Volatile Organic Compounds (VOCs) Removal of Polluted Air Streams Using Ozone in The Presence of Porous Silica Bed

1Ayla Hassani, 2Maryam Keyvanfar, 3Aydin Hassani

1Faculty of Chemical Engineering, Sahand University of Technology, New Town Sahand, Tabriz, Iran.
2Young Researchers Club, Tonekabon Branch, Islamic Azad University, Tonekabon, Iran.
3Department of Applied Chemistry, Faculty of Science, Tabriz Branch, Islamic Azad University, Tabriz, Iran.

Abstract: In this article, the removal of Volatile Organic Compounds (VOCs) from polluted air, by using ozone on silica bed was investigated. Among volatile compounds, toluene a strong and well-known pollutant has been chosen as an index. The oxidation process of VOCs on silica bed is influenced by flow rate of polluted air, concentration of toluene flow rate of ozone, temperature and silica surface area. In this study the flow rate of polluted air and ozone were 25 ml/min and 50 ml/min, respectively. Impacts of temperature and surface areas changes on the amount of removed toluene have been studied. The obtained results revealed that optimum temperature (100ºC) and surface area (400 m²), maximum removal percent of toluene about (97.89%) is achievable. In addition, experiment results showed that when ozone is used, much more toluene is removed compared to using air. Considering that the perfect removal of ozone at lower temperatures is achievable, therefore, removal of volatile compounds in existence of ozone due to its high oxidation ability has a considerable importance in terms of the process and is more economical.

Key words: VOCs, Toluene removal, Silica, Oxidation, Ozon.

INTRODUCTION

Volatile organic components such as benzene, toluene and other organic components (collectively known as VOCs) are key industrial solvents and are frequently required in industrial operations (Zhao et al., 2011). Contamination by VOCs is a common problem because of their wide distribution in the environment, including such places as wastewater treatments plants, dry cleaning and gasoline storage facilities, as well as transportation and other industrial sources (Maliyekkal et al., 2004). These components, also, have a high polluting potential, due to their neurotoxic, carcinogenic and teratogenic properties, representing a high risk to the environment and human health (Jo et al., 2008; Mathur et al., 2007). The United States Environmental Protection Agency (US EPA) has classified these compounds as priority pollutants due to their toxic properties (Jo et al., 2008; Mathur et al., 2007; Semple et al., 1998). There exist different methods for removing VOCs and other pollutants produced from sewage of industrial and petrochemical industries. These methods can be used according to the situation, capacity and present facilities. The most important methods for omitting volatile organic compounds are biological decomposition, adsorption (Chou and Chiou, 1997; Fang and Khor, 1989), filtration, thermal oxidation (Klobucar, 1995; Lewandowski, 2000), catalytic oxidation (Chu and Windawi, 1996; Saracco and Specchia, 2000) and air stripping (Mihelcic et al., 1993; Okoniewski, 1992; Robert, 1984). Oxidation is one of the most common methods for removing VOCs from polluted gas (Ruddy and Carroll, 1993; Leson and Smith, 1997).

In order to run the process with high efficiency, catalytic oxidation needs high temperature and consumes a huge amount of energy. Also, the costs of used catalysts are remarkable (Ruddy and Carroll, 1993, Horvat et al., 1985). A new technology that has been developed for treating waste water and exhausted gases is “advance oxidation by using ozone” as a powerful oxidizer for VOCs abatement. Using ozone as a method for VOC abatement has considerable advantages such as, high VOCs removal efficiency, moderate process temperature and unique oxidizer for all kinds of VOCs, harmless and environmental friendly oxidizer. Ozone attacks stable compounds such as multi ring aromatics and chlorine hydrocarbons and then oxidizes them at low temperature. Moreover, there is no need for its post-process decontamination since it automatically turns into oxygen. One of the advantages of using ozone is reduction of heat during the process which is environmental friendly and a tremendous energy-saver. Also, ozone decomposes after a short residence time in atmosphere and converts to oxygen. Although ozone is an expensive chemical, porous media can be used as a challenge to maximize reaction between ozone and hydrocarbon and minimize ozone waste (Horvat et al., 1985).

In this study, in order to remove toluene (which has been chosen as an index well-known compound) from polluted air, silica has used as a catalyst and ozone as an oxidizer which increases contact area between VOC

Corresponding Author: Ayla Hassani, Faculty of Chemical Engineering, Sahand University of Technology, New Town Sahand, Tabriz, Iran.
and ozone. In addition, the effects of reaction temperature and surface areas of catalyst on removal percentage of toluene have been investigated.

MATERIAL AND METHODS

In order to test the functionality of oxidation process by using ozone the pilot device was used. Figure 1 presents a schematic diagram of oxidation pilot. Precise control of air flow rate, toluene concentration and reactor temperature and ozone concentration is possible. To producing air with constant concentration of toluene, a stream of pure air and a polluted air stream, which is saturated with toluene, were mixed in constant temperature in an ice bath. Toluene concentration is analyzed by GC accurately before and after the reactor.

Fig. 1: schematic diagram of ozonization pilot plant.

These tests study the impact of parameters such as reactor temperature, ozone concentration and surface areas on the toluene removal. At the first stage of the test, the air that contains 800 ppm toluene will be oxidized by oxygen and ozone in the reactor at different temperatures. The maximum capacity of ozonator was 4 g/hr. In each test flow rate of the air that contains toluene is 25 ml/min, flow rate of clean air is 75 ml/min and flow rate of ozone is 50 ml/min. Toluene concentration is analyzed by GC accurately before and after the reactor. The amount of removed toluene is measured at temperatures varying between 50-400ºC and surface area 10-400 m² of silica bed (the absorbent surface is measured by a device using BET equation). The starting temperature is 400ºC and decreases to 50ºC. This is for burning of the coke remains from the previous tests and stopping toluene discharge. According to the fact that at high temperatures toluene will be absorbed and will be discharged at low temperatures, running the tests with an ascending temperature degree would result in toluene discharge from absorbent surface at high temperature and a subsequent severe decline in accuracy of the tests. Each time, the outgoing air will be sent to GC device to measure the concentration of toluene. Analyzing the results of GC device shows a diagram on computer screen. The lower part of diagram represents concentration of toluene. The following equation is used to determine the exact amount of toluene concentration:

\[ C_{\text{toluene}} = 0.0066759 \times A \]  

\( C \) shows the concentration of toluene (ppm) and \( A \) is the surface area below the graph according to the GC device. This equation is derived from linearization of data outputs resulted from calibration of device with different standard solutions. For reproducibility of the results each experiment is repeated at least 3 times.
RESULTS AND DISCUSSION

The Effect of Temperature on Toluene Removal Percentage:
Result of ozonation process in different temperatures and surface areas are presented in Figures 2 to 6. Ozonation of polluted air in presence of 400 m$^2$ silica at different temperatures has been illustrated in Figure 2. As shown in this figure, the amount of removed toluene at different temperatures by using ozone is much more than when oxygen is used as an oxidizer in the absence of ozone. By using ozone as an oxidizer, higher removal by increasing temperature and maximum toluene removal, about 97.89%, was obtained at 100ºC.

By using ozone, increasing temperature to 100ºC, toluene removal percentage increases, but in higher temperature removal percentage decreases with increasing temperature. Ozone is an unstable compound and decomposes to oxygen at high temperature. Thus the ozone concentration diminishes in reactor and removal percentage was reduced. When sufficient surface area is available in reactor, the effect of temperature on toluene removal percentage is marginal. In other words by using adequate surface area, toluene can be removed completely from exhausted gases at low temperature. According to results shown in Figure 2, in presence of ozone, the optimum temperature is 100 ºC in presence of 400 m$^2$ surface area.

Fig. 2: Toluene removal percentage with and without ozone on silica with 400 m$^2$ surface area.

Ozonation of polluted air in the present of silica with 200 m$^2$ surface area has been performed and results are illustrated in figure 3. Similar to the previous case, if air is used as an oxidizer, toluene removal was raised with increasing temperature. Due to the decreasing in silica surface area, toluene removal efficiency (37.53% at 400 ºC) was decreased relative to the pervious case (51.6% at 400ºC). Using ozone at the toluene removal percentage at 100ºC approximately is similar to toluene removal percentage at 400ºC. Therefore, the suitable temperature for removing toluene by using ozone in this surface area is 100 ºC too, according to reasons explained in last part (400 m$^2$ surface area).

Three different steps can be seen from figure 3. In first step, i.e., temperature between 50 and 100ºC, VOCs removal percentage was raised with increasing temperature, in the second step, i.e., temperature between 100 and 300 ºC removal percentage was decreased and in the third step, i.e., temperature between 300 and 400ºC removal percentage increased with increasing temperature. It seems that in the second range of temperature, decomposition of ozone is significant.

In the case of sufficient silica surface area in the reactor, the first step of this curve becomes similar to Figure 2. In other words the effect of temperature will be marginal if enough surface area is available.

Fig. 3: Toluene removal percentage with and without ozone on silica with 200 m$^2$ surface area.
Figure 4 shows the obtained results of 100 m$^2$ silica on amount of toluene removal of polluted air. In the case of sufficient silica surface area in the reactor, steps of this curve becomes similar to Figures 2 and 3. In other words the effect of temperature will be marginal if enough surface area is available.

![Figure 4](image1)

**Fig. 4:** Toluene removal percentage with and without ozone on silica with 100 m$^2$ surface area.

In Figure 5, results of ozonation with 50 m$^2$ surface area were illustrated. These results indicated that the amount of removed toluene at different temperatures by using ozone is much more than when oxygen is used as an oxidizer in the absence of ozone even in the limited surfaces. In this case, the surface area was not enough and the effects of reactor temperature and ozone on toluene removal percentage become more significant. It should be noted that silica surface area is the least in the present case; therefore removal of toluene is the least. On the other hand, by using ozone and without ozone, toluene upward removal trend with increasing temperature is remarkable. The toluene removal with 10 m$^2$ silica bed was investigated too and similar results were obtained. However, the results showed the amount of toluene removal in this case were much less than 50 m$^2$ surface area of silica.

![Figure 5](image2)

**Fig. 5:** Toluene removal percentage with and without ozone on silica with 50 m$^2$ surface area.

**The Effect of Surface Area on Toluene Removal Percentage:**

The effect of surface area of silica on toluene removal by using ozone and without ozone at different temperatures was investigated. Results revealed that the amount of toluene removal approximately has a trend of steady increase at all surface areas with increasing temperature. The obtained results in these series of experiments were summarized in Table 1.
Table 1: Toluene removal percentage with ozone on silica with different surface area.

<table>
<thead>
<tr>
<th>Surface area (m²)</th>
<th>Temperature</th>
<th>T=50 °C</th>
<th>T=100 °C</th>
<th>T=200 °C</th>
<th>T=300 °C</th>
<th>T=400 °C</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td></td>
<td>46.57</td>
<td>55.27</td>
<td>71.69</td>
<td>80.08</td>
<td>84.99</td>
</tr>
<tr>
<td>50</td>
<td></td>
<td>57.09</td>
<td>66.58</td>
<td>74.37</td>
<td>80.88</td>
<td>87.55</td>
</tr>
<tr>
<td>100</td>
<td></td>
<td>62.9</td>
<td>77.31</td>
<td>75.26</td>
<td>77.47</td>
<td>89.94</td>
</tr>
<tr>
<td>200</td>
<td></td>
<td>75.83</td>
<td>88.82</td>
<td>82.99</td>
<td>76.7</td>
<td>92.09</td>
</tr>
<tr>
<td>400</td>
<td></td>
<td>84.46</td>
<td>97.89</td>
<td>84.05</td>
<td>81.9</td>
<td>96.26</td>
</tr>
</tbody>
</table>

It is apparent that due to decomposing ozone in higher temperatures removal percentage decreases a little with increasing temperature. Also, results show that effect of increasing temperature on amount of toluene removal decrease with increasing surface area. Hence, at low temperature is obtained desirable results for removing toluene in high surface area. For example, the suitable temperature for removing toluene by using oxygen is 100 °C in 400 m² silica. It is worthy to say that, if it is possible to reach maximum amounts of removal percentage in lower temperature, it may bring some considerable results especially from economic point of view.

Conclusions:

According to the results of the tests, by using ozone, polluted air can be effectively purified. In all experiments by using ozone, the higher toluene removal was obtained, compared to the air as an oxidizer. At surface area above 200 m² it is impossible to remove toluene higher than 90%.

Choosing optimum surface area and oxidation temperature for complete toluene abatement is undoubtedly necessary. In both case, oxidation with and without ozone, available surface area in reactor significantly influences toluene removal.

If ozone is used as an oxidizer and sufficient amount of Silica available in the reactor, then effect of temperature on toluene removal is marginal while effect of temperature on toluene removal in the presence of air is considerable.

While ozone is used as an oxidizer, toluene can be removed completely at low temperatures. In other words, according to the above-mentioned tests and the fact that silica is both economical and accessible; also, effectiveness of ozone-silica system compared to oxygen-silica system, industrial pollutions (containing VOC) can be removed more efficiently and with less price by using ozone-silica system.

ACKNOWLEDGEMENTS

The authors would like to express their gratitude to Prof. Anvar Khodiev for the technical assistance during this project.

REFERENCES


