



## Original article

Removal of NO<sub>x</sub> using electron beam process with NaOH spraying

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## ARTICLE INFO

## Article history:

Received 11 March 2021

Received in revised form

17 June 2021

Accepted 20 June 2021

Available online 23 June 2021

## Keywords:

Electron beam

NO<sub>x</sub>

Additive

NaOH

NH<sub>4</sub>OH

## ABSTRACT

Nitrogen oxides (NO<sub>x</sub>; NO and NO<sub>2</sub>) are major air pollutants and can cause harmful effects on the human body. Electron Beam Flue Gas Treatment (EBFGT) is a technology that generates electrons with an energy of 0.5–1 MeV using electron accelerators and effectively processes exhaust gases. In this study, NO<sub>x</sub> was removed using an electron beam accelerator with spraying additives (NaOH and NH<sub>4</sub>OH). NO and NO<sub>2</sub> were 100% and more than 94% removed, respectively, at an electron beam absorbed dose of 20 kGy and an additive concentration of 0.02 M (mol/L). In most cases, NO<sub>x</sub> was removed better with lower initial NO<sub>x</sub> concentrations and higher electron beam absorbed doses. As the irradiation strength (mA) of the electron beam increases, the probability of electron impact on the material accordingly rises, which may lead to increase removal efficiency. The results of the present study show that the continuous electron beam process using additives achieved more effective removal efficiency than either individual process (wet-scrubbing or EB irradiation only).

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## 1. Introduction

Many developed countries have burned fossil fuels to obtain the energy, which generates carbon dioxide (CO<sub>2</sub>), nitrogen oxide (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>) and volatile organic compounds (VOCs). They are considered the main culprits of air pollution [1,2]. These toxic substances directly pollute the atmosphere, the water, and the soil by causing smog, acid rain, and eutrophication of lakes. They are known to be harmful not only to humans and other living organisms but also to the environment where we live [1,3,4]. Economical and efficient treatment of these pollutants is therefore required to foster an eco-friendly energy industry and to keep the air clean [4]. Nitrogen oxides (NO<sub>x</sub>) are major air pollutants and can cause cerebrovascular or respiratory diseases including emphysema, bronchitis, and lung cancer [4,5]. In addition, they are the major causes of acid rain, photochemical smog, and depletion of the ozone layer. They play an important role in the formation of PM<sub>2.5</sub> through homogeneous and heterogeneous photo-chemical reactions in the air [4–7]. Accordingly, the importance of removing NO<sub>x</sub> has emerged, and many efforts have been made to improve treatment methods and develop new technologies.

Commonly used post-combustion treatment techniques include calcium-based flue gas denitrification, selective catalytic reduction (SCR), and selective non-catalytic reduction (SNCR). In the SCR process, reducing agents such as NH<sub>3</sub> and urea are mainly used at reaction temperatures below 500 °C to reduce NO<sub>x</sub> to N<sub>2</sub> and H<sub>2</sub>O, and expensive catalysts are used including vanadium, platinum, and Fe-zeolite [7]. Selective non-catalytic reduction (SNCR), also called thermal denitration, causes decomposition by the injection of ammonia or urea-contained reducing agents into a high-temperature furnace at 900–1000 °C. The produced NH<sub>3</sub> and NO<sub>x</sub> react selectively to synthesize N<sub>2</sub>. However, ammonia slip (<900 °C) occurs or NH<sub>3</sub> is oxidized to NO (>1100 °C) and the removal efficiency of NO<sub>x</sub> is reduced outside the temperature range. Although there are advantages of low investment cost, easy construction, and no need of a catalyst, reaction temperature should be tightly controlled and denitration efficiency is relatively low (50–60%). Currently, most large-scale processes use a combination of SCR and SNCR and have high denitrification efficiency of 80% or higher [7]. Other promising process technologies include non-thermal plasma (NTP), dielectric barrier discharge (DBD), and corona discharge (CD) and pulsed corona discharge (PCD) [8].

Electron beam flue gas treatment (EBFGT) is a technology that generates electrons with an energy of 0.5–1 MeV using electron accelerators and processes exhaust gases. Simultaneous treatment

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of nitrogen oxides (NO<sub>x</sub>) and sulfur oxides (SO<sub>x</sub>) is possible, and it has the advantage of producing useable by-products and not requiring wastewater treatment [1,2,9,10]. In addition, it is considered to be an effective method for treating contaminants because it is possible to process the material by inducing a chemical reaction without a pressure drop at an ambient temperature, without using an expensive catalyst [2,11,12]. However, this process requires a shielding system against radioactive rays, and costs for high power consumption [2].

EBFGT was researched and developed by the Japan Atomic Energy Research Institute (JAERI) and Ebara Co. in the United States in the 1970s. In the 1980s and 1990s, research was actively conducted in many countries, including the United States, Japan, Germany, Poland, and China [13,14]. Thereafter, the technology has been successfully implemented industrially in Electropower Station (EPS) Pomorzany [15,16] in northern Poland, and two thermal power plants in Hangzhou [17] and Chengdu [18], China [1,4,9]. The equipment applied to EPS Pomorzany in Poland can handle a flow rate of up to 270,000 Nm<sup>3</sup>/h with an electron beam energy of up to 1 MW and an absorbed dose of 7–12 kGy (kJ/kg) [16]. Equipment applied in Chengdu, China showed a removal efficiency of 50–70% for NO<sub>x</sub> [18]. At the Institute of Nuclear Chemistry and Technology (INCT) in Warsaw, Poland, a wet scrubber was combined with electron beam technology to treat a flow rate of 5 m<sup>3</sup>/h (800 keV, max. Beam power up to 20 kW). An initial concentration of 1500 ppm NO<sub>x</sub> was 49% removed [4]. When the electron beam (EB) is irradiated to NO<sub>x</sub>, NO<sub>x</sub> reacts with OH radicals obtained from radiolysis of moisture (H<sub>2</sub>O) in exhaust gases, and is oxidized to produce nitric acid (HNO<sub>3</sub>). To improve the removal efficiency and to form a useful by-product, ammonia (NH<sub>3</sub>) or sodium hydroxide (NaOH) is added and neutralized to produce nitrates (NH<sub>4</sub>NO<sub>3</sub>, NaNO<sub>3</sub>) as final byproducts. The resulting by-products can be recovered as a dry powder and used as an agricultural fertilizer [2–4,9,19].

In this study the authors sought to remove NO<sub>x</sub> by using the electron beam process, which is one of the new technologies considered to be more efficient and economical than the conventional treatment processes used for denitrification. In order to improve the removal efficiency of NO<sub>x</sub>, the electron beam irradiation process was combined with chemical spraying using additives (NaOH, NH<sub>4</sub>OH). The parameters are the type of additives, the absorbed dose of electron beam, and the initial concentration of the target gas. Optimal operating conditions for achieving high removal efficiency are found according to changes in the initial concentration of target gases. An additive that can replace the NH<sub>3</sub> gas conventionally used as an additive in the existing EB technology is secured.

## 2. Materials and methods

### 2.1. Additives

In the previous research [20,21], eight different types of additives (H<sub>2</sub>O, NH<sub>4</sub>OH, NaCl, NaOH, Ca(OH)<sub>2</sub>, CaCl<sub>2</sub>, NaClO<sub>2</sub>, and HANa) were employed to compare the removal efficiency while electron beam irradiating in the batch system. In the case of NO<sub>x</sub>, NaOH showed the highest removal efficiency when irradiated with 10 kGy, and Ca(OH)<sub>2</sub> showed the second-highest efficiency. The rest of the additives was not efficient to enhance the removal efficiency of NO<sub>x</sub> in the electron beam process. Based on this, NaOH was selected as an additive in this study as it showed the highest removal efficiency, and NH<sub>4</sub>OH was chosen for the comparison with NH<sub>3</sub>, which is generally used in conventional electron beam processes. NaOH (97%, Sigma-Aldrich) and NH<sub>4</sub>OH (25–30%, Duksan) were diluted with distilled water to the desired concentration. Two

liquid additives, sodium hydroxide (NaOH) and ammonium hydroxide (NH<sub>4</sub>OH), at a concentration of 0.02 M were sprayed at 35 ml/min through a nozzle installed inside the reactor where the electron beam was irradiated.

### 2.2. Hybrid electron beam process

A hybrid electron beam process was designed to enhance the removal efficiency of NO<sub>x</sub> (NO and NO<sub>2</sub>). The irradiation of the electron beam and the spray of liquid additive to NO<sub>x</sub> were simultaneously carried out in the reactor. The removal efficiencies were compared between a single process of electron beam irradiation and a hybrid process of electron beam irradiation combined with additive spraying. The effects of type of additive, initial NO<sub>x</sub> concentration, and absorbed dose of electron beam as parameters of the experiments on the removal of NO<sub>x</sub> were evaluated.

NO and NO<sub>2</sub> gas (2000 ppm, N<sub>2</sub> balance, Hankook Special Gas Co. Ltd, Korea) were prepared using a 47 L gas cylinder. For the removal of NO<sub>x</sub>, this study irradiated electron beams using an electron accelerator, ELV-0.6 M (0.6 MeV, 33 mA, Korea Atomic Energy Research Institute, Korea). The absorbed dose of the electron beam was measured using a solid CTA film (Cellulose Triacetate film, FTR-125, Fuji Photo Film Co., Japan) in this study. The gas concentrations before and after electron beam irradiation were measured using an industrial flue gas analyzer, Testo 340 (Testo Korea Ltd., Korea).

The overall experimental process is schematically illustrated in Fig. 1. The NO<sub>x</sub> gases were introduced into the reactor, and the liquid additives (NaOH and NH<sub>4</sub>OH) were finely sprayed through nozzles installed inside the reactor. They simultaneously reacted during the electron beam irradiation. Thereafter, a gas sample was collected in a Tedlar Bag (PVP film, C-type, 3 L, Top Trading ENG, Korea) to measure the concentration after the treatment of target gases. The removal efficiency was calculated by comparing the reduced gas concentration compared to the initial concentrations of NO and NO<sub>2</sub>, and was expressed as a percentage (%).

This study evaluated the removal efficiencies of NO and NO<sub>2</sub> by setting the initial concentration of the gas sample, the absorbed dose of the electron beam, and the presence or absence of an additive reaction as experimental variables in the continuous electron beam reaction. The removal efficiency according to the initial NO<sub>x</sub> concentration was evaluated by setting the target gas diluted to 100 ppm, 200 ppm, and 500 ppm, respectively, at a gas flow rate of 10 L/min, and irradiating an electron beam with an absorbed dose of 5 kGy. For this, the initial NO<sub>x</sub> concentration was made by diluting using high purity air.

The effects of the absorbed dose of the electron beam on the removal efficiency were also studied after irradiating the diluted gas with absorbed doses of 5 kGy, 10 kGy, and 20 kGy, respectively, at a flow rate of 10 L/min. Table 1 shows the detailed conditions of the experiment.

### 2.3. Measurement of absorbed dose

The concept of absorbed dose is generally used extensively in all areas where the interaction of ionizing radiation with substances is involved [22,23]. Absorbed dose refers to the amount of ionizing radiation energy per unit mass actually absorbed through a medium when the electron beam is irradiated to a specific medium. It is a major factor in determining efficient operating conditions in the electron beam process [24,25]. It is represented by Gy (gray) in the radiochemical field and J/kg in the SI unit [8,24]. The energy of 1 MeV in the SI unit is equal to 0.1602177 × 10<sup>-12</sup> J, and 1 J per second (J/s) corresponds to 1 W [24].

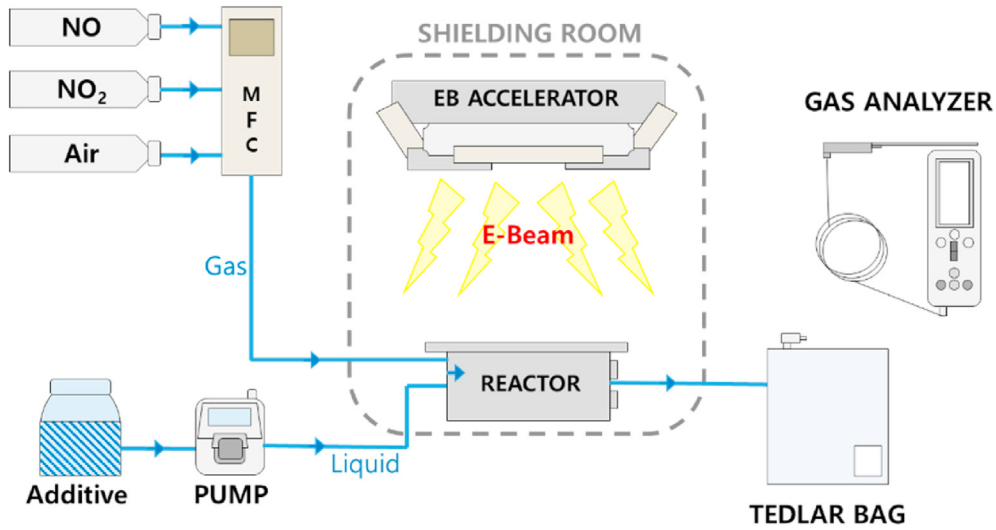


Fig. 1. A schematic diagram of continuous removal process for NO<sub>x</sub> using electron beam and additive treatment system.

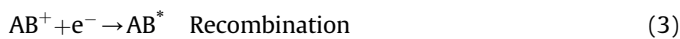
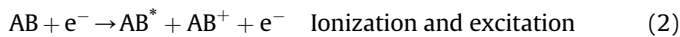
**Table 1**  
Experimental conditions for continuous EB process.

Factor	Experimental conditions
Target gas	NO, NO <sub>2</sub>
Gas flow rate	10 L/min
Initial concentration	100–500 ppm
Absorbed dose	0–20 kGy
Gas analyzer	Testo 340
Additives	NaOH, NH <sub>4</sub> OH
Additives spraying rate	35 ml/min
Gas retention time	15 s

$$\text{Absorbed Dose} = \frac{\text{Energy absorbed per second}}{\text{Product mass per second}} \quad (1)$$

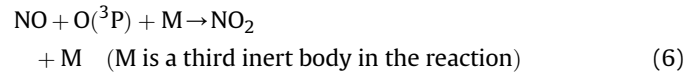
### 3. Results and discussion

When the electron beam, which is an ionizing radiation, is irradiated, the accelerated electrons coulomb interact with atoms or molecules of the target material to generate ions and thermally neutralized electrons. The electron beam generates OH, O, and HO<sub>2</sub> radicals from oxygen and moisture (H<sub>2</sub>O) present in the exhaust gas, and advances the chemical reaction by creating an excited state [1]. At this time, the reaction induced by the irradiation of the electron beam is as follows: Eq. (2)–(5) [26,27].



Unlike photochemical reactions in which one photon causes one reaction, electron beam irradiation can cause numerous responses continuously until all of the energy of high-energy electrons is extinguished.

The removal reaction of NO<sub>x</sub> by an electron beam is as follows:



NO<sub>x</sub> is mainly removed by an oxidation reaction, but reduction reactions by N<sub>2</sub> (via reactions of NO with N, NH<sub>2</sub>) and N<sub>2</sub>O (via reactions of NO<sub>2</sub> with N, NH<sub>2</sub>) also occur [28]. Among them, the most important reactions for the removal of NO<sub>x</sub> are oxidation (see Eq. (13)) by hydroxyl radical (·OH) and reduction (see Eq. (14)) by ammonia (NH<sub>3</sub>) and ammonia radical (·NH) [3].



The reactions described here show the main features of the electron beam process, but the actual removal mechanisms are much more complicated [28].

#### 3.1. Types of additives

NaOH and NH<sub>4</sub>OH were used as additives in this study. The initial concentration of the target gas (NO and NO<sub>2</sub>) was 200 ppm, and the gas flow rate was set to 10 L/min. The concentration of the additive was 0.02 M (mol/L). First, the gas was removed by spraying the additive without irradiation of the electron beam. The results of this experiment are shown in Fig. 2. NO was not removed only by spraying additives. NO<sub>2</sub> was removed at an efficiency of 16.2% when using NaOH and 28.0% when using NH<sub>4</sub>OH. The conventional wet scrubbing works properly when the gas to be removed dissolves

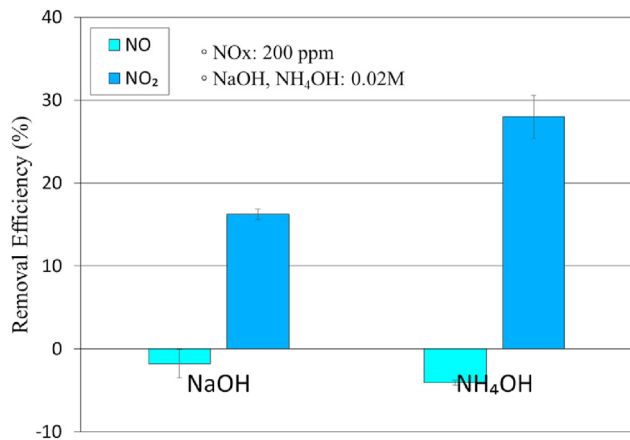


Fig. 2. Removal efficiencies of NO and NO<sub>2</sub> by additive only.

well in water and reacts well with the sorbent of an aqueous solution. NO has a low Henry's law constant (Henry's law constant of NO =  $1.9 \times 10^{-3}$  kmol/m<sup>3</sup>·atm, NO<sub>2</sub> =  $1.0 \times 10^{-2}$  kmol/m<sup>3</sup>·atm (at 25 °C)).

Pourmohammadbagher et al. [29] conducted a study using the existing wet scrubbing type and obtained removal efficiency of 77% for NO, and 88% for NO<sub>2</sub> while SO<sub>2</sub> was fully removed using NaOH (2 wt%). In Hutson's study [30], NaClO<sub>2</sub> (8 mM) and CaCO<sub>3</sub> (10 wt%) were used to remove NO<sub>x</sub> by 50% and SO<sub>2</sub> by 100%. Wang et al. [31] removed 52% of NO and 100% of SO<sub>2</sub> using Fe(II)EDTA (50 mM) and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> (2.5 M).

### 3.2. Initial concentration of target gas

In the existing EBFGT studies, the initial concentration of NO<sub>x</sub> was mainly in the range of 100–240 ppm, but also included a low concentration of 60 ppm and was variously set to high concentrations of 400–420 ppm, 730 ppm, and 820 ppm [1,8,9,18,32–35]. In this study, the initial concentrations of NO<sub>x</sub> were changed to 100, 200, and 500 ppm, and removal efficiencies were measured according to each concentration. As the additive, NaOH and NH<sub>4</sub>OH were used, and the absorbed dose of the electron beam was fixed at 5 kGy. The gas flow rate of the entire process was 10 L/min.

When an electron beam was irradiated and NH<sub>4</sub>OH was simultaneously sprayed, the NO<sub>x</sub> removal efficiency decreased as the initial concentration of NO<sub>x</sub> rose (Fig. 3). At the initial concentration

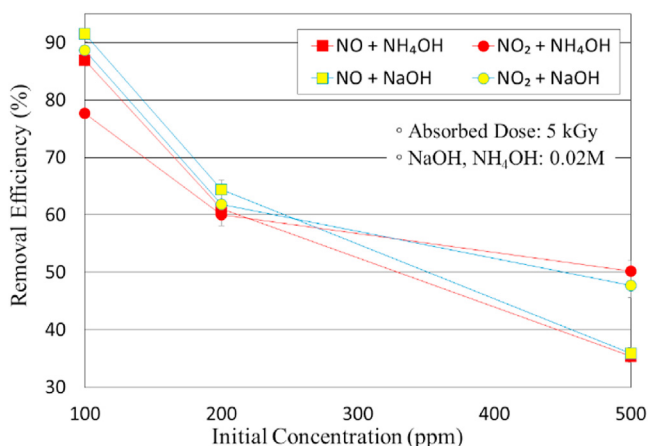


Fig. 3. Removal efficiencies of NO and NO<sub>2</sub> by various initial concentrations at 5 kGy electron beam and 0.02 M additive.

of 100 ppm, NO and NO<sub>2</sub> showed 87% and 78% removal efficiency, respectively. When the initial concentration was increased to 500 ppm, the removal efficiency decreased to 35% for NO, and 50% for NO<sub>2</sub>, respectively. As the initial concentration of the target gas (NO and NO<sub>2</sub>) was increased, the required energy of electron beam increased. And the probability of collision between the target material distributed in the reactor and the emitted electrons decreased. Therefore, as the initial concentration was increased, the removal efficiency decreased. When NaOH was used as an additive, the removal efficiency of NO also decreased as the concentration of the target gas was increased from 100 to 500 ppm (see Fig. 3). As for NO<sub>2</sub>, the efficiency decreased from 89% (100 ppm) to 48% (500 ppm). Table 2 presents the highest NO<sub>x</sub> removal efficiencies according to each initial concentration.

The results of research by Chmielewski et al. [4] revealed that the removal efficiency of NO<sub>x</sub> decreased as the inlet concentration of NO was increased, and that the removal efficiency of NO<sub>x</sub> at 500 ppm was significantly lower with both of these additives compared to the removal efficiency of NO<sub>x</sub> at 100 and 200 ppm. In the study, NO<sub>x</sub> was removed using an electron beam and simulated seawater (3.5% NaCl solution). When the NO inlet was 200 ppm, the NO<sub>x</sub> removal efficiency was about 65% at 10 kGy. As in our study, an increase in the initial concentration of NO (200 ppm–1700 ppm) resulted in a decrease in the NO<sub>x</sub> removal efficiency.

### 3.3. Absorbed dose of electron beam

#### 3.3.1. Removal efficiency of NO<sub>x</sub>

The absorbed dose was changed from a lower dose (5 kGy) to a higher dose (20 kGy) to observe the change in removal efficiency. The zero-dose (at 0 kGy) condition was used for the removal efficiency when only the additive was sprayed into the reactor without irradiation of the electron beam. For comparison with the derived data, the results were also acquired when only the electron beam was irradiated without spraying additives. In Chmielewski [36]'s study on the effect of additives on the removal efficiency of NO<sub>x</sub> in Electron Beam Flue Gas Treatment (EBFGT), the NO<sub>x</sub> removal efficiency was higher when humidity and ammonia were used than when no additives were used for NO<sub>x</sub> with an initial concentration of 500–600 ppm [37]. Due to the oxidation and reduction cycle of NO and NO<sub>2</sub>, it is difficult to obtain an initial NO<sub>x</sub> high removal efficiency of more than 80%.

As the amount of NO<sub>x</sub> removed corresponds to the number of active species produced by electron beam irradiation, higher NO<sub>x</sub> removal efficiencies were achieved at higher absorbed doses [9]. A low concentration of NO could be sufficiently removed even with a low dose electron beam (Fig. 4). As the initial concentration was increased at a specific dose, the NO removal efficiency decreased. This indicates that the concentration of NO that can be processed with the energy of a specific dose of the electron beam is limited. As a result, the NO<sub>x</sub> removal efficiency decreased as the initial concentration increased. Chmielewski et al. [4] also showed the same trend as found in this study. Increasing the absorbed dose and decreasing the initial concentration resulted in an increase in the NO<sub>x</sub> removal efficiency. In addition, the hybrid process (EB + sea water scrubbing) showed higher removal efficiency than when the EB alone was irradiated without scrubbing. In Zhao et al.'s study [38], an increase in the initial concentration decreased the NO<sub>x</sub> removal efficiency. The EB + Wet scrubber (NaClO<sub>2</sub>–NaOH or phosphate buffer-sea water) system achieved significantly higher removal efficiency than EB irradiation alone. The maximum NO<sub>x</sub> removal efficiency achieved in the study of Zhao et al. was 95.3% [38]. The maximum removal efficiency of NO and NO<sub>2</sub> achieved in our study was 100% and 97%, respectively. The approach suggested herein is judged to be more efficient because the electron beam

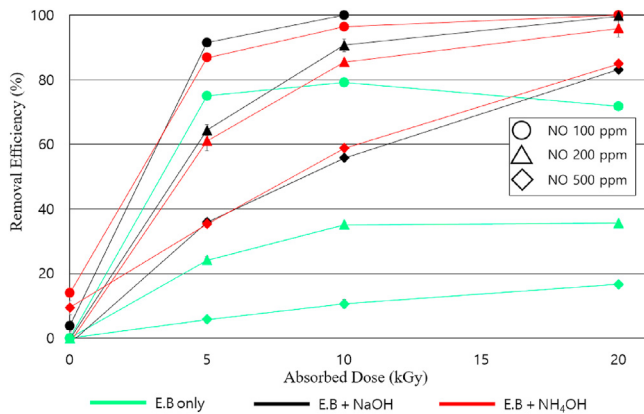


**Table 2**  
Experimental conditions for the highest removal efficiencies of NO<sub>x</sub>.

Gas	Initial concentration of NO <sub>x</sub>								
	100 ppm			200 ppm			500 ppm		
	Additive	A.D. <sup>a</sup> (kGy)	R.E. <sup>b</sup> (%)	Additive	A.D.(kGy)	R.E. (%)	Additive	A.D.(kGy)	R.E. (%)
NO	NaOH	5	100	NaOH	20	100	NH <sub>4</sub> OH	5	100
NO <sub>2</sub>	NH <sub>4</sub> OH	20	94	NaOH	10	95	NaOH	20	97

<sup>a</sup> A.D.: absorbed dose of electron beam.

<sup>b</sup> R.E.: removal efficiency.

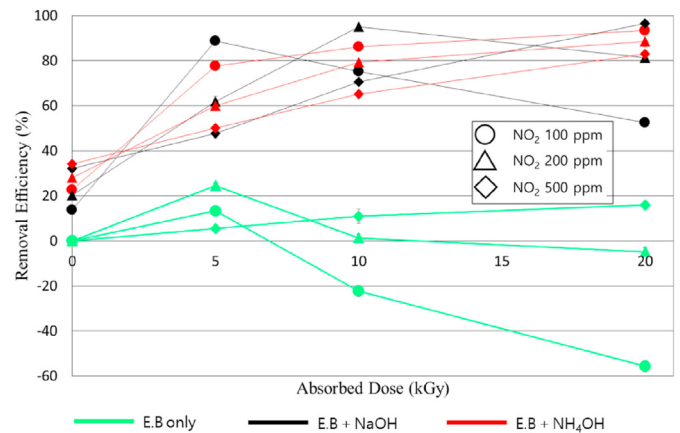


**Fig. 4.** Removal efficiencies of NO by various initial concentrations and changes in absorbed dose (E.B only, E.B with NaOH, E.B with NH<sub>4</sub>OH).

reaction and the additive reaction were performed at the same time inside the reactor in our study.

In this study, the G-value was obtained to consider the experimental results in terms of energy efficiency, not simply the removal efficiency of the target gases (NO<sub>x</sub>) (see Fig. 8 – Fig. 9). Radiation chemical yields were so-called ‘radiation yields’ in the early literature, after then it was defined as a G-value [39]. The G-value refers to the number of radicals (and molecules, or other products) produced or destroyed when the target material absorbs 100 electron volts (eV) of energy in the system [40–43]. This can be expressed as the control efficiency per unit of energy when converted from ionizing radiation to chemical energy. When the material processed by the electron beam has a high molar mass (e.g., a polymer) or a high G-value of the reaction (e.g., a chain reaction), the absorbed dose required for control is relatively low [28]. In most cases, for all target gases, the G-value showed a higher value as the initial NO<sub>x</sub> concentration was increased, and a lower value as the absorbed dose was increased. The decrease in G-value according to the increase in absorbed dose means that the removal efficiency of the gas to be removed has already been saturated in the unit energy and a reduction in the G-value reflects a reduction in energy efficiency [44].

When only an electron beam was irradiated to NO<sub>2</sub> 100 ppm and 200 ppm (‘E.B only’ in Fig. 5), respectively, the removal efficiency showed a maximum value at 5 kGy, and then decreased as the absorbed dose was increased. The degree of this reduction decreased as the initial NO<sub>2</sub> concentration was increased. The amount of NO<sub>2</sub> generated in the high-purity air during electron beam irradiation decreased as the absorbed dose was increased, because a higher initial concentration of NO<sub>2</sub> requires a smaller amount of high-purity air used for dilution. High-purity air, a background gas used for diluting the concentration of the target gas (NO<sub>x</sub>), produced NO<sub>2</sub> when an electron beam was irradiated. In



**Fig. 5.** Removal efficiencies of NO<sub>2</sub> by various initial concentrations and changes in absorbed dose (E.B only, E.B with NaOH, E.B with NH<sub>4</sub>OH).

addition, NO<sub>2</sub> was also generated by an unexpected oxidation reaction caused by electron beam irradiation on the titanium-foil surface used as the irradiation window of the reactor [45]. The amount of NO<sub>2</sub> generated from the high-purity air increased as the absorbed dose increased. The removal efficiency of NO<sub>2</sub> could thereby be reduced as the absorbed dose was increased. Each initial concentration (100, 200 and 500 ppm) of NO<sub>2</sub> was prepared by diluting 2000 ppm NO<sub>2</sub> with high-purity air. Therefore, the lower the initial NO<sub>2</sub> concentration was, the greater the amount of air used for dilution. When NO<sub>2</sub> was removed by only irradiating the electron beam, it was possible to identify a specific point at which the removal efficiency of NO<sub>2</sub> decreased as the absorbed dose increased for each initial concentration (100 and 200 ppm). Although this tendency was confirmed when NaOH was sprayed as an additive, NH<sub>4</sub>OH did not decrease the removal efficiency even when the absorbed dose was increased. NH<sub>4</sub>OH used in this experiment had a slightly lower boiling point of 36 °C. Therefore, when the electron beam was irradiated, NH<sub>4</sub>OH was easily vaporized to NH<sub>3</sub> inside the reactor. It is presumed that NH<sub>3</sub> vaporized (gas-phase) along with NH<sub>4</sub>OH sprayed (liquid phase) further reacts to induce the removal of NO<sub>2</sub>. Performing further studies such as ion analysis of by-products or particle counting could be made clearer.

### 3.3.2. Behavior of total NO<sub>x</sub> during electron beam process

In this study, ‘Total NO<sub>x</sub>’ was defined as ‘NO + NO<sub>2</sub>’. As for NO at 200 ppm, the concentrations of single NO and Total NO<sub>x</sub> all decreased as the absorbed dose was increased up to 20 kGy (see Fig. 6). NO<sub>2</sub>, which was present together with single NO, was generated due to spontaneous oxidation of NO (see Eq. (7)). From Fig. 6, NO<sub>2</sub> did not significantly change compared to the amount of NO removed. Based on these results, it could be seen that the

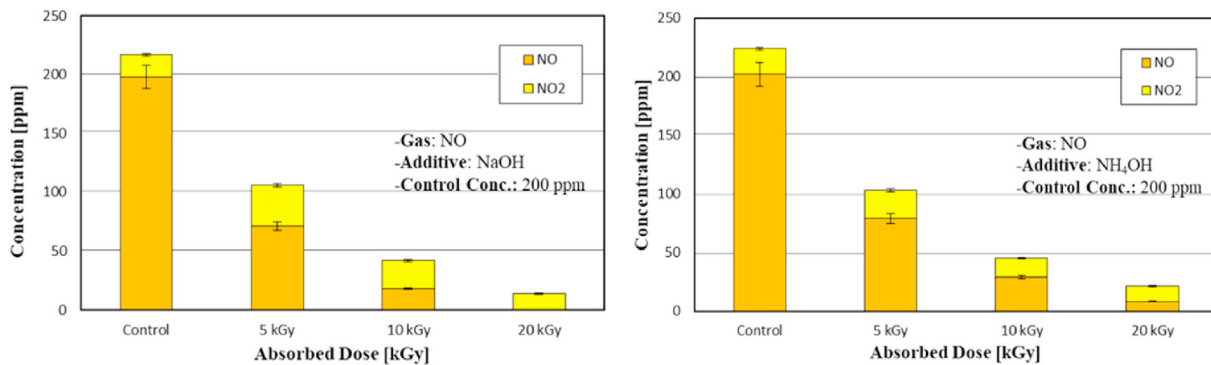


Fig. 6. Behavior of total NO<sub>x</sub> with single NO gas by electron beam with additive.

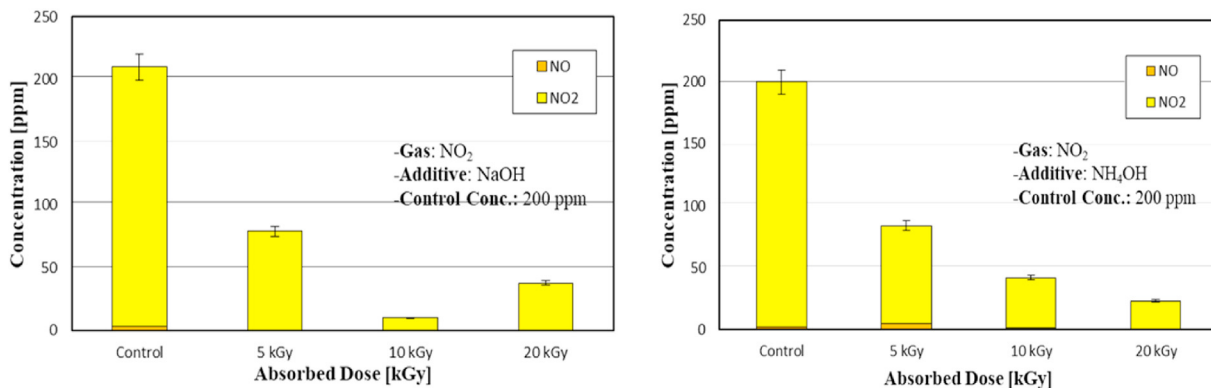


Fig. 7. Behavior of total NO<sub>x</sub> with single NO<sub>2</sub> gas by electron beam with additive.

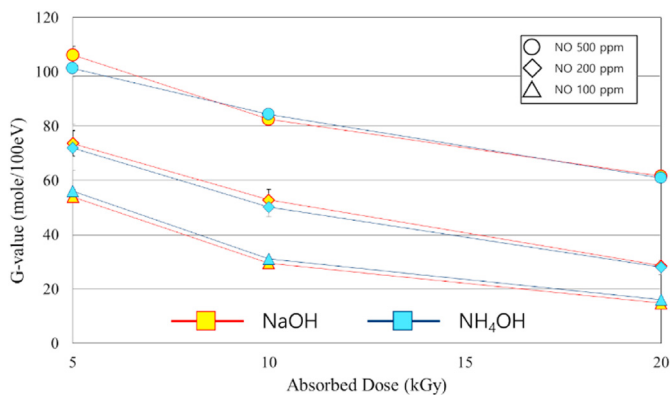


Fig. 8. Changes of G-value during electron beam process of NO by absorbed dose and initial concentration.

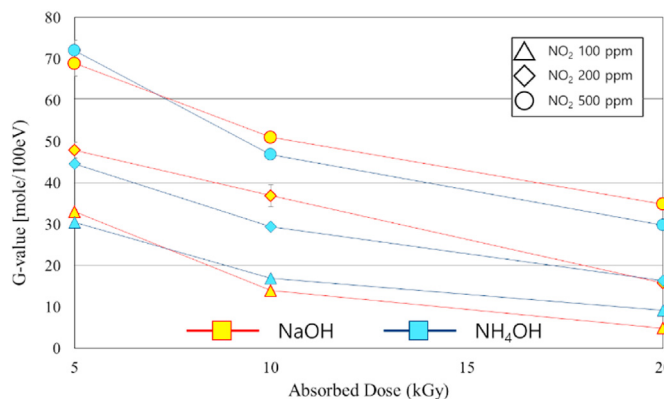


Fig. 9. Changes of G-value during electron beam process of NO<sub>2</sub> by absorbed dose and initial concentration.

reactions delineated in Eq.(6), (8)–(9) occurred preferentially in the NO<sub>x</sub> removal mechanism using the electron beam, and the removal of NO occurred before the removal of NO<sub>2</sub>. In the case of single NO<sub>2</sub>, as NO production theoretically hardly occurred, the behavior of the total NO<sub>x</sub> showed the same tendency as the removal efficiency of NO<sub>2</sub> (see Fig. 7).

#### 4. Conclusion

In this study, a hybrid process incorporating an electron beam and a chemical additive for the effective removal of nitrogen oxides (NO<sub>x</sub>; NO and NO<sub>2</sub>) was suggested. The removal efficiency of NO<sub>x</sub>

was more effective when using a liquid additive (NaOH and NH<sub>4</sub>OH) during electron beam irradiation. When only additives were sprayed without electron beam irradiation, NO was not removed, but NO<sub>2</sub> was removed with an efficiency of 16.2% when using NaOH and 28.0% when using NH<sub>4</sub>OH. In the hybrid electron beam process, the increase in the initial concentration of NO<sub>x</sub> led to a decrease in the removal efficiency. For each target gas, when the initial concentration was increased from 100 ppm to 500 ppm, the removal efficiency decreased by up to 56% for NO and by up to 41% for NO<sub>2</sub>. Finally, with the increase of absorbed dose, NO achieved 100% removal efficiency and more than 94% of NO<sub>2</sub> was removed using this hybrid process.

In most cases, NO<sub>x</sub> (NO and NO<sub>2</sub>) was better removed with lower initial concentrations of NO<sub>x</sub> and higher absorbed doses of the electron beam. As the irradiation strength of the electron beam was increased, the NO<sub>x</sub> removal efficiency increased. However, from the perspective of the G-value, the increase of the absorbed dose of the electron beam resulted in lower energy efficiency even though the decomposition efficiency was increased.

It is shown that the continuous electron beam process with additives achieved more effective removal of NO<sub>x</sub> than either individual process (wet-scrubbing or electron beam irradiation only).

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Acknowledgements

This research was supported by the Nuclear R&D program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science and ICT (NRF-2018M2A2B3A06071698).

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