Toluene photo catalytic degradation by ZnO coated on glass plates under simulated sunlight

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Abstract

In this study the photocatalytic degradation of toluene by ZnO nanoparticles coated on glass plates was studied under simulated sunlight. In order to immobilized ZnO nanoparticles on glass plates heat attachment method was used, then removal performance of these prepared surfaces was evaluated in a rectangular reactor under irradiation of metal halide lamp. The effects of operational parameters including initial toluene concentration, temperature, relative humidity, irradiation time, concentration of zinc oxide suspension on the removal of toluene were evaluated. The structural properties of ZnO nanoparticles and coated glass plates were characterized by scanning electron microscopy, X-ray diffraction and Field Emission Scanning Electron Microscopy. The concentration of toluene was analyzed by gas chromatography with flame ionization detector (GC-FID). Results showed, with variation of operational parameters such as initial toluene concentration, temperature, relative humidity, irradiation time, concentration of zinc oxide suspension, photo activity of ZnO supported on glass plate has different efficiency. Coating of glass surfaces by ZnO resulted in removal of 67% toluene at concentration of 50 ppm at 45 °C, relative humidity of 40% after 240 min irradiation of metal halide light. Results indicated that coating surfaces by ZnO nanoparticles can eliminate low concentration of toluene from polluted air under simulated sunlight.

Keywords: Toluene, Air pollution, Photocatalytic degradation, ZnO, simulated sunlight.

Introduction:

1. Introduction

In recent decades, the environmental pollution caused by volatile organic compounds has attracted a great deal of attention. Because of the high vapor pressure of these compounds, volatile organic compounds can be readily emitted from various sources such as industrial emissions, transport and gasoline service stations [1-3]. All these are the main precursor for the formation of photochemical smog, tropospheric ozone, and secondary aerosols [3,4]. Therefore, removal of these contaminants from the environment is essential to enhance the air quality and remove the adverse effects on human [5].

Many technologies are used to removing VOCs from polluted air such as physical adsorption, chemical absorption, membrane separation, catalytic combustion, and biological treatment. However, these technologies have disadvantages such as production of secondary pollution, high-energy consumption, high cost, and difficult operation [6-8]. Also, these methods are mostly applicable for removing high concentrations of the contaminants produced by industries [7].

Various studies have been carried out to develop techniques, which are both cost-effective and efficient for removing VOCs in urban areas [9,10]. Photocatalytic oxidation is a potential technology to removing organic contaminants in air by using semiconductor materials and presence of light [11,12].

Photocatalytic materials can be immobilized on the external surface of supporting materials, in the presence of light many organic pollutants can convert to less dangerous products such as water (H_2O) and carbon dioxide [13,12]. Currently, various studies have been performed on application of semiconductor materials on different surfaces such as glass, ceramics, color, and cement materials to remove air pollutants [14-16]. Most studies on the photocatalytic processes have focused on degradation of VOCs by using semiconductor oxides such as titanium dioxide (TiO_2), zinc oxide (ZnO), cadmium sulfide (CdS) [17,18]. However, these materials can be activated by ultraviolet (UV) light, which limits use of sunlight as energy source for photocatalytic reactions [19]. Zinc oxide (ZnO) is a semiconductor material with a large excitation binding energy (60 mev) and similar band gap energy to TiO_2 (3.2 ev), but have higher performance due to adsorption of large fraction of sun spectrum [20]. In the present study, ZnO nanoparticles were coated on glass plates to evaluate the removal efficiency of toluene under simulated sunlight irradiation using metal-halide lamp. The effect

of operational parameters, including initial toluene concentration, temperature, relative humidity, irradiation time, concentration of ZnO suspension, on degradation of toluene was evaluated.

2. Materials and methods

2.1. Materials

Toluene (purity 99.9%, CAS No.: 108-88-3) made by Merck Co (Germany) was used to synthesize the standard solution and prepare artificial polluted air entering the reactor. In addition, to extract toluene from charcoal tube, carbon disulfide (CS_2) prepared by Darkozist industrial and mineral research center was used. The ZnO nanoparticles was prepared from US Research Nanomaterials.

2.2. immobilization of ZnO on glass plates

The ZnO powders was immobilized on the glass plates (22*25, 12*25, 17*25 cm^2) by heat attachment method [21]. In this procedure, ZnO suspension with different dosage varying 3 to 12 g/L was prepared by adding ZnO powders to distilled water. Then, the prepared suspension was sonicated in an ultrasonic bath (Elmasonic S 80/H, frequency 37 kHz) to completely separate ZnO nanoparticles and obtains a more uniform solution.

The glass plates were first washed in a NaOH (0.01 M) solution to increase OH groups and for better contact between ZnO and the plates. Afterward, the prepared ZnO suspension was poured on the glass plates and then placed in an oven at 120°C for 1 h. After drying, the glass plates were calcinated in an electronic furnace (Fanazma Gostar Co., Iran, frequency 50Hz) for 3 h at 450°C. The immobilization process was carried out three times to increase the loaded ZnO on the surface of the plates.
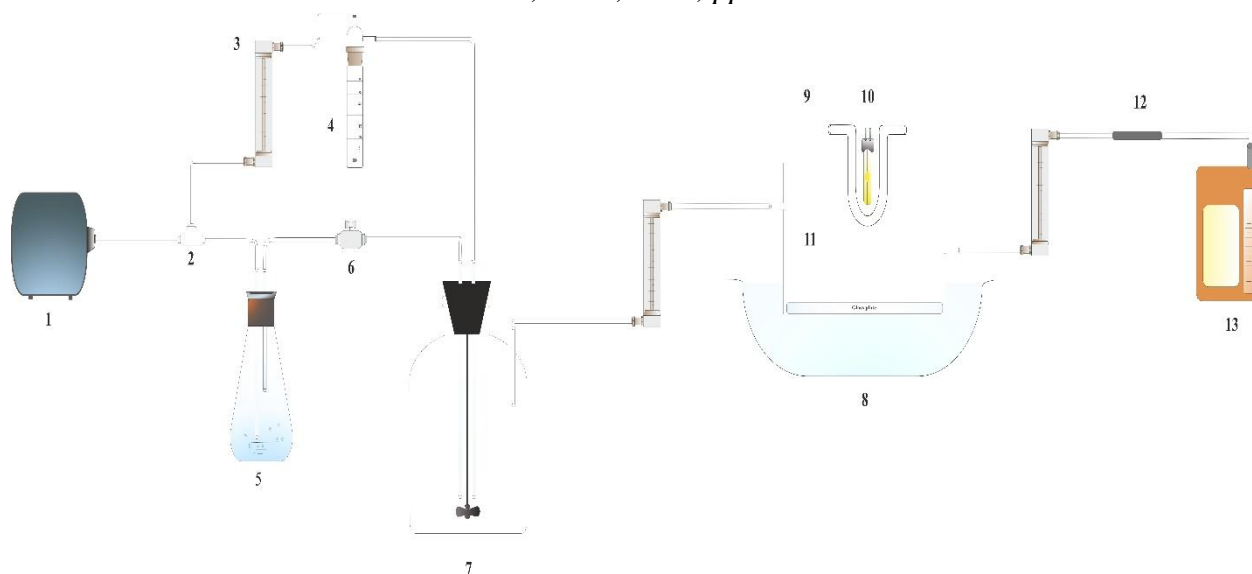
Fig 1. A schematic diagram of experimental set up. (1) air pump, (2) three way valve , (3) rotameter, (4) impinger containing pollutant, (5) humidifier, (6) control valve, (7) mixing chamber, (8) ice bath, (9) double-walled quartz jacket, (10) metal halide lamp, (11) reactor, (12) charcoal tube, (13) sampling pump

2.3. Experimental set up

The photocatalytic degradation experiments were carried out in a rectangular reactor made of Plexiglass with a volume of 15.6 L (30 cm*26 cm*20 cm). A 150 w metal halide lamp (345-800 nm) placed in the central upper part of the reactor and inside a double-walled quartz jacket was used. The intensity of illumination was measured by a luxmeter as 36200 lux. In order to control the temperature of the experiments, the reactor was placed inside a cold water bath. Furthermore, the reactor was covered with aluminum sheet in order to focus the whole lamp radiation into the reactor. The air entering the reactor was supplied by an air pump (Resun AC-9906). The pump's airflow was divided into two parts, part of it entered the impinger containing the pollutant liquid to supply contaminated air and the rest of the flow was entered into the humidifier. The resulting vapors then entered into the mixing chamber (Mariotte's bottle with a volume of 15 L) equipped with a stirrer to uniform the inlet flow to the reactor. The flow resulting from the mixing chamber entered the reactor after decontrolling the flow rate by the Rota meter. All of the experiments were conducted under laboratory hood to better control the environmental conditions of the reaction and prevent contamination of the laboratory environment. Fig. 1 shows the schematic diagram of the experimental set up used in this study.

In all experiments, after the time which had been obtained experimentally considering the flow rate entering the reactor and the volume of reactor, the inlet and outlet of the reactor were closed, and after reaching equilibrium of the flow inside the reactor, the metal halide lamp was turned on.

Before the photo catalytic experiments began, a set of experiments were conducted to evaluate the photolysis of the pollutant and adsorption of pollutant on glass plates, (1) in the presence of light and absence of catalytic plates and (2) in the absence of light and presence of catalytic plates. Furthermore, photocatalytic removal of toluene was conducted under similar conditions with control experiments conditions. All toluene concentrations were analyzed by gas chromatography with a flame ionization detector (GC-FID) (CHROM PACK Co., CP 9001).

2.4. Catalyst Characterization

The x-ray diffraction (XRD) patterns of ZnO powders was obtained on a X Pert MPD diffract meter (PANalytical, Netherlands) using ceramic X-ray tube with Co anode and the accelerating voltage and the applied current were 40 kV and 40 mA, respectively.

The structural properties of the surface and mean size of the nanoparticles were determined by scanning electron microscopy (SEM). Furthermore, after coating the nanoparticles on the glass plates, field emission scanning electron microscopy (FE-SEM) (MIRA3 TESCAN) was employed to identify surface structures of the coated glass plates.

## 3. Results and discussion

3.1. Characterization

The XRD pattern of ZnO powders is shown in Fig. 2. All these diffraction peaks could be index to ZnO crystal phase (JCPDS 5-0664) and there were no other peaks have been determined from other phases, indicating that the crystalline phase is individually related to zinc oxide. The result of SEM image (Fig. 3a) shows that the size of nanoparticles is around 20 nm. As presented in Fig. 3b, the ZnO nanoparticles were successfully coated on the glass plates.
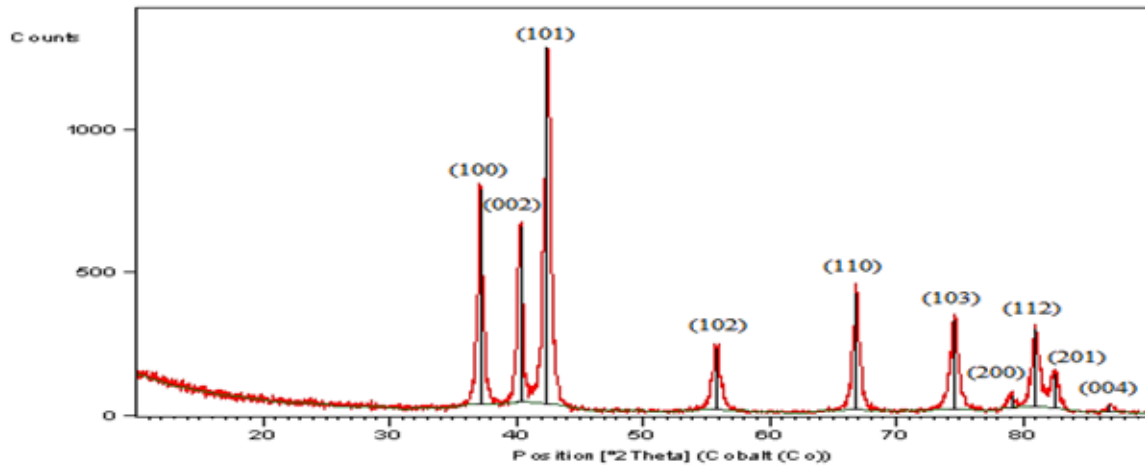
Fig 2. XRD pattern of ZnO
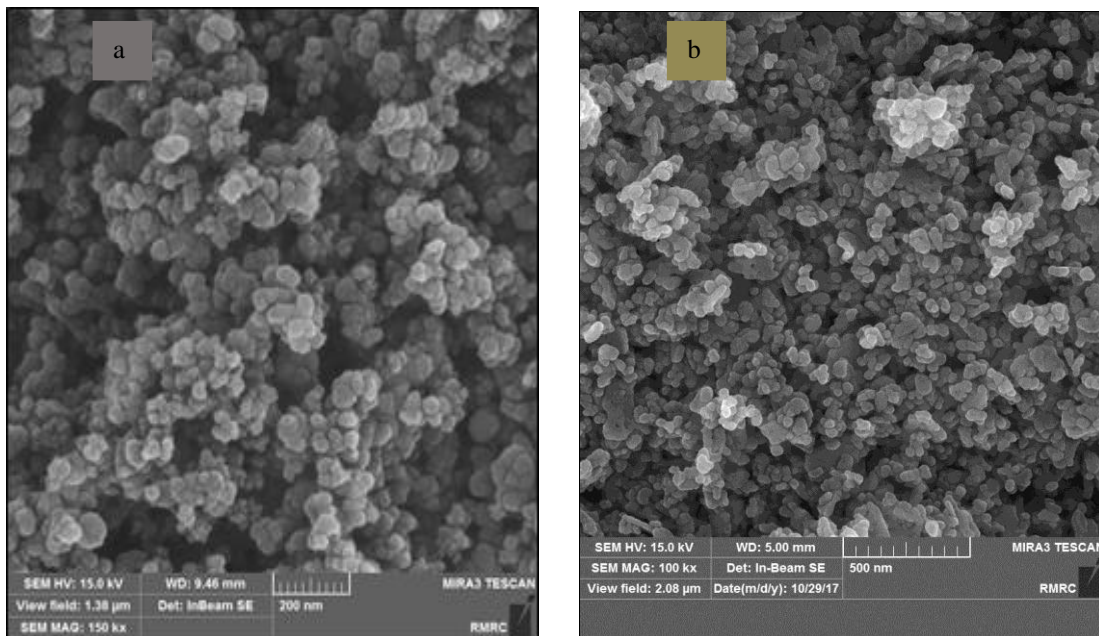
Fig 3. (a) SEM image of ZnO nanoparticles, (b) FE-SEM image of ZnO coated on glass plate
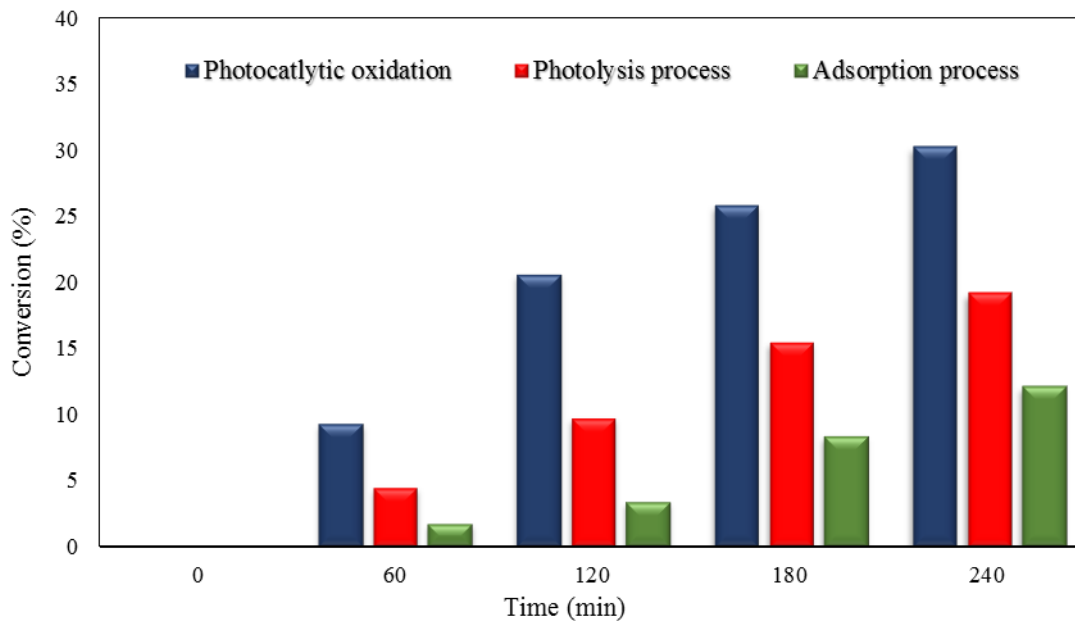
Fig 4. Toluene conversion by photo catalytic oxidation, photolysis, and adsorption process. Initial toluene concentration 50 ppm, at 25 °C, relative humidity 10%, and ZnO suspension dosage 10 g/L

3.2. Comparison of photocatalytic oxidation with controlling experiments

The results of Fig .4 show that irradiation of metal halide lamp only removed 19.74% of toluene. In addition, using catalytic plates alone under dark condition led to 12% of toluene adsorbed. However, photacatalytic degradation of toluene under same condition illustrated that 30.25% of toluene removed after 240 min. It can be found that under irradiation of metal halide lamp, presence of ZnO could accelerate the reaction and more pollutant removed. Masoud rismanchian et al. (2014) indicated that irradiation of UV lamp alone and using uncoated nickel foam resulted in 8% toluene removal, but photo catalytic oxidation of toluene led to 50% of toluene removed after 270 min [22].
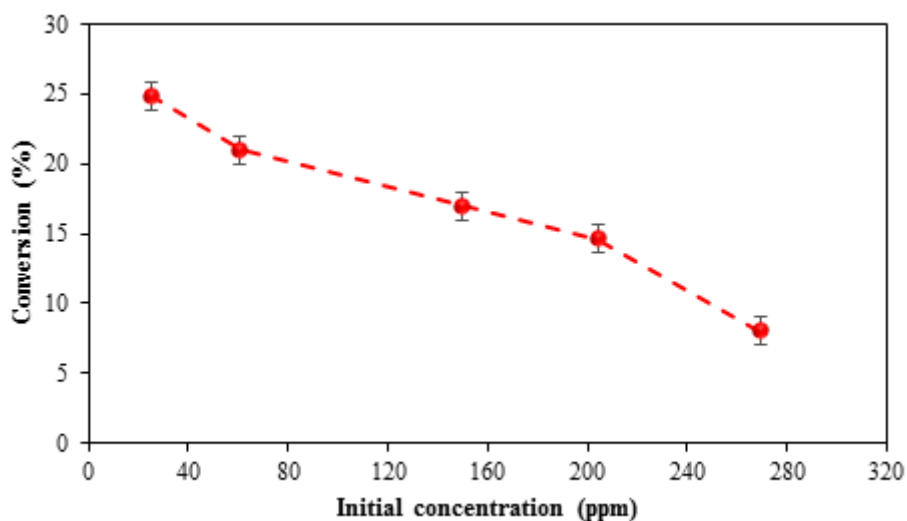
Fig 5. Effect of initial pollutant concentration. at 25 °C, relative humidity 10%, irradiation time 120 min, and ZnO suspension dosage 10 g/L

3.3. Effect of initial concentration of the pollutant

The effect of initial concentration on photocatalytic degradation of toluene was shown in Fig 5. The result shows that with increasing of the initial pollutant concentration, the photocatalytic degradation of toluene decreased. As shown in Fig 5, increasing toluene concentration led to decreased the degradation efficiency of toluene from 24.86 to 8%.

The effect of the inlet concentration of the pollutant in the field of photocatalytic oxidation has been studied by different researchers [23,24]. Abbas Rezaee et al. (2008) showed that increasing of toluene concentration caused reduced photocatalytic efficiency by TiO_2 nanoparticles [25].

Previous studies indicated that as the pollutants concentration increase, more pollutant molecules could adsorb on the photo catalyst surface [26,27]. Therefore, higher active oxidant species such as hydroxyl radical to degrade this high concentration of pollutants also need [28]. In addition, the excess pollutant molecules on the photo catalyst surface prevent adsorption of photons by the photcatalyst. Thereby, the active oxidant species such as hydroxyl radicals (OH·) and superoxide anions ($O_2^{\bullet-}$) are inadequate for photo catalytic oxidation of high concentration of pollutants [29].
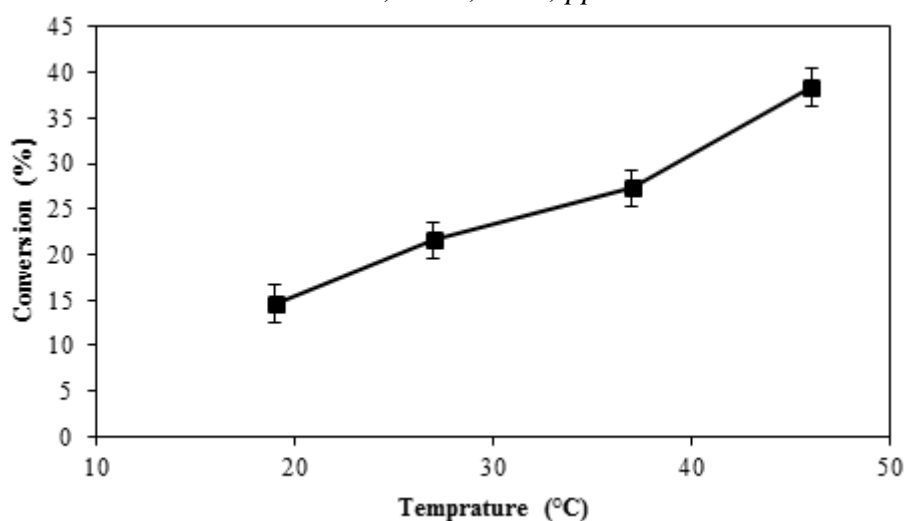
Fig 6. Effect of temperature. Initial toluene concentration 50 ppm, relative humidity 10%, irradiation time 120 min, and ZnO suspension dosage 10 g/L

3.4. Effect of temperature

In the present study, experiments were conducted at different temperatures including 19, 25, 35, and 45°C. As can be seen from Fig 6, increasing the temperature has led to enhanced efficiency of the photo catalytic degradation of toluene, increasing the temperature to 45°C led to 38.374% removal for toluene.

Generally, Temperature could affect the photo catalytic oxidation reaction and adsorption of pollutant on photo catalyst surface [28]. At high temperatures, the rate of chemical reaction and desorption of the product from the surface of the photo catalyst increases. According to Arrhenius temperature dependence coefficient, k, increasing the temperature has a positive impact on photo catalytic oxidation kinetic reaction [30].

$$k = f^{\left(\exp\left(\frac{-E}{RT}\right)\right)} \tag{1}$$

where E is an apparent activation energy, normally greater than 0; T is the temperature and R is the gas constant (8.314×10^{-3} kJ. mol^{-1}. K^{-1}). However, increasing temperature led to reduction of rate of adsorption of the pollutant on the catalyst surface [28]. The adsorption equilibrium coefficient, K, was calculated according to following equation [30].

$$K = f^{\left(\exp\left(\frac{-H}{RT}\right)\right)}/\sqrt{T} \tag{2}$$

Where H is the change in enthalpy adsorption for the adsorbed pollutant. In the present study, as typical air temperatures were investigated, increasing the temperature led to increase the removal efficiency of toluene. It can be implied that reduced adsorption of the pollutant at optimal temperature had no effect on the general efficiency of the photo catalytic process.

Wei Cai et al. (2014) studied the removing of acetone and ethanol by TiO_2 at different temperatures including 14, 31, and 43°C. They observed that higher temperatures led to enhanced acetone and ethanol removal [31]. Fumihide Shiraishi et al. (2005) studied the efficiency of removing formaldehyde by TiO_2 across the temperatures of 45, 55, 70, and 90°C, they observed that the temperature 45°C had the maximum efficiency in formaldehyde removal [32].
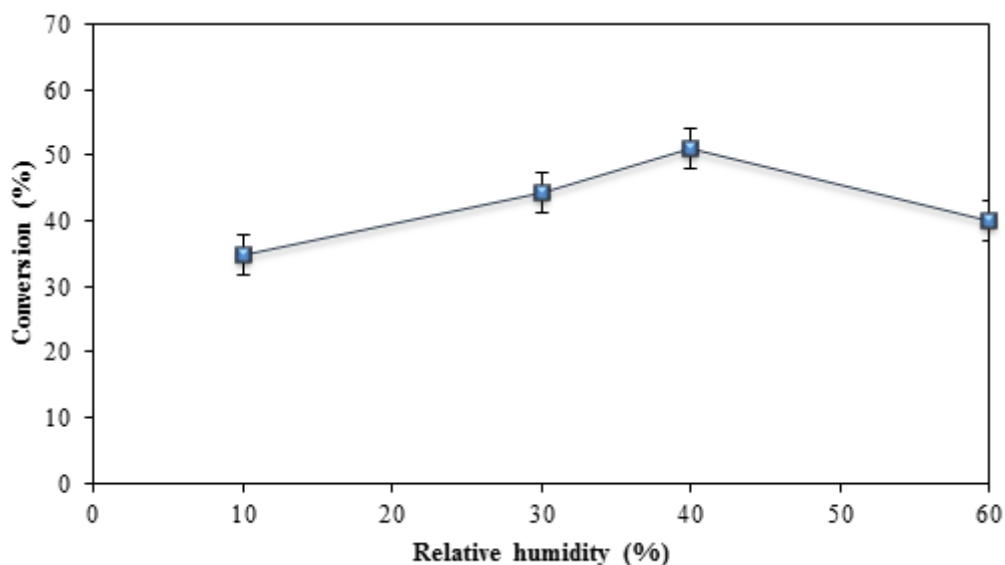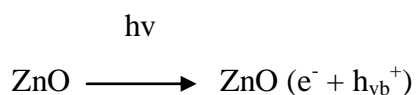

Fig 7. Effect of relative humidity. Initial toluene concentration 50 ppm, 45 °C, irradiation time 120 min, and ZnO suspension dosage 10 g/L
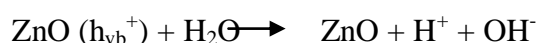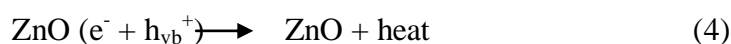
3.5. Effect of relative humidity

The effect of relative humidity on photocatalytic degradation of toluene was examined using different RH including 10, 30, 40, and 60%. The result in Fig 7 shows that with increasing humidity, the removal efficiency of the toluene has increased. However, increasing the humidity from 40 to 60% led to diminished photo catalytic efficiency.
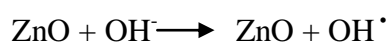
Water vapor is a very important source of hydroxyl radicals in photo catalytic oxidation, therefore presence of water vapor is essential for strengthening the photo catalytic process [29]. The generation of $OH^\bullet$ are described by follow equations:

$$ZnO \xrightarrow{h\nu} ZnO\,(e^- + h_{vb}^+) \tag{3}$$

$$ZnO\,(e^- + h_{vb}^+) \longrightarrow ZnO + heat \tag{4}$$

$$ZnO\,(h_{vb}^+) + H_2O \longrightarrow ZnO + H^+ + OH^- \tag{5}$$

$$ZnO + OH^- \longrightarrow ZnO + OH^\bullet \tag{6}$$

Howover, excessive water molecules on the catalyst surface form multi-layers of water on the catalyst surface and inhibit adsorption of the pollutant on the catalyst surface [29]. Mohamad Sleiman et al. (2009) observed that the maximum conversion of toluene using TiO_2 achieved at the humidity of 30%. However, at 50 and 70%, toluene removal efficiency decreased [33]. Masoud Rismanchian et al. (2014) studied the removal efficiency of toluene by TiO_2 nanoparticles imobilized on nickel metal foam at different humidity 10, 30, 50 and, 80%. They found that increasing humidity first increased removal efficiency of the pollutant then elevation humidity from 30 to 80% resulted in decreased photocatalytic efficiency [22].
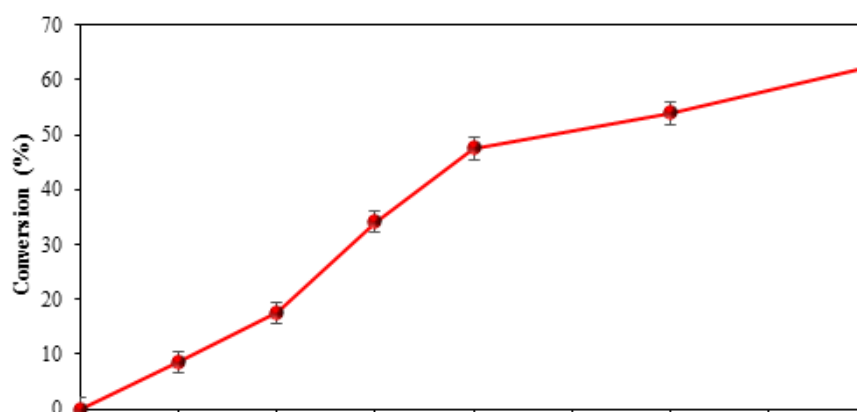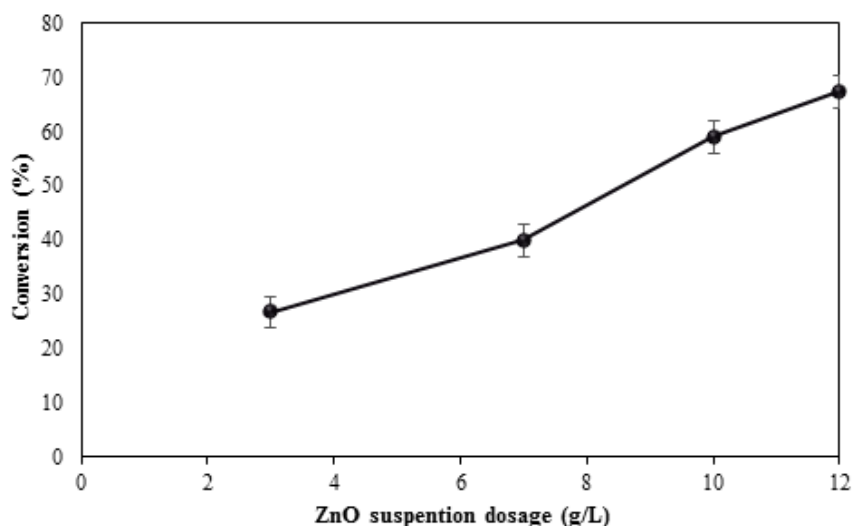
Fig 8. Effect of irradiation time. Initial toluene concentration 50 ppm, 45 °C, relative humidity 40%, and ZnO suspension dosage 10 g/L

3.6. Effect of irradiation time

Investigating the effect of irradiation time on the photocatalytic efficiency of ZnO in removal of toluene implied that, photocatalytic efficiency of ZnO in removal of toluene has increased by increasing the irradiation time. As shown in Fig 8, at the irradiation time 30 min the removal efficiency of toluene was 8.59. However, increasing the irradiation to 240 min has increased the toluene conversion to 62. In addition, higher contact times enhance the mass transfer coefficient between the air and photocatalyst surface. Therefore, more molecules of the pollutant could adsorb on the active sites of the photocatalyst surface and react with the hydroxyl radicals [27].

Previous studies also implied enhanced removal efficiency with increasing the irradiation time. Gholamhossein Pourtaghi et al. (2009) evaluated the photocatalytic oxidation of toluene by TiO_2 at times of 10, 30, and 60 min and at inlet pollutant concentrations of 150, 250, 350, and 450 ppm. They indicated that toluene removal efficiency was maximum at the retention time of 60 min and pollutant concentration of 150 ppm [34].

Masoud Rismanchian et al. (2014) observed that retention times lower than 100 min did not have a good efficiency in removing toluene by TiO_2 nanoparticles immobilized on nickel metal foam. However, with increasing the irradiation time to 270 min, almost 55% of toluene was removed [22].

Fig 9. Effect of ZnO suspension dosage. Initial toluene concentration 50 ppm, 45 °C, relative humidity 40%, and irradiation time of 240 min

3.7. Effect of ZnO suspension dosage

Increasing the ZnO suspension concentration led to increased loading of nanoparticles on the glass surface. Therefore, the surface area of nanoparticles and the number of active sites on the catalyst surface increase which elevates the amount of adsorbed pollutants [29].

Morteza Jafarikojour et al. (2015) showed that by increasing the amount of TiO_2 coated on stainless steel sheets from 0.274 to 0.925 g/m^2, the degradation of toluene enhanced from 26% to 43% [35]. Behnajady et al. (2009) also observed that increasing the ZnO suspension concentration led to further loading of ZnO on the glass plates, thereby enhancing the pollutant removal efficiency. The results indicated that an increase in the ZnO suspension dosage from 2 g/L to 12 g/L showed that the suspension with the concentration of 12 g/L had the maximum efficiency, though no significant difference was observed between the results obtained from the photocatalytic oxidation with suspension concentrations of 10 and 12 g/L[21] .

Hisahiro Einaga et al. (2004) also illustrated that as the amount of TiO_2 loading was changed from 0 to 0.24 g, the averaged catalyst weight was increased to 2 mg/cm^2 and benzene conversion was enhanced [36].

4. Conclusion

In the present study, the efficiency of photocatalytic degradation of toluene by ZnO nanoparticles coated on glass surface under simulated sunlight was evaluated. Results indicated that by variation of operational parameter such as initial toluene concentration, temperature, relative humidity, irradiation time, concentration of ZnO suspension, photocatalytic degradation of toluene changed. It can be stated that the mentioned photocatalytic process had a relatively good efficiency under irradiation of metal halide lamp. Coating of glass surfaces by ZnO suspension, resulted in 67% removal of toluene as concentration of 50 ppm at 45 °C, and relative humidity of 40% after 240 min irradiation of metal halide light.

However, under realistic conditions various parameters affect the process which cannot be considered in the experiment. Therefore, the photocatalytic degradation efficiency may be different under real conditions, illustrating that application of this method requires further investigation. Furthermore, air pollution contains a mixture of contaminant that may inhibit

photocatalytic removal of toluene. Therefore, further studies should be carried out according to removal in a mixture of air pollutants by the photocatalytic oxidation process.

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