World Applied Sciences Journal 37 (4): 242-249, 2019
ISSN 1818-4952

DOI: 10.5829/idosi.wasj.2019.242.249

Influence of Expanded Bed Adsorption in a Combined Oxidation Stripping Process for Removing Phenol from Water

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Abstract: In present study a concentric bubble column with draft tube as synergistic system worked by the effect of combining (oxidation, stripping and adsorption) process to remove dissolved organic pollutants from wastewater. An experimental rig was built up that consist of internal tube of 7.5 cm diameter and 120 cm height extends vertically in the center of outer tube with 150 cm height and 15 cm diameter and 25 L working volume. The present technique was experimentally tested with one of the chemical pollutants namely Phenol. Compressed air is used in air stripping, hydrogen peroxide as oxidation agent in oxidation process and (rice husk, granular activated carbon GAC) as adsorbent materials putted in extended bed (7.5 × 30 cm) at upper end of internal tube. The experiments conducted at different air flow rates range (2-20) (L/min), with various residence times (5-60 min) at a various molar ratio of Phenol to hydrogen peroxide i.e. 1:10, 1:15 and 1:20. The results indicated that the optimum molar ratio between Phenol and hydrogen peroxide is 1:20 with air flow rate at 18 L/min and longer residence time (60 min) having the best performance (83.3%for GAC adsorbent and 81.6 for Rice husk adsorbent) to remove Phenol from the synthetic wastewater. From this research it can be concluded that, the design of the synergistic system are promising for less hazardous compound in the water upon treatment by this system and it is farther safe to reusing the treated water and released to the environment.

Key words: New Design • Bubble Column with Draft Tube • Hydrogen Peroxide • Rice Husk • Phenol.

INTRODUCTION

Water demand is a dynamic issue and must be considered in the development of new treatment technologies that produce an alternative clean water resource. Polluted industrial wastewater streams that are disposed into natural water resources are complex mixtures and require advanced methods rather than the conventional ones that need to be purified. Air lift loop reactors are gas-liquid or gas-liquid- solid contacting equipment's were widely used in chemical, biochemical industries, since they showed a flexibility and ability to ensure high mass and heat transfer processes during many applications [1-4]. In biotechnology (fabrication of microalgae, wastewater treatment) many sensitive operating conditions must be available like power consumption, low shear stress to cells, low contamination risk, the provision of these conditions is an advantage for the air lift loop reactors [5].

The geometric flexibility of bubble columns with draft tube reactors is constantly agent way to improve reactor effectiveness. The improvement of liquid mixing and decreasing the bubble coalescence in addition to uniformity of turbulence in cyclic pattern of flow inside the loop reactor depend on draft tube configuration [3-6]. Pollutants such as phenol may be reach micro scale level when dissolved in water so the conventional wastewater treatment processes are considered weak due the difficulty of treating the microorganisms and non-biodegradable compounds as a result of their short retention times [7].

There are many techniques for treatments of dissolved phenol in effluents wastewater. These techniques have been divided in two main types: degradation processes such as destructive oxidation with ozone, hydrogen peroxide, or manganese oxide and the most frequently used processes such as adsorption into porous solids, membrane separation, ion exchange and solvent [8-10].

The integrated biodegradation of Aerobic/anaerobic process can be applied successfully in lift loop reactor to the treatment of phenolic wastewater [11, 12].

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The high gas-liquid and liquid-solid mass transfer rates in air lift loop reactor showed a good performance in aerobic degradation of phenol and also promote the activity of the biodegradation rate [12].

When the reactor design ensure sufficient supply of oxygen with internal recirculation in sufficient way it will enable to executed successful of complex operations like remove bio-desulfurization of wastewater from oil industry [13-16]. The lack of suitable design to predict the key of the best operational parameters, cause the limited application of larger scale of air loop reactors (liquid circulation velocity, mixing intensity) [17, 18]. Therefore, there is a need to seek an alternative design to effectively remove phenol from water to a less hazardous compound and a cost-effective system.

In this study, synthetic wastewater was used for all the experimental work. Several important parameters have been investigated such as molar ratio of phenol to hydrogen peroxide, air flow rates and residence time. The purposes of this study was to employed and test the efficiency of a synergetic combining design system of oxidation, stripping and adsorption in air loop reactor arrangement at different operating condition.

A series of experiments were conducted to investigate the extent of effects on the percentage of removal phenol from wastewater. In this experimental work, molar ratio of phenol to hydrogen peroxide is 1:10, 1:15 and 1:20, the air flow rates is 7L/min, 10L/min and 18L/min. various residence times (5-60) min was used to investigate the effectiveness for removal phenol from wastewater.

MATERIALS AND METHODS

Materials: The rice husks used were gained from (Al-Shanafia) fields for rice in the south of Iraq. The normal composition analysis of rice husk is listed in Table (1). The rice husk was washed with excess amount of distilled water to remove the dissolved materials present in the rice husk bringing from the field, then boiled to removing color and other fine impurities may be found in the rice husk and then dried at 105°C until reaching the constant weight. The dried husks were stored in desiccators pending to use.

Hydrogen peroxide (H_2O_2) as oxidation agent with 35% concentration in volume/volume, supplied by GmbH Olloweg (Germany).

Analytical grade phenol (crystalline state, 99.9 % purity, formula C_6H_5ClO, 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 10, 20, 50, 100 and 150 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. Commercial activated carbon (AC) (supplied by Didactic company) of purity 99.9% with surface area 1050 (m^2/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 potential efficiency of suitable 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.

Table 1: Rice husk properties

Compound	Composition wt %
SiO_2	90.70
LOI	4.71
K_2O	2.64
P_2O_5	0.73
CaO	0.61
MgO	0.25
Al_2O_3	0.13
Na_2O	0.09
Fe_2O_3	0.06
TiO_2	0.015
S.A (m^2/g)	57.500

Experimental Setup: Figure 1 illustrates the flow diagram of the synergistic system according as proposed design.

The essential design of the synergistic effect unit is shown in Figure 2. Generally the system is a gathering of three operation, stripping, oxidation and adsorption in internal loop reactor.

This system is intended, arranged and tested in an incorporated model. An internal airlift loop reactor (Fig. 2.c) made of Perplex glass was used throughout the experiment. The outer core has a dimension of 15 × 150 cm with a concentrically located inner draft tube of 7.5 × 120 cm dimension. The working volume of the reactor is 25 L. The inner draft tube was fitted with three support legs at the upper and the lower end of the column so as to locate it in central position at any distance above the base. The upper end also containing bed (7.5 × 30 cm) having a porous adsorbent material (granular activated carbon (GAC) or Rice husk) are used to adsorb the phenol pollutant from the wastewater (Fig. 2.a).

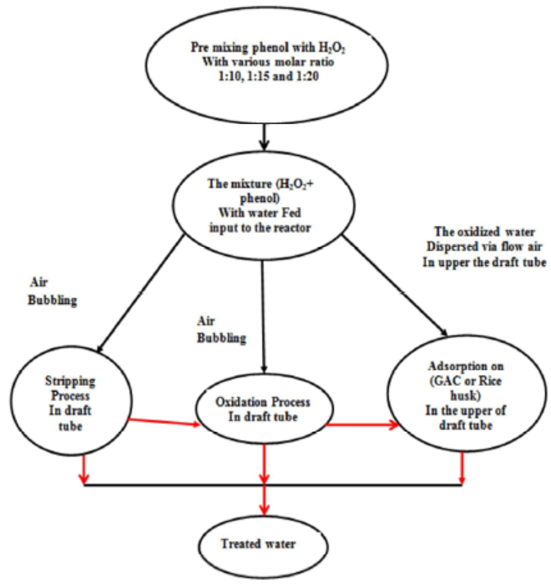


Fig. 1: Flow diagram of synergistic system.

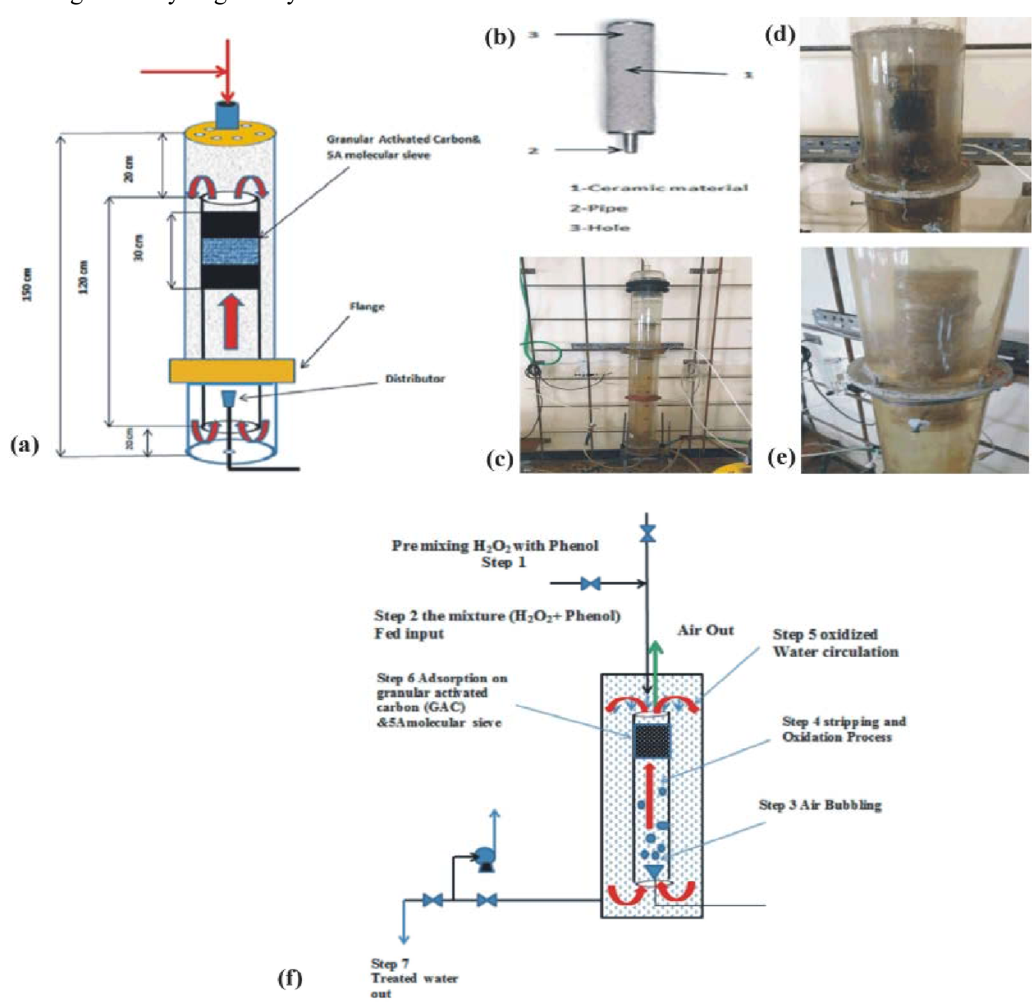
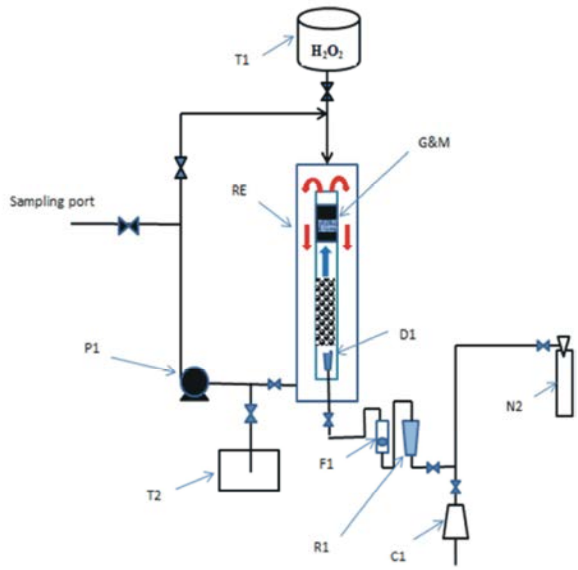


Fig. 2: (a, b, c, d and f) Design, scenario and expanded bed of synergistic system.



	Valves	P1	centrifugal pump	T1	hydrogen peroxide tank
R1	regulator	RE	reactor	D1	distributor
N_2	nitrogen cylinder	C1	compressor	T2	waste water tank
F1	flow meter				

Fig. 3: Schematic diagram of experimental rig.

The gas distributor in Fig. 2.b was constructed from a ceramic material and the type is a multi hole tuyere. The distributor has equivalent pore diameter of 0.15 mm and a free section of 80% located below the inner tube. The phenol was fed to the loop reactor using gravitational flow and the flow is pre mixed with polluted water before entering the reactor and the flow rate was controlled by the gate valves as illustrated in Figure 3. Air flow rate used during the study was range (2-20) L/min. The experiment was conducted in lab temperature (30±2°C).

Experimental Procedure: Synthetic wastewater contained 150 ppm of phenol. The oxidizing agent, 1000 ml of hydrogen peroxide (H_2O_2) was added to 4 liters of water at 25°C in a feed box and a (0.2 Kg) granular activated carbon or rice husk volume was 173.5 cm^3. The molar ratio between phenols to hydrogen peroxide was 1:20. The pump for synthetic wastewater tank was maintained at flow rate of (18 L/min) while hydrogen peroxide tank was maintained at gravitational flow rate. The pressure was kept constant atP (abs) = 2 bar. In order to get steady state for the system, the process required to run for 10 min before conduct the experiments. During the experiments the contains of the reactor was mixed by air flowing through a gas distributor located at the center of bottom of the draft tube (rise zone), causing a difference in the average density of the fluid between both zones (rise &down zone), thus inducing oxidized wastewater circulation with a defined cyclic model, to complete the adsorption process and, at the same time to complete the stripping and oxidation processes which occur simultaneously. Operating conditions like input and output flow rates from the reactor reach steady state after 10 min by using the control on the aperture of the valve located on the pipe uses for exit treated water and thus can control on the residence time. The treated wastewater was collected after 60 min at an outlet port of the loop reactor.

The first experiment was been tested for the 60 minute. Twelve samples of oxidized wastewater in loop reactor was been collected for every 5 minutes in the sample point. After that, the concentration of phenol in these samples was analyzed by a UV - spectrophotometer (U- 1800 spectrophotometer, Hitachi, Japan) at a wavelength of 270nm. The same working procedure is to be repeated with deferent contact time at a different molar ratio of phenol to hydrogen peroxide of: 1:10, 1:15 and 1:20 with air flow rates ranges 2-20 L/min and residence time of 5-60 min respectively to investigate the effectiveness for phenol removal. The experiments were performed at the natural pH of the mixture between 3.5 and 4. The removal efficiency of phenol from water was determined as follows:

The phenol removal percent was calculated for each run by the following equation:

$$E = \frac{C_{in} - C_{out}}{C_{in}} \times 100 \quad (1)$$

where E represent removal%, C_{in} and C_{out} are the initial and final concentrations of phenol in the solution in mg/L.

RESULTS

The results include the effect of synergistic of three processes (stripping, oxidation and adsorption) with the best scenario and the best control factors to obtain highest remove efficiency. These factors include the concentration ratio of the pollutant (phenol) to the concentration of oxidizing agent (hydrogen peroxide), the air flow rate and the contact time between the dissolved organic contaminate in water and the adsorbent substance that present in the packed bed upper the internal tube of the lift reactor. Figure 4 represents the effects of residence time on the removal percentage of phenol from synthetic wastewater. It is shown that the percentage of removal increases in accordance to the contact time. The percentage removal of phenol at 60 minutes for 150ppm, 50ppm and 10 ppm and using GAC as adsorbent were 83.3%, 62.6% and 29.7% respectively. During the same period (60 minutes) and concentration 150 ppm, 50 ppm and 10 ppm with using rice husk as adsorbent substance, the percentage of removal were 81.6%, 60.4% and 29.7% respectively.

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The high phenol concentration in synthetic wastewater fed to treatment process play an important factor to create a driving force for mass transfer step which takes place in both stripping and adsorption stages in addition of presence of hydrogen peroxide which increases destruction rate of organic contaminate in oxidation process by produce highly oxidative hydroxyl radicals. One of the features of this design is that performs of synergistic three operations at the same time making it very difficult to know the effect of each process separately on the percentage of removal phenol from synthetic wastewater. The scenario of the operations and the sequence input of air and the oxidative factor have a critical role in increasing the efficiency of removal. While the air is normally sparged through the distributor which position in the bottom of the center of draft tube the flow is upwards with create the high interfacial area and it is here that most of the gas-liquid mass transfer takes place, stripping process will occur for a fraction of phenol from waste water, at the same moment hydrogen peroxide (H_2O_2) disintegrate and generates a major amount of highly-reactive free radicals, such as OH• which attack the phenol in the water by oxidation reaction to break the hydrocarbon bonds and form degradation products such as alcohols, carbon dioxide and water.

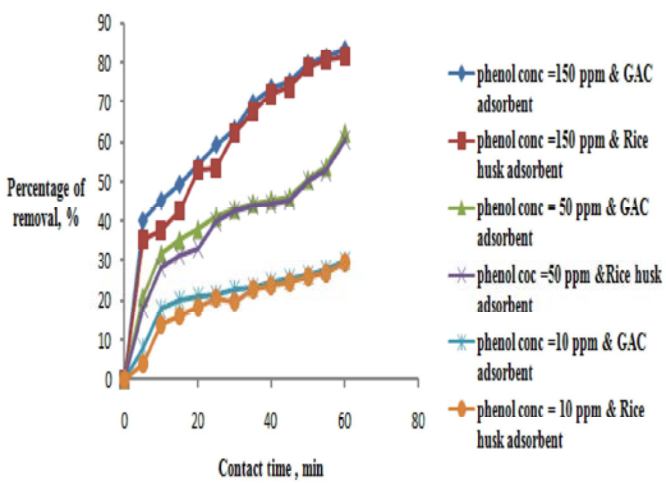

Fig. 4: Effect of contact time on the percentage of removal of phenol with different initial concentration of phenol and different adsorbent materials in air loop reactor.

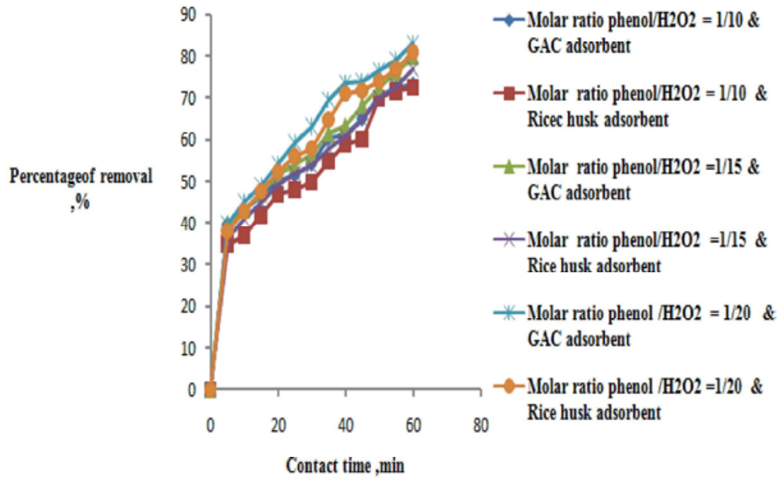

Fig. 5: Effect of contact time on the percentage of removal with different adsorbent materials and molar ratio of phenol to hydrogen peroxide.

At the same time also, when the phenol-contaminated solution passes through the activated granular carbon in the extended packed bed to induce the adsorption process and to complete the last third operation of the synergistic system. From the practical experience, we noticed that the removal rate of phenol begins to decrease after the first five minutes for two reasons. First, when the three processes begin, stripping, adsorption and oxidation, the driving force of the mass transfer is the pollutant concentration between the liquid phase (phenol solution), gas phase (air) and the solid material (the absorbed substance, GAC and Rice husk) is at the highest value and then begins to decrease gradually over time after the first five minutes of the process.

The elimination percentage of phenol reached approximately 83.3%, 81% when using GAC and Rice husk as adsorbent materials with the molar ratio of (phenol /H_2O_2) =1/20, respectively as shown in Figure 5.

The second reason is the unstable chemistry nature of hydrogen peroxide where it’s rapid disintegration in the first minutes of the beginning of the process and generates a large number of strong free radicals that react quickly to break down the phenol bonds, which increases the treatment effectiveness via oxidation process.

The effect of retention time on the percentage of removal of phenol with various molar ratio of phenol to hydrogen peroxide and different adsorbent substance is illustrated in Figure 5. The results showed that the rate of removal of phenol is enhanced at a lower initial molar ratio of phenol to the H_2O_2 when keeping the air flow rate (18 L/min) and initial concentration of phenol (150 ppm) constants. The percentages of phenol removal in 60 min for molar ratios of 1/10 and 1/15 with GAC as adsorbent were 73.2, 79.9 and when used the Rice husk as adsorbent at the same condition for time and molar ratio were 72.6,77.

The key problem in the design of the adsorption bed is the choice of a suitable empty bed contact time, (EBCT) to fully utilize the packed capacity system (GAC or Rice husk in the bed). EBCT is defined as the total volume of the granular activated carbon or Rice husk in the bed divided by the liquid flow rate and is usually expressed in minutes. So was placed (0.2 Kg) granular activated carbon or Rice husk in the top end of the draft tube which containing bed (7.5 × 30 cm) having a porous adsorbent material (granular activated carbon (GAC) or Rice husk) are used to adsorb the phenol pollutant from the wastewater, in the form of the bed rises by (30 cm) as optimal length of the mass transfer zone. The absence of a strong strategy and the credence on Inference may cause to different problems like scanty or not efficient treating. As contaminated water stream passes through a confined bed of GAC or Rice husk, a lusty case improve and it set up a mass transfer zone. This' mass transfer zone' is defined as the GAC or Rice husk bed depth required to minimize the contaminant concentration from the initial to the final level, at a given flow rate. The operation of recycling the liquid as a result of pressure or density difference due to partial or total disengagement of gas at the top of the draft tube gives full opportunity for the adsorption process to occur efficiently.

The (Figure 6) demonstrate the effects of residence time on the percentage of phenol removal from synthetic wastewater at a various flow rate and adsorbent materials.

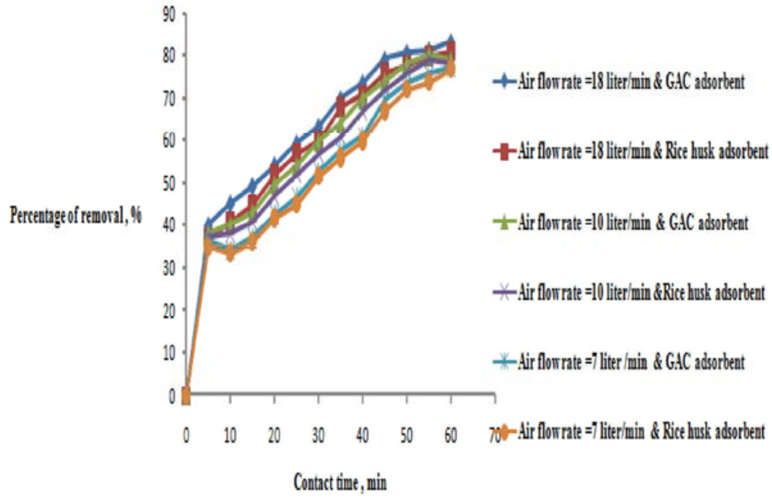

Fig. 6: Effect of contact time on the percentage of removal with various air flow rates and adsorbent material

Nearly no considerable boost in the percentage of removal of phenol from water less than 8% was registered, i.e., it is approximately stable with at contact time (from 78% to 83%).

The consolidated system that has been applied to remove the phenol by the three processes represents the competence of the apparatus. The cost of set up and running of the three equipment's (stripping, oxidation and adsorption) has been reduced to the cost of the installation and working of one device only. The material of makings this device (A Plexiglass) is inexpensive and the cost of utilization and maintenance is very simple, do not occupy a large area with, remediation period is relatively short (60 min) thus in terms of economic feasibility is considered as economically feasible.

CONCLUSIONS

In inference demonstrate the success of suggested design for elimination of organic pollutant from wastewater via employ the synergistic arrangement technique that move trio spontaneous operation (stripping, oxidation and absorption) with technical of internal loop reactor.

Consequently achieved higher percentage elimination of phenol in synthetic wastewater.

Moreover, evidenced through utilize this novel design that increase area and contact time between (phenol) and hydrogen peroxide with granules activated carbon & Rice husk through the recirculation of oxidized water between the internal tube (riser) and annulus (downcomer) in loop reactor drove to an increases in the efficiency of elimination of phenol up to reach 83.3% when using GAC as adsorbent substance and 81% when use Rice husk as adsorbent material. The outcome present that the molar ratio 1/20 for phenol to the hydrogen peroxide with 18 L/min air flow rate and using GAC as adsorbent substance gives the best performance to strip the phenol from the synthetic wastewater.

One more advantage for the current research is building an effective system for the removal of organic pollutants is characterized by low cost and simple maintenance.

The output style of current system is less hazardous compound and farther safe to be released to the environment.

ACKNOWLEDGMENTS

The researcher would like to thank the Faculty of Chemical Engineering, University of technology, Iraq, Baghdad for providing laboratory facilities to complete this Study.

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