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Full Length Article

An integrated system of dielectric barrier discharge combined with wet electrostatic precipitator for simultaneous removal of NO and SO_2: Key factors assessments, products analysis and mechanism

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ABSTRACT

This paper developed an integrated system of Dielectric Barrier Discharge (DBD) with Wet Electrostatic Precipitator (WESP) for simultaneous removal of NO and SO_2. In view of practical application, the effects of some key factors such as peak-peak voltage ($V_{p\text{-}p}$), O_2 and H_2O contents on the simultaneous removal of NO and SO_2 were systematically investigated. The results indicated that, in single DBD process, the additions of O_2 and H_2O significantly promoted the removal of SO_2, especially in presence of $V_{p\text{-}p}$, but remarkably inhibited the conversion of NO, which was due to the regeneration of NO from the radical-induced reactions among N·, O· and OH·. WESP played a key role in capturing the oxidizing products and acid aerosols, and as anticipated, the integrated system of DBD-WESP showed a better performance on the simultaneous removal of NO and SO_2, with the best efficiencies of 98.9% for SO_2 and 87.1% for NO. The interaction mechanism among the components in the presence of $V_{p\text{-}p}$ was speculated, the transformations of SO_2 and NO in this system were also proposed. The removal products of SO_2 and NO by the integrated system were determined as sulfate and nitrate according to the Ion Chromatography (IC) analysis of the spent solution collected in WESP.

1. Introduction

Approximately 45% of coal are used for electric power generation in China [1]. During coal combustion, a large number of pollutants including sulfur dioxide (SO_2) and nitrogen oxides (NO_x) are generated [2], thereafter the secondary derivants generated from the photocatalysis of SO_2 and NO_x in the atmosphere lead to the formation of haze [3]. The toxic chemical smog has threatened the people's health and been received extensive attention in recent years. The mature technologies for removal of SO_2 and NO in coal-fired power plants are Wet Flue Gas Desulfurization (WFGD) [4,5], and Selective Non-catalytic Reduction (SNCR) and Selective Catalytic Reduction (SCR) [6,7]. For dealing with the terrible problem of air pollution, Chinese government has issued the rigorous ultra-low emission standards of SO_2 (35 mg/m^3) and NOx (50 mg/m^3) for coal-fired power plants in 2015 [8]. Afterwards, most of the coal-fired power plants installed the low-temperature economizer and Wet Electrostatic Precipitator (WESP) to conduct the effective capture of dust and deep removal of fine particles after WFGD. However, the stage-treatment strategy of SCR-WFGD-WESP has a series potential problems, such as large floor area, complicated system, high investment and high operational cost [9,10]. Hence, to develop an advanced method for simultaneous removal of NO and SO_2 with low cost and simplified system is one of the hot issues in the field of air pollution control [11,12].

Recently, various advanced methods for simultaneous removal of SO_2 and NO have been studied, including the electrochemistry [13,14], the gas–solid phase adsorption [15,16], the gas–solid phase catalysis [17,18], the complex-absorbent absorption [19] and the liquid phase oxidation [20,21]. Among the methods, the oxidation method is one of the promising ways [22]. As a consequence of the high utilization of energy power and high yield of activated radicals, non-thermal plasma (NTP) technology has attracted widespread attention. Besides, it also has the superiorities of high oxidation rate, small floor area, non-chemical additives, low investment and low operational cost. Moreover, such technology can be applied at atmospheric pressure and room temperature without any obvious secondary pollution [23].

Dielectric Barrier Discharge (DBD) is a kind of NTP reactor. It can generate electrical discharge when the two electrodes are imposed by high voltage alternating current and separated by one or more insulating dielectric. Compared with other NTP reactors, DBD holds the advantages of high degree of technological maturity, low energy consumption, uniform and stable discharge features, and uniform plasma

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spatial distribution [24]. Some scholars have employed the DBD reactor to conduct the degradation of organic or inorganic compounds. Bahri et al. [25] studied the VOCs degradation in DBD-catalytic reactor. Thevenet et al. [26] studied the performance of photocatalysis-DBD hybrid system on oxidizing acetylene. Niu et al. [27] investigated the flue gas components on the oxidation of Hg^0 by DBD. Wang et al. [28] studied the effects of temperature and relative humidity on NO_x removal by DBD with acetylene. The main oxidizing species generated in DBD process are O·, OH· and O_3, which can be used in gas phase oxidations of SO_2 and NO, with forming H_2SO_4/SO_3 and $HNO_3/NO_2/NO_3/N_2O_5$ [29,30]. When H_2O vapor exists in the flue gas, the oxidation products can be further converted to the corresponding acid mist [31,32].

If these acid-mists and the residual radicals can be captured or absorbed by the downstream device, the goal of simultaneous removal of SO_2 and NO will be realized. It is so lucky, as aforementioned, WESP has been equipped in most of the coal-fired power plants in China and considered as an effective way to remove acid aerosol and fine particulate matter [33]. Thus WESP can be used as a supplement for DBD reactor to develop an integrated system of DBD-WESP. It had been reported [34,35] WESP could complete more than 95% removal of sulfuric acid mist, meanwhile a number of NO_2, SO_2 and SO_3 could be also absorbed by alkaline mist. Not only to this, the active species (O· and OH·) generated in the Direct Current (DC) corona in WESP could further induce the conversions of NO and SO_2 [36]. It implies that the hybrid system of NTP-WESP can be considered as a strengthen oxidation approach of two-stage electro-stimulated radical oxidation, not only a fine-particle capture system. Hence, the combination of NTP and WESP is feasible and reasonable.

In actual, Wang et al. [27] had conducted the experiments of simultaneously removing NOx, SO_2, and Hg^0 by a NTP-$(NH_4)_2SO_3$ absorption-WESP (water) hybrid system, and confirmed the oxidation of NO to NO_2 in NTP process. But the oxidation of Hg^0 was significantly restrained since the NO competition, and S(IV) was found to be favorable for the absorption of NO_2, and the addition of $S_2O_3^{2-}$ further enhanced the absorption of NO_2. But one shortage of this system was that it still needed a scrubber to pre-absorb SO_2 and NO_x, which caused a relatively complicated treating process. Moreover, the effects of some key factors on the simultaneous removal process were not assessed, such as peak-peak voltage ($V_{p\text{-}p}$), O_2 and H_2O, which could significantly affect the generations of radicals and the absorption of oxidizing products. For this, the present paper tried to systematically investigate the synergistic effects of $V_{p\text{-}p}$, O_2 and H_2O on the simultaneous removal of NO and SO_2, and to reveal the interaction mechanism among the reactants. Finally, the removal products of SO_2 and NO in the spent solution of WESP were identified by Ion Chromatography (IC).

2. Experimental

A schematic of the experimental setup is shown in Fig. 1. It consists of a simulated flue gas generation part, a DBD reactor supplied by high-voltage alternating-current (AC) power (CTP-2000K, Suman plasma company, Nanjing), a WESP reactor supplied by high-voltage DC power (DW-N603-2ACDE, Dongwen high-voltage power supply company, Tianjin), and an online flue gas analyzer (ECOM-J2KN, RBR Company, Germany). Teflon pipe was used to avoid the erosion of the tube. The pipeline was heated by a temperature-controlled heating belt (XMTG-7411, Shangtong instrument company, Wenzhou) to keep the gas temperature in DBD at 323 K and avoid the condensation of moisture.

The DBD reactor consisted of four coaxial cylinders: a center stainless steel rod, a stainless mesh tube, a corundum tube and a quartz tube. The rod, enveloped by corundum tube with the diameter of 13 mm and a length of 296 mm, was served as inner electrode and ground electrode. The stainless mesh tube with the diameter of 25 mm and a length of 150 mm attached tightly to the outside wall of the quartz tube, was served as the outer electrode and the high-voltage electrode. Simulated flue gas was introduced into a 3.5 mm gap tube between the inner and outer electrodes. $V_{p\text{-}p}$ and frequency were measured by an oscilloscope (DPO2012B, Tektronix, USA).

A home-made WESP was arranged after the DBD reactor. Barbed wire, with a length of 1000 mm, was served as the corona electrode and connected to the negative electrode of DC power supply. Stainless steel tube, with the diameter of 90 mm and a length of 1000 mm, was used as collecting electrode and connected to the ground. A water tank was set under the collecting electrode, and a circulating pump was used for supplying deionized water to the inner wall of the collecting electrode.

During the experiments, the simulated flue gas was prepared by mixing N_2, O_2, NO and SO_2, supplied by compressed gas cylinders (Huawei gas technology company, Baoding). Meanwhile, the carrier gas, N_2, was injected in a bubbler containing deionized water to carry the water vapor into the simulated flue gas. Of note, the temperature of the bubbler was adjusted by a thermostat water bath (DF101S, Yuhua instrument company, Gongyi), so that the content of water vapor in simulated flue gas could be controlled. Then the generated simulated flue gas passed through DBD and WESP at a flow rate of $0.5\ m^3/h$. The inlet and outlet simulated flue gases were detected by the flue gas analyzer. The components in gases could be detected, including O_2 in the range of 0–21% (0.1%), NO in the range of 0–5000 ppm (1 ppm), NO_2 in the range of 0–1000 ppm (1 ppm) and SO_2 in the range of 0–5000 ppm (1 ppm).

The efficiencies of SO_2 removal and NO conversion were calculated from Eq. (1).

$$\eta = (C_{in} - C_{out})/C_{in} \times 100\% \tag{1}$$

where η is the removal efficiency; C_{in} and C_{out} are the inlet and outlet concentrations, respectively.

For the detection of various anions, an ion chromatography (792 Basic, Metrohm AG) was used. The anions such as SO_4^{2-}, SO_3^{2-}, NO_3^-, NO_2^- could be completely separated on a Metrosep A Supp 4 Anion chromatographic column with a mixture of Na_2CO_3 solution (1.4 mmol/L) and $NaHCO_3$ solution (4.0 mmol/L) at a flow rate of 0.8 mL/min, and detected by an electrical conductivity detector, the detection limit was lower than 10 μg/L.

3. Results and discussion

3.1. Effect of O_2 on the SO_2 removal and NO conversion by single DBD

O_2 is the major source for generating O· and O_3 those mainly contribute to the oxidations of NO and SO_2. Thus the effect of O_2 on the removal efficiencies of NO and SO_2 was investigated. The concentrations of NO and SO_2 were 400 and 1000 ppm, respectively. O_2 content was controlled at 0%, 3%, 6% and 10%. The input power of the DBD reactor was adjusted through changing the $V_{p\text{-}p}$ of AC power supply. Since the plasma discharge can be generated only when the $V_{p\text{-}p}$ reaches up to 24 kV, the investigated $V_{p\text{-}p}$ range was selected from 24 to 38 kV in this work. The frequency of the AC power was set at 8 kHz.

The NO conversion efficiencies (η_{NO}) as a function of $V_{p\text{-}p}$ at different O_2 contents are shown in Fig. 2. It can be found that the effect of $V_{p\text{-}p}$ on η_{NO} depends on the content of O_2. Thus we firstly investigated the effect of $V_{p\text{-}p}$ on η_{NO} in the absence of O_2. As shown in the embedded in Fig. 2, the increase of $V_{p\text{-}p}$ exhibits a little tiny promotion on the NO removal. In N_2 atmosphere, N· could be generated from the motivation of N_2 by high-energy electrons (Eq. (2)), then it would attack and restore NO to N_2 via the Eq. (3) [37], resulting in an abatement of NO. And η_{NO} would be increased when more N· were generated with the increasing of $V_{p\text{-}p}$. But it should be noted that, at the same time, the increasing of N· resulting from the increasing of $V_{p\text{-}p}$ would accelerate the regeneration of N_2 due to the N· quenching reactions. Hence, the utilization of N· was decreased, and the strengthening oxidation of NO was inhibited, and the increasing trend of η_{NO} was slowed down. Given the energy cost-efficiency, an appropriate $V_{p\text{-}p}$ was recommended.