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## 1 Removal of phenol from gas streams via combined plasma catalysis

2 Q1 Kuan Lun Pan<sup>a</sup>, Dai Ling Chen<sup>a</sup>, Guan Ting Pan<sup>b</sup>, Siewhui Chong<sup>c</sup>, Moo Been Chang<sup>a,\*</sup>3 <sup>a</sup> Graduate Institute of Environmental Engineering, National Central University, No. 300, Zhongda Road, Zhongli District, Taoyuan City 32001, Taiwan4 <sup>b</sup> Department of Chemical Engineering, National Taipei University of Technology, No. 1, Sec. 3, Zhongxiao E. Rd., Da'an District, Taipei City 10608, Taiwan5 <sup>c</sup> Department of Chemical and Environmental Engineering, University of Nottingham Malaysia Campus, Jalan Broga, 43500 Selangor, Malaysia

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## ABSTRACT

A hybrid system consisting of non-thermal plasma and perovskite-like catalyst is developed and evaluated for the effectiveness in removing phenol from gas streams. For thermal catalysis,  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Mn}_{0.8}\text{Cu}_{0.2}\text{O}_3$  shows high activity for phenol removal. Further,  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Mn}_{0.8}\text{Cu}_{0.2}\text{O}_3$  is applied for combined plasma catalysis (CPC). The results indicate that phenol removal efficiency with CPC remains 100% at applied voltage range of 13–16 kV. Importantly, secondary pollutants ( $\text{O}_3$  and  $\text{NO}_x$ ) and energy efficiency can be inhibited and increased, respectively, as CPC is applied. Overall, this study demonstrates that combining non-thermal plasma with perovskite-like catalyst is effective in removing phenol from gas streams.

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## 6 Introduction

7 Volatile organic compounds (VOCs) are commonly utilized for  
8 commercial and industrial applications such as paints, chemical  
9 processing, and printing industries [1,2]. Among them, phenol has  
10 a high toxicity to human even with a low concentration.  
11 Additionally, phenol has a low odor threshold (0.04 ppm), causing  
12 severe odor problem [3,4]. Although many studies have been  
13 conducted for effective removal of phenol from wastewater,  
14 study on effective removal of phenol from gas streams is still  
15 limited.

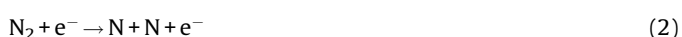
16 Conventional methods including adsorption, combustion,  
17 absorption, catalysis are considered as effective ways for VOCs  
18 removal [5]. However, they are still of some disadvantages for  
19 practical application. For example, adsorbent needs to be replaced  
20 or regenerated regularly, while absorption may cause secondary  
21 water pollution. Catalysis is considered as one of promising  
22 technologies for removing VOCs, and it can effectively oxidize  
23 VOCs to carbon dioxide ( $\text{CO}_2$ ) and water ( $\text{H}_2\text{O}_{(g)}$ ) with appropriate  
24 catalyst. Among them, noble metal catalysts (including Au, Pd,  
25 and Pt) [6] generally possess high activity in removing VOCs at  
26 moderate temperatures (even room temperature), but high cost  
27 has limited their applications. Metal oxides have also been

investigated for VOCs removal, and CuO,  $\text{Co}_3\text{O}_4$ ,  $\text{Fe}_2\text{O}_3$ , and  $\text{MnO}_x$   
are commonly applied [7–10]. Recently, perovskite-like catalysts  
have been demonstrated with good activity to potentially replace  
noble metal catalysts for VOCs removal [11–13]. However,  
perovskite-like catalysts need to be operated at a high temperature  
for good performance [14]. Another technique which has been  
investigated is non-thermal plasma (NTP), and it has been proved  
that NTP can effectively remove VOCs [15]. NTP provides a rapid  
reaction route for converting VOCs to  $\text{CO}_2$  and  $\text{H}_2\text{O}_{(g)}$  even at room  
temperature, and this mechanism is significantly different from  
conventional catalysis. Park et al. reported that conversion  
efficiency of benzene ( $\text{C}_6\text{H}_6$ ) achieved with DBD (dielectric barrier  
discharge) reaches 70% with the applied voltage of 8.5 kV, and  
frequency of 60 Hz [16]. Lee et al. also applied DBD to remove  $\text{C}_6\text{H}_6$   
and indicate that  $\text{C}_6\text{H}_6$  conversion efficiency reaches 90% with the  
applied voltage of 12 kV, and frequency of 900 Hz [17]. Recently,  
Liang et al. applied DBD for the removal of  $\text{C}_6\text{H}_6$ , and concluded  
that  $\text{C}_6\text{H}_6$  conversion efficiency can reach 90% with the applied  
voltage of 100 kV and frequency of 1000 Hz [18]. However, the  
performance of NTP still needs to be improved for better energy  
efficiency and selectivity [19]. Also, it is demonstrated that NTP  
potentially leads to generation of harmful byproducts such as  $\text{O}_3$   
and  $\text{NO}_x$  [20,21]. Formation of  $\text{O}_3$  and  $\text{NO}_x$  in a plasma system can  
be described as Reactions (1)–(10). However, ozone generated may  
be consumed via Reactions (5)–(7) during discharge.



\* Corresponding author. Fax: +886 3 4226774.

E-mail address: [mbchang@ncuen.ncu.edu.tw](mailto:mbchang@ncuen.ncu.edu.tw) (M.B. Chang).



Recently, plasma catalysis has been proposed to potentially improve the defects of catalysis system and NTP [22], which could also be called combined plasma catalysis (CPC). NTP and heterogeneous catalyst are combined to form a hybrid system. Generally, synergistic effects which could bring several advantages on performance such as increasing energy efficiency, improving  $\text{CO}_x$  selectivity, and reducing secondary pollution can be observed within CPC. Based on the location of catalyst, CPC can be distinguished into two configurations, i.e., in-plasma catalysis (IPC) [23,24] and post-plasma catalysis (PPC) [25]. The former is similar to packed-bed reactor, namely, catalyst is directly packed into the discharge zone. The latter implies that catalyst is located downstream NTP reactor. For PPC system, catalysis system and plasma reactor are separated. Hence, active species generated from NPT may not interact well with catalyst surface, because they would disappear before reaching the catalysis system due to their high reactivities and short lifetimes. Therefore, the change of feeding gas composition by NTP is regarded as the major mechanism in PPC, namely, it could pre-convert the pollutants into easier treatable substances for catalysis. Compared with IPC, PPC is relatively simple and its benefits sometimes are limited [26,27]. IPC configuration is more complicated because plasma and catalysis may interact each other. Possible mechanisms can be described as: (1) the influence of catalyst on the plasma, and (2) the influence of plasma discharge on catalyst surface. Therefore, IPC is speculated to bring more positive effects on performance. Various catalysts including Ni/ $\gamma$ - $\text{Al}_2\text{O}_3$  [28],  $\text{TiO}_2/\gamma$ - $\text{Al}_2\text{O}_3/\text{Ni}$  [29], Pd/ $\text{Al}_2\text{O}_3$  [30], Ag-Mn/SBA-15 [31], and Ag-Ce [32] have been investigated with IPC configuration for removing VOCs.

In this study, a hybrid system consisting of NPT and perovskite-like catalyst is developed to remove phenol from gas streams. Previous study indicates that perovskite-like catalysts have a unique physicochemical property of high dielectric constant [33]. Potentially, it would enhance the performance for phenol removal. However, studies concerning the application of perovskite-like catalysts with plasma are limited. It is expected that CPC can significantly increase phenol removal efficiency and improve mineralization efficiency. In this study, Mn-based and Mn-Cu-based ( $\text{LaMnO}_3$ ,  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Mn}_{0.8}\text{Cu}_{0.2}\text{O}_3$ ) perovskite-like catalysts are prepared and evaluated because Mn and Cu

are commonly considered due to their good activities towards VOCs removal. For activity test, various operating parameters are extensively evaluated via a lab-scale experimental setup, and the mechanisms responsible for phenol removal will be elucidated.

## Experimental

### Catalyst preparation

The catalysts ( $\text{LaMnO}_3$ ,  $\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_3$  and  $\text{La}_{0.8}\text{Sr}_{0.2}\text{Mn}_{0.8}\text{Cu}_{0.2}\text{O}_3$ ) were prepared with the Pechini method [34]. First, the corresponding metal nitrates and citric acid were dissolved in deionized water to form 1 M solution as precursor. Then solutions of metal nitrates and citric acid prepared were mixed completely and heated to 80 °C for 1 h with stirring. Afterwards, ethylene glycol was added to the solution, meanwhile temperature was adjusted to 90 °C for 4 h with stirring. The molar ratio of citric acid to the metal nitrates was fixed at 3 to 1, while the molar ratio of the ethylene glycol to citric acid was 1:1. The mixed solution was heated and stirred continuously until water was evaporated. Finally, the residual solid precursor was placed into an oven to dry overnight, and then calcined in air at 700 °C for 5 h. Subsequently, the synthesized particles were milled and sieved to a 30–70 mesh size for use.

### Catalyst characterization

Surface properties of catalysts prepared were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM) with energy-dispersive X-ray spectroscopy (EDS). Brunauer Emmett Teller (BET) surface areas of catalysts prepared were measured using ASAP2010 (ASAP2010 Micromeritics, USA). Morphologies and compositions of catalysts were observed by S80 JEOL (SEM, S80 JEOL, Japan). XRD patterns of catalysts were identified by X-ray diffractometer (D8AXRD BRUKER, Germany), with the operating parameters of at 40 kV and 10 mA, while degree of  $2\theta$  ranged from 10° to 80° with a scanning rate of 6°/min. X-ray photoelectron spectroscopy (XPS) spectra were recorded with monochromatic Al anode X-ray which was equipped with a concentric hemispherical analyzer. Al K $\alpha$  (1486.6 eV) X-ray source was used for excitation. The binding energies were referenced to the C1s line at 284.5 eV.

### Plasma characterization

Dielectric-barrier discharge (DBD) type reactor is applied to generate the nonthermal plasma. The DBD reactor is made of a quartz tube (OD = 20 mm, ID = 18 mm, and L = 300 mm), while stainless steel rod (OD = 2 mm) and stainless steel wire mesh (L = 50 mm) are used as the inner and outer electrodes, respectively. The total discharge volume is 12.56 cm<sup>3</sup>, and a DC pulse power (You-shang, Taiwan) is applied as plasma power source, with the applied voltage and frequency up to 23 kV and 20 kHz, respectively. The discharge power was measured by a digital oscilloscope (Tektronix DPO3014, USA) with a current probe (Tektronix TCPA300, USA) and a high-voltage probe (Tektronix P6015A, USA). Fig. 1(a) shows the appearance of discharge at 15 kV and 10 kHz, indicating that plasma generated is uniform, accompanying with many streamers. In addition, Fig. 1(b) shows the voltage and current waveforms across the discharge, and quasi-pulse waves appear.

### Activity measurement

Phenol removal tests are divided into 3 parts including catalysis, plasma-alone, and combined plasma catalysis (CPC). First, catalytic removal of phenol was performed in a fixed-bed

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