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A new treatment design for water contaminated with phenol

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Abstract

Background: Several important designs have been applied to remove toxic and hazardous organic substances like phenol and phenol compounds from wastewater, but there is a need to seek an alternative design to effectively remove organic pollutants from water to less hazardous compounds and a cost-effective system.

Methods: A modified internal loop airlift reactor was designed to remove the organic pollutants in synthetic wastewater using an efficient and cost-effective treatment technique by means of a synergistic effect of combination oxidation, stripping, and adsorption. The influence of the current style was experimentally examined in the treatment of synthetic phenol contaminated wastewater. The practical device was tested under different airflow rates range (2-15 L/min) through gross difference retention period (5-60 minutes) at a various molar ratio of phenol to hydrogen peroxide ranging from 1:10 to 1:20.

Results: It was revealed that the preferred molar ratio of phenol to hydrogen peroxide equals to 1:20. Moreover, the airflow rate is 15 L/min with longer retention period of 60 minutes, indicating the maximum removal efficiency (89%) of phenol from the synthetic wastewater.

Conclusion: Successful removal of phenol from water by the removal efficiency of 89% boosts the success of the executed design as well as the scenario of conducting the synergistic processes (stripping, oxidation and adsorption) in one device and also increases the chances of solving environmental problems via treating wastewater before recycling and releasing it into natural water sources.

Keywords: Waste water, Hydrogen peroxide, Oxidation, Stripping, Adsorption, Phenol.

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Introduction

The development in chemical and petrochemical industries, especially in recent decades, has led to increased release of organic chemicals and inorganic pollutants into the environment, while they have harmful effects, on human health and environment (1). Many processes and applications including physical, chemical, and biological processes have been developed to treat or minimize the effects of pollution (2,3). Airlift loop reactor is an effective alternative to conventional treatment systems (stirred tank and bubble column contactors) to remove organic pollutants from wastewater (4-6). Airlift loop reactors can be classified according to several criteria, but according to the literature review it can be classified into two main categories the BC and the ALR, consisting mainly of a cylindrical or rectangular column with a gas distributor at the entrance, usually static without mechanical parts (7). Air is injected as a single energy input through a simple sparger (8-10). Problems arising during advanced processes for conventional wastewater treatment can be solved by the basic features of the air circulation reactors and increased processing efficiency as well as the possibility of combining these reactors and other processes (multistage ALR with biofilm carriers, sequencing batch bioreactor, biofilm system, membrane bioreactor, ultrasonic reactor, oxidation ditch, photo bioreactor, electro coagulation/ electrochemical systems etc) (11). For example, the engineering dimensions of the draft tube play an important role in improving fluid mixing, bubble contraction, and regulation of flow disturbance through periodic rotation in one or more rings (12-14). The integrated biodegradation of aerobic/anaerobic process can be successfully applied in the airlift loop reactor for the treatment of phenol-contaminated wastewater (15,16). Pollutants such as phenol may reach micro scale level when dissolved in water, so the conventional wastewater treatment processes are considered weak due to the difficulty of treating the microorganisms and non-biodegradable compounds as a result of their short retention times (17). In the present study, modern design

of a developed internal airlift loop reactor accompanied by a synergistic combine system of oxidation, stripping, and adsorption was used to remove of phenol from wastewater. Various substantial parameters like the molar ratio of phenol to hydrogen peroxide, airflow rate, and contact time have been investigated. The purpose of this study was to examine the effectiveness of the application of a modern design of a synergistic combining of oxidation, stripping, and adsorption for the removal of phenol from wastewater.

Materials and Methods

The granular activated carbon (GAC) was purchased from Didactic Company. With purity of 99.9%, surface area of 1050 (m^2/g), and solid density of 1.153 (g/mL), GAC was used as an adsorbent. The solid GAC was milled and sieved to three size ranges (75-250, 250-600, and 600-1190 µm,). The obtained powder was used in the present study to investigate the impact of particle size of adsorbent on the adsorption process. Activated carbon was washed with distilled water to remove the fine particles and then dried via dryer. Molecular Sieve Type 5A, "(Ca_4, 5Na3 [(AlO_2)12(SiO_2)12] •XH_2O)", (S.P.A Milano/divisione analitica), calcium with crystalline type A is an alkaline silicate. Concentrated hydrogen peroxide 35% volume/volume, was purchased from GmbH Olloweg (Germany). Pure crystalline phenol (purity: 99.9 %, formula C_6H_5ClO, molecular weight: 128.5 g/mol) supplied by Sigma-Aldrich, was used to prepare the phenol stock solution of 1000 mg/L by dissolving 1 g phenol in L distilled water. Five required concentrations of 10, 20, 50, 100, and 150 mg/L were prepared.

The pH of the solutions was adjusted by adding 0.1 M hydrochloric acid or 0.1 M sodium hydroxide solutions as needed in the study. Figure 1 presents the effects of the scenario of operating the synergistic style as a predicted in suggested design.

Figure 2 shows details of the basic design of the effect of the synergistic system for stripping, oxidation, and adsorption processes in the internal airlift loop reactor. The design was implemented, arranged, and tested in an integrated visible model.

A modified airlift reactor (Figure 2C) manufactured from the Perplex glass was used during all experiments. The reactor contains an outer tube had a dimension of 15 × 150 cm with a concentric inner draft tube that dimensions of 7.5 × 120 cm. The active volume of the reactor was 25 L. The inner draft tube was fixed via three support rods at the upper and the lower end of the column to locate it in a focal position at any distance over the base. The upper end of the draft tube with a bed (7.5 × 30 cm) containing a porous adsorbent material (granular activated carbon (GAC) and 5Å molecular sieves) was used to adsorb the phenol in the wastewater. (Figure 2A). The gas dispenser in (Figure 2B) was made from a ceramic materials and the
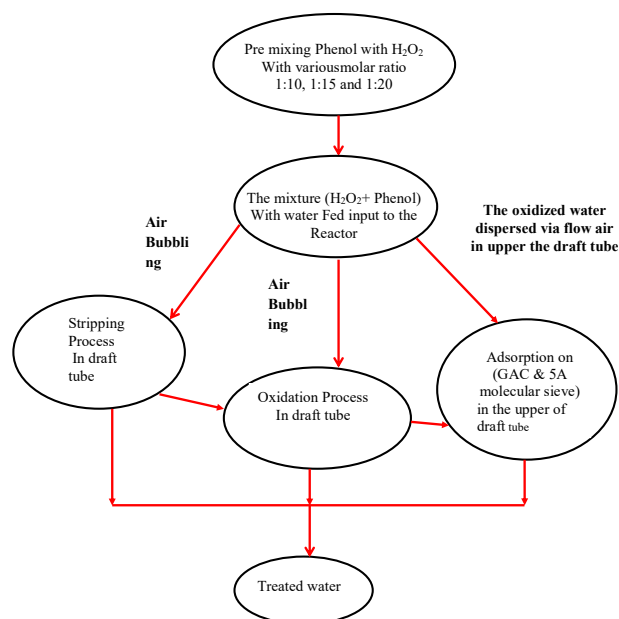

Figure 1. Planning of a synergistic system.

pattern was a multi-puncture stem. The distributor had a valent 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 while the flow was pre-mixed with polluted water before entering the reactor, and the flow rate was controlled by the gate valves as illustrated in Figure 3. Airflow rate used during the study was between 2 and 15 L/min. The experiment was conducted at lab temperature (30 ± 2°C).

Experimental procedure

Synthetic wastewater contained 150 ppm phenol. The oxidizing agent, 1000 mL of hydrogen peroxide (H_2O_2), was added to 4 L water at 25°C in a feed box, so that 0.2 kg of GAC with 5Å molecular sieve occupied space of 173.5 cm^3. The molar ratio of phenols to hydrogen peroxide was 1:20. The pumping of the synthetic wastewater into the tank was continued at a flow rate of 15 L/min while hydrogen peroxide was continuously flowing at a gravitational flow rate. The pressure was kept constant at P (abs) = 2 bar. In order to get a steady state for the system, the process required to run for 10 minutes before 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 and down zone), and inducing oxidized wastewater circulation with a defined cyclic model, to complete the adsorption process, as well as, to complete the stripping and oxidation processes which occur simultaneously. Operating conditions like input and output flow rates from the reactor reached steady
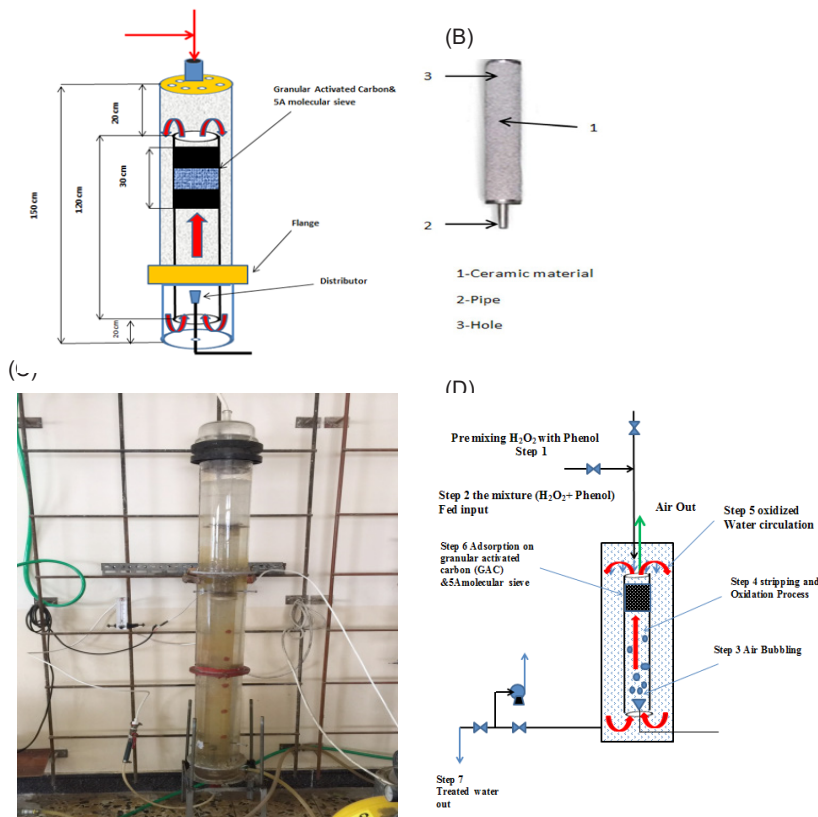
Figure 2. A schematic plan of the synergistic system.

state after 10 minutes by controlling the aperture of the valve located on the pipe using to outflow treated water, and thus, controlling the residence time. The treated

⋈	Valves	P1	Centrifugal pump	T1	Hydrogen peroxide tank
R1	Regulator	RE	Reactor	D1	Distributor
N2	Nitrogen cylinder	C1	Compressor	T2	Wastewater tank
F1	Flow meter				

Figure 3. A Schematic diagram of the experimental rig.

wastewater was collected after 60 minutes at an outlet port of the loop reactor.

The first experiment was tested for 60 minutes. Twelve samples of oxidized wastewater in a loop reactor were collected 5 minutes intervals for 60 minutes Afterwards, the concentration of phenol in the samples was analyzed by a UV-Vis spectrophotometer (UV-Vis1800 spectrophotometer, Hitachi, Japan) at a wavelength of 270 nm. The same working procedure was repeated with different contact times at different molar ratios of phenol to hydrogen peroxide (1:10, 1:15 and 1:20) with air flow rates of 2-15 L/min, and residence time of 5-60 minutes, to investigate the effectiveness for phenol removal. The experiments were performed at natural pH of the mixture between 3.5 and 4. The removal efficiency of phenol from the wastewater was determined as follows:
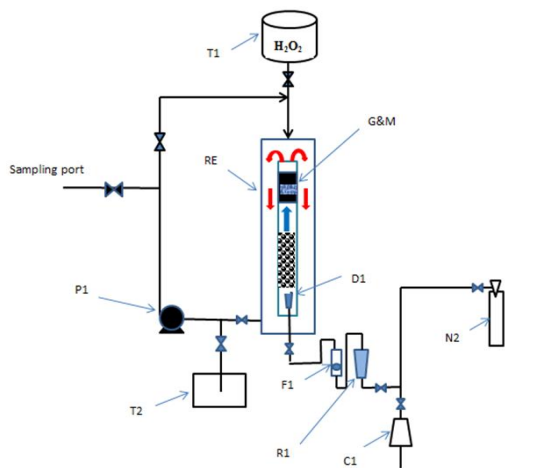
The percentage of phenol removal was calculated for each run using the following equation:

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

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

Results

The effect of synergistic action on stripping, oxidation, and adsorption processes in the scenario of the operation

of the air loop reactor with the variation in retention period on the percentage of phenol removal from synthetic wastewater are presented in Figure 4. As shown in this figure, by increasing the residence time of polluted water in the reactor, the removal rate increased. The percentage of phenol removal at 150, 100, 50, 20, and 10 ppm for 60 minutes was 89.09%, 77.8%, 63.6%, 43.43%, and 29.29%, respectively.

Figure 5 shows the effect of the variation of the molar ratio of phenolic to hydrogen peroxide, as well as the residence time on the oxidation reaction, and thus, the removal efficiency of phenol from the wastewater. As shown in this figure, the removal percentage of phenol reached approximately 89.09% at 1:20 molar ratio of phenol to H_2O_2. Analysis of the results showed that the phenol removal rate improved at the minimum initial molar ratio of phenol to hydrogen peroxide when the airflow rate (15 L/min) and the primary concentration of phenol (150 ppm) remain constants. The percentages of phenol removal for molar ratios of 1:10 and 1:15 at residence time of 60 minutes were 74.2 and 80.1%, respectively (18-21).

Figure 6 shows the effects of changes in air flow rates on the circulation time of the polluted liquid and the contact time of contaminated water with the adsorbent substances in the bed during the adsorption process, and thus, the removal efficiency of the phenol from the polluted water.
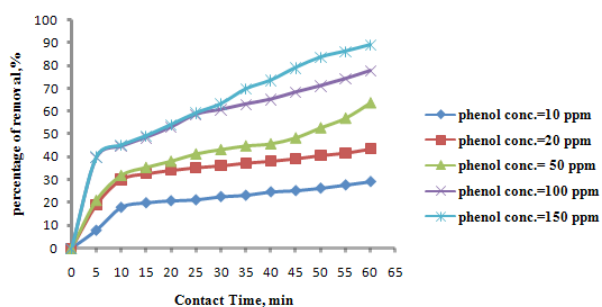
Figure 4. Influence of residence time on the ratio of removal of phenol at different initial concentrations of phenol in an air loop reactor.
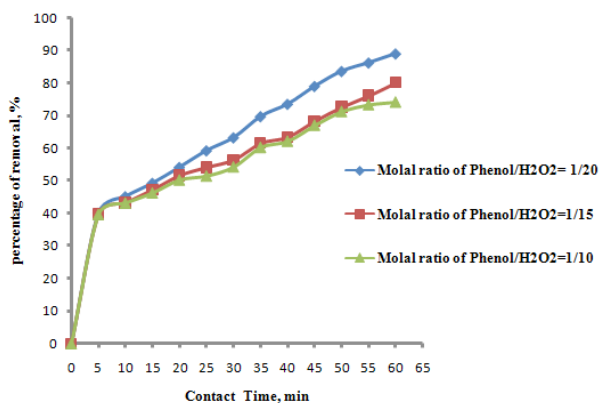
Figure 5. Influence of residence time on the amount of phenol removal with a different molar attribution of phenol to hydrogen peroxide.
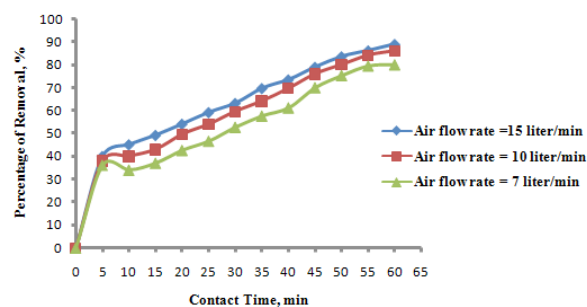
Figure 6. Influence of retention period on the amount of phenol removal with various airflow rates.

There was no increase in the phenol removal rate of more than 8%was recorded during the residence time range between 50 to 60 minutes where the percentage of removal range between 81% to 89%, i.e., its approximately constant. Figures 4 to 6 generally show the effect of the conditions and scenario of three synergistic processes of stripping, oxidation, and adsorption on one device as well as the effect of the use of an oxidizing agent (H_2O_2) alone and exclusive adsorbent material (GAC with 5Å molecular sieve) on the removal efficiency of phenol from wastewater.

Discussion

The process of treatment involves the synergy of two physical processes: stripping and adsorption with one chemical reaction. Therefore, the important factor the primary concentration of the pollutant (phenol) had an effective role in creating a large driving force to transfer the pollutant (phenol) from the wastewater to the air in the process of stripping and from the wastewater to the absorbent material (activated carbon particle and molecular sieves) through the process of adsorption, as well as in increasing the rate of phenol degradation in the oxidation process. The characterization of the design of the reactor in this study (synchronization of occurrence of the three processes, and the scenario of the process) had two important effects: The increase of the removal efficiency of phenol from wastewater was considered as a positive effect and inability to determine the percentage of the impact of each process alone on the removal efficiency was considered as a negative effect. When the air flows into the air loop reactor through the pore distributor to create a good mixture of wastewater and hydrogen peroxide solution, hydrogen peroxide decomposes, forming a large number of OH· free radicals that will attack phenol in the wastewater to break down the hydrocarbon bonds in the oxidation reaction and form the decomposition outputs such as alcohol, water, and carbon dioxide. Air passing through the distributor will dispersed the wastewater, creating a difference in the densities of the air and the dispersed wastewater, resulting in a constant rotation velocity of wastewater that will increase the contact time with the adsorbents materials in the bed extended on the

top of the internal tube of the loop reactor, which leads to an increase in the efficiency of the adsorption process. Figure 4 shows that the removal rate of phenol at different concentrations of the feed input of the internal loop reactor was higher for the first five minutes compared to the next 5 minutes of the occurrence of the triple synergy processes stripping, oxidation, and adsorption because of the highest difference in phenol concentration between wastewater and air in the process of stripping and phenol concentration in wastewater and absorption materials in the adsorption process in addition to the presence of large numbers of free radicals OH$^\cdot$ at the beginning of the dissolution of hydrogen peroxide for the first 5 minutes of the oxidation reaction time.

One of the most important factors for the success of the adsorption process is the capacity of the bed packed with activated substances (activated carbon and molecular sieves), as well as the empty bed contact time (EBCT), therefore, to obtain the highest removal rate of phenol from wastewater, the full size of the packed bed should be used.

Therefore, the engineering dimensions of the bed (7.5 × 30 cm) represent the optimum size of the mass transfer area in the presence of 0.2 kg of activated granular carbon and 5Å molecular sieves. The flow of water contaminated with phenol at an appropriate rate through the confined layer of GAC with molecular sieves (5Å) in the packed bed creates a dynamic state that establishes a mass transport area used to reduce the concentration of phenol from the initial level to the final level through recycling contaminated water with the different pressure or density resulting from the partial or total disengagement of the gas from the contaminated water at the top of the draft tube.

This process gives opportunity to complete the adsorption process efficiently. The design of the phenol removal system with the synergistic effect of three processes of stripping, oxidation, and adsorption reduced the cost of designing, constructing, operating, and maintaining three devices in one single-reactor system. The design has been taken into account the requirements of economic feasibility by using the inexpensive material (Plexiglass) to manufacture the phenol removal reactor, also due to its ease of operation and maintenance as well as the less space occupied at a relatively short treatment period (60 minutes), it can be concluded that this system is economically feasible.

Conclusions

By applying the styling idea in a practical way and according to the results, the following cases can be concluded:

1. Successful design of an air loop reactor that works synergistically for more than a physical or chemical process in one device.
2. The success of the synchronization scenario of the occurrence of the processes of stripping, oxidation,

and adsorption in terms of the selection of operating conditions and the concentration of pollutant to oxidant factor, air flow rate, treatment time, and pre-mixing of contaminated water with the oxidizing agent.
3. The removal efficiency of the organic pollutant (phenol) from the water was obtained to be 89% under the following operational conditions: Air flow rate = 15 L/minutes, the ratio of phenol to oxidation agent = 1:20, and the residence time of 60 minutes.
4. The use of hydrogen peroxide alone as an oxidizing agent is an economically feasible method.
5. In terms of economic perspective, the design is practically done using inexpensive manufacturing materials, it occupies less space, and its maintenance is simple, therefore, it is economically feasible.
6. The outputs of the synergistic system are less hazardous organic compounds in the treated water, which their release in the environment is safer.

Ethical issues

Both researchers involved in the completion of this research and certify that all data collected during the study are presented in this manuscript, and no data from the study has been or will be published separately elsewhere.

Competing interests

The authors emphasize the full compatibility of opinions and interests, which resulted in the completion of the research in its current form.

Authors' contribution

Authors participated in all aspects of the research like data collection, analysis, interpretation, and manuscript approval.

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