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Electron beam treatment of gas stream containing high concentration of NO_x: An in situ FTIR study

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HIGHLIGHTS

- The NO reduction is predominant in the absence of O_2 under EB processing.
- The O_2 presence in the gas stream increases the NO_2 formation during EB operation.
- The moisture content in the stimulated gas found to inhibit the NO_2 formation.
- The Na_2SO_3 scrubbing of the gas during EB operation increases the NO_x removal.
- The NO and NO_x concentration is reduced to the extent of 95% using reactor type II.

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ABSTRACT

The NO_x removal from gas stream by electron beam (EB) technique under flow through condition was investigated using two different reactors. Initially reactor type I was fabricated to investigate the NO_x removal performance, based on its performance, the reactor type II was designed for further investigation on NO_x removal efficiency. The influence of the applied current, O_2, water vapour and gas flow rate on the removal of NO and NO_x was studied. The FTIR spectrum of the NO gas samples acquired after EB irradiation indicated the formation of NO_2 and N_2O. It was observed that the NO_x concentration could be reduced to 95% from the initial NO concentration of 1000 ppm using reactor type II. The possible removal mechanism of NO_x under EB irradiation is discussed. It is inferred that the EB irradiation accelerate the reduction of NO to N_2 and O_2 in the absence of O_2. Addition of O_2 and water vapour in the NO/N_2 gas stream was found to influence the oxidation and reduction reaction, respectively under EB irradiation. The Na_2SO_3 scrubbing of the gas increased the NO_x removal performance during EB processing. The N_2O was observed to form mainly by the reduction of NO_2 during EB irradiation.

1. Introduction

Nitrogen oxides (NO_x), are considered as one of the primary pollutants in the atmosphere. Their emissions into atmosphere not only cause the formation of acid rain and ozone depletion, but also adverse effect to human health [1]. The term NO_x used to denote both nitrogen monoxide (NO) and nitrogen dioxide (NO_2). In the recent decades, abatement of NO_x from the combustion processes has become one of national priorities and greatest challenges in environmental protection. Though the efforts have been made to control the NO_x emission from the combustion process, post combustion NO_x control techniques are mandatory to meet the stringent emission standards.

The most widely studied NO_x control technique is selective catalytic reduction (SCR) with ammonia in the presence of oxygen, in which the NO_x could be reduced to 85% [2–5]. The SCR reaction is promoted by the catalyst which enables the reaction to proceed at low temperature. Nevertheless, the catalyst life time, catalyst poisoning and corrosion are common problems encountered in the SCR technique [2]. The other conventional NO_x control method is selective noncatalytic reduction (SNCR) performed with ammonia or cyanuric acid [5,6,2]. The high temperature window (982–1149 °C) is necessary to operate the SNCR process, which limits its application for the treatment of the diesel engine exhaust at atmospheric pressure conditions. Similarly, the methods include catalytic decomposition [7], adsorption [8,9], ion-exchanged zeolites [10,11], etc., have suffered with their limitations and disadvantages.

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In the recent decades, nonthermal plasma methods such as pulsed corona discharge [12–16], microwave discharge [13,17], dielectric barrier discharge [13,15,16,18–20], and electron beam (EB) irradiation [21,22], shown to be efficient compared to conventional treatment techniques employed in the reduction of NO_x and undesired species from gas stream at atmospheric pressure conditions. The nonthermal plasma or the nonequilibrium plasma is generated by the reaction of high energy electrons produced during EB or discharge processing with background gas molecules at ambient temperature. The plasma consists of the active components viz., free radicals, ions and secondary electrons which are playing vital role in the abatement of the gaseous pollutant. The advantage of the nonthermal plasma method is that the kinetic energy of the electrons mainly utilized for vibrational excitation or dissociation of the gas molecule rather heating up the gas molecule [21].

It is shown that the EB processing is remarkably high energy efficient than the electrical discharge processing. The specific energy consumption for the dissociation of N_2 molecule in the pulsed corona processing is 480 eV while, it is only 80 eV in the EB processing [23]. It is reported that the EB accelerator could reduce the NO_x to the extent of 82% from exhaust gas by combustion of high sulphur fuel oil, containing the NO_x concentration of 150–170 ppmv [22]. In another study, it is shown that pulsed EB treatment on exhaust gas from the combustion of polish light oil could achieve 30% reduction of NO_x from its initial concentration of 532 ppm [24]. The laboratory and the pilot scale test of EB flue gas treatment demonstrated that the process can be prominently applied for the simultaneous removal of three pollutant such as NO_x, SO_x and volatile organic compounds (VOC) [25].

The present research work is aimed to develop the efficient EB technique for the abatement of NO_x, emitted from the diesel engine in ships. Two different reactors named reactor type I and reactor type II was investigated for the removal of the NO_x at the laboratory scale. Initially reactor type I was fabricated to investigate the NO_x removal performance, based on its performance, the reactor type II was designed for further investigation on NO_x removal efficiency. The behaviour of the NO gas in the presence of O_2 and water vapour was studied at different experimental conditions under EB irradiation. The NO and NO_x removal efficiency and its possible mechanisms were discussed.

2. Materials and methods

2.1. EB reactor and experimental set up

The EB accelerator equipped with carbon nanotube (CNT) anode and Ti window cathode was used for the NO_x removal experiments. A DC rectifier with a maximum of 60 kV and 0.05 mA was used as a radiation source for irradiating the pollutant gases. The distance between anode and cathode was fixed at 23 mm. The EB, accelerated from the CNT anode at high vacuum pressure (10^{-5} Torr) projected perpendicular to the process vessel of the gas flow duct through Ti window having the diameter of 1.3 cm and thickness of 0.5 mm. The schematic diagram of the EB reactor experimental setup and the gas flow configuration of the reactor type I and reactor type II are illustrated in Fig. 1. The NO in N_2 standard gas, concentration of 1000 ppm obtained from Sumitomo Seika Chemical Co., Ltd. was used as a source pollutant gas. The O_2 gas with the purity of 99.8% from Taiyo Nippon Sanso Co., was utilized to mix with NO in order to stimulate the conditions of the diesel engine exhaust. The mass flow controller was used to control the gas flow into the EB reactor as given in Fig. 1. The O_2 was used as a carrier gas for the water vapour to study the influence of the water vapour on the removal of NO and NO_x. The O_2 gas from the mass flow controller was purged through the airtight glass container half filled with Milli-Q water placed in a thermoregulated water bath. Thus, the resultant outlet O_2 gas carried the water vapour was facilitated to mix with the NO gas stream as depicted in Fig. 1. The generation of the water vapour could be controlled to desire amount by adjusting the temperature of the thermoregulated water bath. All the EB experiments were conducted at ambient room temperature of 25 °C. The stimulated gas after EB irradiation was routed to flow into the moisture trap kept at 2 ± 1 °C to remove all its moisture prior to the FTIR analysis.

Initially, the reactor type I was designed to examine the NO_x removal performance. The volume of the process vessel for the EB treatment was 2.3 cm^3. The electron beam was directed to apply perpendicular to the gas flow in the process vessel through Ti window cathode. The gas flow configuration of the reactor type I is given in Fig. 1. Based on the NO_x removal performance of reactor type I, the reactor type II was designed and fabricated. The reactor type II (Fig. 1) was accomplished in such a way that to circulate the Na_2SO_3 reductant solution into the process vessel of the gas flow duct to scrub the reactant gas during EB processing. The reductant solution was circulated through the silicon tube with constant flow rate using the peristaltic pressure pump from the external Na_2SO_3 reservoir tank. The volume of the process vessel was 3.2 cm^3.

2.2. Analysis

The concentration of NO and other by-products in the gas stream obtained after EB irradiation were analyzed using JASCO FT/IR-4200 analyzer. The FTIR instrument is equipped with a glass cell to analyze components of the resultant gas from the EB processing. Forty scans were collected on the gas samples at a resolution of 4 cm^{-1}. The N_2 gas with a purity of 99.8% obtained from Toei Kagaku Co., Ltd., was used for background correction. The concentration of NO, NO_2 and N_2O was calibrated using commercial NO in N_2, NO_2 in N_2 and N_2O in N_2 standard gas, respectively. The absorbance band assigned at 1903, 1625 and 2235 cm^{-1} in the FTIR spectra (Fig. 2) of NO, NO_2 and N_2O standard gas, respectively was used to prepare the calibration curves.

3. Results and discussion

3.1. NO_x removal performance of reactor type I

Primarily, the feasibility of the EB technique using CNT anode for the treatment of the gas stream containing the high concentration of NO_x was investigated using reactor type I. The systematic preliminary investigations on behaviour of NO in N_2 gas and the stimulated NO in N_2 gas with O_2 and water vapour under EB irradiation were carried out to understand the removal mechanism of NO_x. The NO in N_2 gas stream was stimulated with O_2 and water vapour as it reflects in the real diesel combustion exhausts.

3.1.1. Effect of EB irradiation

The behaviour of the NO gas under EB irradiation was studied in a flow-through configuration using reactor type I (Fig. 1). The effect of the electron beam on dry nitrogen gas stream containing the high initial NO concentration of 1000 ppm was examined at two different dose of 186 and 400 J/L. The gas flow rate was fixed at 6.0 L/h. The reduction in the concentration of NO, NO_x and the formation of NO_2 and N_2O with respect to time is given in Fig. 3a. It was observed that the removal efficiency of NO is attained to 40% at the applied dose of 186 J/L and further, the removal is enhanced to 50% while increasing the EB dose to 400 J/L. In a nonequilibrium technique, the electron mean energy is considerably higher and efficiently utilized on selective decomposition of the