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# Degradation of gaseous 1,2-dichloroethane using a hybrid cuprous oxide catalyst

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## ABSTRACT

A hybrid catalyst (CZ50) of cuprous oxide and zeolite was prepared and used in a catalytic system in which gaseous 1,2-dichloroethane (1,2-DCE) was degraded. The effects of relative humidity (15 and 65%) and wavelength of irradiation (ultraviolet, blue, green and red light) on the degradation of 1,2-DCE were studied. The experimental results reveal that the hybrid catalyst degraded the hazardous material under visible light. A comparison of the results obtained under ultraviolet and red irradiation at a relative humidity of 15% revealed almost equal degradation efficiencies of 83.8 and 82.2%, respectively. In the system with hybrid catalysts, a lower relative humidity favored 1,2-DCE degradation, indicating that relative humidity critically affects the degradation of hazardous materials. The most important result was that the intermediates of the degradation of 1,2-DCE included ethane, carbon dioxide, and formic acid yielded, which are mostly harmless and non-toxic materials. The experimental results indicated that the method could effectively be used to degrade hazardous materials to provide a healthy and safe environment for workers.

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**Keywords:** Cuprous oxide; Degradation; Visible light; Hybrid catalyst; 1,2-Dichloroethane

## 1. Introduction

Volatile organic compounds (VOCs) are used extensively in industrial processes, such as those associated with the petrochemicals, plastics and semiconductor sectors. VOCs are important and hazardous carcinogens, mutagens and teratogens. 1,2-Dichloroethane is a chlorinated volatile organic compound (ClVOC) that is utilized as an industrial solvent, and is used mostly in the production of vinyl chloride, which is used to make polyvinylchloride (PVC). 1,2-DCE decomposes into products that remain in the atmosphere for a long period (Vogel et al., 1987). The International Agency for Research on Cancer (IARC) has classified 1,2-DCE as a suspected carcinogen (Group 2B), reporting that genes and chromosomes can be mutated by exposure to 1,2-DCE under unsettled conditions (Gwinn et al., 2011). 1,2-DCE represents an important hazard to the working environment and human health.

VOCs control technology includes condensation, absorption, adsorption, incineration, and biological treatment.

However, these VOCs control methods generate hazardous byproducts or secondary pollutants and so are not ideal (Yan et al., 2010). Therefore, photocatalytic degradation has recently been developed as a promising strategy for controlling hazardous VOCs (Vincent et al., 2011) and it has been confirmed to have unlimited potential for degradation of the hazardous material.

Zeolite consists of silicon tetrahedra and aluminum tetrahedra that are linked via oxygen atoms. The important characteristics of zeolite include its being hydrated, cation-exchangeable and catalytic, and that it has voids in its crystal structure (Breck, 1974). In this study, zeolite is used as a carrier that is mixed with cuprous oxide ( $\text{Cu}_2\text{O}$ ) to prepare hybrid catalyst.

Cuprous oxide is a p-type semiconductor with a direct band gap of 1.9–2.2 eV, and can be activated by irradiation of visible light; it has been used in solar cells (Chen, 2013), the synthesis of catalysts (Zhu et al., 2012) and treating bioaerosols and other chemicals.  $\text{Cu}_2\text{O}$  is non-toxic, highly stable, readily available

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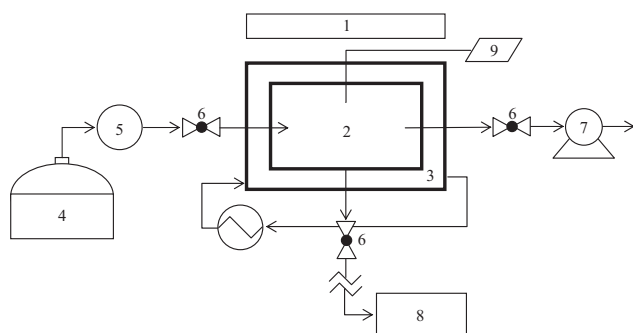


Fig. 1 – Schematic diagram of experimental set-up.

and environmentally friendly; it can also degrade organic pollutants to form  $\text{CO}_2$  and  $\text{H}_2\text{O}$ . In recent years,  $\text{Cu}_2\text{O}$  has been used in the photocatalytic degradation of hazardous material in various studies.

In this study, cuprous oxide is mixed with zeolite as a carrier to prepare a hybrid catalyst, which is used in a catalytic system to degrade the gaseous 1,2-DCE. The effects of relative humidity of 15 and 65% and of the wavelength of irradiation (ultraviolet, blue, green and red light) on the degradation of 1,2-DCE were studied.

## 2. Experimental methods

### 2.1. Preparation and characterization of catalysts

The  $\text{Cu}_2\text{O}$ /zeolite hybrid catalyst was prepared using an immobile method. Y-zeolite is zeolite that consists of silicon dioxide and sodium aluminum silicate in a ratio of 65/35 wt%. Y-zeolite powder was thoroughly mixed with 50 wt%  $\text{Cu}_2\text{O}$  powder with 0.75 N sodium hydroxide as a gelling agent. The mixture was shaped into strips using a granulator and then placed at  $105^\circ\text{C}$  for 12 h in a muffle furnace to dry it. Each strip was cut into lengths of 1.5–2.5 cm. The granular hybrid catalyst (CZ50) of cuprous oxide and zeolite was thus obtained, and CZ00 was obtained by following the above method without adding  $\text{Cu}_2\text{O}$ .

The prepared catalysts were characterized to demonstrate the phase and energy gap by X-ray Diffraction (BRUKER-AXS D8-Advanced, XRD) and UV-visible Spectrophotometer (JASCO, V-670) at wavelength range of 200–8020 nm.

### 2.2. Photodegradation experiments

Fig. 1 depicts the experiments system for degradation. The photocatalytic experiments were performed in a double-layered glass cylindrical batch reactor tank with an internal volume of 2.85 L, diameter 19 cm and height 12 cm, and an external diameter 25 cm and height 18 cm. The outer layer of the reactor tank included a constant-temperature water bath to control the temperature of the internal environment at  $35 \pm 2^\circ\text{C}$ . It was mounted on a magnetic stirrer, with a rotational speed of 800 rpm to maintain the uniformity of the gas. The relative humidity was monitored and controlled at  $65 \pm 5\%$  or  $15 \pm 5\%$  using a temperature-humidity detector. The top of the reactor is covered the quartz glass of thickness 0.5 cm. Before the experiment was carried out, 45 g CZ50 was placed 6 cm from the top of the internal reactor. The pump was used for 3 min to release air from the reactor. Gaseous 1,2-DCE was extracted using a gasification device. Subsequently, the gasification of bag was connected to a nitrogen source to prepare initial concentration of 1,2-DCE. The initial

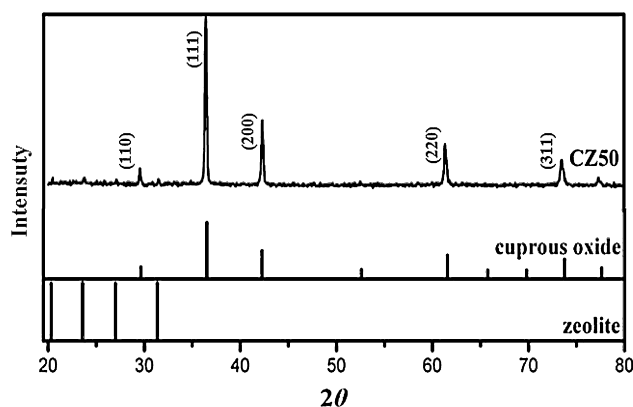


Fig. 2 – XRD patterns of CZ50.

concentration of gaseous 1,2-DCE in the reactor was  $20 \pm 5$  ppm. 1,2-DCE was adsorbed by  $\text{Cu}_2\text{O}$ /zeolite catalysts under dark conditions 30 min before irradiation; then 1,2-DCE was irradiated under an ultraviolet (365 nm), blue (460–465 nm), green (520–525 nm) or red (620–625 nm) lamps at a power of 36 W for 120 min.

The concentration of 1,2-DCE was analyzed using GC-ECD. The degradation efficiency of 1,2-DCE was calculated by  $C/C_0$ , where  $C_0$  is the initial concentration of 1,2-DCE and  $C$  is the concentration of 1,2-DCE after irradiation at the sampling time. The intermediate and product after degradation of 1,2-DCE were qualitatively analyzed using FTIR with gas cell. The gas cell was constituted by 6 cm glass cylinder and two covers and it was filled the 15 mL of gas sample to analyze.

## 3. Results and discussion

### 3.1. XRD analysis of catalyst

Fig. 2 presents the XRD pattern of the CZ50. The crystallinity of cuprous oxide and zeolite was verified with reference to No. 05-0667 of the data bank of the International Center of Diffraction Data (ICDD) – formerly the Joint Committee on Power Diffraction Standards (JCPDS). The five peaks correspond to the crystal planes (1 1 0), (1 1 1), (2 0 0), (2 2 0) and (3 1 1) of crystalline cuprous oxide. One peak is observed at a  $2\theta$  of  $35.6^\circ$ – $36.5^\circ$ , and is characteristic of cuprous oxide. The most intense peak was at  $23.73^\circ$ – $23.77^\circ$ , corresponding to zeolite. No other diffraction peak from any possible impurity, such as  $\text{CuO}$ , was obtained, confirming that the cuprous oxide was not oxidized to copper oxide during drying.

### 3.2. Optical properties of catalyst

The UV-vis reflectance ( $R_\infty$ ) of the catalyst was converted to an absorption value ( $F$ ) using the equation, Eq. (1). Fig. 3 plots  $\text{SQRT}[F(R) \times h\nu]$  as a function of the energy gap. The figure also presents the UV-vis absorption spectra of the photocatalysts. The energy gap can be found from the tangent of the curve in the figure (Yoneyama et al., 1989). The energy gaps of CZ00, CZ50 and  $\text{Cu}_2\text{O}$  were 2.75, 1.94 and 1.60 eV, respectively. Eq. (2) was utilized to determine the absorption bands of CZ00, CZ50 and  $\text{Cu}_2\text{O}$ , which were 451.2, 460.2 and 775.5 nm, respectively. Cuprous oxide is well known to be able to be activated as a photocatalyst under irradiation by visible light. The band gap of CZ50 became smaller and the absorption band shifted toward visible wavelengths as the amount of  $\text{Cu}_2\text{O}$  on the zeolite increased. Therefore, the CZ50 hybrid photocatalyst was

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