A MODIFICATION OF A TRIO EFFECTS PORTABLE WATER TREATMENT SYSTEM

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ABSTRACT: *The present work is an attempt to planning and implementation of an innovative design to treat organic pollutants in wastewater at an efficient and inexpensive treatment techniques by apply the synergistic system technique to the processes used in any industrial application (oxidation, stripping and adsorption). The present work introduced a novel and combined design that merges between the two methods (pulsation bubble column and air lift loop reactor) a portable assembly that is suitable for domestic and rural areas use. The effectiveness of the present technique was experimentally tested with one of the chemical contaminants namely methyl tert-butyl ether (MTBE). The experimental rig experiments conducted at different air flow rates 6, 8 and 10 (L/min), with total difference residence time (pulsation time in bubble column and retention period in air lift reactor) (2-25 min) at a difference molar ratio of MTBE to hydrogen peroxide i.e. 1:10, 1:15 and 1:20. The results indicated that the optimum molar ratio between MTBE and hydrogen peroxide is 1:20 with air flow rate at 10 L/min and longer residence time (25 min) having the best performance (90%) to remove MTBE from the synthetic wastewater. From this research it can be concluded that, the overall design achieves the objectives by completion. The combine system effectively removes MTBE from waste water. Hope the environmental problem also will be reduced by treating the waste water contain MTBE before it is released to natural water sources.*

Keywords: *New design, air loop reactor, oxidation, stripping, adsorption, methyl tert-butyl ether.*

1. INTRODUCTION

Water and wastewater treatment processes become increasingly important with diminishing natural water resources, the gradual increase in chemical pollutants in aqueous systems due to the rapid growth of population and industrial activities. Generally, the treatment technologies and design to handle industrial wastewater can be divided into three categories, namely: physical, chemical, and biological processes. Usually, physical treatment may include several processes such as sedimentation, flotation, filtering, air stripping, ion exchange, adsorption, and other processes; that accomplish the removal of dissolved and undissolved substance without necessarily changes in their chemical structure.

Air stripping is an effective technique utilizing the flow of air jet through the contaminated water that allows the volatile organic compound to transfer from the liquid phase to the gas phase. This technique is considered to be complicated due to high air-to-water ratio requirement (>200/1 L for 95% removal) [1-6]. On the other hand, adsorption of chemical contaminates through the use of solid materials such as activated carbon powders is another technique that already applied commercially in many industrial fields. In certain applications (removal of MTBE for example), the main concerns of using such technique for highly purified water resulted high process costs [7-10]. Feed stream have proven to be problematic; as the bacteria are only accumulated in the entry region of the bed and bio film is developed which leads to loss of bioreactor performance due to blockage of the bed, flow channeling and bypassing [17, 18]. Current technologies, including ozonation, microbiological methods, and traditional activated carbon filtration are not sufficient to remove MTBE from wastewater or drinking water [19, 20].

This encourages many authors through the past decades to change, modify and combine the existing wastewater treatment techniques to fulfill the growing requirements and regulation. The bubble column and airlift loop reactors are pneumatically agitated and often employed in bioprocesses where gas-liquid contact is important.

The role of the gas is to provide sufficient contact with the liquid for efficient mass transfer processes such as absorption or desorption and to provide energy through gas expansion or bubble buoyancy for liquid mixing.

In these two pneumatically agitated reactors, gas is usually sparged through the bottom and the buoyancy of the ascending gas bubbles creates mixing. Airlift loop reactors can be classified into two groups: internal-loop and external-loop airlift reactors. An internal-loop airlift reactor contains a vertical baffle or a draft tube by which a loop channel for fluid circulation is formed in the reactor. An external-loop airlift reactor consists of two vertical tubes (a riser and a down comer) which are connected by horizontal connections at the top and bottom [21].

Several modifications have been proposed to improve the conventional airlift loop reactor, and some of them have already been adopted. Modified airlift loop reactors include the inverse fluidized airlift, reactors with static mixers, helical flow promoters, and perforated draft tube; as you can find in any text book "Biochemical Engineering and Biotechnology". These designs are such that there is a continuous generation of bubbles, rupture and regeneration. This system was equipped with different spargers (multiple orifice and single orifice) together with a screen at various stages to encourage the continuous regeneration of bubbles. MTBE is a suspected carcinogenic agent, which poses a significant health threat to people. In addition, it gives a very unpleasant taste to drinking water when present even in very low concentrations and reduces the water quality. The potential environmental hazards associated with MTBE have led (US-EPA) to issue a Drinking Water Advisory on it whose levels do not exceed 20-40 μg/L [22].

The present work introduced a novel and synergistic design that merges between the two methods (pulsation bubble column and air lift loop reactor) is introduced to remove MTBE pollutants from drinking water. Several important parameters have been investigated such as the molar ratio of MTBE to hydrogen peroxide, air flow rate, pulsation time and hydraulic retention time.

The aim of this investigation was to check the feasibility of using a new design of a combined system of pulsation bubble column and air lift loop reactor that containing stripping, oxidation, inverse fluidized and adsorption processes for the removal of organic pollutants from wastewater.

2. EXPERIMENTAL DETAILS

Raw Materials

The following chemicals were used:

(a) Methyl tert-butyl ether (MTBE) ($C_5H_{12}O$), with a molecular weight of 88 g/gmol, and purity greater than 99.8%, supplied by chemical material store in the laboratory of UMP, Permula Chemicals Sdn.Bhd. (Malaysia) was employed as adsorbate.

(b) Hydrogen peroxide (H_2O_2) as oxidation agent with 30% concentration in volume/volume, supplied by Wellmex Sdn. Bhd. Company (Thailand).

(c) Granular activated carbon (GAC) (HmbG chemicals Activated Carbon Corporation, China) as adsorbent is having a particle size, 18-30 mesh fraction, particle density, 0.69 g/cm^3 and true density, 1.17 g/cm^3.

Experimental set-up

Figure 1 shows the flow diagram of the synergistic system function as expected from the proposed design. The main design of the trio-effect unit is shown in Figure 2. Generally the system is a combination of pulsation bubble column and internal loop reactor processes. This system is designed, arranged, and tested in an integrated model. The system comprises (presented in Figure 2) a combination of pulsation bubble column and internal loop airlift reactor, in the pulsation column two main process take place first is the air stripping and the second is oxidation process, the column is equipped with solenoid valve which opens and closes at the top, and which in turn connected electrically with at least two timers. The internal loop reactor, connected to pulsation column at the base through a perforated pipe. The wastewater and hydrogen peroxide feed into the pulsation bubble column continuously by two dosing pumps (P1,P2) and 2 bar of compressed air (2 bars) provided to the pulsation column through a diffuser located at 50 cm above the base. When the solenoid valve opened, the compressed air mixed the wastewater and H_2O_2 vigorously simultaneously perform the air stripping and chemical oxidation processes. When the solenoid valve closed, the compressed air creates pressure difference between pulsation column and the air lift loop reactor, and as a result the oxidized wastewater flows from pulsation bubble column to the air loop reactor through the perforated pipe. The open and close solenoid valve positions were repeated periodically in a steady manner, and the quality of the treated water was observed in a sampling point (1). The internal loop airlift reactor, having a draft tube vertically extended from a center of the reactor, and an absorbent material bed circumference the draft tube with a space is provided at a bottom of the reactor. The draft tube has an opening at a first end and a second end. To create the space between the draft tube and the bottom of the reactor, a support is fitted on the side walls of the reactor such that there is a distance above the bottom of the reactor.

The position of the support is adjustable to a required height for the purposes to achieve a space between the draft tube and the bottom of the reactor.

Each support contains 16 holes the diameter of each hole is 1 mm these holes allow water to pass through it and does not allow the passage of granulated carbon because it has small size 1 mm compared to the size of granulated carbon 3-4 mm. Supports working on installing the inner tube from the side and to carry the granulated carbon by another. An inlet port of the combination of inverse fluidized and internal loop airlift reactor for entering a contaminated water with hydrogen peroxide into the reactor.

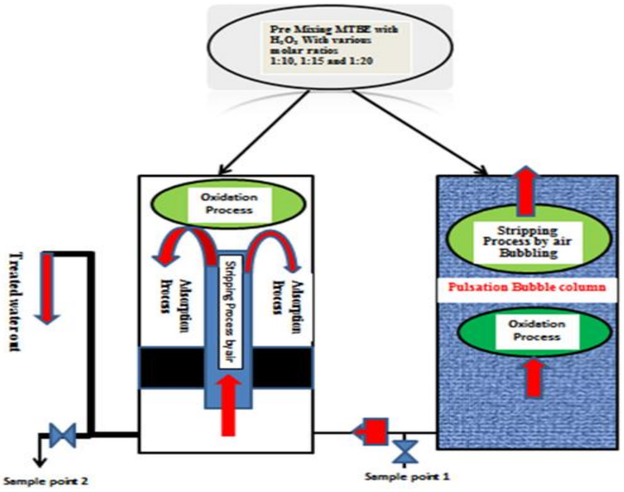
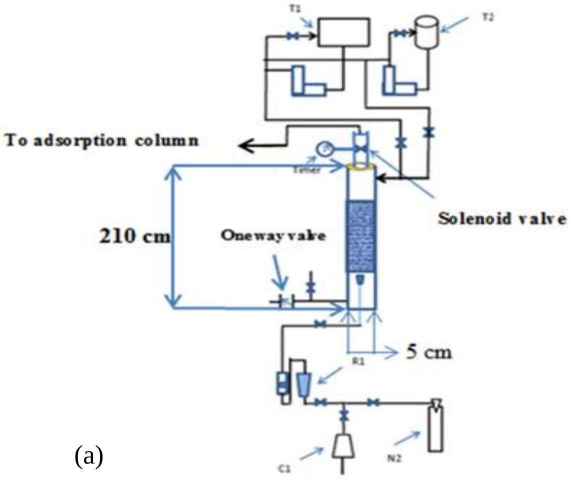


Figure 1: Flow diagram of synergistic system.



(a)

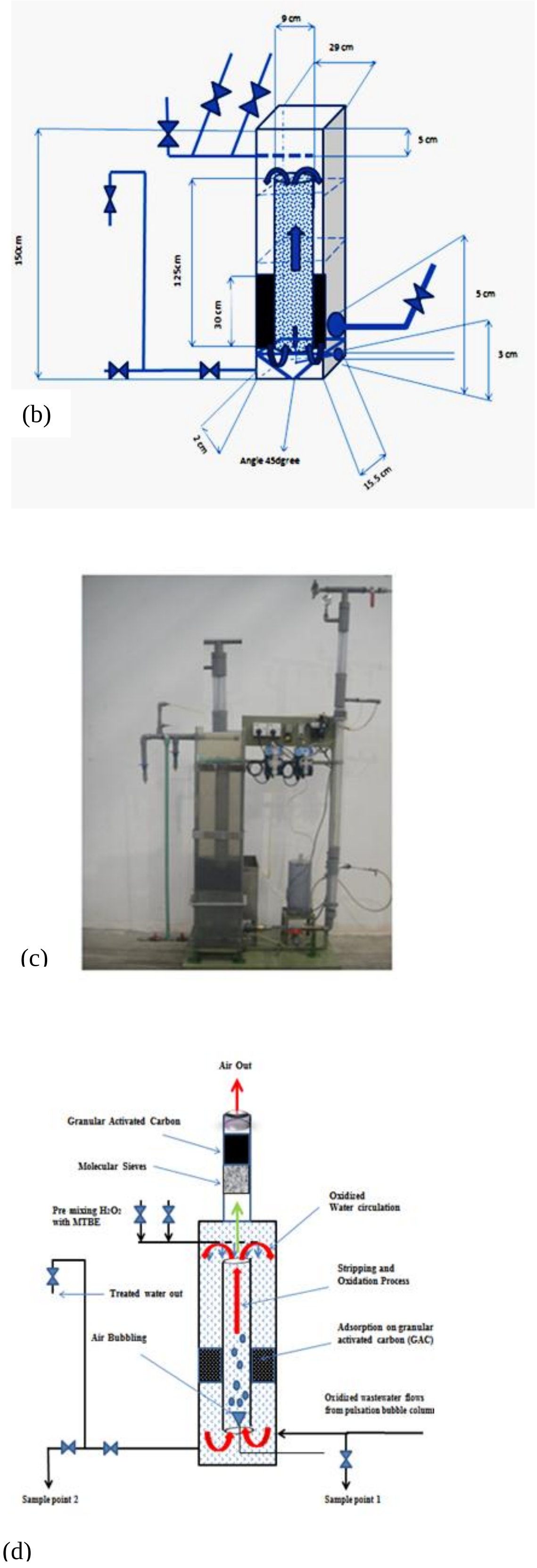
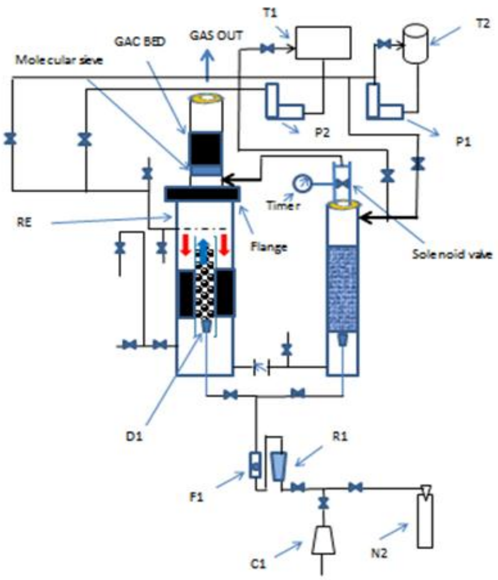


Figure 2: Design and scenario of synergistic system.



⋈	Valves	P1, P2	Dosing pumps	T1	waste water tank	F1	flow meter
R1	regulator	RE	reactor	C1	compressor	T2	hydrogen peroxide tank
N2	nitrogen cylinder	D1	distributor				

Figure 3: Schematic diagram of experimental rig.

A dosing pump (P1, SEKO, chemical dosing pump, China (mainland)) is used to provide hydrogen peroxide into the pulsation bubble column and the loop reactor in order to mix and react with the oxygenate compound in the water before the inlet port as illustrated in Figure 3. During the same period, another pump (P2, SEKO, chemical dosing pump, China) is used for receiving oxygenates contaminated water and providing oxygenates contaminated water with hydrogen peroxide into the pulsation bubble column and loop reactor by the inlet port.

Oxygenates contaminated water, and hydrogen peroxide are fed continuously into the pulsation bubble column and loop reactor. The mixture oxygenates contaminated water with hydrogen peroxide is fed to the draft tube by the opening of the first end in a gravity direction of the reactor. The purpose of introducing the air into the draft tube of the reactor is to provide the mixing of the mixture of oxygenated contaminated water with hydrogen peroxide in a counter direction. In the present study, a multi holes distributor is used for allowing compressed air to spare at the bottom of the draft tube. Air compressor (C1, Hitachi, super oil free bebicon, 4LP-7SAOA, Japan) is provided to supply the required amount of compressed air, particularly nitrogen gas into the draft tube by a gas flow meter (F, LZM-4T.6T.8T Panel Mounted Acrylic Flow Meter, Malaysia). The compressed air (2 bar) was used to mixed, the mixture of contaminated water with hydrogen peroxide vigorously, and, as a result, the air stripping and oxidation reaction between MTBE and hydrogen peroxide occurred within the draft tube. During the air stripping and chemical oxidation processes, the mixture of contaminated water and hydrogen peroxide is overflow from the draft tube due to the compressed air in a counter direction. The mixture of contaminated water and hydrogen peroxide is then directed from the draft tube to the absorbent material bed which extends in the circumference of draft tube via gravity force. It is desirable that a height of the draft tube is positioned relatively higher than the absorbent material bed such that a density difference is formed between the draft tube and the absorbent material bed for circulating contaminated water within the combination of inverse Fluidized and internal loop airlift reactor. With the assistance of the valves, the level of the mixture of contaminated water with hydrogen peroxide as well as air is maintained at a constant level within the combination of inverse fluidized and internal loop airlift reactor.

Experimental procedure

Synthetic wastewater contained 2 ppm of MTBE. The oxidizing agent, 100 ml of hydrogen peroxide (H_2O_2) was added to 8 liters of water at 25°C in a feed box, and a (5 Kg) granular activated carbon volume was 4273.5 cm^3. The molar ratio between MTBE to hydrogen peroxide was 1:20. The pump for synthetic wastewater tank was maintained at 100% flow rate (0.3 L/min) while the dosing pump for hydrogen peroxide tank was maintained at 80% flow rate (0.24 L/min). The pressure was kept constant at P (abs) = 2 bar. In order to get steady state for the system, the process required to run for 90 min before conduct the experiments. The first experiment will be tested for the timer at 30 seconds open valve + 5 seconds closed valve. Five samples of oxidized wastewater in pulsation column will be collected for every 2 minutes in the sample point 1. After that, the concentration of MTBE in these samples was analyzed by a UV - spectrophotometer (U- 1800 spectrophotometer, Hitachi, Japan) at a wavelength of 340 nm. The same working procedure is to be repeated with timer using 40 seconds open valve + 5 seconds closed valve, 50 seconds open valve + 5 seconds closed valve, 60 seconds open valve + 5 seconds closed valve, 2 minutes open valve + 5 seconds closed valve and 3 minutes open valve + 5 seconds closed valve.

During the experiments when the oxidized wastewater pass to the air lift loop reactor the density gradient between the aerated water in the draft tube and the down-comer creates the liquid circulation at stable form and forces the liquid to flow downward into the annular space through granulated activated carbon (GAC) 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 90 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 110 min at an outlet port of the combination of inverse fluidized and internal loop airlift reactor. After 110 min, ten samples were collected for every 2 min at sample point 2. The concentration of MTBE in these samples was analyzed by a UV - spectrophotometer (U- 1800 spectrophotometer, Hitachi, Japan) at a wavelength of 340 nm. The procedure was repeated with different residence time at a different molar ratio of MTBE to hydrogen peroxide of: 1:10, 1:15 and 1:20 with air flow rates of 7, 8 and 10 L/min and residence time of 2-25 min respectively to investigate the effectiveness for MTBE removal. The experiments were performed at the natural pH of the mixture between 6.5 and 7.5. The removal efficiency of MTBE from water was determined as follows:

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

Where E is MTBE removal efficiency (%), C_{in} and C_{out} are the initial and residual concentrations of MTBE in ppm, respectively. The pH was measured, and all samples were collected in the cube vet for analysis.

3. RESULTS AND DISCUSSION

The first part of the results includes the study of the effects of the ratio of MTBE to H_2O_2, the concentration of the MTBE in synthetic wastewater, pulsation time and the volumetric flow rate of air on the efficiency of the removal process. The effect of synergistic of two process stripping and oxidation process take place in pulsation bubble column. Figure 4 shows the effects of pulsation time on the removal percentage of MTBE from synthetic wastewater. It is shown that the percentage of removal increases in accordance to the pulsation time. The percentage removal of MTBE at 7 minutes for 2ppm, 1.5ppm, 1ppm, 0.8ppm and 0.5ppm were 8.09%, 6.78%, 6.36%, 3.43% and 1.29% respectively. For higher concentration of MTBE in synthetic wastewater, the percentage of removal is higher than compared to the low concentration of MTBE. The high concentration of MTBE in contaminated water helps to create a great driving force to the mass transfer process which takes place in stripping process. In addition to increasing the degradation rate in the oxidation process therefore the efficiency of removal MTBE increasing with increases the concentration of MTBE in contaminated water.

The figure also represents the impact of the effectiveness of the removal of MTBE from synthetic wastewater for two operations at the same time, which are: oxidation and stripping.

It is very important to clarify the scenario of occurring of two processes and the synergy quotient in these processes at the same time, therefore the features of this new design is the absence of the possibility to know the impact of each individual process on the percentage of removal MTBE from synthetic wastewater. At the moment that the air passes through the porous distributor to the pulsation bubble column to make a good mixing of the contaminated waste water with hydrogen peroxide, and according to the principle of the difference in concentration of the organic contaminant between the aqueous phase and the air phase (between the concentration of the MTBE in waste water and that in the air), stripping process will occur for a fraction of MTBE from waste water at the same time hydrogen peroxide (H_2O_2) decomposes and generates a large amount of highly-reactive free radicals, such as OH• which attack the MTBE in the water by oxidation reaction to break the hydrocarbon bonds and form degradation products such as alcohols, carbon dioxide, and water.

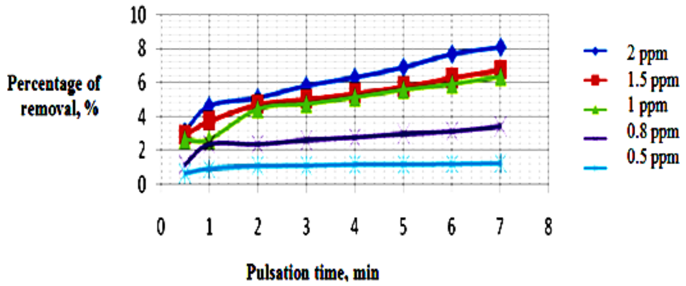


Figure 4: Effect of pulsation time on the percentage of removal of MTBE with different initial concentration of MTBE.

The effect of pulsation time on the percentage of removal of MTBE with different molar ratio of MTBE to hydrogen peroxide is illustrated in **Figure 5**. The results showed that the rate of removal of MTBE is enhanced at a lower initial molar ratio of MTBE to the H_2O_2 when keeping the air flow rate (10 L/min) and initial concentration of MTBE (2 ppm) constants. The percentage removal of MTBE at 7 minute for molar ratio 1:10, 1:15, 1:20 were 11.01%, 10.23% and 9.61%. The levels of H_2O_2 had two opposing effects on the reaction rate [24-26].

Increasing initial hydrogen peroxide concentration enhances the oxidation process up to a certain level at which hydrogen peroxide inhibits the degradation. As a result of higher hydrogen peroxide concentration, hydrogen peroxide acts as a free radical scavenger itself, thereby decreasing the hydroxyl radical's concentration. Hence, there is an optimum H_2O_2 dosage, for example, the molar ratio of H_2O_2 to the contaminant (MTBE). A great reduction in the degradation rate is expected at much higher H_2O_2 concentrations. This depends on the hydrogen peroxide presence indicating that oxidation by radical attacks (i.e. OH• and • OOH) is the major mechanism by which MTBE is degraded.

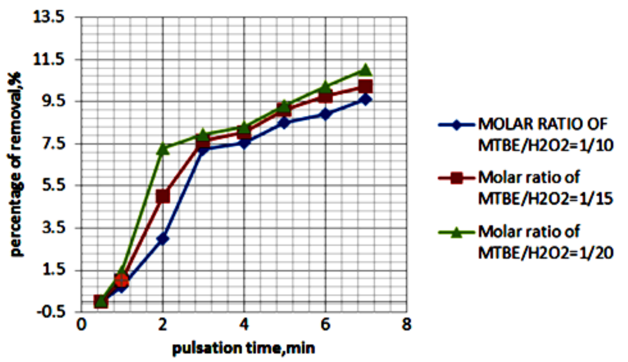


Figure 5: Effect of pulsation time on the percentage of removal with different molar ratio of MTBE to hydrogen peroxide.

The presented data in **Figure 6** shows the effects of pulsation time on the percentage of MTBE removal from synthetic wastewater at a different flow rate. Almost no significant increase in the percentage of removal of MTBE from water was recorded, i.e., it is approximately constant with at pulsation time. The percentages of MTBE removal at 7 min for air flow rate of 10 L/min were 42.14%, 43.57%, 45%, and 46.43% respectively. The results of the second part represent the effects of increasing the percentage of removal of the MTBE through the continuous stripping and oxidation processes of the treated water in the pulse column, which was passed to the air loop reactor and synergistic with adsorption of the side products that produced from the oxidation process, and increase the percentage of removal of the MTBE by adsorption it use granular activated carbon. The attempt to gain a complete mineralization and dismantling of another part of the MTBE in contaminated water represent the supplement another step from the total summation of steps of integration and synergy to the process of oxidation and stripping. The oxidation of MTBE to carbon and water is a multistep, multi-path process in which each step has different equilibrium and kinetic factors that govern the extent and rate that each reaction can take place. Not all oxidants have proven successful in mineralization of MTBE, leaving by-products such as tert-butyl formate (TBF) and tert-butyl alcohol (TBA). The full spectrum of possible reaction intermediates and governing criteria have not been determined for MTBE and the other oxygenates. However, in general, the greater number of carbon atoms in the oxygenate, the greater stoichiometry's proportion of oxidant that will be required (under the same conditions) to fully oxidize it.

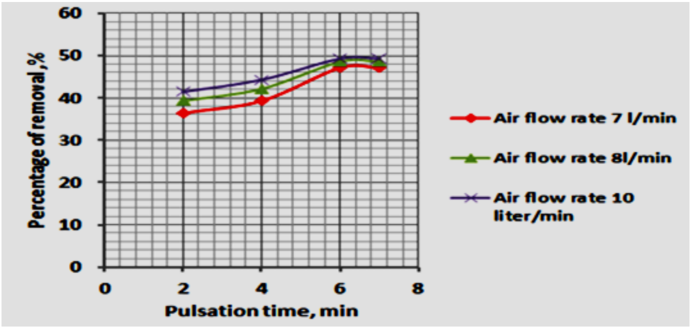


Figure 6: Effect of pulsation time on the percentage of removal with different air flow rates.

It is noticed that the percentage of removal of MTBE at 25 min for 2 ppm is 90% (Figure 7) and that is due to the axial dispersion of air in the contaminated water inside the draft tube upside will creates the difference in density or pressure between the contaminated water in the draft tube and annulus causing a constant liquid circulation and forced the water to passed down through granular activated carbons and based on the same principle as the difference in concentration focus of MTBE in the water and on the surface of granulated carbon, adsorption process occurs in effective way depending on the high surface area of contact between the particles of MTBE and the activated carbon also the large pore size of the activated carbon, this step will be represent the completion of the last step of the summation of the three integration steps and synergies.

In general, the goal is to maximize the extent of contact (maximum rate of mixing, highest specific surface area) while minimizing energy costs associated with the equipment design.

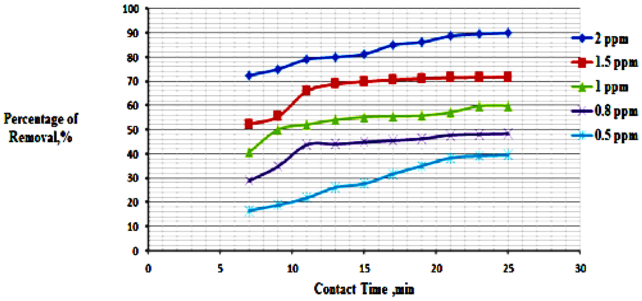


Figure 7: Effect of contact time on the percentage of removal of MTBE with different initial concentration of MTBE in air loop reactor.

The results showed (Figure 8) that the rate of removal of MTBE is enhanced at a lower initial molar ratio of MTBE to the H_2O_2 when keeping the air flow rate (10 L/min) and initial concentration of MTBE (2 ppm) constants. The percentages of MTBE removal in 25 min for molar ratios of 1/15 and 1/20 were 75, 80.1.

The selection of the proper empty bed contact time (EBCT) is one of the important factors in the design should bed contact time (EBCT) is one of the important factors in the design should be taken into consideration to fully utilize the GAC capacity [27].

EBCT is defined as the total volume of the activated carbon bed divided by the liquid flow rate and is usually expressed in minutes.

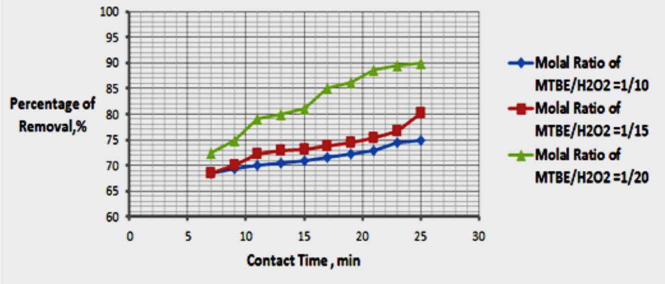


Figure 8: Effect of contact time on the percentage of removal with different molar ratio of MTBE to hydrogen peroxide in air loop reactor.

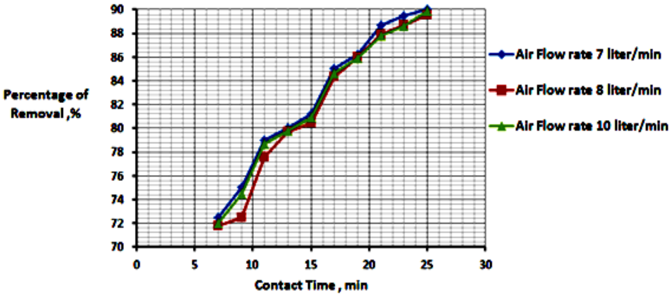


Figure 9: Effect of contact time on the percentage of removal with different air flow rates.

So was placed 5 kg of granules of activated carbon in the space between the inner and outer tube (the annulus) in the form of the bed rises by (0.3 m) and the optimal length of the mass transfer zone.

As a contaminated water stream passes through a confined bed of activated carbon, a dynamic condition develops and it establishes a mass transfer zone. This' mass transfer zone' is defined as the carbon bed depth required to reduce the contaminant concentration from the initial to the final level, at a given flow rate. The process 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 presented data in (**Figure 9**) shows the effects of contact time on the percentage of MTBE removal from synthetic wastewater at a different flow rate. Almost no significant increase in the percentage of removal of MTBE from water was recorded, i.e., it is approximately constant with at contact time(90%). The synergistic system that has been used to remove the MTBE by the three operations represents the efficiency of the device, the cost of installation and operation of the three devices (stripping, oxidation and adsorption) has been reduced to the cost of the installation and operation of one device only. The material of manufacturing this device (A Plexiglass) is cheap, and the cost of operation and maintenance is very simple, do not occupy a large space with the ease of being transferred (portable), the treatment period is relatively short (25 min) thus in terms of economic feasibility is considered as economically feasible.

4. CONCLUSIONS

In conclusion proved the success of proposed design for removal of organic pollutant from wastewater by using the synergistic system technique that apply three spontaneous processes (stripping, oxidation and absorption) with the integration of two technical, pulsation bubble column and internal airlift reactor.

Thus achieved higher percentage removal of MTBE in synthetic wastewater.

Furthermore, evidenced by using this new design, that increase area and a contact time between (MTBE) and hydrogen peroxide with granules activated carbon through the recycling of oxidized water between the draft tube and annulus in loop reactor led to an increase in the efficiency of removal of MTBE up to reach 90% .

Results showed that the molar ratio 1/20 for MTBE to the hydrogen peroxide with 10 L/min air flow rate gives the highest efficiency to remove the MTBE from the synthetic wastewater .

Another advantage of the present study is that the setting up a system for removing is low and less maintenance .

The method of the present study produces less hazardous compound in the water upon treatment by the system, and it is farther safe to be released to the environment.

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