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Treatment of petroleum refinery effluent using ultrasonic irradiation

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Ultrasonic irradiation is one of the advanced oxidation methods used in wastewater treatment. In this study, ultrasonic treatment of petroleum refinery effluent was examined. An ultrasonic homogenizator with a 20 kHz frequency and an ultrasonic bath with a 42 kHz frequency were used as a source for ultrasound. The effects of parameters such as ZnO amount, ozone saturation time, and type of ultrasound source on the degradation of petroleum refinery effluent were investigated. The degradation of petroleum refinery effluent was measured as a change in initial chemical oxygen demand (COD) and with time. According to the results, degradation increased with the addition of ZnO in an ultrasonic probe. There was also a positive effect of ozone saturation before sonication then applying ultrasound on the degradation for an ultrasonic probe. It was observed that there was no positive effect of ZnO addition and ozone saturation on degradation for an ultrasonic bath.

Keywords: degradation; ultrasound; petroleum refinery effluent.

INTRODUCTION

Large volumes of wastewater are constituted from the production stage of a petroleum refinery, such as extraction and refining. Refinery wastewater contains high concentration of aliphatic and aromatic pollutants. These pollutants are toxic and poor biodegradability. The discharge of refinery wastewater could cause serious environmental problems and affect human health. Traditional treatment of refinery wastewater constitutes two main treatment stages. The first stage is called pre-treatment step and it consists of mechanical and physicochemical treatment. The second stage is advanced treatment of the pre-treated effluent. In the pre-treatment step, heterogeneous compounds of the effluent, such as suspended solids and colloids, immiscible liquids, heavy metals, and solid particles are removed. Oil water separation, coagulation/flocculation, and dissolved air flotation units are some of the pre-treatment units. The aim of the advanced treatment is to reduce the level of hazardous compounds in the effluent. Biological systems are widely used to treat petroleum effluent. Petroleum effluent contains toxic and recalcitrant compounds. It is difficult to remove these pollutants effectively by using biological methods. Therefore, efficient water treatment processes are needed to meet increasingly stringent discharge standards and legislation on pollution control[1–6].

Recalcitrant pollutants in wastewater degrade rapidly by using advanced oxidation processes (AOPs). AOPs generate hydroxyl radical (•OH). •OH radical has a high oxidation potential and destroys organic molecules in wastewater[6–8].

Some of the AOP treatments used for petroleum refinery effluent are photo-catalysis[1, 8–12], ozonation[2, 13], Fenton[5, 14, 15], electro-Fenton[4], wet air oxidation[16], catalytic wet peroxide oxidation[17] and ultrasound[18–20].

The use of ultrasound in wastewater treatment has increased in recent years. Sonochemistry is the application of ultrasound to chemical reactions. Contaminants in wastewater can be treated by using ultrasonic irradiation due to cavitation phenomena. During cavitation phenomena, cavities are formed, growth and collapse with releasing large magnitudes of energy. Due to these cavitation conditions, oxidizing species, mainly hydroxyl radicals and hydrogen peroxide, are generated and localized high temperatures and pressures are obtained. The decomposition of the pollutant may be pyrolysis within the cavity and oxidation by hydroxyl radicals. There are several factors affecting the ultrasonic decomposition of pollutants, such as type of sonochemical reactors, frequency of the operation, ultrasonic power, addition of catalyst, presence of gases, and physicochemical properties of the liquid medium[21–25].

There are a few studies about the ultrasonic treatment of petroleum refinery effluent. Sponza and Oztekin[18, 19] investigated the ultrasonic destruction of poly-cyclic aromatic hydrocarbons (PAHs) in a real petrochemical industry wastewater in Izmir (Turkey). The authors examined the effects of sonication time, temperature, NaCl amount, CCl_4 amount, pH, hydrogen peroxide, and dissolved oxygen concentration on the destruction of PAHs. In one study, Ramteke and Gogate[23] investigated the treatment of wastewater containing toluene, benzene, naphthalene and xylene using an ultrasound/Fenton method as a pre-treatment step before biological treatment. These chemicals came from a petroleum refinery. In another study, Rasheed et. al.[20] investigated the treatment of petroleum wastewater by using ultrasound and iron particles.

The aim of the study is to investigate the ultrasonic treatment of petroleum refinery wastewater. An ultrasonic bath and ultrasonic probe were used as ultrasound source. The effects of ZnO addition and ozone saturation time before sonication on the degradation of petroleum refinery wastewater was examined for each ultrasonic device. The degradation rates in the two ultrasonic devices compared.

MATERIAL AND METHODS

The effluent used in this study was received from one of the petroleum refineries located in Turkey. A sample was taken after a pre-treatment unit and stored in a refrigerator at 4°C. The supplied petroleum refinery effluent had a COD of 150–200 mg/l.

In the study, two different types of ultrasonic irradiation devices were used. One of them was a probe type processor with a 20 kHz frequency and 200 W power supplied

by Bandelin, model HD2200. Another ultrasonic device was an ultrasonic bath with a 42 kHz frequency, 50 W power and 1600 ml volume. Its model was DSA50-SK-1 and was supplied by Fuzhou Desen Precision Instrument Co. Ltd. Figure 1 shows the schematic diagram of the experimental set up for the ultrasonic treatment of petroleum refinery effluent.

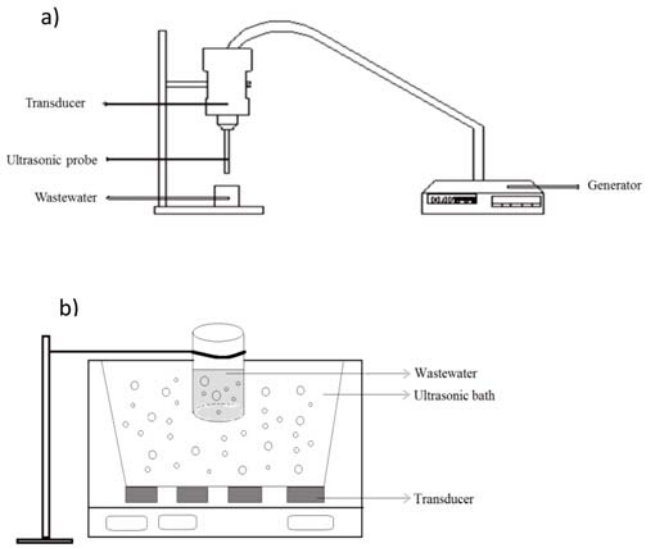

Figure 1. Experimental set up: a) Ultrasonic probe, b) Ultrasonic bath

For the ozonation experiment, an ozone device obtained by Or-cev ozonation system was used. In this device, there is a relationship between the electric voltage and the amount of ozone supplied. The electric voltage is set to give 1 ppm ozone.

The COD of the samples were measured by using a DR2400 spectrophotometer. Hach DRB 200 COD reactor was used to heat the samples and then COD of the sample was measured. Both the spectrophotometer and COD reactor were supplied by Hach.

The ZnO with a 98% purity was supplied from Merck.

The refinery wastewater may contain suspended particles. Blue ribbon filter paper (Schleicher and Schuell) was used to filter these particles before use in the experiments. A reaction vessel has a 250 ml volume, and a cylindrical shape. It is made of glass. It was filled with 250 ml of wastewater and then an ultrasonic probe was placed into the reactor. There was a 1 cm distance between the probe and the bottom of the reactor. The power of the ultrasonic probe was adjusted to 80 W and pulsed at a cycle of 30%. One experiment took 20 minutes. The samples were withdrawn from the reaction mixture and centrifuged at 4000 rpm for 10 min to remove any suspended particles. The same procedure was applied for the experiments using an ultrasonic bath. A reaction vessel was placed in the center of the ultrasonic bath.

COD was the key parameter and COD of the sample was measured using the DR 2400 spectrophotometer following the instructions for the Hach higher range test. A 2 ml centrifuged sample was placed into a special vial containing dichromate solution and then a DRB 200 COD reactor was used to heat the vial according to the directions. After cooling the vial, COD of the sample was read using a DR 2400 spectrophotometer.

The degradation efficiency was calculated as:

$$\text{degradation, } \% = [(COD_0 - COD_t)/COD_0] \times 100 \quad (1)$$

where COD_0 is the initial value, COD_t after any irradiation time.

In this study, the temperature of the reaction mixture was not controlled. An experiment was started at ambient temperature and a 15°C temperature increase was observed at the end of 20 minutes. The pH of the effluent was 5–6 and there was no pH change during the sonication reaction.

RESULTS AND DISCUSSIONS

Effect of ultrasound on degradation by using an ultrasonic probe

In the study, degradation of refinery wastewater was investigated. An experiment took one hour and a sample was taken at 15 min intervals. As shown in Figure 2, degradation was 13% at the end of the one hour. There was no significant difference between the 15 min and 45 min results.

According to Sangave and Pandit[22] and Gogate et. al[24], the degradation of pollutants in wastewater is difficult using ultrasound alone, in particular for the case of a mixture of pollutants. By applying ultrasound with low frequency, the pollutant molecules in petroleum refinery effluent degraded into smaller molecular weight compounds rather than complete mineralization. Wastewater used in this study consists of pollutants with a complex structure. For this reason, it is difficult to break these pollutants.

When the energy consumption is taken into consideration, the conversion value obtained at the end of 60 minutes is low, so other experiments were carried out with a time of 20 min.

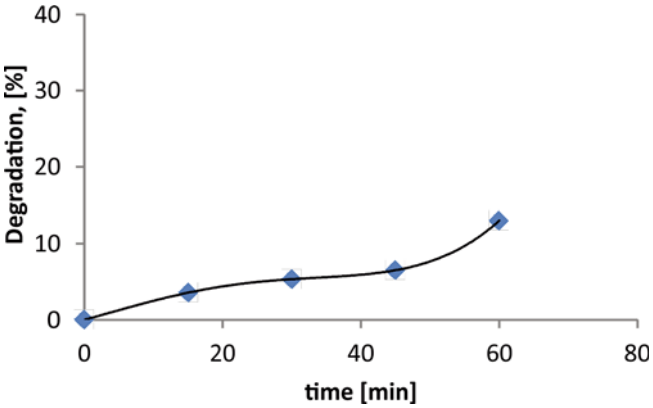

Figure 2. Degradation of refinery wastewater by using an ultrasonic probe

Effect of ultrasound and ZnO addition on degradation by using an ultrasonic probe

Before investigating a solid catalyst effect, experiments were also performed to test adsorption. A total amount of 250 ml of wastewater was poured into the reactor. After the addition of a solid catalyst, the reactor was stirred magnetically at an ambient temperature. COD of the sample was measured at the end of 20 min. There was no change in the COD value. These results showed that there was no adsorption with stirring.

The addition of solid catalyst affects the ultrasonic treatment in two different and adverse ways. The addition of catalyst particles improves the number of cavities by providing additional nuclei. On the other hand, catalyst particles act as a barrier for the transmission of sound waves, and energy dissipated into the system decreases[25].

In the study, to increase degradation, ZnO was used with ultrasound. ZnO was chosen due to its stability, non-toxicity, easily available and low cost. The effect of ZnO amount on ultrasonic treatment of petroleum refinery effluent has been investigated by using an ultrasonic probe.

Figure 3 shows the effect of ZnO amount on degradation. The addition of ZnO has a positive effect on degradation rate. As shown in Figure 3, degradation is greater with the presence of ZnO than without ZnO.

The presence of a heterogeneous catalyst, such as ZnO could accelerate dissociation of water molecules to form hydroxyl radicals. Due to the formation of more hydroxyl radicals degradation of pollutants in wastewater increases[26–27].

By using 0.04 g ZnO, 16% degradation was obtained at the end of 20 min. An increase in ZnO amount causes a decrease in degradation rate, and with an amount of higher than 0.05 g, there was no degradation. An excessive amount of ZnO may inhibit the dissipation of ultrasound in the reaction fluid, so generation of radicals decreases.

The authors Jamalluddin & Abdullah[26] and Abdullah & Liang[27] obtained similar results. At high loading, excess quantity of catalyst particles cause a mutual screening effect among the catalyst particles. The ultrasonic energy could not effectively reach into the system; as a result, generation of active radicals decreases. Anju et. al.[28] investigated the effect of ZnO amount on the sonocatalytic degradation of phenol. The degradation of phenol increases with an increase in the catalyst amount. After reaching the optimum amount of catalyst, degradation decreases. The introduction of more catalyst particles in the solution provides more nucleation sites for cavitation bubbles at their surface. On the other hand, a higher concentration of catalyst particles may also disturb the transmission of ultrasound in water medium. As a result, there is no further increase in the degradation of the pollutant after the optimum dosage. In another study, Yılmaz and Fındık[29] investigated the effect of TiO_2/ZnO composite amount on sonocatalytic treatment of industrial wastewater. According to the results, decolorization efficiency decreases after optimum catalyst loading.

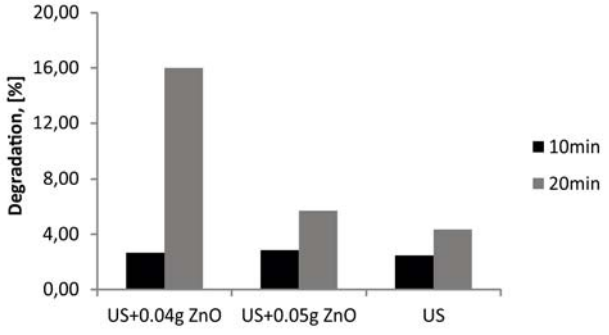

Figure 3. The effect of ZnO amount on the degradation of refinery wastewater by using an ultrasonic probe

Effect of ozonation and ultrasound on degradation by using ultrasonic probe

Ozone is a strong oxidizing agent used in the treatment of wastewater. Organic matter reacts with either molecular ozone or free hydroxyl radicals formed from an ozone self-decomposition cycle[30].

To increase degradation with ultrasound, petrochemical refinery wastewater was saturated with 1 ppm ozone before sonication. After expose to 5, 10 or 15 min ozone, ultrasound was applied for 20 min. Figure 4 shows the combined effect of ozone saturation before sonication and then applying ultrasound. Degradation was increased with ozone saturation before sonication. A positive effect of ozone saturation was observed.

The combined operation of ultrasonic treatment and ozonation provides synergistic effects in wastewater treatment and mass transfer of ozone in the wastewater increases due to ultrasonic effects. Dissolved ozone in a liquid may react directly with organic pollutants in wastewater or thermally decompose. After thermal decomposition reactions, hydroxyl radicals are formed[31].

On the other hand the passage of gas has two main effects on ultrasonic degradation. Due to the introduction of gas, energy released at the end of the cavity collapse decreases or the number of cavities increases in the presence of gas, due to the presence of gas bubbles as additional nuclei. Thus, the proportions of these two effects will decide the net effect on the degradation of petroleum refinery wastewater[24].

As seen in Figure 4, an increase in ozone saturation time before sonication causes a reduction in degradation rate. At 5 min ozone saturation before sonication, degradation was highest. There is a negative effect of increase in ozone saturation time due to the decrease in energy released at the end of the cavity collapse. In another study, Teo et. al.[32] said that cavitation bubbles can possibly be interrupted or destroyed by sparking gas.

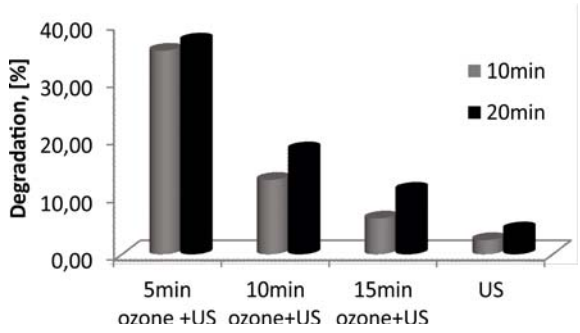

Figure 4. The effect of ozone saturation time on the degradation of refinery wastewater by using an ultrasonic probe

Effect of ozonation, ZnO addition, and ultrasound on the degradation by using an ultrasonic probe

In this part of the study, the effect of ZnO addition and ozone saturation on ultrasonic degradation was investigated. Figure 5 shows the comparison of different applications. First, by using ZnO, the effect of ozonation time was examined. As shown in Figure 5, there is no positive effect with the increase in ozone saturation time on ultrasonic treatment. As explained in the previous section, the negative effect of an increase in ozonation

time was observed due to the decrease in energy released at the end of the cavity collapse.

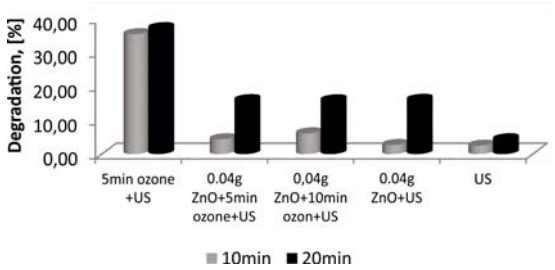

Figure 5. The effect of ozone saturation, ZnO addition on the degradation of refinery wastewater by using an ultrasonic probe

As shown in Fig. 5, there is no significant difference between 0.04 g ZnO combined ultrasound and ozone saturation then applying ultrasound with 0.04 g ZnO. According to the results, 5 min of saturation with ozone before sonication gave the highest degradation rate. The effect of saturation with ozone was more dominant than the presence of catalyst particles in the conditions studied.

Effect of ultrasonic bath on the degradation

An ultrasonic bath with 42 kHz was used as a source of ultrasound. The reaction vessel was placed in the center of the bath, and wastewater was indirectly sonicated. Figure 6 shows the results for the different combinations. There is no positive effect of ZnO addition and saturation with ozone. As said before, cavitation bubbles can possibly be destroyed by ozone and catalyst particles inhibit transmission of ultrasound in wastewater. Degradation was highest using only the ultrasonic bath. At the end of the 20 min, 38.4% degradation was obtained.

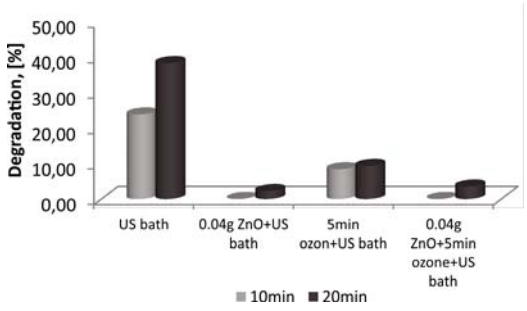

Figure 6. Effect of an ultrasonic bath on the degradation of refinery wastewater

Comparison of the results using ultrasonic bath and ultrasonic probe on the degradation

In this study, an ultrasonic bath with a 42 kHz frequency and an ultrasonic probe with a 20 kHz frequency were used as a source of ultrasound. The reaction vessel contains wastewater was immersed in an ultrasonic bath and wastewater was indirectly exposed to ultrasound. On the other hand, in the ultrasonic probe wastewater was directly exposed to ultrasound.

Ultrasonic irradiation rate depends on types of ultrasonic device, frequency, properties of pollutants, presence of gas, and presence of catalyst. Although an ultrasonic bath is commonly used as a source of ultrasound, an ultrasonic probe provides better energy dissipation due to the larger irradiation surface, so it produces a better sonochemical effect[25, 31, 33]. On the other hand, as the frequency of irradiation increases, the rate of degradation also increases. Due to an increase in frequency, cavitation occurs more rapidly and more violently, so the number of the ·OH radicals increases[24].

Figures 7–9 show the comparison of the ultrasound sources on the degradation of petroleum refinery effluent.

As shown in Figure 7, an ultrasonic bath gave a better result than the ultrasonic probe. The frequency of ultrasonic bath being used in this study is greater than that of the ultrasonic probe. The rate of ultrasonic degradation increases as the frequency of irradiation increases. The collapse pressure and temperature of the cavity increases with frequency. Due to an increase in energy released at the end of the cavitation, cavitation occurs very rapidly and more violently and higher concentrations of hydroxyl radicals are formed. Therefore ultrasonic degradation rate increases[24, 25, 31, 33]. The frequency of the ultrasonic bath has the predominant effect based on the conditions studied.

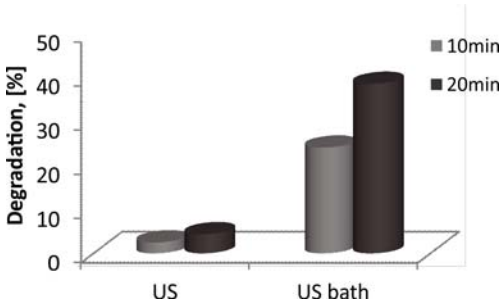

Figure 7. Comparison of an ultrasonic probe and ultrasonic bath on the degradation

Figure 8 shows the effect of 5 min ozone saturation before sonication, and Figure 9 shows the effect of ZnO addition with 5 min ozone saturation before ultrasonic irradiation using a different ultrasound source. As shown from Figures 8 and 9, the ultrasonic probe gave a better result.

In an ultrasonic probe, 250 ml of wastewater was directly exposed to ultrasound. Reaction vessel with 250 ml wastewater was placed in the ultrasonic bath and effluent in the reaction vessel was indirectly exposed to ultrasound. The reaction vessel causes energy loss due to the indirect sonication and may affect the passage of ultrasound. The combined effects of reaction vessel, and ozone saturation, causes a decrease in transmission of sound waves; therefore the degradation rate may decrease by using an ultrasonic bath.

In addition to this, energy dissipation due to a large irradiation surface is more effective than a high frequency of ultrasonic bath at the conditions studied. Another factor that affects the ultrasonic treatment is power density. Power density is defined as a ratio of power dissipated into the system to volume of the reaction mixture. An increase in power density enhances the cavitation effects, so degradation of the reactants increases[24, 31, 33]. There was a coupling liquid inside the ultrasonic bath. The volume of the liquid ultrasonically treated in the ultrasonic bath was larger. In the case of ultrasonic probe, power density is greater than ultrasonic bath.

As shown in Figure 9, usage of ultrasonic probe gave greater degradation than the ultrasonic bath. As said earlier, in an ultrasonic bath reaction vessel acts

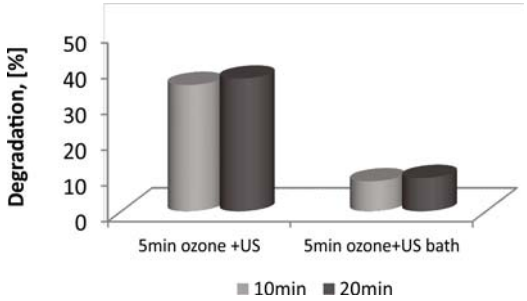

Figure 8. Comparison of an ultrasonic probe and ultrasonic bath on the degradation with 5min ozone saturation

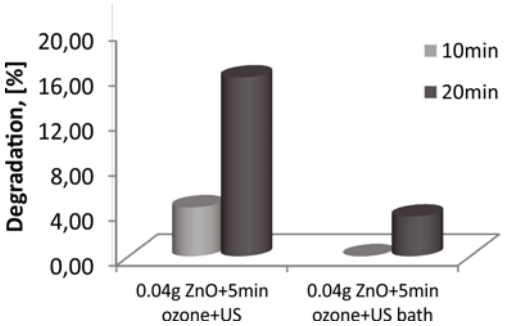

Figure 9. Comparison of an ultrasonic probe and ultrasonic bath on the degradation with 5min ozone saturation and 0.04g ZnO

as a barrier for the transmission of sound waves and dissipated energy into the system decreases. Due to the combined effects of the reaction vessel, ZnO addition, and ozone saturation, transmission of sound waves decreases in the ultrasonic bath. In addition, the power density of the ultrasonic probe is greater than the power density of the ultrasonic bath. The power density of the ultrasonic probe has the predominant effect based on the conditions studied.

CONCLUSIONS

In this study, ultrasonic treatment of petroleum refinery wastewater was investigated. The degradation of petroleum refinery wastewater was examined by using two different types of ultrasonic devices. An ultrasonic bath with 42 kHz frequency and an ultrasonic probe with 20 kHz frequency were used as a source of ultrasound. The effects of ZnO addition, ozone saturation, and source of ultrasound were examined.

By using an ultrasonic probe, the degradation increased with the addition of ZnO. There was also a positive effect of ozone saturation before sonication then applying ultrasound on the degradation. When the ultrasonic bath was used, negative contribution of ozone saturation before sonication and ZnO addition was observed.

An ultrasonic bath gave higher degradation than that of the ultrasonic probe when only ultrasonic irradiation was applied. On the other hand ultrasonic probe gave better results with ozone saturation before sonication and with addition of ZnO plus ozone saturation before sonication than the ultrasonic bath at the studied conditions.

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