor with a 100-ml solution of water containing 10 g of ZnCl<sub>2</sub>. The chemical ratio (activating agent/precursor) was 100% in this case. After mixing, the slurry was subjected to vacuum drying for 24 h. These chemically loaded precursors were then carbonized between 350 and 550°C in the absence of air in a furnace. The temperature was raised at an increment of 5°C/20 min. The samples were collected and the weight was measured before each individual temperature increment. The precursors were ground into powder in a ball mill and the powder was screened in a 200 mesh screen to collect the under flow. The experiments were carried out for different chemical ratios, carbonization times, and temperatures.

Weight loss of the carbon samples was calculated on a chemical-free basis and chemical recovery (CR) was estimated according to:

$$CR = \frac{W_{Pi} - W_{Pf}}{W_C} \times 100 \quad (1)$$

where  $W_{Pi}$  and  $W_{Pf}$  are the weight of product before and after washing and  $W_C$  is the weight of chemical used. Chemical recovery and weight loss are regarded as indicators of the process efficiency in the chemical activation process and are reported on % basis.

Characterizations of the activated carbons were determined by nitrogen adsorption at  $-196^{\circ}\text{C}$  with the help of Micromeritics Flowsorb-2300 and Quantachrome Autoscan Mercury Porosimeter. The BET surface area was calculated from  $\text{N}_2$  adsorption isotherms by using the Brunauer-Emmett-Teller (BET) equation. The details of the procedure are described elsewhere [2].

## 2.2. Phenol removal studies in the multistage bubble column adsorber

The experimental setup shown in Figure 1 has been used for the removal of phenol from aqueous solutions. The bubble column adsorber (volume: 70 L) is a tall vertical column fitted with a gas-liquid separator at the top and a gas sparger at the bottom. The space between these has been formed into three vertical stages which in effect are operated in series. To achieve the staging effect, the column has been fitted with four disks (orifice with one large opening at the centre known as a contraction disk) and three screens (four mesh known as expansion disk). Relatively large sized bubbles were generated at the bottom of the first stage with the help of a multi-orifice sparger. As the bubbles tend to move upward they are ruptured and coalesced while passing through the first contraction disk. These coalesced bubbles are then made to pass through an expansion disk thereby creating a swarm of smaller bubbles. Above this expansion disk another contraction disk is fitted. When these smaller bubbles try to pass through this contraction disk, they are again ruptured and coalesced. The section of the column consisting of an expansion disk positioned between two contraction disks comprises of one stage. At every stage bubbles lose their individual identities and new bubbles are generated. Thus the continuous generation, breakage, and regeneration of bubbles not only creates more interfacial area for mass transfer but also helps in faster transport of pollutants to the surface of the adsorbents.

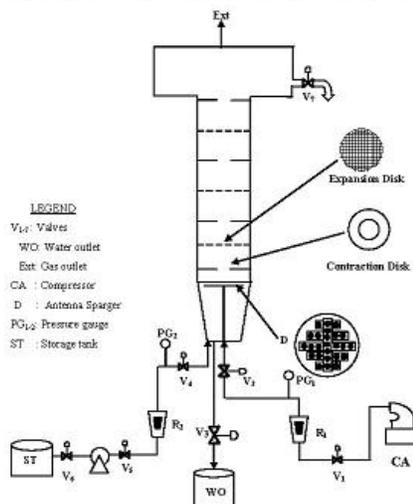


Figure 1. Schematic diagram of the experimental setup of multistage bubble column adsorber for removal of phenol from dilute aqueous solution

Experiments for phenol removal were carried out by using synthetic wastewater prepared by dissolving the necessary amount of phenol in tap water to obtain standard solutions of concentrations ranging between 10 and 100 mg/l and kept in a storage tank. This solution was fed to the bottom of the column until a particular level was reached. Compressed air (CA) from the compressor, after removing oil and moisture, was fed to the sparger at a controlled rate, so that a desired flow pattern was established in the column. The experiments were conducted at gas flow rates of  $3.33$  to  $10.0 \times 10^{-4} \text{ m}^3/\text{s}$ . A known amount of powdered activated carbon prepared from *Tamarindus indica* wood was added to the wastewater from the top of the column. It is a well known fact that solution pH is the most important parameter governing the rate of phenol removal. From some initial batch experiments, we found that 3.2 was the optimum pH at which maximum adsorption took place. Since this is highly acidic range, a lot of acid would be required to make the solution pH 3.2, which is not cost-effective. To make the process cost-effective as well as efficient, the activated carbon used was itself acidified using concentrated sulphuric acid. For this, a quarter part of concentrated sulphuric acid was mixed with one part of the activated carbon. All the adsorption experiments were carried out at a constant pH of 3.2, adsorbent loading of 0.25 g/l, and superficial gas velocity of 0.0263 m/s. After the establishment of steady-state, flow samples were collected from the bottom of the column at suitable time intervals, filtered using 0.45 micron filter papers, and analyzed for residual phenol concentration in a spectrophotometer (Model: UV-2100, make Shimadzu). All the data presented in this manuscript are the average of three experimental runs.

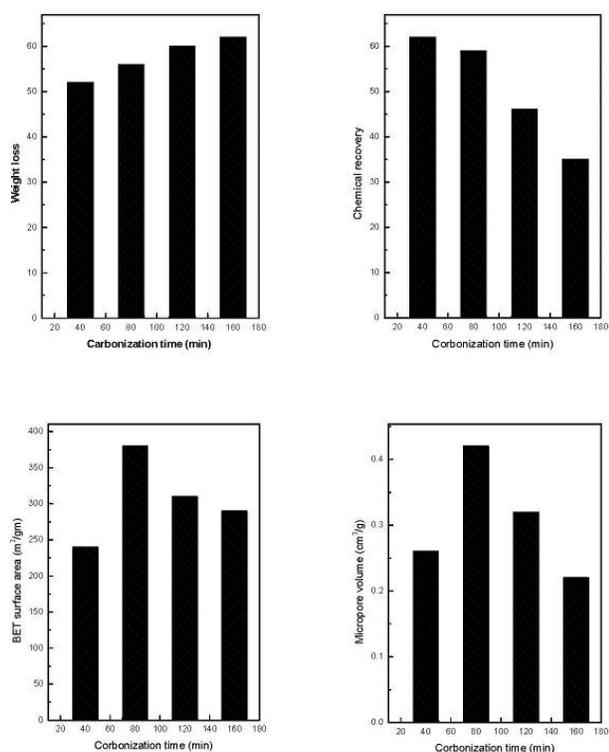
## 3. Results and Discussion

### 3.1. Characterization of the activated carbon

The results for the characterization of the activated carbon prepared from *Tamarindus indica* wood are reported here. The tests for characterization include weight loss, chemical recovery, BET surface area, and micropore volume. Effect of the carbonization time and temperature, the two most important variables during the preparation of activated carbon on these characterization parameters, are discussed.

*Effect of carbonization time.* The effect of carbonization time on various parameters like weight loss, chemical recovery, BET surface area, and micropore volume is shown in Figure 2. For these, experiments were conducted at a fixed carbonization temperature of  $500^{\circ}\text{C}$  and a chemical ratio of 100%, as some initial experiments found these are the optimum temperature and chemical ratio. It can be seen from Figure 2 that carbonization time does not have much effect on weight loss. The marginal increase in weight loss with increasing carbonization time is, however,

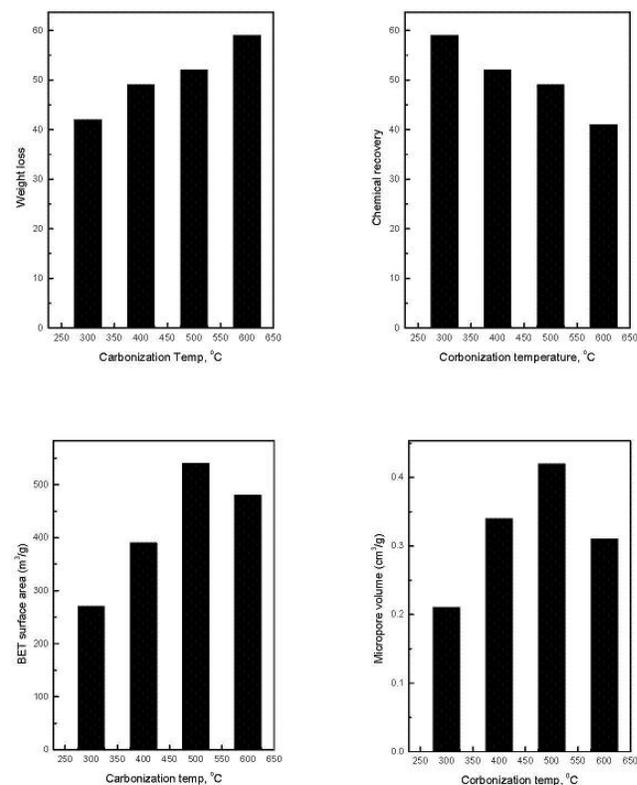
due to the release of more volatiles. The effect of carbonization time on chemical recovery has a reverse trend. It can be seen that chemical recovery values decrease with carbonization time. This might be due to the evaporation of  $ZnCl_2$  from the precursor at a longer carbonization time. However, the effect of carbonization time on BET surface area has a different trend. The surface area first increases with carbonization time and reaches its maximum at 80 min, thereafter decreasing. This decrease is possibly due to some of the pores being sealed off as a result of sintering at excessive time duration. Generally, a longer carbonization time is needed to enhance porosity as well as to clear blocked pore entrances before detrimental effects set in at prolonged times. From an initial high surface area, it deteriorated with increasing carbonization time. A similar trend is reported for the micropore volume.



**Figure 2.** Effect of carbonization time on weight loss, chemical recovery, BET surface area, and micropore volume of the activated carbon prepared from *Tamarindus indica* wood

*Effect of carbonization temperature.* The effect of carbonization temperature on weight loss, chemical recovery, BET surface area, and micropore volume is shown in Figure 3. Carbonization temperature also does not have much effect on weight loss. Overall, weight loss was found to increase with increasing temperature, resulting in decreasing yield of char as the temperature increased. This weight loss was essentially due to the devolatilization of the tamarind wood upon heating and, as expected, the quantity of volatiles increased with increasing temperature. The reason for a decreasing trend

in chemical recovery is possibly due to the evaporation of  $ZnCl_2$  at high temperature. The effect of carbonization temperature on BET surface area is also shown in Figure 3. When the carbonization temperature was 300°C, pyrolysis reactions had just commenced, thereby producing a very small BET surface area. This phenomenon was due to the inadequacy of heat energy produced at this low carbonization temperature for any substantial evolution of volatiles necessary for pore development. As the temperature was increased to 400°C and then to 500°C, more volatile matters were released progressively during carbonization, resulting in the development of some new porosity; hence, the BET surface area increased progressively. The decrease in surface area with further increase in temperature to 600°C might be due to a sintering effect at high temperature, followed by shrinkage of the char, and realignment of the carbon structure, which resulted in reduced pore areas. The trend for micropore volume was also due to the same reason given for BET surface area.



**Figure 3.** Effect of carbonization temperature on weight loss, chemical recovery, BET surface area, and micropore volume of the activated carbon prepared from *Tamarindus indica* wood

### 3.2. Removal of phenol in the multistage bubble column adsorber

The results for the removal of phenol from dilute aqueous solutions using acidified activated carbon prepared from *Tamarindus indica* wood are reported here. The effect of

the three most important parameters (contact time, activated carbon loading, and superficial gas velocity) on the removal of phenol is discussed.

*Contact time study.* To study the effect of contact time on phenol removal and to calculate the equilibrium time, experiments were carried out with constant superficial gas velocity of 0.0263 m/s, and an adsorbent dose of 1 g/l. Four different initial phenol concentrations were used for this study. It can be seen from Figure 4 that an increase in initial phenol concentration results in a decrease in phenol removal. It is clear from the figure that the rate of adsorption of phenol onto the surface of activated carbon is higher during initial stages, and gradually decreases and becomes almost constant after 160 min. It is worth mentioning that this equilibrium time (160 min) is much less compared to that of batch adsorption of phenol (300 min) reported by several other researchers [1-2]. This difference in equilibrium time can be attributed to the continuous supply of gas phase, which serves the dual purpose of agitation as well as aeration.

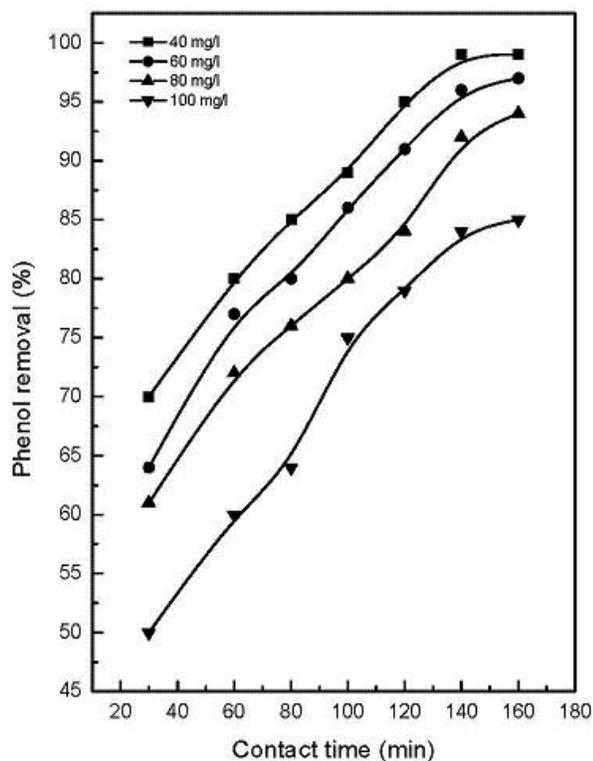


Figure 4. Effect of contact time on the removal of phenol in the multistage bubble column adsorber, superficial gas velocity = 0.0263 m/s, activated carbon loading = 1 g/l

*Effect of activated carbon loading on removal of phenol.* Figure 5 shows the effect of activated carbon loading on the removal of phenol in the multistage bubble column adsorber for four different initial phenol concentrations. These experiments were carried out at a constant

superficial gas velocity of 0.0263 m/s and equilibrium time of 160 min. It can be seen from the figure that maximum removal of phenol occurs at an activated carbon loading of 1 g/l. Further increase in loading has no effect of phenol removal as equilibrium was reached. The results have shown a sharp increase in phenol removal with increasing the carbon loading from 0.25 to 1 g/l. This is due to the increase in the number of surface active sites for phenol adsorption. At lower loading, the significantly small adsorption is possibly due to the saturation of surface active sites with the adsorbate molecules. The optimum activated carbon loading was thus found to be 1 g/l, much lower compared to the batch adsorption studies carbon loading (5 g/l) reported in the literature [1-2]. This is another advantage of treating phenolic wastewater in this multistage bubble column adsorber.

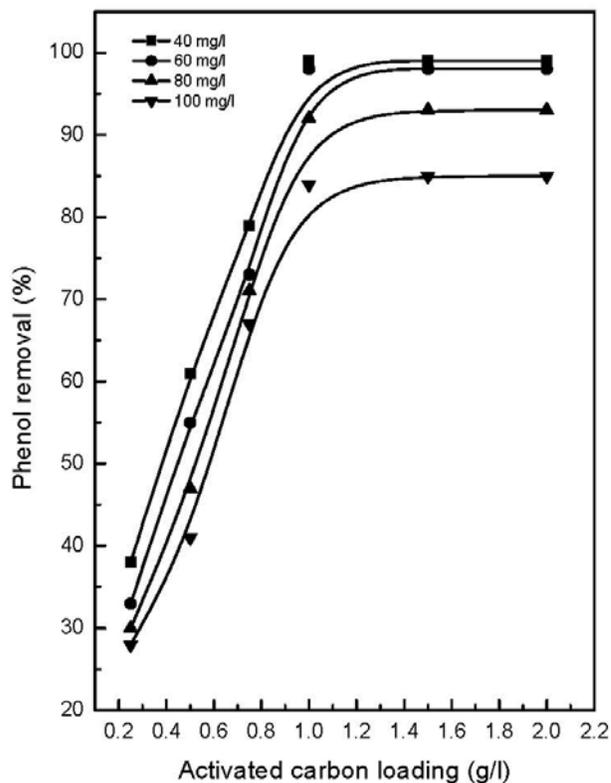


Figure 5. Effect of activated carbon loading on the removal of phenol in the multistage bubble column adsorber, superficial gas velocity = 0.0263 m/s, contact time = 160 min.

*Effect of superficial gas velocity on phenol removal.* The effect of superficial gas velocity on phenol removal is reported in Figure 6. These experiments were carried out for a carbon loading of 1 g/l. It can be seen from the figure that phenol removal increases with increasing superficial gas velocity, for a particular initial concentration of phenol. Phenol removal increases sharply at lower values of superficial gas velocity and reaches its maximum at 0.0263 m/s, after which it remains almost constant. Phenol

removal in this type of system is directly related to the interfacial area and mass transfer coefficient. In the region of lower gas velocities, due to formation of a large number of bubbles, gas holdup and in turn the interfacial area and mass transfer coefficient increases sharply. In the region of higher gas velocities, however, due to formation of larger bubbles and bubble coalescence, the rate of increase of gas holdup with gas velocity decreases, thereby decreasing the interfacial area as well as mass transfer coefficient. To check the effect of air stripping, phenol removal is studied in the bubble column adsorber without using activated carbons. However, it was found that for the range of operating superficial gas velocity, the phenol removal was 4-11%. Though there is an effect of air stripping, it is much less compared to that of adsorption by activated carbons. Usually air stripping is effective at very high gas velocities and high phenol concentrations.

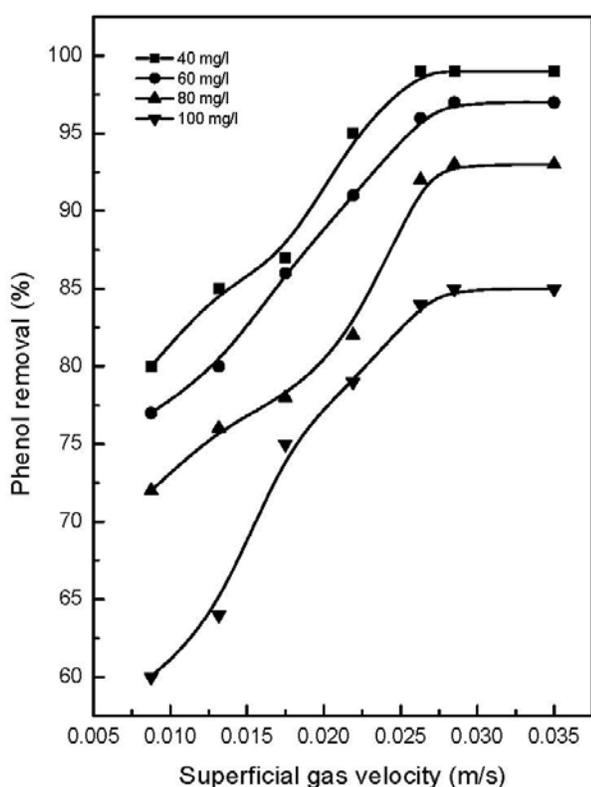


Figure 6. Effect of superficial gas velocity on the removal of phenol in the multistage bubble column adsorber, superficial gas velocity = 0.0263 m/s, activated carbon loading = 1 g/l

#### 4. Conclusions

Activated carbon prepared from *Tamarindus indica* wood by chemical activation of zinc chloride was used as the adsorbent for removal of phenol in a multistage bubble column adsorber. The results from the preparation and characterization of activated carbon showed that carbonization time and temperature are the two most important parameters that affect the quality of activated

carbon. The maximum BET surface area and pore volume obtained at optimum conditions are 540 m<sup>2</sup>/g and 0.42 cm<sup>3</sup>/g, respectively. Four different initial phenol concentrations of 40, 60, 80, and 100 mg/l were used for the study and the removal efficiency of phenol for these concentrations were 99, 97, 94, and 85%, respectively. It was found that the contact time, activated carbon loading, and superficial gas velocity are the three most important operating parameters that affect phenol removal. The equilibrium contact time and activated carbon loading for this system were found to be 160 min and 1 g/l, respectively, which were much lower compared to that of batch adsorption studies. It has been observed that the adsorption mechanism of phenol removal on activated carbon is mostly by physico-chemical adsorption. Effect of adsorbent dose, contact time, and superficial gas velocity enhances the removal efficiency. The results obtained were encouraging and confirmed that phenol can be successfully removed by adsorption in this multistage bubble column adsorber.

#### 5. References

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