



## Trio Effects Portable Water Treatment System

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### ABSTRACT

The main motivation of the present work is the efficiency and the high cost of the three major water treatment techniques used in many industrial applications (oxidation, adsorption and air stripping). The present work introduced a novel and combined technique that merges between the three major water treatment techniques, namely: stripping, oxidation and adsorption that are 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 experiments were conducted at different air flow rates 7, 8 and 10 (L/min), with difference residence time (2-20 min) at a different 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 was 1:20 with air flow rate of 7 L/min, having the best performance (90%) to remove MTBE from the synthetic wastewater. From this research it can be concluded that, the overall design achieved the objectives. The combined system effectively removed MTBE from wastewater. The combined process should be able to solve the environmental problems and reduce pollutants in wastewater contained MTBE, before it is released to natural water sources.

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## 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 allow 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) [1-5]. On the other hand, adsorption of chemical contaminants 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 [6-9].

In many occasions chemical methods were applied as an effective and commercially feasible water treatment technique to remove chemical contaminants through several approaches such as precipitation, oxidation or reduction and other chemical reactions that involved exchange of electrons between atoms [10-13]. In an advanced oxidation processes (AOPs), chemical, electrical or irradiative energy is supplied to the water matrix to produce highly reactive oxidants, which can attack and destroy the target compound. Usually AOPs are operating at ambient temperature treatment processes which are primarily based on the formation of highly reactive radicals, have emerged as an important class of technologies for accelerating the oxidation and

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reduction of a wide range of organic contaminants in polluted water and air [14]. Ideally AOP systems are designed to completely mineralize the organic contaminants of concern to  $\text{CO}_2$  and  $\text{H}_2\text{O}$ , but this may require more energy and greater chemical dosage and ultimately may prove to be cost prohibitive in certain applications [15]. However, hazardous bromate and biodegradable organic carbon are byproducts of these processes; as pollutants have to be removed. Meanwhile, biological treatment methods are those that involve living organisms using organic or in some instances, inorganic substances in food, completely changing their chemical and physical characteristics. Many different chemicals are discharged into the aquatic environment. Some of them are not only toxic, but also partly biodegradable; therefore, they are not easily removed in biological wastewater treatment plants. However, attempts to inoculate packed beds of granular activated carbon (GAC) with bacteria by conventional means of introducing bacteria into the packed bed of GAC together with a contaminated water feed stream have proven to be problematic; as the bacteria are only accumulated in the entry region of the bed and biofilm is developed which leads to loss of bioreactor performance due to blockage of the bed, flow channeling and bypassing [16, 17]. Current technologies, including ozonation, microbiological methods, and traditional activated carbon filtration are not sufficient to remove MTBE from wastewater or drinking water [18, 19]. This encourages many authors through the past decades to change, modify and combine the existing wastewater treatment techniques to fulfil 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 [20]. 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  $\mu\text{g/L}$  [21].

In the present work, a new design of combined stripping, oxidation, inverse fluidized, and internal loop reactor system (SOIAAI) 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, and hydraulic retention time. The aim of this investigation was to check the feasibility of using a new design of a combined system of stripping, oxidation, inverse fluidized, adsorption and internal loop reactor for the removal of pollutants from wastewater.

## METHODOLOGY

### Materials

The following chemicals were used:

- Methyl tert-butyl ether (MTBE) ( $\text{C}_5\text{H}_{12}\text{O}$ ), with a molecular weight of 88 g/gmol, and a purity greater than 99.8%, supplied by chemical material store in the laboratory of UMP, Permula Chemicals Sdn.Bhd. (Malaysia) was employed as adsorbate.
- Hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) as oxidation agent with 30% concentration in volume/volume, supplied by Wellmex Sdn. Bhd. Company (Thailand).

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  $\text{g/cm}^3$  and true density, 1.17  $\text{g/cm}^3$ .

### Experimental set-up

Figure 1 shows the flow diagram of the integrated 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 stripping, oxidation, inverse fluidization, adsorption 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 inverse fluidized and 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.

A dosing pump (P1, SEKO, chemical dosing pump, China (mainland)) is used to provide hydrogen

peroxide into the draft tube and reactor in order to mix and react with the oxygenate compound in the water before the inlet port as illustrated in Figure 3.

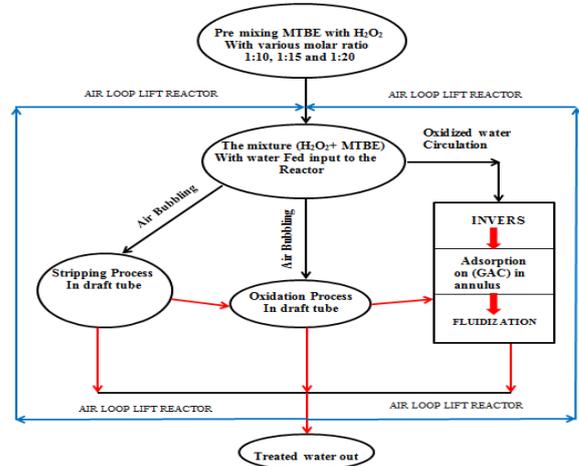


Figure 1. Flow diagram of integrated system.

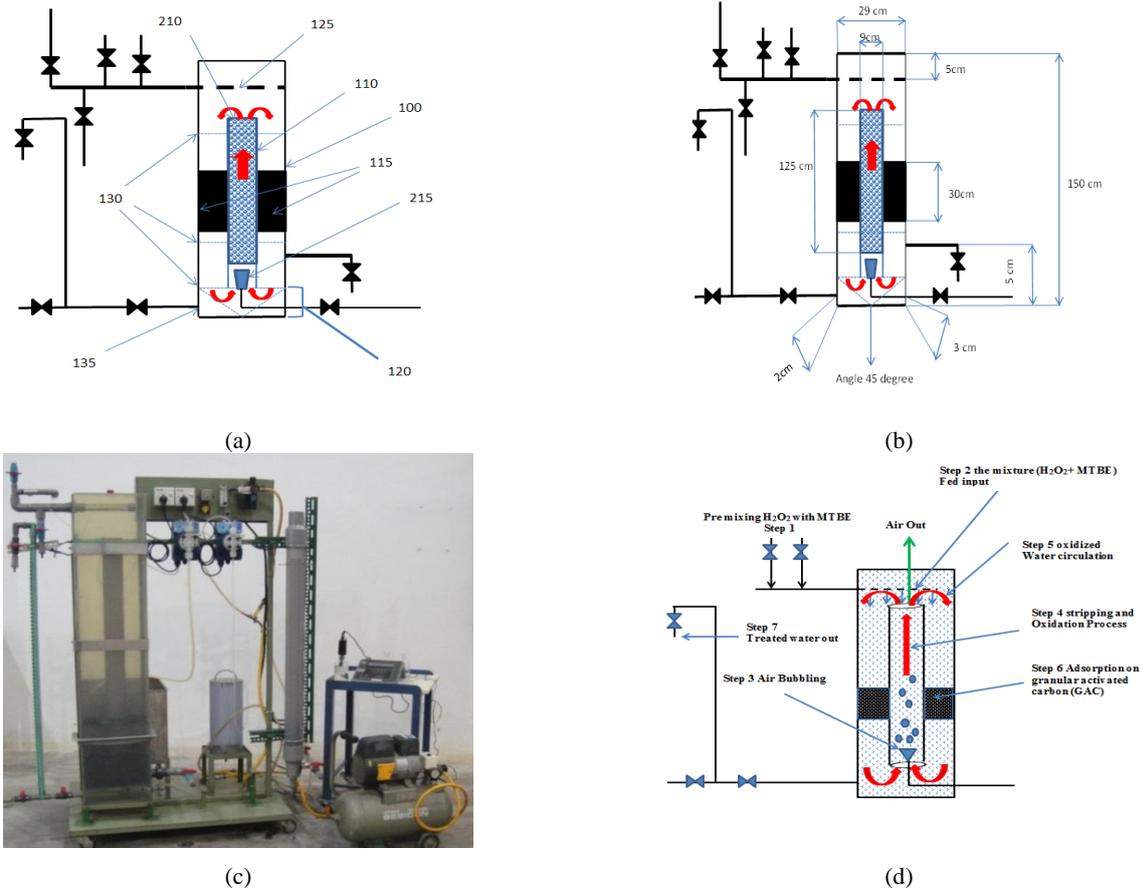
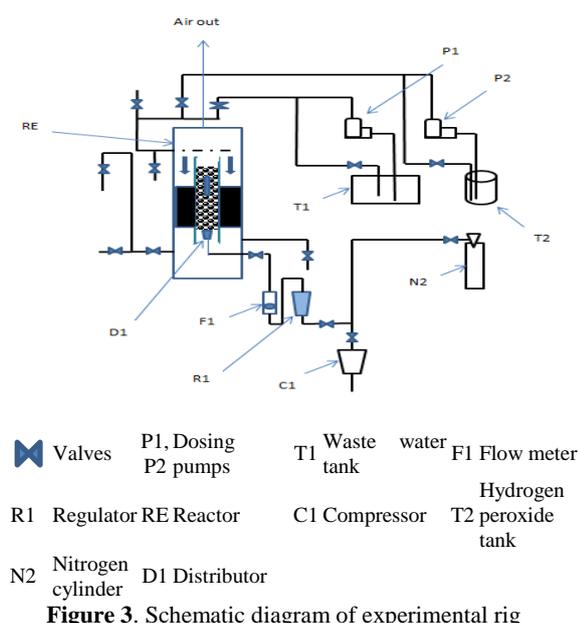


Figure 2. Design and scenario of internal loop reactor system (SOIAAI)



**Figure 3.** Schematic diagram of experimental rig

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 draft tube and reactor by the inlet port. Oxygenates contaminated water, and hydrogen peroxide are fed continuously into the 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 extend 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  $25^\circ C$  in a feed box, and a (5 Kg) granular activated carbon volume was  $4273.5 \text{ cm}^3$ . The molar ratio between MTBE to hydrogen peroxide was 1:20. During the experiments 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. 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  $P \text{ (abs)} = 2$  bar. To achieve the steady state of the system, the synthetic wastewater was pumped for 90 min to fill the reactor before the introduction of hydrogen peroxide to initiate the reaction. 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. 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 2-20 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:

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

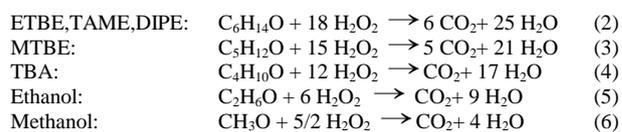
Where  $E$  is MTBE removal efficiency (%),  $C_{in}$  and  $C_t$  are the initial and residual concentrations of MTBE in ppm, respectively. The pH was measured, and all samples were collected in the cube vet for analysis.

## RESULTS AND DISCUSSION

Figure 4 shows the effects of contact time on the removal percentage of MTBE from synthetic wastewater. It is shown that the percentage of removal increases in accordance to the contact time. The percentages of removal of MTBE at 20 minutes for 2 ppm, 1.5 ppm, 1 ppm, 0.8 ppm and 0.5 ppm were 90%, 71.70%, 59.95%, 48.32% and 39.47%, respectively. For higher concentration of MTBE in synthetic wastewater, the percentage of removal is higher as compared to the low concentration of MTBE. The high concentration of MTBE in contaminated water helps to create a great driving forces to the mass transfer process which take 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 five operations at the same time, which are: oxidation, stripping, inverse fluidization, adsorption and air lift loop reactor.

It is very important to clarify the scenario of occurring of three 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 center of the draft tube 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  $OH\cdot$  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. Air stripping relies on an equilibrium phase transfer process where the contaminant partitions between the aqueous phase and the air phase. The equilibrium partitioning coefficient is called the Henry's constant which, in dilute solutions, is determined by Raoult's law using the vapor pressure of the pure compound and its water

solubility. In general, the higher the Henry's constant for a contaminant, the more effective air stripping will be for that contaminant. The results from the subsequent experiments showed that the removing rate of MTBE for combined processes (stripping, oxidation and adsorption) gradually decreases after 10 min. As shown in Figure 4, there are the two-stages for removing of MTBE. The percentage of removal of MTBE rate within the first 4 min in the first stage was higher than of those that followed after 4 min (approximately for all types of the feed input to the combination of inverse fluidized and internal loop airlift reactor). There are two main reasons for this: the first is the high MTBE concentration difference between the liquid phase ( $H_2O$ ) and gas phase (air) at the beginning of the stripping process, as well as that between the liquid phase (oxidized water in the draft tube) and solid phase (GAC in the annulus) in the start of the adsorption process. This provides a high driving force for mass transfer between the water-gas and water-solid phases. The second reason is that the  $H_2O_2$  decomposed very quickly to produce a large amount of  $OH\cdot$ . The produced  $OH\cdot$  can react rapidly with organic substance (MTBE) and cause the increasing of the MTBE oxidation efficiency. The elimination percentage of MTBE reached approximately 90% when the molar ratio of ( $MTBE/H_2O_2$ ) = 1/20, as shown in Figure 5a. If a sufficient amount and strength of oxidant ( $OH\cdot$ ) and enough time are provided, all of the ether- and alcohol-based fuel oxygenates can be mineralized to carbon dioxide and water. For example, the reactions (2-6) below shows the equations for the stoichiometries mineralization of some of the common oxygenates through oxidation using hydrogen peroxide. The oxidation of MTBE to carbon and water is a multi-step, 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. Chang [22] stated that the kinetics of the advance oxidation process are dependent on initial concentrations of starting organics (MTBE) and the oxidant ( $H_2O_2$ ). The experimental results showed that the initial rates of MTBE oxidation depended on hydrogen peroxide concentration as well as the initial MTBE concentration.



The oxidation of MTBE to carbon and water is a multi-step, 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 stoichiometries proportion of oxidant that will be required (under the same conditions) to fully oxidize it.

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.

It is noticed that the percentage of removal of MTBE at 20 min for 2 ppm is 90% 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.

The effect of contact time on the percentage of removal of MTBE with different molar ratio of MTBE to hydrogen peroxide is illustrated in Figure 5a. 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 (7 L/min) and initial concentration of MTBE (2 ppm) constants. The percentages of MTBE removal in 20 min for molar ratios of 1/10, 1/15 and 1/20 were 75, 80.1

The levels of  $H_2O_2$  had two opposing effects on the reaction rate[23-25]. 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.  $OH\cdot$  and  $\cdot OOH$ ) is the major mechanism by which MTBE is degraded. The selection of the proper empty

bed contact time (EBCT) is one of the important factors in the design should be taken into consideration to fully utilize the GAC capacity[26]. EBCT is defined as the total volume of the activated carbon bed divided by the liquid flow rate and is usually expressed in minutes. 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.

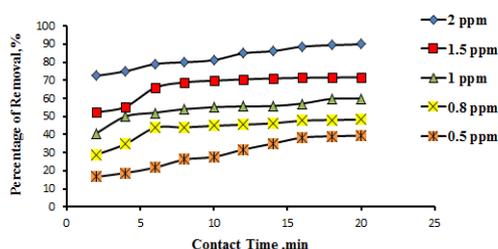
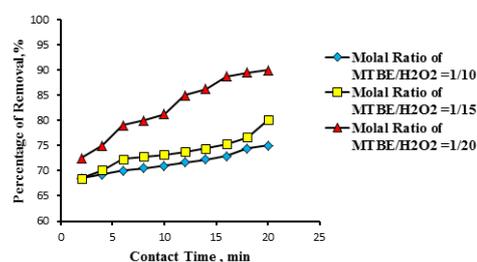
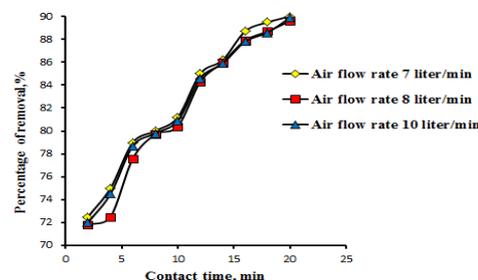


Figure 4. Effect of contact time on the percentage of removal of MTBE with different initial concentration of MTBE.



(a)



(b)

Figure 5. a) Effect of contact time on the percentage of removal with different molar ratio of MTBE to hydrogen peroxide and b) Effect of contact time on the percentage of removal with different air flow rates.

The presented data in Figure 5b shows the effects of contact time on the percentage of MTBE removal from synthetic wastewater at a different flow rate. No significant increase in the percentage of removal of MTBE from water was recorded, i.e., it is approximately constant with at contact time. The percentages of MTBE removal at 20 min for air flow rate of 7, 8, 10 L/min were 90, 89.6 and 89.9%, respectively.

The sum of what has been removed from the MTBE for 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 (20 min) thus in terms of economic feasibility is considered as economically feasible.

## CONCLUSIONS

In conclusion proved the success of proposed design for removal of organic contaminants from wastewater by using three spontaneous processes (stripping, oxidation and absorption) with the integration of two technics, inverse fluidized and internal airlift reactor, thus achieved higher percentage removal of MTBE from industrial 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 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 7 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 the pollutant removal is low cost and it requires 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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#### چکیده

انگیزه اصلی از انجام کار علمی حاضر بازده و هزینه بالای سه روش اصلی تصفیه آب است که در بسیاری از صنایع کاربرد دارد (اکسیداسیون، جذب و عاری سازی هوایی). کار حاضر روشی جدید و ترکیبی از سه تکنیک اصلی تصفیه آب: عاری سازی، اکسیداسیون و جذب که مناسب برای مصارف خانگی و روستایی است ارائه می دهد. کارایی روش حاضر به صورت عملی با استفاده از ماده شیمیایی متیل تری-بوتیل اتر به عنوان آلاینده مورد آزمایش قرار گرفته است. آزمایش ها در دبی های هوای ۷ و ۸ و ۱۰ لیتر بر دقیقه با زمان ماندهای ۲ تا ۲۰ دقیقه در نسبت های مولی ماده شیمیایی آلاینده به هیدروژن پر اکسید ۱ به ۱۰ و ۱ به ۱۵ و ۱ به ۲۰ انجام شده است. نتایج نشان می دهد که نسبت مولی بهینه ماده شیمیایی آلاینده به هیدروژن پر اکسید برابر ۱ به ۲۰ با دبی هوای ۷ لیتر بر دقیقه؛ بهترین بازده برابر ۹۰٪ جداسازی ماده شیمیایی آلاینده از پساب ساخته شده است. با استفاده از این تحقیق می توان ادعا کرد که طراحی کلی به هدف رسیده است. سیستم ترکیبی به صورت موثر ماده شیمیایی آلاینده را از پساب جدا کرده است. روش ترکیبی می تواند مشکلات زیست محیطی را حل نموده و آلاینده های حاوی متیل تری-بوتیل اتر را قبل از رهايش در طبیعت از پساب جدا نماید.