



Sustainable removal of particulate matter from diesel engine exhaust at low temperature using a plasma-catalytic method



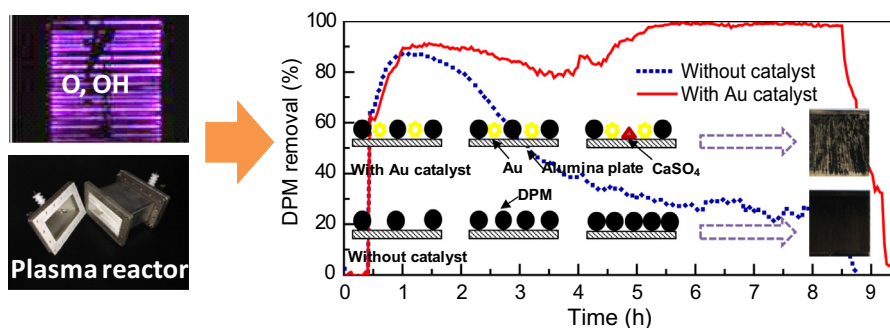
Shuiliang Yao, Xing Shen, Xuming Zhang*, Jingyi Han, Zuliang Wu, Xiujuan Tang, Hao Lu, Boqiong Jiang

School of Environmental Science and Engineering, Zhejiang Gongshang University, Hangzhou, Zhejiang 310018, China

HIGHLIGHTS

- We can sustainably remove DPM using plasma without external heating.
- Au is the suitable noble-metal catalyst for the plasma-catalytic oxidation of DPM.
- The cost of Au catalyst only occupies ~1% of the total cost of plasma reactor.
- Maintained DPM precipitation results in an enriched CaSO_4 deposition.
- CaSO_4 has positive effect on the plasma-Au catalytic oxidation of DPM.

GRAPHICAL ABSTRACT



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ABSTRACT

There is a great need for a sustainable diesel particulate matter (DPM) removal approach to effectively control the emission of DPM over long term and to improve the efficiency of diesel engine. However, methods that are currently available either require a high operation temperature or an external device. Here, we report an optimized plasma-catalytic method for the sustainable removal of DPM at low temperature. Au catalyst, which was the most effective in the enhancement of DPM oxidation among the tested noble-metal catalysts, was introduced into the plasma reactor for the diesel engine test. We found that after 1.1 h plasma processing, the performance of DPM removal in the condition of sole plasma (without a catalyst) was reduced due to the accumulation of precipitated DPM within the plasma reactor. As a result of an enhanced DPM oxidation efficiency due to the use of Au catalyst, the accumulation of precipitated DPM was inhibited and the performance of DPM removal was maintained until plasma was switched off (8 h processing). Importantly, the maintained DPM removal caused an enriched CaSO_4 deposition on the plasma-catalytic reactor, which further enhanced DPM oxidation efficiency. Thus, a sustained DPM removal performance over long term could be ensured.

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1. Introduction

Diesel particulate matter (DPM) is mainly constituted by carbonaceous particles and metal sulfates [1]. It can lead to various

detrimental effects on human health [2,3], and it has been classified by International Agency for Research on Cancer (IARC) as group I carcinogenic to humans [4]. For this reason, the emission standard of DPM has become stricter globally. Diesel particulate filters (DPFs) are conventionally employed for the DPM emission control. To enhance the sustainability of DPM removal process, one of the most important factors for practical application,

* Corresponding author.

E-mail address: Miraclezhang918@163.com (X. Zhang).

periodic regeneration of DPFs is required as it can avoid back-pressure increase that causes reduction in engine efficiency [5]. The regeneration process, however, results in additional heating requirement because the thermal DPM combustion requires a temperature at least 560 °C [6], while the temperature of light-duty diesel exhaust gas is typically in the range of 150–360 °C. The use of catalysts, such as noble-metals [7,8], perovskite-type oxides [9] and transition metal oxides [10], can facilitate the DPM oxidation. Nevertheless, the required combustion/oxidation temperature is still higher than 400 °C, which may cause a fuel penalty of typically 6–11% [11].

Non-thermal plasma (NTP), which can be conveniently produced by electrical discharge [12–16], has been considered as an alternative technology to control the emission of DPM at low-temperature conditions [17–20]. It can be used as an oxidative radical (e.g. O and OH radicals) source for the low-temperature combustion of DPM [21,22], or used as an ion source for the electrostatic precipitation of DPM [23]. It is also possible to take full advantage of NTP by utilizing plasma produced ions and oxidative radicals in a single treatment process. In previous works, we have developed a pulsed dielectric barrier discharge (DBD) reactor [24], in which the DPM removal was proposed due to two steps [25]: (1) the electrostatic precipitation of charged DPM (through ion attachment), and (2) the oxidation of the precipitated DPM into CO and CO₂. By optimizing the reactor configuration and plasma specification, low DPM emission (5 mg/km) with a very competitive fuel penalty (3%) have been achieved for the light-duty diesel vehicles [11]. The remaining major challenging task is to enhance the sustainability of removal process for maintaining the DPM removal in high performance over long term.

The performance of DPM removal could be kept by preventing the accumulation of DPM on the surface of electrodes. Water washing is a useful way to continuously clean the precipitated DPM from electrodes (e.g. wet electrostatic precipitator) [23], however, it needs water supply system which may prevent the development of compact system for vehicle applications. Another effective way is to enhance the oxidation of precipitated DPM. This could be achieved by increasing the plasma energy or the temperature of exhaust gas, but may consume a large amount of electrical energy [26]. Previous works have reported that the introduction of catalysts into the plasma can cost-effectively improve the plasma DPM oxidation. Liu et al. found that the oxidation efficiency of simulated DPM on MnO_x/CeCO₂ catalysts could be as high as 85.2% at temperature of 20 °C and discharge power of 18.0 W [21]. Burachaloo et al. reported that, as compared to the plasma alone process, the oxidation efficiency of simulated DPM can be enhanced up to 76% (increased from 3.4 g/kWh to 6.0 g/kWh) when the Co₃O₄ was added into the plasma reactor at temperature of 300 °C and discharge power of 3.9 W [22]. Taking these observations into account, it is possible to continuously remove DPM over long term using a proper plasma-catalytic reactor.

Herein, we report the sustainable removal of DPM at low temperature using plasma-catalytic method for the first time. Noble-metal catalysts, which are commonly used for promoting the low-temperature catalytic combustion of DPM [7,8,27,28], were used to facilitate the plasma DPM oxidation. The activity of plasma-catalytic oxidation was evaluated by DPM oxidation efficiency. A screened noble-metal catalyst was then used in a diesel engine test, in which the temporal variation of DPM removal was investigated. Results suggested that in the plasma-catalytic DPM removal process, a different catalyst from noble-metal could be activated after several hours of plasma processing. In addition, the cost increase due to the use of noble-metal catalyst was estimated.

2. Experimental section

The activity of plasma-catalytic oxidation was evaluated in a single-cell DBD (SCDBD) reactor as shown in Fig. 1(a). The SCDBD reactor was composed of two alumina plates (115 × 115 × 1 mm³, 96% α-Al₂O₃ with 0.6 m²/g specific surface area, 12.6 nm pore size and 0.0019 cm³/g pore volume), two stainless steel plates (95 × 95 × 0.1 mm³) and two alumina spacers (10 × 115 × 0.80 mm³, 96% α-Al₂O₃). Its discharge volume was 95 × 95 × 0.80 mm³. The temperature of the SCDBD reactor was controlled by a feedback-controlled electrical heating furnace (CX-GW-1, Oubo). Mass flow controllers (D07 series, Sevenstar) controlled each flow rate of the supplied gases, nitrogen and oxygen. The total flow rate was fixed at 1 L/min. The activity of plasma-catalytic oxidation was evaluated by DPM oxidation efficiency. For a fairly evaluation, the tested DPM was collected from a diesel engine that was used for the diesel engine test as subsequently mentioned. The DPM (20 mg) was dispersed in 2 ml liquid ethanol (99.7%, Kelong) using an ultrasonicator for 2 min. The DPM-containing liquid ethanol was uniformly loaded to the surface of alumina plate within a designated area (95 × 95 mm²) where a catalyst was pre-deposited (also an area where DBD took place), and was dried at 80 °C for 1 h.

For the diesel engine test [Fig. 1(b)], we employed a multi-cell DBD (MCDBD) reactor which was designed for practical application. The MCDBD reactor had 20 discharge cells connected in parallel. The number of discharge cells was optimized based on the trade-off between energy efficiency and reactor cost [29]. The configuration of each discharge cell was the same with that of the aforementioned SCDBD reactor. It resulted in total discharge volume of 20 × 95 × 95 × 0.80 mm³. All discharge cells were carefully assembled to maximize the discharge uniformity. It is worth mentioning that the MCDBD reactor is as compact as the conventional DPF, and thus could be installed in the exhaust pipe line for the practical application. The MCDBD reactor was installed at the downstream (1.2 m) of a diesel engine (YT6800T-ATS, Yiten) that was fueled with diesel oil (China III standard, Sinopec) and used two electrical heaters (HC2038S, Aimeite) as a load. The temperature at the inlet and outlet of the MCDBD reactor was monitored using thermocouples (K-type, Hengfeng).

Noble-metal catalysts, Au, Pt, Pd and Ag, were deposited onto the pre-processed (cleaned and dried) alumina plates by an impregnation method using AuCl₃·HCl·4H₂O (Au ≥ 47.8%, Zhanyun), H₂PtCl₆·6H₂O (Pt ≥ 37.6%, Zhanyun), PdCl₂ (Pd ≥ 59%, Zhanyun) and AgNO₃ (AR, ≥99.8%, Zhanyun) as precursors, respectively. Each precursor was dissolved into deionized water with vigorous stirring. 2 ml noble metal-containing solution was uniformly deposited (by writing brush) onto one surface of each alumina plate within a designated area (95 × 95 mm²), where DBD took place. The alumina plates were then submitted for drying at 110 °C for 5 h and calcinations at 500 °C for 3 h.

The DBD reactors were energized by a pulsed power source (HV-10-08, Ketai) with 70 μs in full width at half maximum (FWHM), 30 μs in rise time, and a typical range of applied peak voltages of 5–6 kV (typical V-I waveform is shown in Fig. S1 of Supplementary Material). A digital oscilloscope (DPO 3034, Tektronix) was used for voltage-current characterization. The applied voltage was measured with a 1000:1 high voltage probe (P6015A, Tektronix), and the discharge current was monitored with a current sensor (TCP 0030, Tektronix). Discharge power (P_{dis}) was determined from the product of the discharge energy and the applied frequency, where the discharge energy was obtained from numerical integration of the product of voltage and current.

For experiments on the activity of plasma-catalytic oxidation, the DPM deposited alumina plates were weighted using an

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