



Catalyst screening for acetone removal in a single-stage plasma-catalysis system



Xinbo Zhu^{a,b}, Xiang Gao^{a,**}, Xinning Yu^a, Chenghang Zheng^a, Xin Tu^{b,*}

^a State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou 310027, Zhejiang Province, China

^b Department of Electric Engineering and Electronics, University of Liverpool, Liverpool, L69 3GJ, UK

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ABSTRACT

Plasma-catalytic removal of acetone over $\text{MO}_x/\gamma\text{-Al}_2\text{O}_3$ ($M = \text{Ce, Co, Cu, Mn}$ and Ni) catalysts has been investigated in a dielectric barrier discharge (DBD) reactor. The influence of different catalysts on the plasma-catalytic process has been examined in terms of acetone removal efficiency, CO_2 selectivity and the formation of by-products. The combination of plasma with the $\text{MO}_x/\gamma\text{-Al}_2\text{O}_3$ catalysts significantly enhances the removal efficiency of acetone and CO_2 selectivity, whilst substantially reducing the formation of by-products (HCHO and HCOOH). The maximum removal efficiency of 94.2% and CO_2 selectivity of 80.1% were achieved when using the $\text{CuO}_x/\gamma\text{-Al}_2\text{O}_3$ catalyst. Introducing the $\text{CuO}_x/\gamma\text{-Al}_2\text{O}_3$ catalyst into the plasma system also shows the lowest formation of by-products. Different catalyst characterization techniques have been used to understand the effect of catalyst properties on the plasma reactions. It has been found that the reducibility of the $\text{MO}_x/\gamma\text{-Al}_2\text{O}_3$ catalysts plays a dominant role in the plasma-catalytic oxidation of acetone, whilst the specific surface area and pore properties of the catalysts have a very weak effect on the performance of the plasma-catalytic reaction. In addition, a simplified kinetic model has been developed, showing the effect of different catalysts on the acceleration of the plasma-catalytic removal of acetone.

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1. Introduction

The emission of volatile organic compounds (VOCs) in the atmosphere is associated with the formation of fine particle matter, such as haze and photochemical smog, and consequently causes air pollution [1,2]. VOCs have also been found to impose significant negative effects on human health. Acetone, as one of the most abundant oxygenated VOCs in the atmosphere, is mainly released from automobile exhaust, tobacco smoke and incineration of waste materials. The increasingly stringent emission regulations and growing public concern for the environment is the main drive to develop new and cost-effective gas cleaning processes for environmental pollution control.

A variety of different pollution control technologies have been developed including adsorption, thermal processes, vapour condensation, and catalytic oxidation and combustion [3]. Conventional methods for the removal of VOCs in high volume waste gas

streams are not cost-effective because of the large flow rates to be treated and high operating costs due to the energy consumption of these methods. For instance, thermal processes require large amounts of energy for heating a high volume of gas flow in which only low concentrations of environmental pollutants need to be removed.

Non-thermal plasma technology offers an attractive and promising alternative as means for the removal of environmental pollutants such as VOCs in high volume waste gas streams. In non-thermal plasma, the overall plasma gas temperature can be as low as room temperature, whilst the electrons are highly energetic with a typical electron temperature of 1–10 eV [4,5]. The initially generated electrons collide with the gas molecules (e.g. carrier gas, pollutant molecules) to produce chemically reactive species including free radicals, excited atoms, ions and molecules for plasma reactions. The use of a plasma discharge has shown that a wide range of VOCs can be effectively decomposed and oxidized at low temperatures [6–8]. However, the main drawback of plasma processes for pollution remediation is the formation of unwanted and hazardous by-products, especially organic by-products. For example, Lyulyukin et al. found acetone (205 ppm) in air can be effectively oxidized in a negative corona discharge at a discharge power of 60 W, with the main products being HCHO, CO

* Corresponding author. Tel.: +44 1517944513.

** Corresponding author. Tel.: +86 571 87952443.

E-mail addresses: xgao1@zju.edu.cn (X. Gao), xin.tu@liv.ac.uk, tuxinjz2000@gmail.com (X. Tu).

and CO₂ with a maximum CO₂ selectivity of 67.6% [9]. Narengerile and Watanabe reported the formation of 0.2–0.9% CH₄, HCOOH and HCHO in the effluent using a direct current (DC) water plasma torch for the decomposition of acetone [10]. The combination of plasma and heterogeneous catalysis has been regarded as a promising and effective solution to overcome the drawback of plasma processes [7,11,12]. Introducing a catalyst into a plasma reactor could change the electron energy distribution function (EEDF) and generate more highly energetic and reactive species, which favour the plasma-induced reactions towards deep oxidation of VOCs [13]. The combination of plasma with catalysis has great potential to lower the activation temperature of the catalysts, enhance the removal of pollutants, and minimize the formation of unwanted by-products: all of which may contribute in different ways to the enhancement of the energy efficiency of the plasma process [7].

Different metal catalysts have been investigated in single-stage plasma-catalytic gas cleaning processes for the oxidation of acetone, including supported precious metal catalysts (Ag, Pd and Pt) due to their excellent activity even with low metal loading [14–16]. However, the high cost of precious metals is a major barrier which limits their use in plasma systems for industrial applications. Transition metal oxide catalysts have gained growing interest in combination with plasma for pollution abatement because of their comparable performance and low cost. Trinh and Mok reported that introducing ceramic supported MnO₂ catalysts into a dielectric barrier discharge (DBD) reactor significantly enhanced the removal efficiency of acetone, whilst the presence of ceramic supported ZnO in the discharge weakly affected acetone removal efficiency [17]. In our previous work, we found that the combination of plasma with γ -Al₂O₃ supported Cu, Co and Mn catalysts improved the removal efficiency of acetone and drove the reaction towards deep oxidation in humid conditions [18]. Similar improvement in acetone removal efficiency was achieved using a LaCoO₃ catalyst in a packed-bed plasma reactor [19]. To the best of our knowledge, far less has been done to investigate the effect of a range of supported metal oxide catalysts on the plasma-catalytic removal of low concentration acetone. The fundamental understanding of catalyst properties in the plasma-catalytic oxidation of acetone is still very limited [17,20]. It is of primary interest to screen a range of supported metal oxide catalysts to get new insights into the roles of different metal oxides and related structures in the plasma-catalytic removal of acetone and to establish a relationship between the process performance and catalyst properties.

In this work, the plasma-catalytic removal of acetone over MO_x/ γ -Al₂O₃ (M = Ce, Co, Cu, Mn and Ni) catalysts has been investigated in a coaxial DBD plasma reactor. The effect of these catalysts on the plasma-catalytic removal of acetone as a function of specific energy density (SED) has been evaluated in terms of acetone removal efficiency, CO₂ selectivity and by-product formation. A wide range of catalyst characterization techniques, including N₂ adsorption–desorption, X-ray diffraction (XRD) and temperature-programmed reduction by H₂ (H₂-TPR), have been used to establish links between the reaction performance and catalyst properties and to get a better understanding of the role of catalyst properties in the plasma removal of acetone. Based on the experimental results, a steady-state approximation method has been employed to represent the reaction kinetics in the plasma-catalytic process for acetone decomposition.

2. Experimental

2.1. Catalyst preparation and characterization

The MO_x/ γ -Al₂O₃ (M = Ce, Co, Cu, Mn and Ni) catalysts with metal loading of 10 wt.% were prepared by incipient wetness

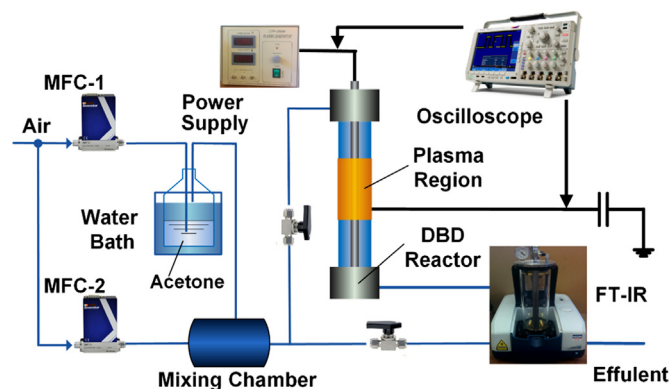


Fig. 1. Schematic diagram of the experimental setup.

impregnation using nitrate salts (Alfa Aesar, 99.5%) as the metal precursor. Initially, the metal nitrates were dissolved in deionized water and stirred for 1 h at room temperature to get 0.1 M solutions. The appropriate weight of support (γ -Al₂O₃) was added to the solution. The slurry was continuously stirred at 80 °C for 4 h and then dried at 110 °C overnight, followed by the calcination at 500 °C for 5 h. All the catalysts were sieved in 35–60 meshes prior to the plasma reaction. Pure γ -Al₂O₃ support was also used for comparison in this study.

The N₂ adsorption–desorption experiment was carried out using a Quantachrome Autosorb-1 instrument at 77 K to get the specific surface area, pore size distribution and average pore diameter of the catalysts. X-ray diffraction (XRD) patterns of the catalyst samples were recorded by an X-ray diffractometer (Rigaku, D/max-2200) using a Cu-K α radiation source in the 2 θ range from 10° to 80°. The reducibility of the catalysts was investigated by temperature-programmed reduction with hydrogen (H₂-TPR) using a GC-1690 chromatograph (Kexiao). Prior to each run, a 50 mg catalyst sample was pre-treated at 200 °C in an N₂ flow for 60 min. After cooling to room temperature, the sample was heated from 20 °C to 900 °C with a heating rate of 10 °C min⁻¹, using 5% H₂/Ar at a flow rate of 40 ml/min. The amount of consumed H₂ was quantitatively measured by the integration of H₂-TPR signals.

2.2. Experimental setup

The experiment was carried out in a coaxial DBD reactor, as shown in Fig. 1. A 60 mm-long aluminium foil (ground electrode) was wrapped around a quartz tube with an outer diameter of 10 mm and wall thickness of 1 mm. A stainless steel rod with an outer diameter of 4 mm was placed inside the quartz tube to act as a high voltage electrode. The reactor was supplied by an AC power supply with a maximum peak voltage of 30 kV and a frequency of 10 kHz. Zero grade air (BOC) was used as carrier gas, whilst acetone (99.5%, Fisher Scientific) was introduced into the DBD reactor by passing a dry air flow through a water bubbler kept in a water bath (20 °C). All gas streams were premixed prior to the DBD reactor. Catalyst samples (100 mg) were placed at the end of the discharge region and were held there using quartz wool. The total flow rate was kept at 1 l min⁻¹, resulting in a gas hourly space velocity of 600,000 ml g⁻¹ h⁻¹, with the initial concentration of acetone fixed at 1184 ppm.

The applied voltage was measured by a high voltage probe (Testec, HVP-15F, 1000:1), whilst the voltage on the external capacitor (0.47 μ F) was measured by a Tektronix P5100 probe. All the electrical signals were sampled by a four-channel digital oscilloscope (Tektronix 3034B). The discharge power (P) of the DBD was calculated using the Q–U Lissajous method. An online power measurement system was used to monitor and control the discharge

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