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# Modified Sequencing Batch Airlift Reactor Capability in MTBE Removal

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#### PAPER INFO

ABSTRACT

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## **INTRODUCTION**

Methyl tertiary butyl ether (MTBE) is one of the petroleum compounds with the chemical formula of  $C_5H_{12}O$ , which is used as an oxygenated fuel additive and a substitute for tetraethyl leading to achieve better combustion [19]. Because of MTBE resistance to biodegradation along with its high water solubility, poor absorption to soil particles, high mobility in soil and water, it moves towards groundwater. In this regard, one of the most important environmental problems is its aggregation in underground water and thus, in agricultural wells and wells that supply urban drinking water; it has adverse effects on the health of humans and animals that use them [1]. There are various physical, chemical, and biological methods for MTBE treatment. Ozone ultrasonic irradiation [8], hybrid oxidation [11], ozonation [20], UV/chlorine advanced oxidation process [14], electro-photocatalytic processes such as TiO<sub>2</sub> [21, 23, 27], ZnO-AgCl nanocomposite photocatalyst [18] and oxidation with per-sulfate to produce activated carbon for the removal of MTBE [10] are some of the methods used to reduce and eliminate this product. The most common methods of removing MTBE from drinking water are aeration [13], application of natural and modified diatoms [2, 3, 33] optical dispersion with

The aim of this study was to investigate MTBE removal efficiency using sequencing batch airlift reactor (SBAR) and to determine the share of aeration and adsorption processes during the operation. The present study was conducted with a new design of the system (cubic area and embedded baffle). The reactor was applied in 4-h cycles, which included 2 min filling, 210 min aeration, 5 min sedimentation, 8 min draw, and 15 min idle time. One week after start-up, the initial brown granules were observed. During the operation, some granules were formed with the size of 2–6 mm, average settling velocity and density of 0.66 cm/s and 0.06 g/mL, respectively. The results showed that COD removal efficiency was over 94 percent.

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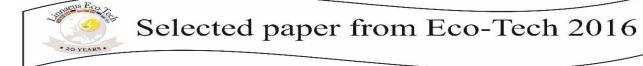
Research

Note

nano- zeolite combinations ZnO [34], adsorption by activated carbon [26], advanced oxidation processes (using a fluoride ion,  $O_3$ , and  $H_2O_2$ ), Fenton and pseudo-Fenton [5, 12], UV- Fenton [17] and using  $H_2O_2$ in the presence of Fe-zeolite catalysts [24]. Although there are many physical and chemical methods for the removal of pollution caused by MTBE, biodegradation can be a good alternative that can reduce the harmful and adverse effects on the environment at lower costs due to some reasons including high expenses, by-products formation and low efficiency [15].

Aerobic bio-granular production process is a new biotechnological method that has recently been developed to biologically purify wastewaters with high organic loading rates or wastewaters that contain toxic aromatic pollutants such as phenol, toluene, pyridine, dyes from textile industries, as well as nitrogen, phosphorus, and sulfate [25]. Granules consist of a thick mass of various species of microorganisms whose morphological structures (dense particles with large diameters) have some advantages over the conventional activated sludge; they have higher sedimentation ability and are more able to withstand high biomass. It results in reducing the capacity of the reactor and a possibility of treating wastewater with high loading rates [2]. Smooth and thick texture of granules, effective removal of nutrients (nitrogen and phosphorus) and sludge retention

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time (SRT) are other advantages of this system [31]. Some researchers consider biogranules as a suspended spherical biofilm, which consists of microbial cells, neutral particles, degradable particles, and extra-cellular polymeric materials [30]. This technology is applied in wastewater treatment systems and sequencing batch reactor (SBR) systems [29]. An SBR along with an internal riser is called sequencing batch airlift reactor (SBAR); it has a similar structure to SBR and purifies wastewater with a certain temporal cycle in a single reactor [16]. The SBAR system, which is used along with granules to treat wastewater, is known as granule sequencing batch airlift reactor (GSBAR). Using this system for biodegrading requires a high concentration of biomass (aerobic granules) [19, 28].

In the present pilot study, the baffled cube model, as a modified SBAR system, was examined to evaluate its performance in the production of granules and removal of MTBE. Owing to its different geometrical shape and to having baffles with similar sizes, this pilot provided conditions for a good mixture and suitable hydraulic flow for the creation of biological granules and prevented the decay of the granules. Moreover, because a suitable length was designed to move granules by the baffles, the length of the pilot decreased thus resulting in saving energy consumption by the aeration pumps. Therefore, the aim of this study was to investigate the capability of the SBAR system to remove MTBE using aerobic granules.

#### **MATERIAL AND METHODS**

In this study, SBAR consisted of plexiglass and was 4.8  $\times$  3.19 cm. System useful size was 3.5 L and it was 8 cm high. The reactor's work period was six 4-h cycles; 2 min for infusion, 210 min for aeration (reaction), 5 min for sedimentation, 8 min for discharge, and 15 min for rest. To activate the SBAR system, microbial seed was prepared from activated sludge of aeration tank of Ekbatan urban wastewater treatment plant in Tehran. The SBAR system schema is presented in Fig. 1. During the test, temperature was set at 25°C, the amount of dissolved oxygen was 2-5 mg/L, and the pH was 7-8. To make sludge compatible with sewage presented in this research, wastewater containing various concentrations of MTBE was injected in several stages. After adaptation stage, the amount of organic load entering the system after reaching a stable condition increased gradually (25 mg/L of the organic load was added to the concentration up to 700 mg/L of input COD; from 700 to 900 mg/L, 100 mg/L was added to the concentration).

To calculate density of biological granules, some granules were passed through a strainer for 3.5 min and were dewatered. To determine their volume, they were placed in a measuring cylinder containing distilled water.In the next step, samples were kept at 105°C for 24 h; then, density of granules was calculated [9]. To



Figure 1. Schematic SBAR system used in experimental study

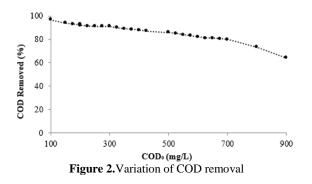
determine the fall speed of granules, a measuring cylinder containing distilled water was used [22]. To study the formation of granules formed in the reactor during operation, Bar- plot and Box- plot methods were used. Samples were collected every three days from the seventh day when the granules were formed. This statistical analysis was performed using SPSS. The values of pH, MLSS, MLVSS, F/M, SVI<sub>30</sub>, SVI<sub>5</sub>, ORP, and DO of the reactor were measured regularly during system operation period. Input pH and dissolved oxygen were set to 7-8 and 2-5 mg/L, respectively. ORP and DO changes were proportional to each other. Moreover, changes in the trend of granule size, sedimentation rate, and distribution and dispersion of granules were analyzed periodically and systematically during the operation period. During system operation period, the amounts of MLSS, MLVSS, and SVI were monitored regularly so that average MLSS, MLVSS, and SVI were 3480, 2623, and 102 mL/g respectively. Moreover, the ratio of F/M and 1/d was 31.0-25.0. It should be noted that all experiments were performed according to standard methods for examination of water and wastewater [4].

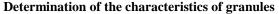
Glucose [6, 32] and MTBE were used as carbon sources; nutrients were used to regulate the ratio of carbon to nitrogen in the system (equal to 1:5:100 =C:N:P). Fine-material nutrients, including 5.202 mg/L of CaCl<sub>2</sub>, 10 mg/L of MgSO<sub>4</sub>, 25.0 mg/L of ZnCl<sub>2</sub>, 1.37 mg/L of KH<sub>2</sub>PO<sub>4</sub>, 15.80 mg/L of urea, and 208.04 mg/L of FeSO<sub>4</sub> were also used. To perform experiments and determine the main and control parameters, the following equipments were used: Spectrophotometer Hach-DR4000 to determine COD, Hach-DRD200 COD reactor with 16 tubes, DO meter, ORP meter to measure dissolved oxygen (HQ30D Hach), pH meter to measure the pH (made by Metrohm Co., model 300 PJ) and centrifuge to remove suspended and colloidal particles from solutions (made by Sigmo Company).

#### **RESULTS AND DISCUSSION**

#### **Changes in COD removal**

The results of loading from 100 to 900 mg/L during 3 months are shown in Fig. 2. According to the change chart, removal percentage of COD compared to the initial COD indicates that this process had a constant trend up to COD of 700 mg/L and after that it decreased due to increased load. Maximum and minimum removal efficiency of COD of 100 and 900 mg/L was 97.3 and 67, respectively. Based on the observations, the system could achieve a higher efficiency in a short period of time when the amount of organic load was 500 mg/L; it took longer at CODs less than 500 mg/L, so it was not optimal due to increased energy consumption. Moreover, as organic load increased by more than 500 mg/L, removal efficiency decreased to less than 90%. Thus, COD = 500mg/L with removal efficiency of 90% was chosen as the optimal level.





After one week of operation, under the same conditions and feeding with glucose and MTBE, granules were formed and some fine granules with 0.4 mm diameters were observed (Figs. 3 and 4). At the beginning of the operation, sludge seed had a fluffy, feathery, irregular, and loose gray structure; however, after about a month and after the production of granules, they became brown [30].

During the adjustment period, which lasted for 65 days, granules formed in the system were 4.3 mm in size. After a while when organic load increased, the granules were affected and some changes were observed in their size and color; after 3 months, they were 4.6 mm in size, and finally they reached the maximum diameter of 6 mm. According to the results, the density and size of granules increased with time, and their density was between 0.0505 and 0.06607 g/mL (Fig. 5). In a study conducted by Taheri et al. [29], the density was 0.0598 g/L. Concerning sedimentation rate, it can be said that as granule size increased, sedimentation rate increased as well. Granule fall speed was 0.3–1.03 cm/s (Fig. 6). In studies conducted by Bao et al. [7], the sedimentation rate

was 0.31-1.085 cm/s. In another study by Taheri et al. [29], the sedimentation rate was 0.97-1.35 cm/s.

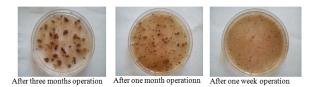
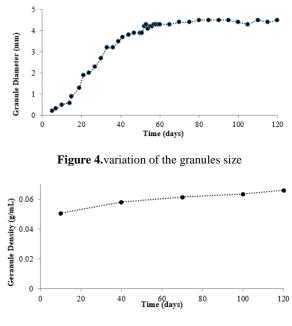
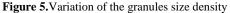
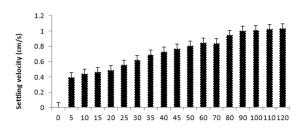


Figure 3. Size distribution of the granules at different operation







Time (days) Figure 6. Settling velocity of granules

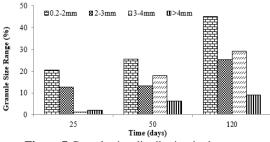


Figure 7. Granule size distribution in the system

#### **Determination of the size distribution of the granules**

According to graph 5, after 25 days of system operation, 26.1% of the granules were 0.2–2 mm in size and only 1.5% of them were 4 mm in size. Similarly, 2 months after operation, 54.7% of granules were 4 mm in size and 9% were 0.2–2 mm in size. It reflects the fact that granule size increases with time (Fig. 7).

# CONCLUSION

This study was conducted based on the production of biogranules for MTBE removal. The results showed that the granules produced with appropriate speed and density can biodegrade MTBE with more than 90% efficiency. Therefore, it is recommended that GSBAR process be used for non-biodegradable compound treatment.

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Persian Abstract

# چکیدہ

هدف از این تحقیق، بررسی راندمان حذف متیل ترشیاری بوتیل اتر با استفاده از سیستم SBAR بود که توسط یک سیستم بافل ¬دار مکعبی با طراحی جدید انجام شد. راکتور در سیکل ¬های ۴ ساعته شامل ۲ دقیقه خوراکدهی، ۲۱۰ دقیقه هوادهی، ۵ دقیقه ته نشینی، ۸ دقیقه تخلیه و ۱۵ دقیقه سکون به کار گرفته شد. یک هفته بعد از راه اندازی، گرانولهای اولیه به رنگ قهوه ای مشاهده شدند. طی دوره، گرانولهائی با قطر ۶-۲ میلی ¬متر، سرعت ته ¬نشینی و دانسیته به ترتیب ۱/۶۶ سانتی متر بر ثانیه و ۲۰/۶ گرم بر میلی ¬لیتر تشکیل شدند. طبق نتایج، راندمان حذف بیش از ۹۴ درصد COD حاصل شد.