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The optimization of SF_6 decomposition process using an electron beam

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ABSTRACT

This study was carried out to apply the decomposition process of SF_6 using an electron beam to the actual field by optimizing the process. To do this, we selected Inconel 625, which has excellent corrosion resistance and durability, as an optimal reactor film to prevent damage from by-products (Hydrogen fluoride). Among additive gases, H_2 gas was used to enhance the removal efficiency (RE) of SF_6 because it has the greatest contribution to the RE. Additionally, the initial concentrations of SF_6, the currents, and the flow rates, considered the main optimal factors, were 1%, 15 mA, and 10 L/min, respectively. Based on the above experimental results, the long-term operating test was performed for 3.5 h at 10 L/min and for 8 h at 5 L/min. The removal efficiency of SF_6 was constantly maintained at approximately 80% at a flow rate 10 L/min. On the other hand, when the flow rate was adjusted to 5 L/min, the RE continuously remained at a level of about 90%.

1. Introduction

Sulfur hexafluoride (SF_6), a non-CO_2 warming material, has the highest GWP (Global Warming Potential: 23,900) of all the greenhouse gases that contribute to the global greenhouse effect. It is a thermally and chemically stable gas that disappears slowly from the atmosphere, with a long lifetime of 3200 years. Emissions of SF_6 are smaller than those of other global warming gases, but the damage that this gas is liable to be large because it constantly accumulates in the atmosphere (Chang and Lee, 2004; EPA, 2015; Ryu et al., 2012a; Tsai et al., 2007). The major emission sources of SF_6 in the world are electrical transmission and distribution, magnesium production and processing, and semiconductor manufacture (EPA, 2018). The emission sources of SF_6 in Korea are composed of 62.5% from LCD production, 32.2% from production, use and disposal of heavy electric machinery, 5.2% from semiconductor production, and 0.4% from magnesium production (GIR, 2017). Atmospheric emissions of SF_6 in Republic of Korea have risen approximately threefold from 2.9 CO_2 eq. in 2000 to 8.2 CO_2 eq. in 2015 (GIR, 2016, 2017). Because SF_6 is a permanent greenhouse gas once released, complete removal of SF_6 is required (KEITI, 2010).

Technology to decompose SF_6 chiefly consists of non-destructive and destructive emission controls, both of which have been studied (Lee et al., 2009, 2011). Especially, methods of destructive emission control such as plasma decomposition, pressure swing adsorption, arc plasma process and high ionization energy (electron beam technology) have been developed to control SF_6 more efficiently (Kim et al., 2013a, 2013b; Lee et al., 2009, 2011; Park et al., 2013; Son et al., 2016). It is reported that electron beam technology is able to treat target materials quickly and operate economically at industrial facilities because this process can operate at lower temperatures than can be used with other technologies such as combustion or the plasma process (Hirota et al., 2004; Park et al., 2005).

In previous studies, Kim et al. (2013a, 2013b) and Ryu et al. (2012b) reported that the destruction and removal efficiency (DRE) of SF_6 (less than 1000 ppm) when using an electron beam increased with the addition of materials such as H_2O and H_2. Son et al. (2016) showed that the DRE of SF_6 with a high concentration (2–10%) is affected by absorbed doses and retention times. Also, the DRE of SF_6 increased up to approximately 20% when H_2 (SF_6/H_2 ratio of 1:4) was added to the reactor. However, for all of these decomposition processes using electron beam irradiation, by-products such as hydrogen fluoride (HF) are formed when H_2 gas is used as an additive. Ryu et al. (2015) reported that 4803 ppm of HF was generated when 3000 ppm of H_2 was added to 1000 ppm of SF_6 decomposition process using an electron beam. This causes corrosion and damage of the Ti foil, which is mainly used as transmission window for the reactor. In order to overcome this problem, materials such as Monel and Inconel, which have excellent corrosion resistance, have been used as transmission windows and are

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installed on the upper part of the reactor. Both Monel and Inconel are nickel-based alloys containing Cu, Cr, Mo, etc.; these materials were developed to improve the corrosion and heat resistance of metals (Davis, 2000; Shankar et al., 2001). To investigate whether these materials are suitable for use in the transmission window of a reactor, the absorbed doses of materials were evaluated to determine the energy transmittance. Also, durability tests of the materials were conducted for up to 12 h under the maximum irradiation condition.

In the previously performed decomposition studies, which attempted to use electron beam irradiation to control SF_6, there was a limit in terms of field application because the main purposes of the studies were to determine the factors to consider for enhancing of the removal efficiency of SF_6. Also, those studies did not observe that the removal efficiency of SF_6 remained constant (Kim et al., 2013a, 2013b; Ryu et al., 2012a; Son et al., 2016). Therefore, factors for optimizing the decomposition process of SF_6 were identified by considering the possibility of field application. Furthermore, for application to the field, a long-term operating test for a maximum of 8 h was conducted to confirm whether the decomposition rate was stable. To do this currents with various additive gases (mA: H_2, N_2, Ar, He, Air, O_2,), initial concentrations (%), flow rates (L/min), and currents with 1% SF_6 (mA) were the main factors used to find the optimal conditions.

2. Experimental methods

2.1. Experimental devices

A mobile EB accelerator (0.6 MeV, 33 mA, Korea Atomic Energy Research Institute, Korea) was used for this study. The reactor, made of stainless steel (SUS-316L, L: 680 mm, W: 190 mm, H: 150 mm and V: 19 L), is of rectangular duct-type. To withstand the internal pressure and transmit an electron beam, a metallic thin film with a thickness of 0.05 mm (Titanium-grade 2, Monel-400, and Inconel-625) is installed on the upper part of the reactor; this film is designed so that it can be replaced. To prevent temperature rise and maintain a constant temperature inside the reactor, cooled water is circulated in a water jacket.

All systems were continuous flow systems; this was decided on to allow us to investigate the applicability of the practical treatment process. In this system, we used mass flow controllers (MPR-3000, MKP, Korea) to control the flow rate, two mixing chambers (SUS-304, 2 L) to mix SF_6 with the additive gases and a PTFE tube (Poly Tetra Fluoro Ethylene, ID 6.35 mm) to connect all systems. The flow rate was regulated by MFC and examined by a flow meter (Bios defender 510 H, Dry Cal Masa Labs, USA) to determine the accuracy of the flow rate. Standard gases such as 10% SF_6 (Ar balance, RIGAS, Korea), H_2 (99.999%, KOREA NOBLE GAS, Korea), and He (99.999%, DEOKYANG, Korea) were used to adjust the initial concentration.

2.2. Experimental conditions

The experimental conditions such as the currents with the additive gases (mA: H_2, N_2, Ar, He, Air, O_2,), initial concentrations of SF_6 (%), flow rates (L/min), and currents with 1% SF_6 (mA) were investigated; results are presented in Table 1.

Table 1
The experimental conditions.

Factors	Experimental condition
Current with background gases[a] (mA)	0, 1, 5, 15, 20, 30
Initial SF_6 concentration (%)	1, 2, 4, 8, 20
Current with 1% SF_6 (mA)	0, 2.5, 5, 6, 8, 10, 12, 15
Flow rate (L/min)	2.5, 5, 7.5, 10

[a] Additive gas: H_2, N_2, O_2, He, Ar, Air.

2.3. Sampling and analysis

All samples were obtained using a sample bag (Tedlar bag PVF C-type, 1 L and 3 L, Top Trading E&G Co., Korea); samples were taken from the sampling ports, which are at locations before and after the reactor. Samples collected at the location before the reactor were used to measure the inlet concentration of SF_6. Samples taken from the sampling port after the reactor were used to determine the outlet concentration of SF_6 after this gas had passed through the irradiated reactor. HF was analyzed by gas detector tube (NO.17, Gastec, Japan) and diluted 100–2500 times as needed.

A cellulose triacetate (CTA) film dosimeter (FTR = 125, Fuji, Japan), which is mainly used because it is easy to handle and store (Choi et al., 2013; Ryu et al., 2012b), and a UV/VIS spectrophotometer (UVIKONxs, SECOMAN, France) were used to measure the absorbed dose in the reactor. The absorbed dose is represented by the formula below.

$$AD_{film}[kGy] = \frac{\Delta OD}{k - value} \quad \left(k - value : 0.0063\right)$$

△OD is the absorbed does difference of the CTA film before and after irradiation. The analysis conditions of the CTA firm were 280 nm of wavelength range after 90 min of irradiation. At that time, the irradiation conditions were 0.6 MeV of power, 1 mA of current, and 10 s of irradiation time.

A gas chromatograph-mass spectrometer (GCMS-QP2000 Ultra, Shimadzu, Japan), equipped with a GC-GASPRO (90 m (L) *0.32 mm (I.D.), J&W Scientific, USA), was used to analyze the concentration of SF_6.

2.4. Reactor film

The films used in this study consisted of titanium, Monel, and Inconel. Generally, experiments using electron beam irradiation to remove air pollutants utilized thin films made of titanium as a transmission window. In order to improve the durability of the reactor film, three kinds of materials including titanium (Grade-2), Monel (Monel-400), and Inconel (Inconel-625), which are well known for having good corrosion resistance, were tested for up to 12 h under the maximum irradiation condition (approximately 23,000 kGy).

2.5. Long-term operating test

In order to confirm the probability of success in the actual field, a continuous performance test was carried out using an electron beam. This test verified the stability of the removal efficiency (RE) of SF_6 when continuous test operations were performed for 3.5 and 8 h using the optimal conditions (1% SF_6, 15 mA, 5 and 10 L/min) as determined above.

The removal efficiency of SF_6 is represented by the formula below,

$$\mathrm{RE}(\%) = \frac{Inlet\ concentraion\ of\ SF_6 - Outlet\ concentration\ of\ SF_6}{Inlet\ concentraion\ of\ SF_6} \times 100$$

3. Results and discussion

3.1. Improvement of reactor film

The physical properties and the corrosion factors (against hydrogen fluoride) of the materials used in this experiment are shown in Table 2. The newly selected Ni-based alloys, Monel 400 and Inconel 625, are superior to Ti alloy in terms of physical properties and corrosion resistance against hydrogen fluoride (Special metals, 2005, 2006; Welsch et al., 1993). Prior to the durability test, the absorbed doses, used to evaluate the electron beam irradiation efficiency, are calculated based on the measuring of the absorbance of the CTA film; these data are