



## Improving the Removal Efficiency of Elemental Mercury by Pre-Existing Aerosol Particles in Double Dielectric Barrier Discharge Treatments

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### ABSTRACT

Plasma technology has been employed for the removal of gaseous elemental mercury ( $\text{Hg}^0$ ) from simulated flue gases without pre-existing airborne particles. This study developed a double dielectric barrier discharge (DDBD) treatment system, in which two coaxial electrodes were covered by quartz dielectrics, for removing mercury with the presence of aerosol particles. The increase in pre-existing aerosol surface concentration can improve  $\text{Hg}^0$  removal efficiency up to 160% in the DDBD device. Inorganic aerosol particles (sodium chloride) perform better than organic ones (sucrose) in improving  $\text{Hg}^0$  removal efficiency. These aerosol particles can be collected in the DDBD system. For sodium chloride particles, a collection efficiency of more than 90% was observed in the tested diameter range of 10–100 nm. The improvement in  $\text{Hg}^0$  removal with the presence of particles is possibly due to that (i) aerosol particles provide additional surface for surface-induced  $\text{Hg}^0$  oxidations, (ii) reactive species (such as Cl) generated by plasma etching particle surface rapidly react with  $\text{Hg}^0$ , and (iii) charged particles can in-flight adsorb mercury species.

**Keywords:** Mercury removal; Aerosol particles; Plasma; DDBD; Pollution control.

### INTRODUCTION

Mercury pollution has attracted growing concern owing to its negative effects on the environment and human health (Parks *et al.*, 2013). Many technologies have been developed for mercury control (Biswas *et al.*, 1999), such as injecting powdered activated carbon to adsorb mercury (Sjostrom *et al.*, 2010; Clack, 2012), injecting halogen reagents (Zhao *et al.*, 2006) and using UV light and ozone to realize mercury oxidation (Wu *et al.*, 1998; Granite and Pennline 2002; Lee *et al.*, 2004; McLarnon *et al.*, 2005; Wang *et al.*, 2007; Yang *et al.*, 2012). Non-thermal plasma operated at atmospheric pressure is one of the promising technologies for converting gaseous elemental mercury ( $\text{Hg}^0$ ) to oxidized mercury ( $\text{Hg}^{2+}$ ) and particulate bound mercury ( $\text{Hg}_p$ ) (Chen *et al.*, 2006; Byun *et al.*, 2011a), which can then be effectively removed by conventional air-pollution control devices (Clack, 2006). Most of the electrical energy of non-thermal plasmas is consumed to produce UV and reactive species such as  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ , radicals of OH,  $\text{HO}_2$ , and O (Jia *et al.*, 2013; Li *et al.*, 2012). These reactive species and UV

can oxidize  $\text{Hg}^0$  at a high reaction rate. These species have also been recognized for potentially oxidizing NO and  $\text{SO}_2$  from flue gas (Liang *et al.*, 2002; Jeong and Jung 2007; Xu *et al.*, 2009), destructing carbonyl sulfide (Tsai *et al.*, 2007), and removing organic compounds (Chang and Hsieh 2013; Lin *et al.*, 2013).

The application of non-thermal plasma technology for mercury removal has been mainly studied with corona discharge (Ko *et al.*, 2008; Byun *et al.*, 2011b) and dielectric barrier discharge system (Chen *et al.*, 2006; Jeong and Jung, 2007; Byun *et al.*, 2011a). Both kinds of plasma treatment systems employed one naked metal electrode (commonly as the high voltage electrode), which can partially heat the flue gas, resulting in the unnecessary consumption of electrical energy. In a recent study, surface discharge plasma injection approach (naked electrode for discharge in the reactor) was proposed for oxidizing  $\text{Hg}^0$  (An *et al.*, 2014). Its challenge is that the naked metal electrode will be etched over a long-term continuous operation. In addition, to the best of our knowledge, previous investigations on non-thermal plasma treatment of simulated flue gases did not include aerosol particles, which always co-exist with mercury in real flue gases.

The objective of this study was to develop a non-thermal plasma device with double dielectric barrier discharge (DDBD) configuration, in which two coaxial electrodes

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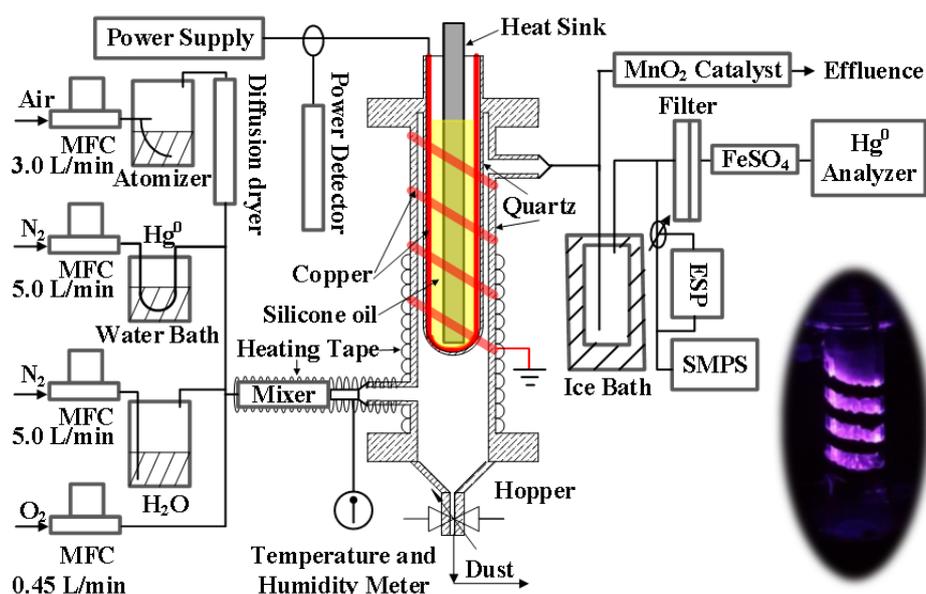
were covered by quartz dielectrics, for a low-consumption of electrical energy and no-consumption of metal electrodes. Aerosol particles were included in the simulated flue gases to examine their role in mercury removal. Both inorganic (sodium chloride) and organic (sucrose) particles were tested. Possible mechanisms for the role of aerosol particles in mercury removal were discussed. Furthermore, the collection efficiency of these aerosol particles by the plasma device was investigated.

## EXPERIMENTAL DESCRIPTION

Fig. 1 shows a schematic diagram of the experimental setup and a typical discharge photo. The DDBD device is made by two quartz cylinders with the height between the inlet and the upper lid of  $\sim 18$  cm, while the gap (discharge region) between the two cylinders is 2.0 mm. A copper band with the width 2.0 cm surrounds the outside surface of the outer cylinder in four loops as the ground electrode, which is covered with silicon vacuum grease to avoid discharge in the outside surface. Another copper band fully covers the inside surface of the inner cylinder as the power electrode, which is connected to a high voltage power supply (CW1003, Nantong Sanxin, China) with a fixed voltage frequency of  $10.0 \pm 0.6$  kHz. Its output power, monitored by a power meter, can be controlled by adjusting the output voltage amplitude in the range of 0.5–20 kV. The input energy density was obtained by dividing the output power with the volume of the discharge region. A stainless steel heat sink is placed in the middle of the inner cylinder for cooling the power electrode. The gap between the heat sink and the power electrode is filled with silicone oil to avoid discharge inside the inner cylinder. The inlet for incoming flue gas is at the outside bottom while the outlet for outgoing flue gas is at the outside top. A hopper is

joined in the bottom to collect dust and cooling water from the flue gas. This design of DDBD configuration, only discharging between two quartz dielectric surfaces, ensures a long-term continuous operation with no-consumption of metal electrodes.

The flow rates of incoming simulated flue gases were controlled by mass flow meters. Aerosol particles were generated by an atomizer. Both inorganic (sodium chloride) and organic (sucrose) particles were produced. Aerosol surface area concentration was adjusted by using solutions with different concentrations.  $\text{Hg}^0$  was introduced by passing nitrogen through a mercury permeation tube. Its concentration was controlled by the water bath temperature ( $15$ – $38^\circ\text{C}$ ). Humidity and temperature of the incoming gases were adjusted by a moistener and a heating tape, respectively. The tubing temperature was fixed at  $65^\circ\text{C}$  by controlling the power supplied to the heating tape surrounding the tubing. All the incoming gases were mixed before entering the DDBD device. The residence time of the simulated flue gases in the DDBD chamber was  $\sim 0.25$  s. After the DDBD treatment,  $\text{Hg}^0$  concentration ( $C_{\text{Hg}^0}$ ) was monitored by an  $\text{Hg}^0$  analyzer (Lumex RA-915+, Ohio Lumex Company, Inc., St. Petersburg, Russia). Its lower detection limit is  $\sim 0.1$   $\mu\text{g}/\text{m}^3$ . An ice bath was used to cool down the flue gas after the plasma device. A membrane filter was used to remove particles prior to  $\text{Hg}^0$  measurement. Since residue ozone generated by plasma can absorb ultraviolet with the wavelength of 254 nm and affect  $\text{Hg}^0$  measurement. Melanterite ( $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ) powders, converting ozone to oxygen (Logager *et al.*, 1992), were used for adsorbing residual ozone ( $\text{O}_3$ ) prior to  $\text{Hg}^0$  measurement. Elementary mercury concentration was monitored at the outlet of the DDBD device with it being on and off.  $\text{Hg}^0$  removal efficiency was then defined as  $1 - (C_{\text{Hg}^0} \text{ when DDBD turns on}) / (C_{\text{Hg}^0} \text{ when DDBD turns off})$ .



**Fig. 1.** Schematic of experimental setup and a typical plasma discharge photo (in the lower right corner). The red color lines denote electrodes made of copper, while the yellow color band denotes silicon oil in the inner quartz tube. MFC: mass flow controller; ESP: electrostatic precipitator; SMPS: scanning mobility particle sizer.

A scanning mobility particle sizer (SMPS 3936, TSI Inc., USA) was used to measure aerosol size distributions at the outlet of the DDBD device. Tygon tubing was used between the reactor outlet and the SMPS inlet to minimize aerosol losses (Liu *et al.*, 1985). Fig. 2 shows typical size distributions of sodium chloride aerosol generated in this study. Sucrose aerosol had similar size distributions. Total aerosol surface concentrations were estimated by assuming that particles are spherical. Total surface concentrations of  $0.1 \times 10^{11}$ ,  $0.4 \times 10^{11}$ , and  $1.2 \times 10^{11} \text{ nm}^2/\text{cm}^3$  were tested in this study, while their corresponding peak diameters were about 40, 49, and 67 nm, respectively. A homemade electrostatic precipitator (ESP; a coaxial cylinder with the inner electrode diameter of 0.45 cm, the gap of 0.1 cm, and the length of 40 cm; its collection efficiency for sub-200 nm charged particles was higher than 95% when a high voltage of 1 kV was supplied) was set as a bypass before SMPS. The collection efficiency at a given particle diameter is defined as  $1 - (C_{\text{particle}} \text{ when DDBD turns on}) / (C_{\text{particle}} \text{ when DDBD turns off})$  with ESP turns off, while the enhanced collection efficiency by the ESP at a given particle diameter is defined as  $1 - (C_{\text{particle}} \text{ when DDBD turns on}) / (C_{\text{particle}} \text{ when DDBD turns off})$  with ESP turns on. Particle concentration,  $C_{\text{particle}}$ , was derived from aerosol size distribution measured by the SMPS.

## RESULTS AND DISCUSSIONS

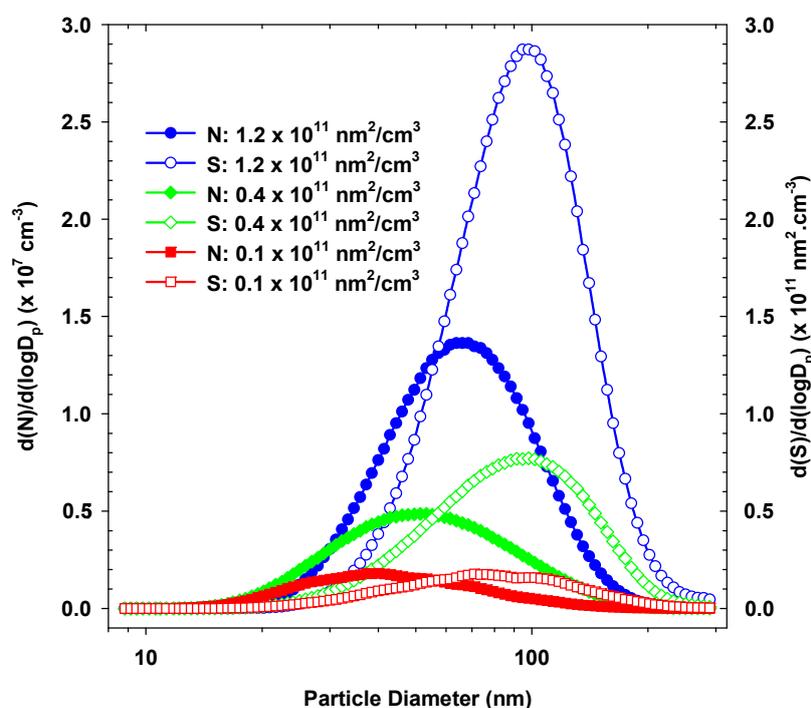
### Improving Mercury Removal Efficiency by Adding Aerosol Particles

Fig. 3(a) shows  $\text{Hg}^0$  removal efficiency as a function of

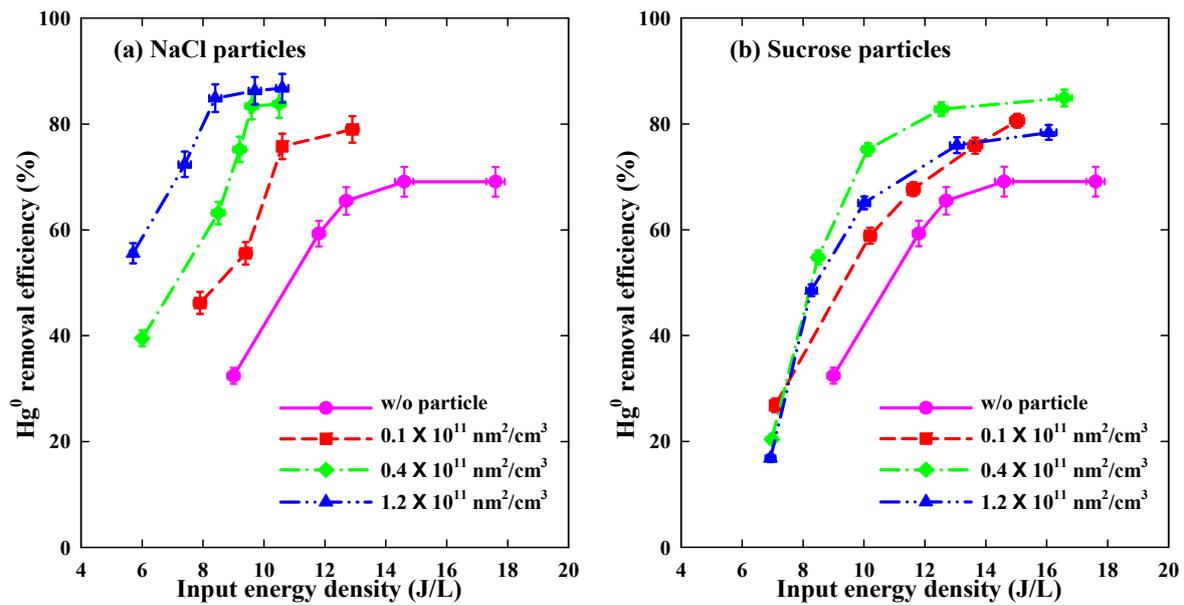
input energy density in the DDBD device with different NaCl aerosol surface concentrations. An increase in input energy density results in a higher  $\text{Hg}^0$  removal efficiency. However, the removal efficiency appears to reach a plateau at a certain input energy density, e.g.,  $\sim 15, 11, 9.5, 8.5 \text{ J/L}$  for NaCl aerosol surface concentration of  $0, 0.1 \times 10^{11}, 0.4 \times 10^{11}$ , and  $1.2 \times 10^{11} \text{ nm}^2/\text{cm}^3$ , respectively. A continuous increase in input energy increases electrical energy cost. These findings are consistent with results reported in plasma systems using corona discharge and single dielectric barrier discharge (Chen *et al.*, 2006; Jeong and Jurng, 2007; Ko *et al.*, 2008; Wang *et al.*, 2010). Furthermore, these findings show that pre-existing NaCl particles improve  $\text{Hg}^0$  removal efficiency. The efficiency enhancement is  $\sim 160\%$  at the input energy density of  $\sim 9 \text{ J/L}$ . An increase in aerosol surface concentration leads to a higher  $\text{Hg}^0$  removal efficiency.

Fig. 3(b) shows  $\text{Hg}^0$  removal efficiency as a function of input energy density in the DDBD device with the existence of sucrose particles. The tendency with increasing input energy density is similar. However, the enhancement in  $\text{Hg}^0$  removal by sucrose particles doesn't increase monotonically with increasing aerosol surface concentration.  $\text{Hg}^0$  removal efficiencies at sucrose aerosol surface concentration of  $1.2 \times 10^{11} \text{ nm}^2/\text{cm}^3$  are lower than those at a lower aerosol surface concentration of  $0.4 \times 10^{11} \text{ nm}^2/\text{cm}^3$ .

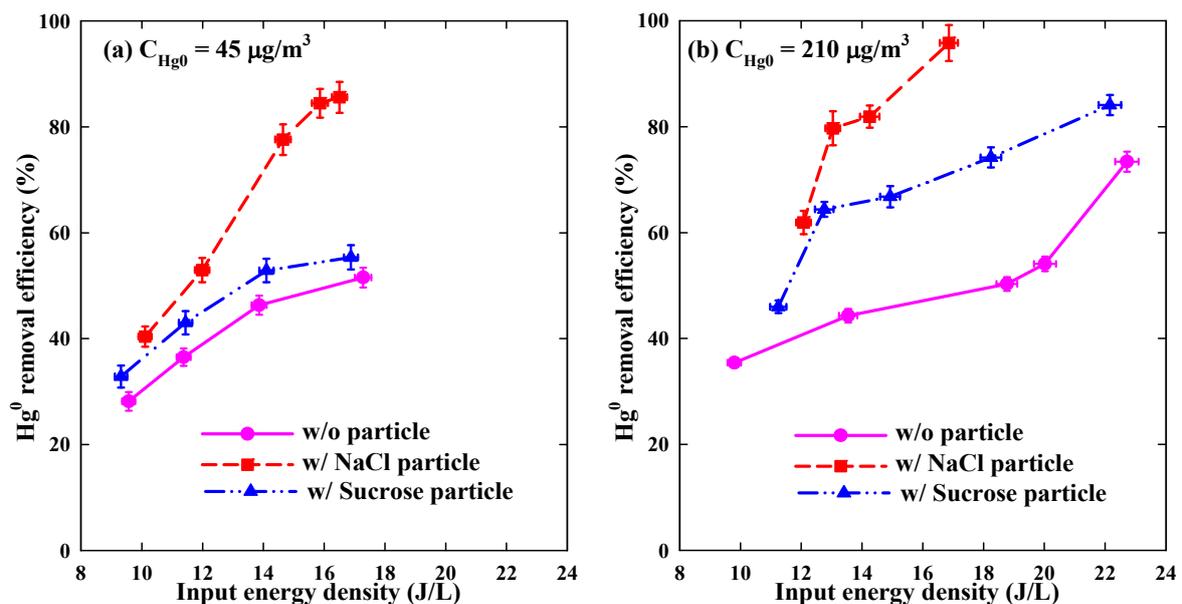
Results shown in Fig. 3 were obtained at the incoming  $\text{Hg}^0$  concentration of  $110 \mu\text{g}/\text{m}^3$ . Fig. 4 compares the effect of sodium chloride and sucrose particles at other  $\text{Hg}^0$  concentrations, i.e., 45 and  $210 \mu\text{g}/\text{m}^3$ . Consistent results were observed. Pre-existing NaCl particles perform better in improving  $\text{Hg}^0$  removal efficiency than sucrose particles,



**Fig. 2.** Typical distributions of sodium chloride particle number (N) and surface (S) concentrations with the function of particle diameter for three aerosol surface concentrations:  $0.1 \times 10^{11}$ ,  $0.4 \times 10^{11}$ , and  $1.2 \times 10^{11} \text{ nm}^2/\text{cm}^3$ , respectively.



**Fig. 3.**  $\text{Hg}^0$  removal efficiency as a function of input energy density in the plasma process with different (a) NaCl and (b) sucrose aerosol surface concentrations. For comparison, the solid pink line shows the result without pre-existing aerosol particles. Gas conditions: 8%  $\text{O}_2$  and 4%  $\text{H}_2\text{O}$  in  $\text{N}_2$  balance; incoming  $\text{Hg}^0$  concentration fixed at  $110 \mu\text{g}/\text{m}^3$ .



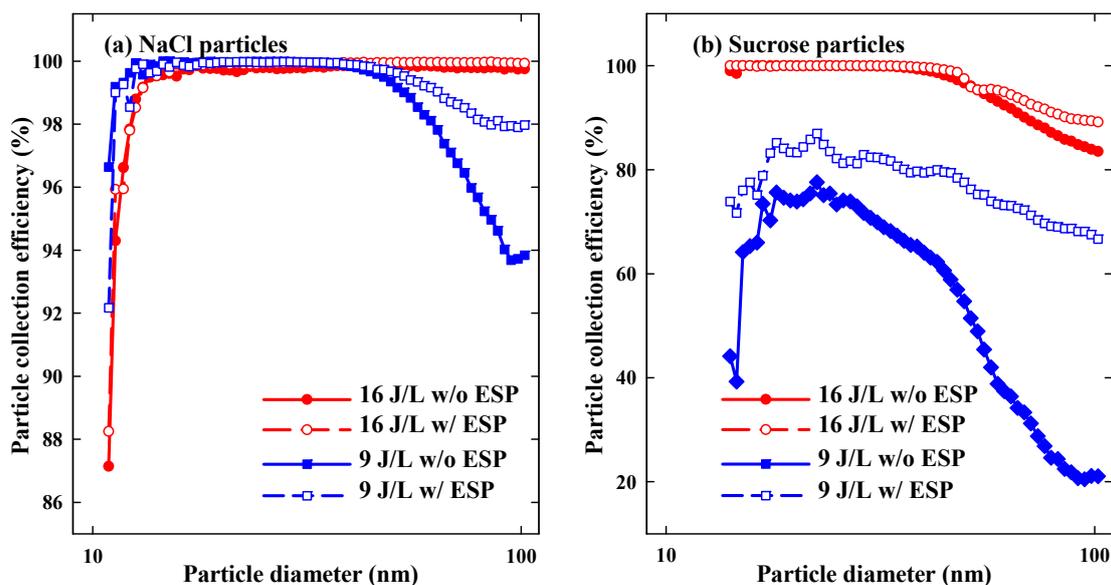
**Fig. 4.** The effect of aerosol type on mercury capture in the plasma system. The incoming  $\text{Hg}^0$  concentration was (a)  $45 \mu\text{g}/\text{m}^3$  (b)  $210 \mu\text{g}/\text{m}^3$  with aerosol surface concentration of  $0.4 \times 10^{11} \text{nm}^2/\text{cm}^3$ . The solid pink line shows the result without pre-existing aerosol. Gas conditions: 8%  $\text{O}_2$  and 4%  $\text{H}_2\text{O}$  in  $\text{N}_2$  balance.

especially at high input energy density (such as  $> 14 \text{J/L}$ ). Another inorganic particles,  $(\text{NH}_4)_2\text{SO}_4$ , were also tested in this study and showed similar characteristics as NaCl particles with a slightly lower  $\text{Hg}^0$  removal efficiencies.

#### Collection Efficiency of Aerosol Particles

Fig. 5(a) shows collection efficiencies of sodium chloride particles by the DDBD device with and without the downstream ESP. A higher energy input in the DDBD device results in a higher particle collection efficiency. When

the downstream ESP was used, most charged aerosol particles from the reactor outlet were captured. When particle diameter is smaller than  $\sim 15 \text{nm}$ , the collection efficiency drops significantly because of their low charging efficiency (Jung and Kittelson, 2005; Jiang *et al.*, 2007a, b; Suriyawong *et al.*, 2008) and possibly related to the generation of new particles in the plasma region as well (Romay *et al.*, 1994; Guo *et al.*, 2014). Most pre-existing NaCl particles ( $> 90\%$  in particle number) were collected. The charged ratio of aerosol particles in the outgoing gases was over 50% in the



**Fig. 5.** Particle collection efficiency by the double dielectric barrier discharge device with and without a downstream electrostatic precipitator. The incoming  $\text{Hg}^0$  concentration was  $110 \mu\text{g}/\text{m}^3$ . Pre-existing (a) NaCl and (b) sucrose aerosol surface concentration was  $0.4 \times 10^{11} \text{nm}^2/\text{cm}^3$ .

particle diameter range of 30–100 nm, according to results with and without the ESP.

Fig. 5(b) shows collection efficiencies of sucrose particles by the DDBD device. The result for sub-15 nm particles is not shown due to bad repetition in experiment. The tendency of the collection efficiency as a function of particle diameter is similar to that of NaCl particles. However, the collection efficiencies are much lower than those of NaCl particles, especially at low input energy density, e.g., 9 J/L.

#### **Possible Mechanisms for Improving Mercury Removal in the DDBD System**

Schematic illustration for possible  $\text{Hg}^0$  removal mechanism in the DDBD system with pre-existing aerosol particles is shown in Fig. 6, i.e.,  $\text{Hg}^0$  oxidized by plasma species directly in gas-phase and on aerosol surface, and  $\text{Hg}^0$  in-flight adsorbed by charged aerosol particles.

Firstly,  $\text{Hg}^0$  can be directly oxidized by plasma species (e.g.,  $\cdot\text{OH}$  and  $\text{O}$  radicals) in discharge and post-discharge regions to form oxidized mercury ( $\text{Hg}^{2+}$ ). This process is the major removal pathway of  $\text{Hg}^0$  when there are no suspended particles in the flue gas. When aerosol particles are present, the etching of particle surface by plasma may generate reactive species as well. For instance, Cl might be produced when NaCl particles are present. As one of the most common composition in flue gas from coal combustion (Saarnio *et al.*, 2014), Cl has been reported for rapidly reacting with  $\text{Hg}^0$  in gas phase and on the particle surface to form oxidized mercury (Zhao *et al.*, 2006; An *et al.*, 2014). Possibly due to this mechanism, NaCl aerosol performs better in improving  $\text{Hg}^0$  removal than sucrose aerosol.

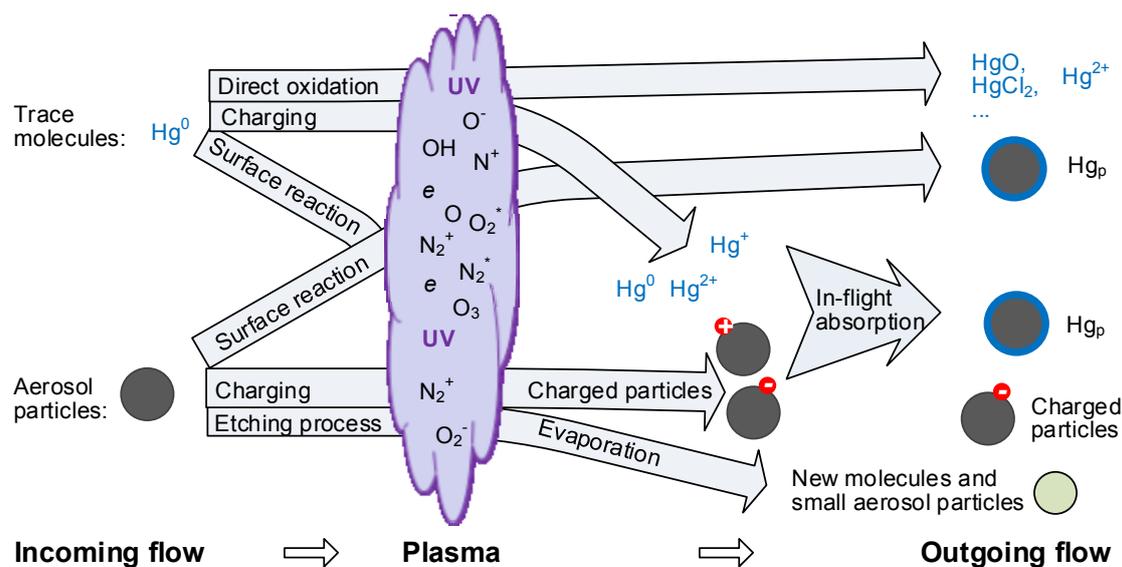
Secondly, pre-existing aerosol particles provide additional surfaces for  $\text{Hg}^0$  oxidation reactions. Experimental investigation without pre-existing aerosol particles found that most of the removed  $\text{Hg}^0$  deposits on plasma reactor

surface in the form of oxidized mercury (Byun *et al.*, 2011a; An *et al.*, 2014). Surface-induced reaction has higher reaction rate than that of gas-phase oxidation reaction (Pal and Ariya, 2004). The effect of surface-induced reaction can be enhanced due to the addition of surface area by suspended aerosol particles in the plasma region.

Thirdly, suspended particles possibly in-flight adsorb mercury, especially for charged particles and mercury ions. The plasma process can lead to aerosol charging (Manirakiza *et al.*, 2013), while  $\text{Hg}^0$  can be ionized to mercury ions ( $\text{Hg}^+$ ,  $\text{Hg}^{2+}$ ) due to the lower first ionization energy of elemental mercury (10.4 eV) than that of most common flue gas molecules such as  $\text{N}_2$  and  $\text{O}_2$ . The charged aerosol particles can then in-flight adsorb mercury (Clack, 2006), especially for mercury ions. Our experimental results reveal that NaCl particles of higher charged fractions have better mercury removal efficiencies than sucrose particles of relatively lower charged fractions.

Pre-existing aerosol particles provide reaction surface for heterogeneous nucleation and increase condensation of condensable species (McMurry *et al.*, 2005; Kuang *et al.*, 2010). Formed  $\text{Hg}_p$  can be collected by the DDBD device and/or removed by the downstream conventional air pollution control system such as ESP. This enhanced removal of mercury is possibly magnified in stage ESP systems which include a dielectric barrier discharge configuration for pre-charging aerosol particles (Byeon *et al.*, 2006).

Except for the above processes, the reaction of plasma species with aerosol particles cannot be ignored, especially for organic particles whose surface oxidation can compete for the reactive species. The highly reactive species can oxidize and etch aerosol surface, resulting in aerosol evaporation and shrink to form new molecules and smaller aerosol particles. The newly formed molecules and/or particles can also compete for plasma-generated reactive species. It is



**Fig. 6.** Schematic illustration of  $\text{Hg}^0$  removal mechanism with pre-existing aerosol particles in the plasma system. Three possible kinetic processes are responsible for  $\text{Hg}^0$  removal, i.e., (i) directly oxidation by active species (such as  $\bullet\text{OH}$  and  $\text{O}$  radicals) and etched out species (such as  $\text{Cl}$  from  $\text{NaCl}$  particle) to form oxidized mercury ( $\text{Hg}^{2+}$ ); (ii) oxidation on particle surface to form particulate bound mercury ( $\text{Hg}_p$ ); and (iii) in-flight adsorption by charged particles to form  $\text{Hg}_p$ .

possible that organic particles consume more reactive species than inorganic particles. This may explain different behaviors of inorganic particles ( $\text{NaCl}$ ) and organic particles (sucrose) in improving  $\text{Hg}^0$  removal efficiency. The practical kinetic processes can be more complex and remains to be further addressed.

## SUMMARY

A double dielectric barrier discharge plasma device was developed for removing elemental mercury with the presence of aerosol particles. The existence of particles improves  $\text{Hg}^0$  removal efficiency in plasma treatment process. Inorganic  $\text{NaCl}$  particles perform better than organic sucrose ones in improving  $\text{Hg}^0$  removal efficiency. The plasma system also serves as an aerosol charger and an electrostatic precipitator such that these particles can be efficiently removed in the reactor. Three kinetic processes are possibly responsible for the improving  $\text{Hg}^0$  removal efficiency: (i) rapid mercury oxidation by those etched out radical species from particle surfaces, (ii) mercury oxidation on the aerosol particle surface, and (iii) in-flight mercury adsorption by charged aerosol particles to form particulate bound mercury. These findings underline the important role of aerosol particles in mercury pollution controls. Further mechanism investigations are needed to reveal the role of suspended particles in the removal of mercury and other pollutants such as  $\text{NO}$  and volatile organic compounds.

## ACKNOWLEDGEMENTS

We thank Qiang Zhang, Yang Qiao, Jiixin Li, and Qingru Wu from Tsinghua University for their experimental supports. Q. Li thanks Profs. Reinhard Niessner, Dong Nam Shin, Herek L. Clack, Jie Li, and Da-Ren Chen for

their helpful insights and discussions. Financial supports from the National Key Basic Research and Development Program of China (2013CB228505 & 2013CB43005), National High Technology Research and Development Program of China (2013AA065101 & 2013AA065004) and National Natural Science Foundation of China (21221004) are acknowledged.

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*Received for review, January 4, 2015*

*Revised, March 12, 2015*

*Accepted, March 13, 2015*