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Novel electrostatic precipitator using unipolar soft X-ray charger for removing fine particles: Application to a dry de-NO_X process

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HIGHLIGHTS

• Unipolar soft X-ray charger and collection part were applied to by-product particle removal in a dry de-NO_X process.

• NO_X was converted to a NH₄NO₃ aerosol with a peak diameter of ~100 nm.

• Greater than 90% of particle collection efficiency was observed.

• Soft X-ray charger has the compact size, easy operation, and non-interruptive charging mechanism.

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ABSTRACT

The novel electrostatic precipitator (ESP), consisting of a soft X-ray charger and a collection part, was demonstrated and applied to a dry de-NO_X process to evaluate its performance in by-product particle removal. NO_X gas was oxidized by ozone (O₃) and neutralized by ammonia (NH₃) sequentially, and finally converted to an ammonium nitrate (NH₄NO₃) aerosol with ~100-nm peak particle diameter. The unipolar soft X-ray charger was introduced for charging the by-product particles in this dry de-NO_X process. For the highest particle collection efficiency, the optimal operating conditions of the soft X-ray charger and collection part were investigated by adjusting the applied voltage of each device. The results showed that ∼99% of NO_X was removed when the O₃/NO_X ratio was increased to 2 (i.e., the maximum production conditions of the NH_4NO_3 by-product particles by the gas-to-particle conversion process). The highest removal efficiency of particle (∼90%) was observed with operating conditions of positive polarity and an applied voltage of ∼2–3 kV in the charger chamber. The unipolar soft X-ray charger has potential for particle removal systems in industrial settings because of its compact size, ease of operation, and non-interruptive charging mechanism.

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1. Introduction

Nitrogen oxides (NO_X) are major contributors to many adverse environmental and health effects because they have roles in global warming, smog, acid rain, ozone destruction, and the incidence of respiratory diseases $[1,2]$. As the use of diesel engines has increased in both industry and transportation, diesel exhaust gases, which contain large amounts of NO_X are considered an increasing

problem. Thus, NO_X emissions from diesel engines have been regulated, such as by European emission standards and the Tier standards [\[3\].](#page--1-0) Specifically, the regulation of heavy-duty diesel engines, which are used for large vessels or power plants, will tightened considerably as environmental and health concerns continue to grow.

As part of an effort to remove NO_X emissions, non-thermal plasma techniques known as "dry de-NO_X processes" (e.g., electron beams, pulsed corona discharges, corona radical shower systems, and indirect ozone (O_3) injection) have been developed [\[4–10\].](#page--1-0) These dry de-NO_x processes have unique advantages, such as simple operating conditions (at room temperature and atmospheric pressure) in comparison with the wet-chemical de- NO_X process

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[\[11,12\],](#page--1-0) and the ability to also remove other pollutants, such as $SO₂$ and heavy metals. Additionally, the collected by-product particles after the dry de-NO_X process may be reused as agricultural fertilizer (e.g., ammonium nitrate, NH_4NO_3 , particles resulting from de-NO_X process with indirect O_3 injection) [\[13\].](#page--1-0)

Generally, particulate filters, such as metal foam and electrostatic filters, have been used for eliminating the by-product particulate matter after dry de-NO_X processes. However, these filtration methods have several disadvantages: frequent replacement of filters due to dust loading and high energy consumption with the filter pressure drop. To overcome these shortcomings, electrostatic precipitators (ESPs) have been used widely for the removal of particulate pollutants in flue gases as the most effective option at plants and with heavy diesel engines. Conventionally, an ESP has a two-stage configuration, consisting of a separate particle charger and collection electrode sections to improve particle collection efficiency [\[14\].](#page--1-0)

In the relationship between particle charge and mechanical mobility, particles in the size range of 0.1–1 μ m are difficult to control and capture because mobility under typical charging conditions has a minimum value in this size range [\[15\].](#page--1-0) Particulate matter (PM) from diesel engines and after-treatment processes has low resistivity and very small sizes, around 100 nm [\[16,17\].](#page--1-0) Thus, various chargers, such as packed bed plasma [\[18\],](#page--1-0) corona discharge [\[19\],](#page--1-0) and ionizers [\[20\]](#page--1-0) have been investigated to increase the particle charging efficiency. Despite their high charging abilities, these chargers produce additional ozone in the exhaust gas with a high-voltage source; also, corrosion of the electrode is a major drawback. UV and soft X-ray emissions have been investigated to overcome these problems [\[21,22\].](#page--1-0) UV irradiation has been studied using laboratory-synthesized nanoparticles and ultrafine particles as a charging method [\[23,24\].](#page--1-0) Bipolar charging [\[25\]](#page--1-0) and unipolar charging [\[26\]](#page--1-0) by soft X-rays have been introduced for nanoparticle control [\[27,28\].](#page--1-0)

In this study, we demonstrate a new ESP system using a unipolar soft X-ray charger and applied it to the removal of by-product particles produced from a dry de-NO_x process. The dry de-NO_x process was conducted using O_3 oxidation and NH₃ neutralization. In this process, NH_4NO_3 aerosols were generated as a by-product. The novel ESP system was introduced to remove this by-product. The unipolar soft X-ray charger was used for charging the NH_4NO_3 aerosol. The optimal operating conditions for ESP using the soft X-ray charger were investigated in terms of various applied voltages in the charger and collection plates.

2. Materials and methods

2.1. Experimental apparatus

The experiment was conducted in the following four steps: the generation of a simulated flue gas, the gas-to-particle conversion by gas oxidation in a reaction chamber, particle charging and removal by the collection part, and the analysis of gas and particle characteristics (Fig. 1(a)).

The simulated flue gas of a diesel engine was prepared by a mixture of the following gas species: air, oxygen, NO (0.2% NO with 98.8% N₂ balanced), NO₂ (0.2% NO₂ with 98.8% N₂ balanced). The ratio of NO to $NO₂$ was set to 9:1, which simulates the real flue gas of diesel engines [\[29\].](#page--1-0) The flow rate of each gas species was controlled through a mass flow controller (MFC; FC-280SAV; Mykrolis, Billerica, MA, USA) and the NO_X concentration in the carrier gas was adjusted by controlling the mixing ratio between each gas. The relative humidity and temperature of the simulated flue gas were 20% and 25 ◦C, respectively.

The reaction chamber was made of stainless steel and was of 30-cm length and a circular cross-section with 7.6-cm inner diameter. O_3 gas was introduced into the chamber, generated by an O_3 generator (LAB-S; Ozonetech Co., Korea). O_3 has a high oxidation potential for eliminating NO_X in the flue gas. To convert the reacted and converted NO_X (e.g., $NO₂$ and $HNO₃$) gases into nano-sized ammonium nitrate (NH₄NO₃) particulate matter (a chemically neutral by-product that can be recycled as an agricultural fertilizer), additional gaseous $NH₃$ was injected into the reaction chamber with the simulated flue gas. The concentration of $NH₃$ was set to the same molar concentration as NO_X in the flue gas.

The nano-sized NH_4NO_3 particle removal system consisted of two major parts. One was a particle charger for the electrical charging of airborne particles by means of a soft X-ray module (SXN-10UE; Invenix, Gumi, Korea) that emits 'soft' X-rays, below \sim 10 keV. As shown in Fig. 1(b), the hole, located between the soft X-ray module and the charger chamber, makes the X-ray emissions enter the chamber and was sealed with a polyamide film (Kapton 30HN, DuPont Corp., 30-µm thick), which has high penetrability for soft X-rays (∼90%). In particular, the hole in the chamber has

Fig. 1. (a) Schematic of the experimental set-up, (b) unipolar soft X-ray charger.

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