

Research Article

# Performance Evaluation of an Internal-Loop Airlift Reactor for Phenolic Wastewater Treatment by Adsorption Onto Activated Carbon

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## Abstract

*In present work, an internal loop concentric tube airlift reactor unit has been designed and constructed to remove phenol from wastewater by adsorption onto commercial activated carbon (experimental reactor with internal tube of 8 cm diameter and 60 cm height while the outer tube was 80 cm height and 12 cm diameter with 6.5 L working volume). The performance of this reactor was evaluated by performing a set of experiments where the percent removal of phenol was studied at different operating variables such as inlet air flow rate, initial concentration of the phenol present in wastewater, contact time, particle size, pH and the activated carbon loading. The results show that the percent removal of phenol from wastewater increase with increasing inlet air flow rate and activated carbons loading, decreasing initial phenol concentration and slightly effected by solution pH and particle size. About 98% removal of phenol was reached in one condition (air flow rate of 12 L/min, pH= 3.5, solid loading=2v% and dp=250-600µm).*

**Keywords:** Airlift reactor, Phenolic wastewater, adsorption, Activated carbon.

## 1. Introduction

Environment pollution, especially with the dangerous and toxic chemicals, is one of the major problems faced by the developing countries. Phenolic compounds are such toxic pollutants discharged into the environment by various industrial processes, which include petroleum refineries, petrochemicals, textile, dyeing, phenolic resin manufacturing, glass fiber units, varnish industries and smelting related to metallurgical operations and involved in production of steel, resins, ceramics, fiberglass, fungicides and herbicides and metallurgical extraction, and textile (P. Saravanan *et al.*, 2009) (K. Mohanty *et al.*, 2008) (X. Jia *et al.*, 2010). Phenolic compounds may have harmful effects on both public health and aquatic life. It has high toxicity and they are carcinogenic in nature. They cause considerable damage and threat to the ecosystem. Also they are aromatic organic compounds which are rather resistant to natural biodegradation and continue to persist in the environment for a longer time. Moreover, the World Health Organization (WHO) has specified a maximum upper limit of these compounds as 1 mg/L of in drinking water. Elimination or degradation phenol components from the wastewater completely have thus become a challenging task for the scientific community working in the related field. (P. Saravanan, *et.al*, 2008) (P. Saravanan *et al.*, 2009)

Various treatment processes such as polymerization, Electro coagulation Extraction, precipitation, ozonation, adsorption, ion exchange, reverse osmosis and advanced oxidation processes have been used for the removal and recovery of phenolic compound from polluted water and wastewater.

Most of these methods have been found to be limited, since they often involve high capital and operational costs. Among the possible techniques for water treatments, the adsorption process by solid adsorbents shows potential as one of the most efficient methods for the treatment and removal of organic contaminants in wastewater treatment. Adsorption has advantages over the other methods because of simple design and can involve low investment in term of both initial cost and land required. The adsorption process is widely used for treatment of industrial wastewater from organic and inorganic pollutants and meets the great attention from the researchers. Also used in the field of removing especially for small quantities of pollutant present in large volume of fluid, this can be carried out in batch wise or continuous manner of operation. (M.N.Rashed, 2013)

In late years, the search for low-cost adsorbents that have pollutant –binding capacities has strengthened. Materials locally available such as natural materials, agricultural wastes and industrial wastes can be utilized as low-cost adsorbents. Activated carbon produced from these materials can be used as adsorbent for water and wastewater treatment (M.N.Rashed, 2013)

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Early uses of activated carbon were reported for water filtration and for sugar solution purification. Activated carbons ability to remove a large variety of compounds from contaminated waters has led to its increased use in the last thirty years (A.R.Shepherd, 1992). Activated carbon is by far the most common adsorbent used in wastewater treatment. Since, during adsorption, the pollutant is removed by accumulation at the interface between the activated carbon (adsorbent) and the wastewater (liquid phase) the adsorbing capacity of activated carbon is always associated with very high surface area per unit volume. Adsorption is a natural process by which molecules of a dissolved compound collect on and adhere to the surface of an adsorbent solid.

Airlift reactors are a special type of multiphase, pneumatic contactors. The commercial-scale effectiveness of these reactors has been proven in numerous applications, including animal cell cultures, aerobic microbial fermentations, flue gas desulphurization, water and wastewater treatment, coal liquefaction, chemical synthesis, and petroleum refining.

Airlift reactors have attracted considerable attention due to their simple construction without internal moving parts, excellent heat and mass transfer capacity, and excellent mixing properties with low power requirements. Good mixing is essential for some processes to enhance the chemical reaction rate and decrease side reactions caused by non-uniform concentration or temperature profiles. Mixing behavior is also crucial for reactor scale-up and optimal operation. (Behin, 2010), (M. L.Cancino, 2013)

Airlift systems represent a very convenient technical solution for cases where liquid circulation needs to be achieved; the system is made up of two separate sections, a riser and a downcomer. In the riser, gas is blown in at the bottom, resulting in a difference in static pressure in the two sections brought about by the different concentrations of the gas phase; this results in an overall circulation of the liquid and, depending on the operating parameters, also of part of the gas. Specific applications may require the presence of a solid phase also, which is charged in the riser column and again, depending on the operating parameters, could form a packed bed at the base of the column, be fluidized in the riser or, for the extreme case, circulate in the system. These various modes of operation have already been classified for a draft tube system but that classification can be equally applied to external airlift reactors and will not be repeated here. (R.D. Felice , 2005)

## 2. The Objective

The main objective of the thesis was the study the removing process and treatment of phenol from contaminated water using adsorption method onto commercial activated carbon in the three phase airlift reactor. Several variables during this process has been studying on the percent phenol removal such as inlet

air flow rate, initial concentration of the phenol present in wastewater, contact time, particle size, and the activated carbon loading. And to investigate three phase internal-loop airlift reactor hydrodynamic properties and to establish the mathematical model for the relationship between the gas holdup, gas flow rate, liquid circulation velocity and liquid recycle rate based on experimental data.

The purpose of using this reactor is to take advantage of the unique characteristics of this type of reactors such as internal recycling of the liquid as well as the involvement of air in the removal the process by oxidation of phenol which increases the efficiency of removal of phenol.

## 3. Experimental

### 3.1 Materials

#### 3.1.1 Activated carbon

Commercial activated carbon (AC) (supplied by Didactic company) of purity 99.9% with surface area 1050 (m<sup>2</sup>/g) and solid density 1.153 (g/ml) was used as an adsorbent. The GAC solid was milled and sieved to three size ranges 75-250, 250-600, and 600-1190 μm. the sieved activated carbon of granular shape was used in the present work to study the effect of particle size of adsorbent on the adsorption process. The activated carbon was washed before being in distilled water to remove fines and dried and drying.

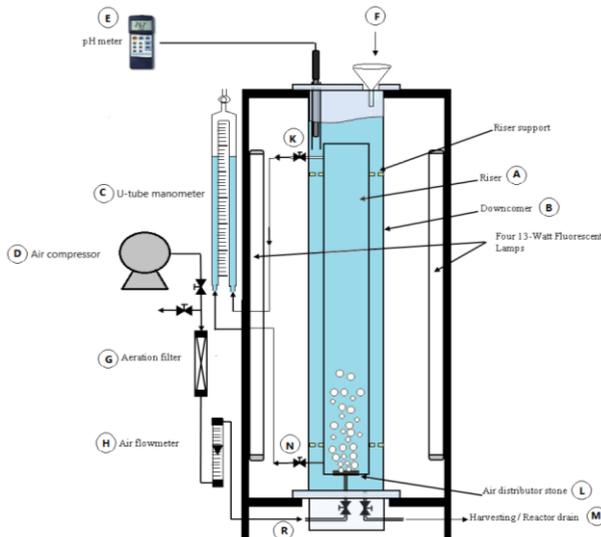
#### 3.1.2 Phenol

Analytical grade phenol (crystalline state, ≥ 99.0 % purity, formula C<sub>6</sub>H<sub>5</sub>ClO, molecular weight 128.5) was provided from Sigma-Aldrich, Inc. and used without any further treatment. Firstly phenol solution was prepared by dissolving 1 gm of phenol in 1 L of distilled water to obtain stock solution of concentration 1000 mg phenol/l. The stock solutions were diluted respectively if necessary to get the required concentrations of 50, 100, 200, and 400 mg/l.

The pH of the solutions was adjusted by addition of 0.1M HCl or 0.1M NaOH solutions as needed at initial solution pH study.

### 3.2 Experimental unit setup

The main unit of the process is the airlift reactor used in this work was internal-loop airlift reactor, shown schematically in Figure (1) consisted of two concentric columns which made up of transparent Perspex material, the ratio of the cross-sectional area of the downcomer to the riser equal to (0.29), working heights (80cm) and 3mm wall thickness, The internal tube (which is the riser(A)) is (8 cm) diameter and (60 cm) height while the outer tube(which is the downcomer (B)) is (80 cm) height and (12 cm)diameter.



**Fig.1** Schematic diagram representation of experimental equipment A. riser, B. Downcomer, C. U-tube manometer, D. Air pump and, H. Air flow meter, K,M,N-4 way valve, E. PH meter, L. Air distributor stone, G. Aeration filter, F. addition material.



**Fig.2** Photographic view of the experimental airlift reactor in use

The air is passed through the internal column to become the riser this was done by 80W, 2 bar pressure air compressor (D). The air distribution was done by small air porous stone (L) place in the center draft-tube. The height of internal tube (riser) from the bottom is 5 cm that give a free area about 251 cm<sup>2</sup>. The inlet air was passed into charcoal filter (G) before entered to the air flow meter. The air flow meter (H) using was rang (3–25 L/min), the inlet air flow rate to reactor was controlled by a valve (R) mounted on the bottom of reactor. One valve (M) was provided located at the bottom of the reactor for taking samples and monitoring the performance of the system in removal of phenol. In addition, a valve was to withdraw the processed waste and drain the reactor, as needed.

Differential pressure apparent in U-tube manometer (C) to measure the three phase pressure drop through airlift reactor, pressure taps were drilled in the reactor top (K) and in the bottom (N) of the riser in reactor.

To measure the pH for solution through the experimental using the pH meter (F) electronic mounted on the top of reactor. Figure (2) shows a photographic view of the experimental airlift reactor in use.

### 3.3 Procedure

The phenol removal experiments were performed according to the following procedure:

- 1) At the beginning of the experimental distilled water was added to the airlift reactor.
- 2) The activated carbon (AC) used in this study was purchased from local market and use in the batch experiments. Before to use, the carbons were washed several times with distilled water. Afterward they were dried and stored in desiccators until used. The experiments were conducted at solid loading from activated carbon were (0.5–4%v) added to the aqueous solution from the top of the reactor.
- 3) Artificial phenolic aqueous solution was prepared by dissolving the necessary amount of phenol (1gm) in flask contain(1L) water to obtain standard solutions of concentration (1000 ppm) and diluting to obtain initial phenol concentration ranging as 25, 50, 100, and 200 ppm and kept in a storage tank. Phenol found in wastewater from various sources in Industrials in the same range of initial concentrations as mentioned above. This solution was fed to the top of the reactor until a high of mixture (distilled water + solutions of phenol + activated carbon v %) in reactor become working level (71cm).
- 4) Air compressed from the compressor after removing oil and moisture by passing the air through charcoal filter before entered to the reactor, was fed to the stone at a controlled rate by air flow meter at air flow rates of (3–12 L/min) by valve , so that a desired flow pattern was established in the reactor.
- 5) The lowest part of the reactor below has the air distribution, where in bubbles are generated at relatively high velocities. At the beginning of bubbles have been ruptured and coalesced by imposing a flow disturbance using a stone place in the center riser. The gas-in-liquid dispersion passing through the riser is subjected to dynamic instabilities in the form of flow expansion caused circulation the fluid in the reactor.
- 6) From literature, it was found that solution pH is an important parameter that effect on the rate of removal of phenol [ (K. Mohanty *et al.*, 2008) (D.K.F.C.Al-Jiboury, 2013)]. Although from Several experiments was reported that, maximum phenol

adsorption took place at the pH of 3.5. Since, this is highly acidic range; hence, a lot of acid will be required to make the solution pH 3.5, which is not cost-effective. Hence, the pH of the reactor content was adjusted using dilute HCl added to the reactor. Also all the experiments were conducted at room temperature.

- 7) After the establishment of steady-state flow with liquid recirculation, samples were collected from the bottom of the downcomer in each run to evaluate the change of phenol concentration at suitable time interval. The samples were filtering by filtration paper and were analyzed for residual phenol concentration.
- 8) The concentrations of phenol were determined based on the spectrophotometer analysis. The phenol that have occurred calibration curve of UV-spectrophotometer was checked to correct deviations in peak height phenol calibration curve was made by collecting samples of stocks solution in the range of (10-200mg/l) and measuring the absorbency against each concentration in wave length (270nm) then plotting concentration values against.
- 9) The amount of phenols absorbed onto activated carbon was calculated according to the equation:

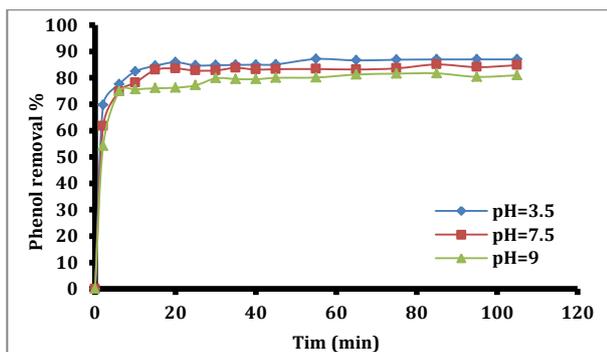
$$q_e = \frac{(C_0 - C_e)V}{W} \tag{1}$$

Where  $C_0$  and  $C_e$  are the initial and equilibrium concentrations of phenol (mg/l),  $V$  is the volume (l) and  $W$  is the weight (g) of the adsorbent.

#### 4. Results and discussion

##### 4.1 Effect of pH

The effect of pH solution at adsorption of phenol from aqueous solutions on AC was studied at three pH values (3.5, 7.5 and 9) with maintaining the other factor constant and the results are shown in Fig.(3).The other variable were fixed as superficial gas velocity at air flow rate ( $Q=6$  L/min), particle diameter ( $d_p=250-600\mu\text{m}$ ) and solid loading= $2$  v %.

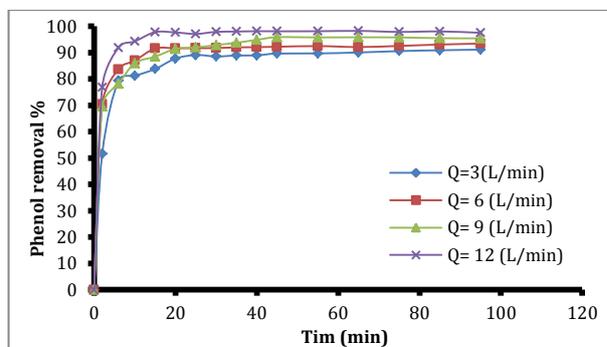


**Fig.3** Effect of pH solution on phenol removal with varied time (particle diameter =250-600 $\mu\text{m}$ , solid loading=2%v and air flow rate =6 L/min)

It is clear from this figure that the adsorption percentage removal increasing to maximum (87.12% with pH 3.5, while 85.06% for pH 7.5 and 81.69% for pH 9), the results approximated and was not much affected with pH variation and there is a slighted decrease in adsorption removal with pH increasing, this is mainly due to the dissociation of phenol molecules in the basic medium (the dissociation of phenolic compounds start at pH value between 7.13 and 9.6 (M. Ahmaruzzaman and D.K. Sharma, 2005). So all followed experiments were used at pH 3.5, this because they found that the phenols are adsorbed as monolayers by different types of activated carbon both porous and non-porous (Belo, et. al., 2013).The monolayer adsorption capacity at pH 3 is ten times more than that at pH 9 and the non-hydrolyzed molecule is more easily adsorbed on the surface of the activated carbon than the hydrolyzed molecule. Also, effect of pH might suggest a possible mechanism of chemical reaction between the reactive groups (-OH) of phenol and the surface of the activated carbon according (Z. Al-Qodah and R. Shawabkah, 2009 ).

##### 4.2 Effect of air flow rate

The effect of inlet air flow rate is showed in Fig. 4. It may be seen from this Figure that the removal percentage of Phenol increases with increasing air flow rate, At air flow rate is  $Q=3$ L/min the maximum value of removal percentage of Phenol is 91.13% , while 93.43% with  $Q=6$  L/min , 95.85% for  $Q=9$  L/min and 98.25% for  $Q=12$  L/min, also it is clear from the results that best removal percentage was in high air flow rate, therefore; air flow rate of  $Q=12$  L/min was chosen because from studied the hydrodynamic the airlift reactor that the limited the reactor maximum of air flow rate  $Q=12$  L/min. These experiments were carried out for a pH of 3.5, solid loading of 2 v% and initial concentration of phenol 50 ppm.



**Fig.4** Effect of air flow rate on phenol removal % with increase time, pH= 3.5, solid loading=2v% and  $d_p=250-600\mu\text{m}$

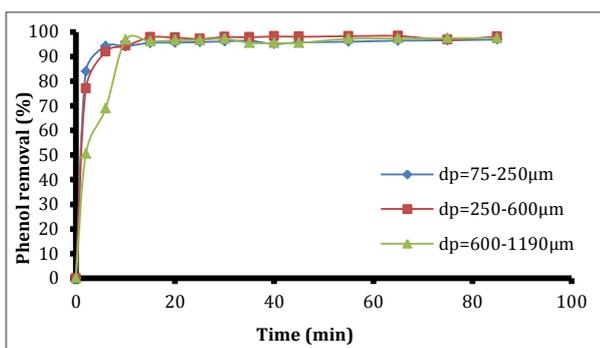
Beginning the experimental the removal increases relatively sharply with the increase time, until reaching to maximum after which it remains almost constant, the removal was occur result an increase in air flow rate carry out to increase gas velocity results in the

formation of a large number of bubbles thereby increasing the gas holdup, which in turn increases the interfacial area as well as the liquid side mass transfer coefficient (K. Mohanty *et al.*, 2008). To check the effect of air stripping phenol removal is studied in the airlift reactor without using activated carbons. It was found that for the range of operating air flow rate, the phenol removal was 5–11%. Though there is an effect of air stripping, but it is very less compared to that of adsorption by activated carbons. Usually air stripping is effective at very high gas velocities and high phenol concentration.

### 4.3 Effect of particle size

Figure 5 shows the effect of particle size on the percent removal, it is clear that the removal percentage of phenol from wastewater was with the impact of a very few when change the particle size at three rang particle size (75-250, 250-600 and 600-1190), These experiments were carried out constant pH of 3.5, solid loading of 2v%, Q=12 L/min and initial concentration of phenol of 50 ppm.

Enlargement of mean particle size does not improve the intraparticle diffusion because external mass transfer had been note the limited step of process adsorption. Therefore; the time needed to achieve equilibrium cannot be decreased at mean particle size rise than 75µm. Then the results agree with (M.L.Cancino, 2013).



**Fig.5** Effect particle size of activated carbon on phenol removal %, pH 3.5, solid loading =2v% and Q=12 L/min

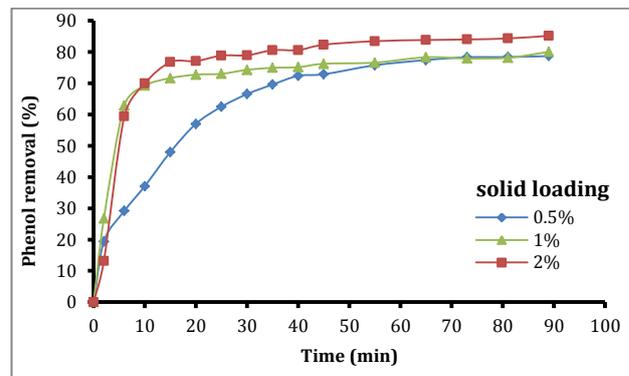
### 4.4 Effect of solid loading of AC (v%)

Figures (6, 7, 8 and 9) indicates the effect of solid loading of activated carbon (ranged as 0.5, 1 and 2 v%) on the percentage removal of phenol in an internal loop airlift reactor for four initial concentration of phenol (25, 50, 100 and 200 ppm) with pH of 3.5, Q=12 L/min and particle size (600-1190µm).

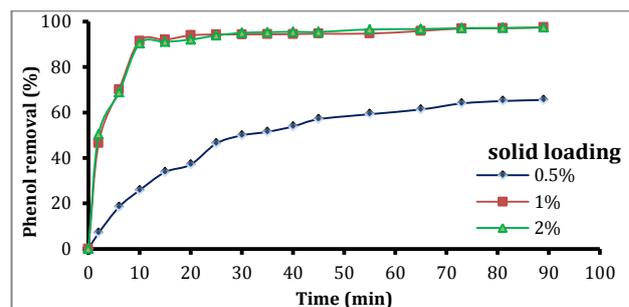
It was observed from these figures that maximum removal occur at solid loading = 2 v% of 97.37% and in general with various initial concentration of phenol when increase the solid loading from 0.5 v% to 2 v% increase the totals present removal phenol, as well as arrive the equilibrium case quickly, any sense that

whenever increase the solid loading decrease time required to removals and to arrive the equilibrium case due to increase in the number of active sites resulted in more available sites to adsorb phenol from aqueous solution.

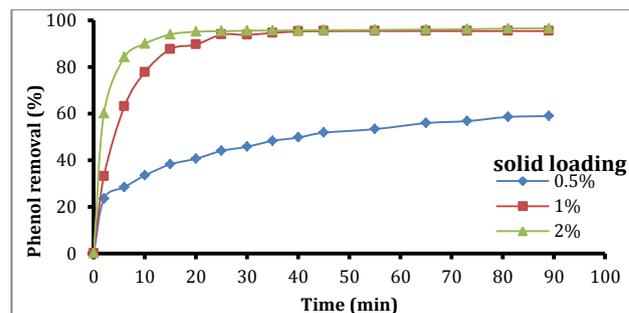
At lower doses, the significantly small adsorption is possibly due to the saturation of surface active sites with the adsorbate molecule (K. Mohanty *et al.*, 2008 and M.L.Cancino, 2013), This shows that for phenol removal in internal loop airlift reactor need the activated carbon loading quite less as compared to other batch systems.



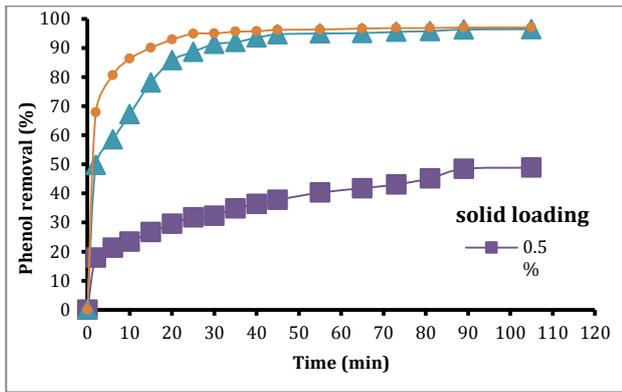
**Fig.6** Effect solid loading of activated carbon (v%) on phenol removal (%) with varied time (particle diameter =600-1190µm, pH= 3.5, Phenol conc. =25 ppm and air flow rate =12 L/min)



**Fig.7** Effect solids loading of activated carbon (v %) on Phenol removal (%) with varied time (particle diameter =600-1190 µm, pH= 3.5, Phenol conc. =50ppm and air flow rate =12L/min).



**Fig.8** Effect solids loading of activated carbon (v %) on phenol removal (%) with varied time (particle diameter =600-1190µm, pH= 3.5, phenol conc. =100 ppm and air flow rate =12 L/min)

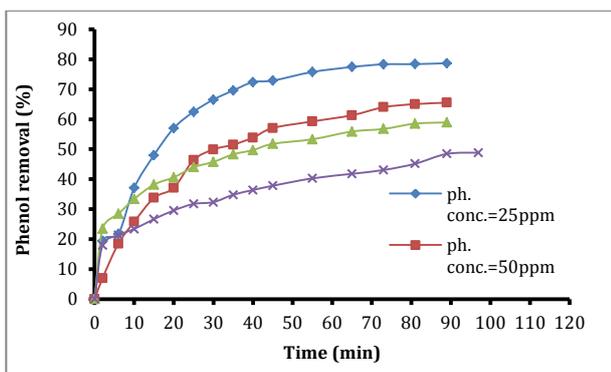


**Fig.9** Effect solids loading of activated carbon (v%) on phenol removal (%) with varied time (particle diameter =600-1190 μm, pH= 3.5, phenol conc. =200 ppm and air flow rate =12 L/min)

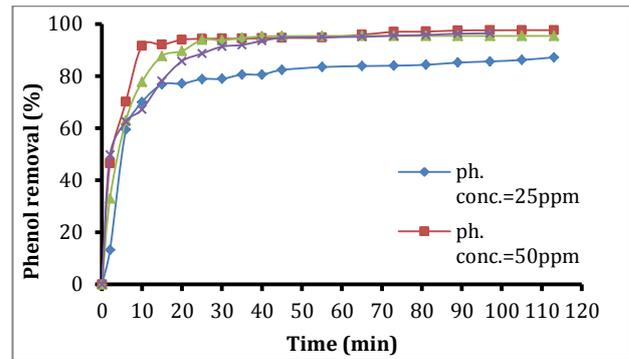
4.5 Effect of initial concentration of phenol

Fig.10 shows the percentage removal of phenol at four levels of phenol initial concentrations (25, 50, 100 and 200 ppm). The initial phenol concentration determines both the equilibrium concentration and the adsorption rate of phenol. It is clear from this figure that when increase initial concentrations at low solid loading 0.5v % decrease phenol removal percentage, because the active sites in the surface of activated carbon become full.

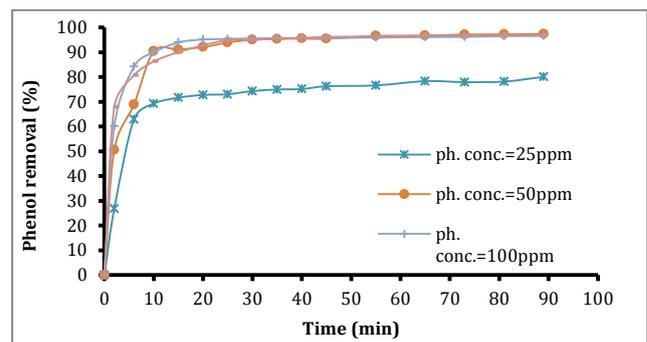
while in Figs.11 and 12 at high solid loading that an increased the time required to attain equilibrium whenever increase initial phenol concentration at approximately 25min at initial concentration of 50ppm, and 35min at 100 and 45min at 200ppm. It was also observed that rising initial phenol concentration from 25 to 200ppm increased doubled the equilibrium percentage removal. The increase of the equilibrium percentage removal is due to the increase concentration gradient which is the driving force for mass transfer of phenol through the film and pores of adsorbent particles (M.L.Cancino, 2013).



**Fig.10** Effect initial concentration of phenol on phenol removal (%) with varied time (particle diameter =600-1190 μm, pH= 3.5, solid loading = 0.5 (v %) and air flow rate =12 L/min)



**Fig.11** Effect initial concentration of phenol on phenol removal (%) with varied time (particle diameter =600-1190 μm, pH= 3.5, solid loading = 1 (v %) and air flow rate =12 L/min)



**Fig.12** Effect initial concentration of phenol on phenol removal (%) with varied time (particle diameter =600-1190μm, pH= 3.5, solid loading = 2 (v %) and air flow rate =12 L/min)

Conclusions

In the present work, the internal loop concentric tube airlift reactor was used to remove phenol from wastewater. It is claimed that the use of this type of three phase contactor was very successful for this purpose. The presence of air which in addition to being in charge of proper mixing it believed that contributes to oxidize phenolic and thus increase the removal efficiency. As compared with other batch contactors, airlift reactor needs the activated carbon loading quite less for phenol removal. About 98% as a maximum removal of phenol was achieved in the conditions of air flow rate of 12 L/min, pH= 3.5, solid loading=2v% and dp=250-600μm.

References

M. Ahmaruzzaman and D.K. Sharma, (2005). Adsorption of phenols from wastewater. Journal of Colloid and Interface Science 287, 14-2.  
 Z. Al-Qodah and R. Shawabkah, ( 2009). Production and characterization of acticated carbone from activated sludge. Brazilian Journal of Chemical Engineering, Vol. 26, No. 01, 127 - 136.  
 R. S. Austin, (1992), Granular activated carbon for water & wastewater treatment., 10-92.

- J. Behin, (2010). Modeling of modified airlift loop reactor with a concentric double-draft tube. *chemical engineering research and design*, 919-927.
- I. A. Bello, M. A. Oladipo, A. A. Giwa, and D.O. Adeoy, (2013). Adsorptive Removal of Phenolics from Wastewater; A Review. *International Journal of Basic and Applied Science*, Vol. 02 , 79-90 .
- F.C. Khalid, (2013). Adsorption of Phenol from Industrial Wastewater using Commercial Powdered Activated Carbon . *Cappadocia* , 114-123.
- M. Kaustubha, D. Debabrata and Manindra N., (2008). Treatment of phenolic wastewater in a novel multi-stage external loop airlift reactor using activated carbon . *Separation and Purification Technology* 58 , 311–319.
- L.C. Margarita, S. R. Eduardo, María T. G. G., María M. A. T., Felipe J.C.C., Oscar L.C.M., Refugio B.G.R. and Ricardo G.G., (2013). Artificial neural network modelling of phenol adsorption onto barley activated carbons in an airlift reactor. *Thsis. Universidad Autónoma de Nuevo León* , p. 89.
- N.R. Mohamed, (2013). Adsorption Technique for the Removal of Organic Pollutants from Water and Wastewater. In *Organic Pollutants - Monitoring, Risk and Treatment*, 167-194.
- S. Pichiah, Pakshirajan K., Prabirkumar S. (2008). Biodegradation of phenol and m-cresol in a batch and fed batch operated internal loop airlift bioreactor by indigenous mixed microbial. *Bioresource Technology* , 8553-8558
- S. Pichiah, K. Pakshirajan, Prabirkumar S., (2009). Treatment of phenolics containing synthetic wastewater in an internal loop airlift bioreactor (ILALR) using indigenous mixed strain of *Pseudomonas* sp. under continuous mode of operation. *Bioresource Technology* 100, 4111–4116.
- D. F. Renzo (2005). Liquid circulation rates in two- and three-phase external airlift reactors. *Chemical Engineering Journal* 109 , 49–55.
- J. Xiaoqiang, Xue W., Jianping W., Wei F., Yan J., (2010). CFD modelling of phenol biodegradation by immobilized *Candida tropicalis* in a gas–liquid–solid three-phase bubble column. *Chemical Engineering Journal* 157 , 451–465.