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Nanoparticle removal and exhaust gas cleaning using gas-liquid interfacial nonthermal plasma

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ABSTRACT

We investigate the removal of air pollutants containing nanoparticles with a wet-type plasma reactor that generates a gas-liquid interfacial nonthermal plasma. To evaluate the performance of this reactor, we prepare a simulated exhaust gas using polystyrene latex particles, as well as gas cylinders of synthesized air and mixed gas of nitric oxide (NO) and sulfur dioxide (SO_2). We achieve a partial collection efficiency of more than 99% for nanoparticles. Moreover, we obtain removal efficiencies exceeding 99% and 81% for SO_2 and NO_x, respectively. We confirm that the proposed reactor is useful for the simultaneous removal of nanoparticles, NO_x, and SO_x.

1. Introduction

Particulate matter (PM), nitrogen oxides ($NO_x = NO + NO_2$), and sulfur oxides ($SO_x = SO_2 + SO_3$) emitted from diesel engines, thermal power generation plants, and general industrial factories have had an adverse effect on the global environment and humans. Therefore, there is a need to treat them. Recently, one of the treatment methods that have attracted much attention is nonthermal plasma technology [1–7]. In particular, in the Asian region, the presence of nano-sized particles suspending in the atmosphere along with harmful air pollutants has become an international problem in terms of its negative impact. $PM_{2.5}$ represents particles that have an aerodynamic diameter less than 2.5 μm. In particular, smaller particles or nanoparticles having an aerodynamic diameter of approximately 100 nm induce the onset of respiratory diseases because they are proceed to the distal regions of the lung and deposited there. In the EU, PM control regulations for motor vehicles are hence conducted by not only mass standard but also particle number (PN) standard [8].

Under these circumstances, various kinds of studies have been conducted worldwide, especially in Asian countries, to develop indoor air cleaners for the removal of $PM_{2.5}$. Generally, PM that is suspended in the atmosphere can be collected using air cleaners, where polymer filters (HEPA or ULPA filters) are combined with a fan apparatus. These types of air cleaners were used in nuclear power plants after the March 2011 nuclear power plant accident occurred. However, radioactive materials that are collected inside the filters after filtration are problematic, and should be treated properly. Furthermore, it is difficult to achieve a highly efficient removal of gaseous air pollutants using polymer filters, and the pressure drop, which increases with the deposition of particles in the filter, induces energy losses in the fan utilized. Therefore, to remove gaseous harmful radioactive air pollutants such as radioactive iodine, I 131, and cesium, Cs 137, which were emitted from the nuclear power plant and suspended in the atmosphere, as well as suspended nanoparticles that combined with these harmful matter, there is a need for a compact air-cleaner that does not require the use of filters.

Considering this background, in the present research, we propose a system where a wet-type electrostatic precipitator [9–11] is combined with an atmospheric-pressure nonthermal plasma (NTP) generator for the removal of nanoparticle pollutants suspended in the atmosphere. This system was named a “wet-type plasma

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reactor [12–16]." The proposed wet-type plasma reactor has an advantage in that there is no decrease in the collection efficiency for PM having low electric resistance such as soot due to the re-entrainment observed in a dry electrostatic precipitator [17], and we achieve a high collection efficiency for nanometer-sized particles. In the apparatus, we applied a high-voltage pulse between the electrodes, and positive ions are induced. As a result, PM in the channel is positively charged and collected inside the water film on the grounded electrode. After the collection, pollutants are cleaned with high efficiency by treating the wastewater from the reactor.

In this paper, as a basic study of this type of real-sized system, we describe the experimental model and principle of PM and gaseous air pollutants cleaning of the proposed wet-type plasma reactor. Following the previous paper [16], we carried out the experiments of nanoparticle removal and the simultaneous removal of NO_x and SO_x using the proposed wet-type plasma reactor. Furthermore, we evaluated the partial collection efficiency for nanoparticle using an SMPS (scanning mobility particle sizer) as a function of a gas flow rate and the removal efficiencies of NO_x and SO_x as a function of Na_2SO_3 concentration.

1.1. Principle of PM and gaseous air pollutants cleaning

In this section, we describe a mechanism and principle for the simultaneous removal of PM, NO_x, and SO_x using the wet-type plasma reactor with the aqueous solution of Na_2SO_3 or a mixture of Na_2SO_3 and NaOH [14–16,18–21]. The principle of charging and collecting the PM is illustrated in Fig. 1. The high-voltage pulse power supply is used for generating NTP to treat gaseous air pollutants in the reactor. More than 95% of nanoparticles can be collected, and this approach has superior performance. On the other hand, gaseous air pollutants flowing in the channel are treated by the NTP. Further, electrostatic wind induced by the moving ions stirs the flow [22] to enable absorption into the aqueous solution film. Wastewater solution should be treated and vaporized, condensed, and chemically treated (in the case of radioactive particles), and then it finally becomes solid. However, for low-level harmful pollutants, it can be drained to sewage as is. As an aqueous solution, the following reducing agents are suitable for the simultaneous removal of NO_x and SO_x.

NO gaseous component passing through the wet-type reactor reacts with nitrogen and oxygen radicals induced by the NTP. It is then partially reduced to nitrogen, but mostly oxidized to NO_2.

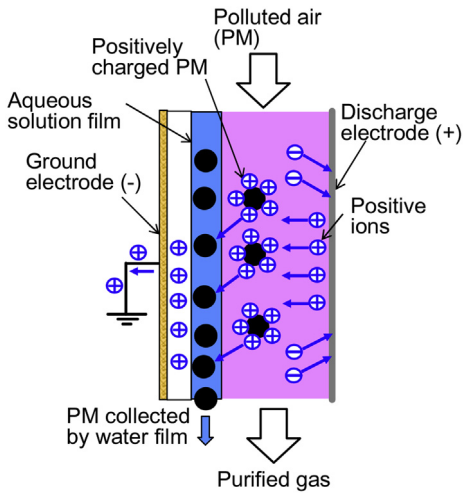

Fig. 1. Principle of particle collection using wet-type plasma reactor [16].

$$N_2 + e \rightarrow \bullet N + \bullet N + e \quad (1)$$

$$NO + \bullet N \rightarrow N_2 + \bullet O \quad (2)$$

$$NO + \bullet O + M \rightarrow NO_2 + M \quad (3)$$

where •N and •O refer to nitrogen and oxygen radicals, respectively, e denotes an electron, and M represents third materials (N_2 and O_2). The reaction (3) progresses by NTP in the presence of oxygen radicals •O. NO_2 obtained by the oxidation reaction (3) is water soluble and can be removed by the reaction in Na_2SO_3 aqueous solution; the NO_2 is converted and reduced to N_2 and non-toxic water-soluble Na_2SO_4 according to the chemical reaction.

$$2NO_2 + 4Na_2SO_3 \rightarrow N_2 + 4Na_2SO_4 \quad (4)$$

To remove SO_2 included in the exhaust gas and enhance the reducing reaction (4) for NO_2, the mixture solution of Na_2SO_3 and NaOH is used. Na_2SO_3 is generated by the chemical reaction (5) between SO_2 and NaOH.

$$SO_2 + 2NaOH \rightarrow Na_2SO_3 + H_2O \quad (5)$$

On the other hand, SO_2, SO_3, and NO_2 are water soluble, and as a result, H_2SO_3, H_2SO_4, HNO_2, and HNO_3 are induced in the solution.

$$SO_2 + H_2O \rightarrow H_2SO_3 \quad (6)$$

$$SO_3 + H_2O \rightarrow H_2SO_4 \quad (7)$$

$$2NO_2 + H_2O \rightarrow HNO_2 + HNO_3 \quad (8)$$

The pH in the solution decreases as reactions (6)–(8) progress. However, NaOH added in the solution contributes to the neutralization and increases the pH. On the other hand, when the pH of the aqueous solution is more than eight (alkaline), carbon dioxide (CO_2) is also absorbed by the solution film. Generally, because the CO_2 concentration in the exhaust gas is several vol%, the solution becomes acidic or enters a low-pH state. However, when NaOH is added to the solution, sodium hydrogen carbonate ($NaHCO_3$) and sodium carbonate (Na_2CO_3) are generated, and the solution becomes a pH buffer in order to keep it as an alkali. Furthermore, it is known that the denitration based on the reaction (4) is effective for alkali solutions.

From these explanations, we deduce that the simultaneous removal of NO_x and SO_x based on the reactions (1)–(8) is a highly consistent technique. Next, we discuss some laboratory-scale experimental results on the removal of PM, NO_x, and SO_x based on the principle and the experimental model.

2. Experimental apparatus and method

Fig. 2 shows schematics of the wet-type plasma reactor used in the experiment. In the research, we combined the electrostatic precipitator for particulate removal with the wet-chemical reactor for solution scrubbing. This reactor is a laboratory-scale model with a cylindrical channel having a diameter of 20 mm, a stainless-steel discharge wire having a diameter of 2 mm, and a grounded electrode having a length of 260 mm. This apparatus consists of a pair of electrodes and an aqueous solution film on the inner wall of the channel. The solution overflows from an overflowing reservoir in upper region of the reactor and flows on the inner wall of the reactor to generate the solution film. The solution film uniformly flows between electrodes because the inner wall of the flow channel is hydrophilic because of the plasma. The air pollutants are