



# Removal of ethylene from air stream by adsorption and plasma-catalytic oxidation using silver-based bimetallic catalysts supported on zeolite



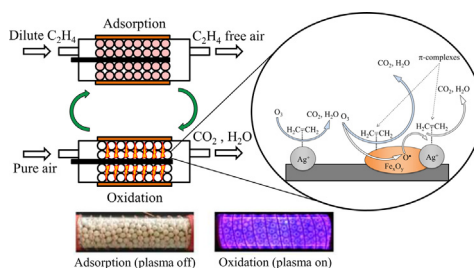
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## HIGHLIGHTS

- Energy-efficient VOC abatement by cyclic adsorption/plasma-catalytic treatment.
- 13X zeolite-supported Ag and metal oxides as dual functional adsorbent/catalyst.
- Adsorption enhancement by the co-incorporation of Ag and metal oxide into 13X zeolite.
- Bimetallic Ag–Fe<sub>2</sub>O<sub>3</sub> capable of effectively reducing O<sub>3</sub> and increasing VOC oxidation.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Dynamic adsorption of ethylene on 13X zeolite-supported Ag and Ag–M<sub>x</sub>O<sub>y</sub> (M: Co, Cu, Mn, and Fe), and plasma-catalytic oxidation of the adsorbed ethylene were investigated. The experimental results showed that the incorporation of Ag into zeolite afforded a marked enhancement in the adsorptivity for ethylene. The addition of transition metal oxides was found to have a positive influence on the ethylene adsorption, except Fe<sub>x</sub>O<sub>y</sub>. The presence of the additional metal oxides, however, appeared to somewhat interrupt the diffusion of ozone into the zeolite micro-pores, leading to a decrease in the plasma-catalytic oxidation efficiency of the ethylene adsorbed there. Among the second additional metal oxides, Fe<sub>x</sub>O<sub>y</sub> was able to reduce the emission of ozone during the plasma-catalytic oxidation stage while keeping a high effectiveness for the oxidative removal of the adsorbed ethylene. The periodical treatment consisting of adsorption followed by plasma-catalytic oxidation may be a promising energy-efficient ethylene abatement method.

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## 1. Introduction

Volatile organic compounds (VOCs) cause serious problems for environment and human health [1–5]. Among VOCs, ethylene is an important raw material widely used in various industrial reactions [6]. Besides, many agricultural commodities produce ethylene

during storage and transportation [7]. Ethylene can bring about physical and chemical changes in the appearance and texture of fruits and vegetables such as yellowing and softening [8], thereby considerably degrading the quality.

Non-thermal plasma (NTP) generated by dielectric barrier discharge (DBD) in combination with catalysts has been reported as an effective tool for VOCs abatement [9–12]. Among heterogeneous catalysts, silver has broadly been used not only for commercial production of ethylene oxide but also for ozone decomposition and deep VOC oxidation [13–22]. In recent years, the

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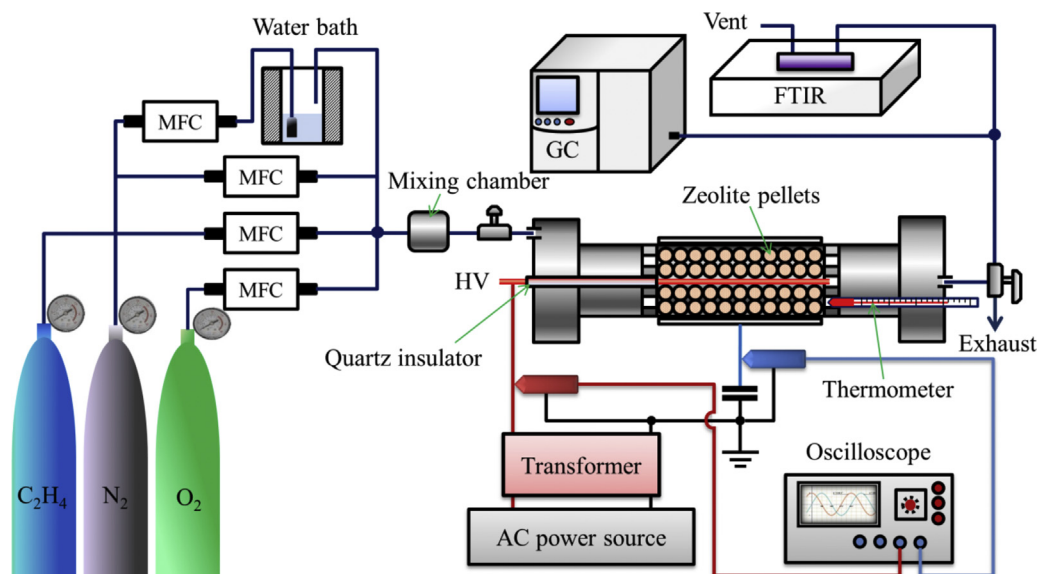


Fig. 1. Experimental setup for the cyclic treatment of ethylene.

plasma-catalytic oxidation in combination with the enrichment of VOCs by adsorption has been investigated elsewhere to enhance the energy efficiency, particularly for dilute VOCs [23,24]. Zeolite as an adsorbent for VOCs like ethylene has received much attention, so far [25–27]. The strength of interaction between cations existing in zeolite framework and ethylene double bond is known to be responsible for its excellent adsorption capabilities [28,29]. The adsorption of ethylene on silver was also investigated in previous studies, mainly for catalytic epoxidation of ethylene [30–32]. According to the previous article [33], the presence of subsurface oxygen on Ag(110) enhances the binding energy between adsorbed oxygen and ethylene.

The objectives of this work are to investigate the dynamic adsorption of ethylene over 13X zeolite-supported Ag and Ag- $M_xO_y$  (M: Co, Cu, Mn, and Fe), and influences of the second transition metal oxides incorporated together with Ag on the catalytic oxidation of adsorbed ethylene under plasma-activation conditions. The oxidation activities of the above catalysts and the formation of unwanted byproducts during the plasma-catalytic oxidation of the ethylene enriched in the zeolite were evaluated. The dependences of the oxidation time required to completely oxidize the adsorbed ethylene on the applied voltage and adsorption time were also examined. Finally, the mechanisms for the adsorption of ethylene and plasma-catalytic oxidation of adsorbed ethylene were explained.

## 2. Experimental

### 2.1. Apparatus and materials

The adsorption and oxidation of ethylene were conducted in a packed-bed DBD reactor depicted in Fig. 1. The reactor consisted of a quartz tube (inner diameter: 25.5 mm; thickness: 2 mm) wrapped outside with a 100-mm long aluminum foil. A 5.5-mm threaded steel rod serving as the high voltage electrode was coaxially inserted into the quartz tube, resulting in a 10-mm discharge gap. The reactor was energized by an alternating-current voltage of 400 Hz.

Commercial 13X pelleted zeolite (diameter: 4.2 mm, Cosmo Fine Chemicals) with a BET surface area of  $460 \text{ m}^2 \text{ g}^{-1}$ , which was measured by a surface area analyzer (AUTOSORB-1, Quantachrome),

was coated with silver and with silver-metal oxides ( $M_xO_y$ , M: Co, Cu, Mn, and Fe). The dual function catalysts were prepared by incipient wetness impregnation. An appropriate amount of metal nitrate was dissolved in deionized water (20 mL water for 40 g zeolite) under stirring condition for 30 min. For the preparation of the bimetallic catalysts, mixtures of silver nitrate and the nitrate of each corresponding metal were used. The prepared solution was added dropwise onto the zeolite pellets. Then, the impregnated zeolite was dried overnight at  $110^\circ\text{C}$  before calcined at  $500^\circ\text{C}$  for 5 h in ambient air. For convenience, the catalyst samples were named as Ag(*a*%)/13X and Ag- $M(b-c\%)/13X$ , such as Ag(1.0%)/13X and Ag-Co(1.0–0.7%)/13X, in which *a–c* stand for the weight percentages of the corresponding metals. The reactor was packed with 29 g of each catalyst, unless otherwise mentioned.

Synthetic ethylene-contaminated gas was prepared by mixing pure ethylene with air ( $O_2$ : 21%;  $N_2$ : 79%) using mass flow controllers (MFCs). So as to examine the effect of water vapor on the adsorption of ethylene, the feed gas was humidified to 50% relative humidity by bubbling water with  $N_2$  in a temperature-controlled water bath. All the adsorption experiments were conducted at room temperature. For the cyclic treatment of ethylene, the ethylene-air mixture was first enriched in the adsorbent for a certain time period. After the adsorption step, the dilute ethylene was closed, and pure air was fed to the reactor with plasma switched on to oxidize the adsorbed ethylene.

### 2.2. Analyses and measurements

The structures of the zeolite before and after incorporating active metallic components were analyzed using an X-ray diffraction (XRD) system (D/MAX 2200H, Bede 200, Rigaku Instruments). The dispersion of active phases was characterized using transmission electron microscopy (TEM, JEM-2100F, JEOL). The surface chemistry of the catalysts was characterized by X-ray photoelectron spectroscopy (XPS, Theta Probe AR-XPS System, Thermo Fisher Scientific) with monochromatic Al  $K\alpha$  radiation (1486.6 eV) operated at 15 kV and 150 W X-ray excitation sources. The binding energies were calculated with reference to C1s peak (284.6 eV).

The electrical measurement has been described elsewhere [12]. The power consumed in the plasma reactor was determined using

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