



# Catalytic performance of plasma catalysis system with nickel oxide catalysts on different supports for toluene removal: Effect of water vapor

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## ABSTRACT

The effect of water vapor on the performance of a combined non-thermal plasma catalysis (CPC) system with nickel oxide catalysts loaded on different supports was investigated. The catalysts NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, NiO/SBA-15 (Santa Barbara Amorphous-15), and NiO/TiO<sub>2</sub> were prepared and their activities were tested in the absence and presence of water vapor. Complete destruction of toluene was achieved in the absence of water vapor at ambient temperature and pressure. The activities of catalysts for the toluene conversion in dry air decreased in the following order: NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> > NiO/SBA-15 > NiO/TiO<sub>2</sub>. The presence of water vapor in the feed stream had a significant negative impact on the performance of the CPC systems. This reduction in performance was primarily due to the quenching by water vapor of active species in the plasma and the competitive adsorption of water vapor on the catalyst surfaces. A novel *in situ* FTIR system was constructed and used to obtain *in situ* FTIR spectra of the reactive surfaces of the catalysts, revealing that the water molecules that adsorbed on the catalyst surfaces came from both water vapor present in the gas stream and from water vapor formed during the oxidation of toluene. H<sub>2</sub>O-TPD results indicated that the activation energies of water desorption from the catalysts decreased in the following order: NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> > NiO/SBA-15 > NiO/TiO<sub>2</sub>. The catalyst with lower water vapor desorption activation energy had higher resistance to water vapor. Therefore, the durability towards water vapor poisoning of these catalysts followed the order of: NiO/TiO<sub>2</sub> > NiO/SBA-15 > NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.

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## 1. Introduction

Volatile organic compounds (VOCs) emitted from various industrial processes is an important sources of air pollution. VOCs are the precursors for the formation of photochemical smog, ground level ozone and fine particles in the air [1]. Some of air-borne VOCs have been associated with adverse health effects. For example, toluene found in gasoline, paints, glues, rubber and varnishes, causes numerous effects, including eyes and respiratory tract irritation, nausea and headaches. Therefore, considerable research attention has been focused on the removal of VOCs from the atmosphere.

Various techniques have been widely used in treating VOCs emissions from industrial plants, including carbon adsorption, catalytic oxidation, thermal decomposition, and condensation.

More recently, a new technique, combined non-thermal plasma catalysis (CPC), is being developed and offers an innovative approach to solving the problem of VOCs pollution, especially when present in low concentrations [2]. A CPC system has the advantage of operating at ambient temperature and pressure. In the CPC system, solid catalysts are often placed inside the non-thermal plasma zone [3], where catalytic reactions can be induced by the plasma without external heating. VOCs are quickly oxidized under ambient conditions through the synergetic effect of the combination of the plasma and catalyst [4]. A catalyst plays an important role in determining the overall efficiency of VOCs removal in the CPC system [5,6]. Khacef et al. [7] tested Ag, Au, Cu, and Co catalysts supported on Al<sub>2</sub>O<sub>3</sub> for toluene decomposition in a catalytic dielectric barrier discharge (DBD) reactor and found that Au/Al<sub>2</sub>O<sub>3</sub> had the highest level of toluene removal. Zhao et al. [8] applied a cycled storage-discharge (CSD) plasma catalytic process using HZSM-5, Ag/HZSM-5, Cu/HZSM-5 and AgCu/HZSM-5 catalysts to remove low concentrations of formaldehyde, and reported that AgCu/HZSM-5

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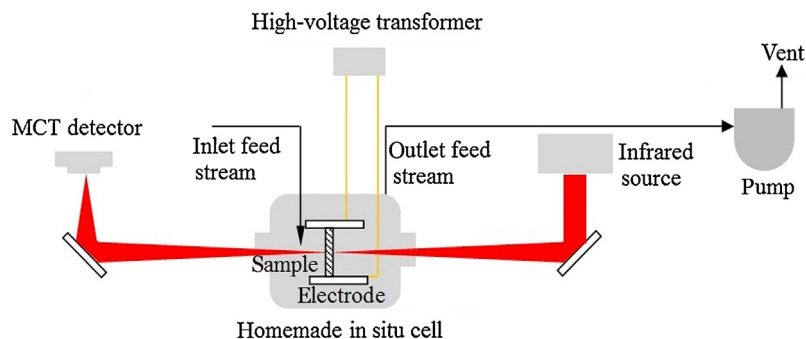


Fig. 1. Scheme of setup for FTIR measurements with a custom-made *in situ* cell.

performed best. Karuppiah et al. [9] used a non-thermal plasma reactor with a catalytic electrode modified with Mn and Co oxides to oxidize isopropanol and found that MnOx performed better than CoOx.

The nickel catalyst is an important catalyst for complete oxidation of VOCs [10]. Kim et al. [11] demonstrated that benzene conversion in a CPC system using Ni/TiO<sub>2</sub> was higher than when using Ag/TiO<sub>2</sub>. Ogata et al. [12] also reported that NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub> showed good performance for VOCs decomposition in plasma.

In recent years, it has been noticed that the presence of water vapor in the gas stream may affect the performance of the CPC systems. Fan et al. [13] investigated the effects of relative humidity (30%, 50% and 80%) on the removal of low concentration of benzene in air by a CPC system fitted with a MnOx/Al<sub>2</sub>O<sub>3</sub> catalyst and found that increasing humidity inhibited the production of O<sub>3</sub> in plasma, thus reducing the benzene conversion. Guo et al. [14] studied the effect of relative humidity on toluene removal using a MnOx/Al<sub>2</sub>O<sub>3</sub>/nickel foam catalyst in the plasma and found that toluene removal decreased with increased humidity. Thevenet et al. [15] investigated the influence of water vapor on acetylene oxidation in a CPC system with a TiO<sub>2</sub> catalyst and reported that the presence of water vapor in the gas stream greatly reduced the removal of acetylene. Van Durme et al. [16] studied the abatement of toluene in indoor air using a CPC system fitted with TiO<sub>2</sub>, and found that for an inlet gas stream with 27% relative humidity, the degradation of toluene was only 40% of the decomposition observed in the absence of water.

In each of these previous studies, only one catalyst was included. From our perspective, different supports for the metal oxide catalysts could have different susceptibilities to water vapor. Therefore, this study was undertaken to examine the effect of water vapor on the performance of a CPC system with nickel oxide catalysts loaded on different supports, with the aim of widening the utility of the CPC systems in practical situations.

In this work, three types of the nickel based catalysts with different supports –  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, SBA-15 and TiO<sub>2</sub> – were prepared using an impregnation method. The catalysts were evaluated and characterized under both dry and moist conditions. The effects of different supports and water vapor on the activities of catalysts for the toluene conversion are discussed and reported below.

## 2. Experimental

### 2.1. Catalyst preparation

Three different materials –  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, SBA-15 and TiO<sub>2</sub> (40–60 mesh) – were used as the supports. The  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> and TiO<sub>2</sub> were supplied by Alfa Aesa; and the SBA-15 was supplied by Shanghai Novel Chemical Technology Company, China. Nickel oxide catalysts were prepared by impregnation using Ni(NO<sub>3</sub>)<sub>2</sub> aqueous solutions,

followed by drying for 24 h at 30 °C. The samples were then heated to 120 °C for 1 h and calcined in air at 400 °C for 3 h. The resulting catalysts contained 5 wt% nickel loadings and were denoted as NiO/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>, NiO/SBA-15, and NiO/TiO<sub>2</sub>.

### 2.2. Catalyst characterization

X-ray diffraction (XRD) patterns were measured with a Bruker D8 Advance diffractometer using Cu K $\alpha$  radiation. The X-ray tube was operated at 40 kV and 40 mA.

Hydrogen temperature programmed reduction (H<sub>2</sub>-TPR) was performed in a Micromeritics Autochem 2920 instrument. The gas mixture (10% H<sub>2</sub> and 90% Ar) was passed through the catalyst sample (0.05 g) at a flow rate of 30 ml/min while the temperature was increased from room temperature to 800 °C at a heating rate of 10 °C/min.

Carbon monoxide temperature programmed desorption (CO-TPD) was employed to estimate the dispersion of the active component of the catalyst [17]. CO-TPD was measured in a Micromeritics AutoChem 2920 instrument. The samples were reduced in a flow of 10% H<sub>2</sub>/Ar (30 ml/min) at 650 °C for 1 h, and then flushed with pure helium flow (30 ml/min) for 0.5 h at the same temperature. After being cooled to room temperature, the samples were exposed for 2 h to a gas flow of 99.99% CO. The samples were then heated to 650 °C in pure helium flow (10 ml/min) at a heating rate of 5 °C/min. The desorbed gas was analyzed with a thermal conductivity detector (TCD).

Water temperature programmed desorption (H<sub>2</sub>O-TPD) experiments were conducted at different heating rates of 2–6 °C/min. In each experiment, the sample that had adsorbed water vapor was packed in a quartz tube and then heated in a flow of high-purity N<sub>2</sub> at a constant rate of 10 ml/min. The desorbed water vapor was measured in a Micromeritics Autochem 2920 instrument equipped with a TCD. The effluent curves (also known as TPD curves) were recorded. The experimental procedures have been reported in detail elsewhere [18–20].

*In situ* Fourier transform infrared (*in situ* FTIR) spectra were recorded using a Nicolet 6700 spectrometer equipped with a mercury-cadmium-telluride (MCT) detector cooled by liquid nitrogen. A custom-made flow cell with a path length of 10 cm and capped at both ends by IR-transparent KBr windows was used for the *in situ* measurements (Fig. 1). Two copper sheets separated by quartz glass plates acted as the electrodes. The discharge gap was 4 mm. An applied voltage of 8000 V was supplied by a high-voltage transformer (50 Hz). The calculated electric field intensity in the discharge gap was 20 kV/cm. A 10 mg sample of catalyst was pressed into a wafer that was approximately 1 cm in diameter. After being placed in a sample holder between the electrodes, the sample was heated to 300 °C for 1 h in pure Ar before measurements. Then, the gas steam (10 ml/min) containing 0.8 vol% water vapor

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