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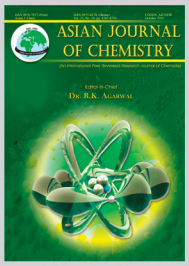

Removal of Toluene from Polluted Air Using Ozone and Pt Catalyst

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In this study, removal of volatile organic compounds (VOCs) from polluted air by using ozone based Pt catalyst were investigated. Highly active Pt catalysts for toluene oxidation could be obtained by using colloid-deposition method. The type of the support is an importance of supported Pt/MeOx (Me = Fe, Ni, Zn, Al) catalysts. The results indicated that process of toluene oxidation based on Pt catalyst is dependent on the type of catalyst, temperature and concentration of ozone. As can be seen, in the absence and presence of ozone, Pt/Fe_2O_3 catalyst has the best efficiency for the removal of toluene in the various temperatures. Furthermore, at 50-200 ºC, the rate of toluene omission would be higher when the concentration of applied ozone increased.

Key Words: Polluted air, Ozone, Organic compounds, Catalyst.

INTRODUCTION

Over the last 25 years, health complaints related to indoor climate have increased[1]. Poor indoor air quality can be attributed to physical (humidity), chemical (organic and inorganic), physical-chemical (particulate matter) and biological (molds) agents. Buildings are being sealed more tightly to reduce thermal energy losses. Concentrations of indoor pollutants can build up because there is a low turnover rate of indoor air[2]. It is proved that the level of indoor pollutants is 2-10 times higher than outdoor pollutants. In general volatile organic compounds (VOCs) can be given off by office products, insulating materials, synthetic furniture, cleaning and maintenance products, pressed wood, *etc*. or may originate from tobacco smoke[3]. Much effort has been devoted to characterize the levels of indoor air pollutants. Concentrations are known to be random variables because of their dependence on several sources and the fluctuations of emission variables[4]. Associations to adverse health effects such as allergic reactions, headache, eye, nose and throat irritation, dry cough, dizziness and nausea, concentration problems, tiredness[5] and even cancer[6] have been made to a poor indoor air quality[7]. These symptoms affect human health severely and lead to economic losses[8]. It is estimated that the 'sick building syndrome' results in a productivity decrease of average 6.5 % in offices. Mechanical or electronically filters can effectively trap particulate contaminants and remove them from the circulating air. Ionic air purifiers emit ions enhancing the agglomeration of smaller particles into larger ones, which then gravitationally settle. Ionization may also cause attraction between particles and grounded surfaces resulting in electrostatic precipitation[9]. It has been proven that accumulation of pollutants, may serve as nutrient source for moulds and bacteria. These organisms are able to produce volatile organic compounds such as aldehydes, ketones, alcohols, furans, esters and acids[10,11]. Sorption media (*e.g.*, activated carbon) only transfer volatile organic compounds to another phase rather than eliminating them[12]. The use of adsorbents implies the change, disposal or regeneration of the adsorbents[13]. The self oscillatory behaviour in toluene oxidation on zeolite-NaX[14] and metallic mixed oxides (Pt, Mn or Cr) as catalysts for the gas-phase toluene oxidation[15] has been reported.

Einaga and Futamora[16] have reported that benzene is oxidized on supported manganese oxide catalyst to form CO_x and various types of partially oxidized by products at room temperature (295 K) and that the build-up of these by products on the catalyst surface is the cause of catalyst deactivation. As an extension of the work, they carried out benzene oxidation with ozone over ion-exchange zeolite catalysts, Mn-Y, Mn-B, Mn-MoR and Mn-ZSM-S to further investigate the effect of catalyst support on the reaction[17].

In this work, we tried to synthesize a series of supported Pt catalysts (Pt/MeOx, Me = Fe, Zn, Al, Ni) by using colloid-deposition method and their catalytic performances on the toluene oxidation in the absence and presence of ozone are investigated.

EXPERIMENTAL

The schematic flow chart of pilot employed for the omitting toluene, as shown in Fig. 1. Catalytic oxidation of

toluene was carried out with a fixed-bed reactor. Reaction was carried out by using mixture of toluene with air. The toluene concentration, catalyst weight and gas flow rate, were 500 ppm, 0.05 g and 1000 mL/min, respectively. Ozone was synthesized from O_2 by a silent discharge ozone generator (Ebara Jitsugyo Co., Ltd. O_2SD-3000A). Ozone concentration in O_2 flow was controlled at 35, 55 and 65 ppm by monitoring ozone concentration with an ozone generator. Reactor diameter was 7 mm. Before the catalytic reaction, the sample in a pyrex glass reactor was heated in N_2 flow for 1 h.

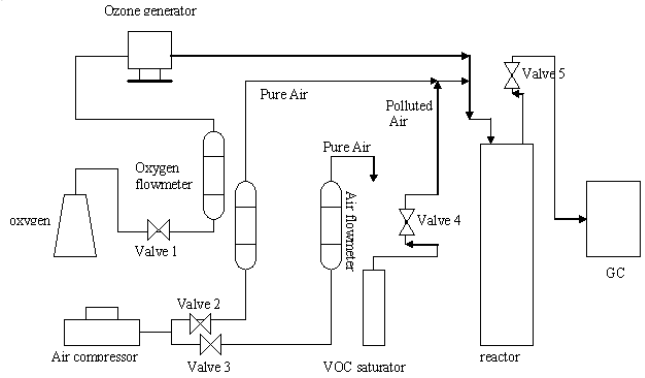


Fig. 1. Schematic diagram of the experimental apparatus

Catalyst preparation: The supported Pt catalysts were prepared by a colloid deposition method. The colloidal platinum was prepared according to the literature procedure[18]. Typically a glycol solution of NaOH was added into a glycol solution of H_2 $PtCl_6 \cdot 6H_2O$ (100 mL, 2 g/100 mL) was stirring for 1 h to form a brown solution. The colloidal solution was obtained by heating the brown solution at 140 ºC for 3 h under the protection of Ar.

Various precipitates of $Me(OH)_x$, (Me = Fe, Ni, Al or Zn), which are the precursors of oxide supports, were prepared by precipitation method using corresponded nitrate and Na_2CO_3 solution. After filtering, the hydrate precipitate was washed several times with distilled water.

The colloid-deposition process was carried out as following: the hydrates precipitate was re-suspended in a certain amount of distilled water, then the Pt colloidal solution was added to the suspension under stirring. The mixture was heated at 80 ºC under moderate stirring until total deposition of Pt colloid occurred. The precipitate was isolated and washed thoroughly with distilled water until free of chloride ions ($AgNo_3$ test). Finally, the product was dried at 80 ºC for 24 h and calcined at 300 ºC for 4 h in a flow rate of 20 % O_2/Ar to obtain the Pt supported catalysts (denoted as Pt/MeO_x). For the purpose of comparison, a reference sample of supported Pt/Fe_2O_3 catalyst (denoted as Pt/Fe_2O_3 -DP) was also prepared by a classical deposition-precipitation (DP) method[19,20].

In these tests, effects of reactor temperature, ozone concentration and different catalyst on the toluene omission percentage were inspected. At first contaminated air with toluene analyzed by GC machine. Then toluene oxidized with oxygen and ozone using different temperatures. The determination of toluene concentration, exiting air sent to GC machine. The concentration of toluene was obtained by using the following equation:

$$\text{Concentration of toluene} = 0.007 \times A$$

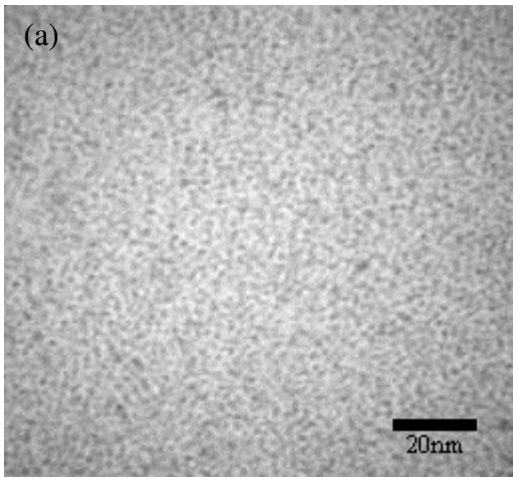
The concentration of toluene in ppm, A under surface of graph in GC machine that its unit is square meter.

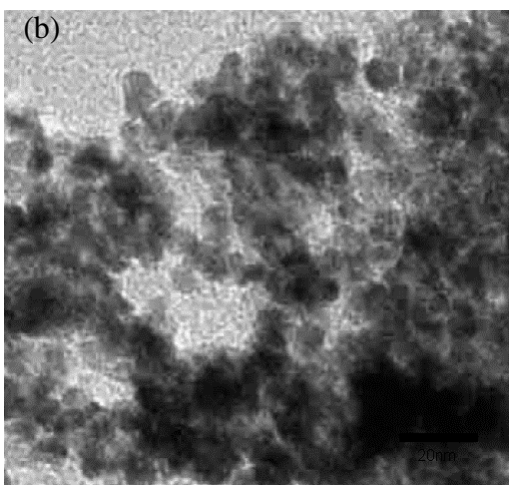
This function obtained from linearity of data in calibration of machine with standard solutions. In each test, input and output of toluene measured by GC machine. The tests repeated approximately twice and toluene omission percentage was measured by using ozone at 50 to 200 ºC.

The oxidation in natural air was measured 50 to 300 ºC. Every test began in high temperature and finished in lowest temperature. The main aim of this work was burnt the remaining probable fleas from the last test and prevent from toluene discharge. Also absorption occurs in the low temperatures. If the flow temperature can be used with toluene ascending disposal of high-temperature absorption level we are facing the disposal of toluene reduces test accuracy.

RESULTS AND DISCUSSION

TEM images of the native Pt colloidal solution and various Pt supported catalysts are shown in Fig. 2. As one can see, the Pt clusters in the native solution are uniform sphere with particle size in range of 1-3 nm (Fig. 2a). After deposited on the supports, it is clear that platinum nanoparticles are homogenously dispersed on the surface of supports (Fig. 2b-e). For the supported samples of Pt/Al_2O_3, Pt/NiO and Pt/ZnO, it can be seen that the particle size of cluster seems no obvious change in comparison with the colloidal Pt particles.





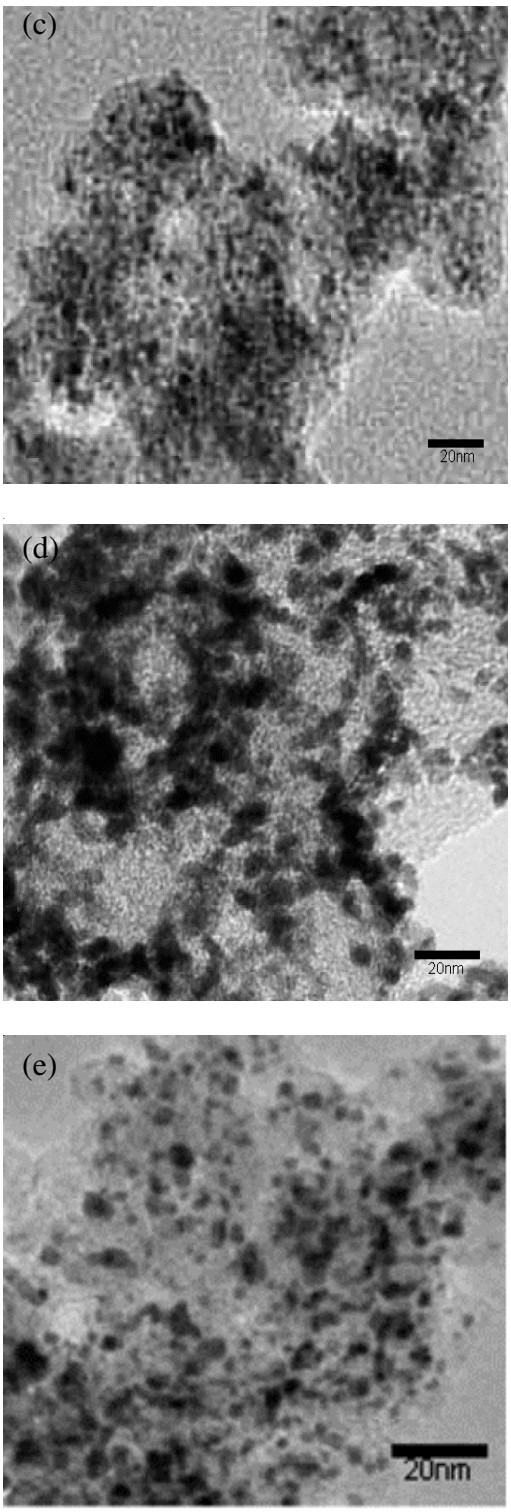


Fig. 2. Transmission electron micrograph of a) Pt and Pt catalyst based on b) Fe_2O_3, c) Al_2O_3, d) ZnO and e) NiO

For all the supported Pt catalysts, it can be seen, deposition of Pt particles on the oxide supported could slightly change the shape of these particles. The change might be caused by interaction between the Pt particle and the supported catalyst[21]. Previously, it has been claimed that the change in geometrics of noble metal particles, may lead to defect formation on the clusters of noble metals, thus may bring active sites on with the catalytic reaction could take place[22,23].

The catalytic performance of toluene oxidation in the various temperatures in the absence of ozone over various Pt/MeO_x catalysts is shown in Fig. 3. T_{100} (Temperature at, which 100 % toluene conversion has been achieved) is 200 ºC for Pt/Fe_2O_3, while it is higher than 200 ºC for others catalyst. Among them, Pt/Fe_2O_3 shows the best catalytic activity, it can convert toluene completely at about 200 ºC. It should be pointed out here, that the catalytic property of the reference sample of Pt/Fe_2O_3-DP (prepared by deposition-precipitation method) has also been investigated. Under the same condition, toluene can not be oxidized completely until the reaction temperature reach to 330 ºC over this Pt/Fe_2O_3-DP catalyst, which is much higher than the corresponding Pt/Fe_2O_3 prepared by colloid-deposition method. These results suggest that highly active supported Pt catalysts can be obtained by colloid-deposition method and the type of the support is also an important factor on influencing the catalytic performance for toluene oxidation. Concerning the fact that Pt particle size is almost identical on different supports, it can be concluded that the metal support interaction could considerably influence the catalytic properties of the supported Pt catalysts.

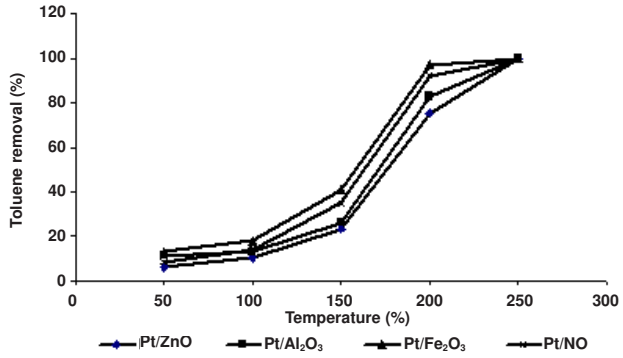


Fig. 3. Removal percentage of toluene at different temperatures in absence of ozone

The removal percentage of toluene at different temperatures in the presence of ozone are shown in Figs. 4-6. As can be seen in Fig. 4, T_{100} is 110, 130, 140 and 170 ºC for Pt/Fe_2O_3, Pt/NiO, Pt/Al_2O_3 and Pt/ZnO respectively, while ozone concen-tration was 36 ppm. It can be seen, according to data and results in all tests, Pt/Fe_2O_3 show the best catalytic activity in this study.

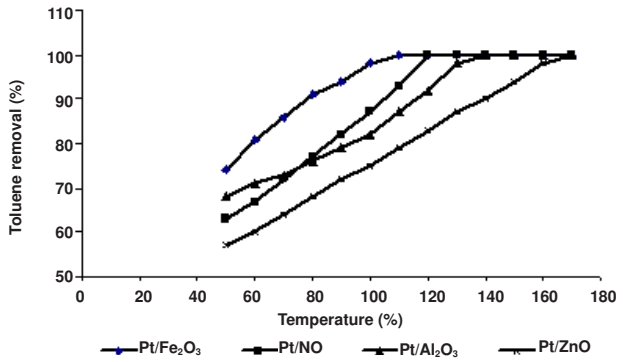


Fig. 4. Removal percentage of toluene at different temperatures in presence of ozone (35 ppm)

The results indicate that at 200 ºC and higher, the rate of omitted toluene was not dependent on the concentration rate of ozone, but in 50-200 ºC, the rate of toluene omission would be higher when the concentration of applied ozone increased. It can be seen in Figs. 4-6 at 100 ºC, the removal efficiency reached to 100 % with the concentration of 55 and 65 ppm of ozone and with concentration of 35 ppm, removal efficiency is more than 80 % because by increasing ozone concentration, effective surface and probability, effective collision of ozone with toluene increased.

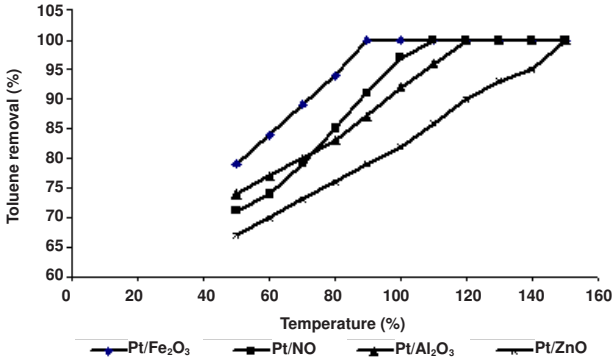


Fig. 5. Removal percentage of toluene at different temperatures in presence of ozone (55 ppm)

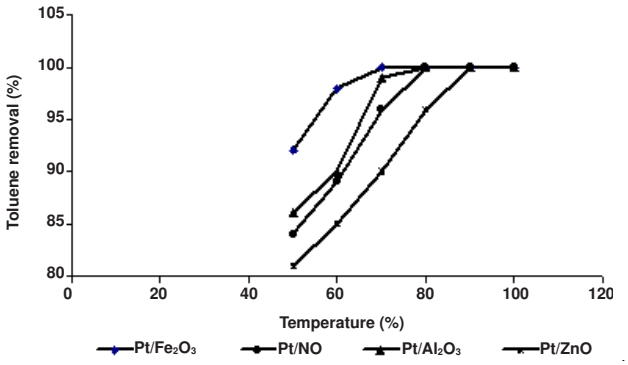


Fig. 6. Removal percentage of toluene at different temperatures in presence of ozone (65 ppm)

Conclusion

In this work, Pt/MeO_x catalysts (Me = Fe, Ni, Zn, Al) were investigated for the toluene oxidation in the absence and presence of ozone. Highly active Pt catalysts for toluene oxidation could be obtained by using colloid-deposition method. The type of the support is an importance of supported Pt catalysts. It was found that type of catalyst and concentration of ozone has the considerable effect on the toluene oxidation. Among the tested catalysts, Pt/Fe_2O_3 exhibits the highest activity.

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