

# Evaluation of a Prototype SF<sub>6</sub> Purification System for Commercialization

Hai-Kyung Seo, Jeong Eun Lee, Kwang Sin Kim, Kyeongsook Kim

## Abstract

Korea Electric Power Corporation (KEPCO) uses large amount of SF<sub>6</sub>, one of the potent greenhouse gases, in electric equipment for electrical insulation. KEPCO is developing SF<sub>6</sub> recovery and purification technology to minimize the release of SF<sub>6</sub> into the environment, to secure certified emission reduction, and to save purchase cost of new SF<sub>6</sub> by reusing the refined SF<sub>6</sub>. A prototype SF<sub>6</sub> purification system using cryogenic solidification technology has been built in demonstration scale. To evaluate the feasibility of the commercialization, the system has been operated to purify large amount of used SF<sub>6</sub> in a long-term operation and the performance has been economically evaluated. The system was stable enough for commercial operation such that it was able to purify 5.4 tons of used SF<sub>6</sub> from power transmission equipment in 2-month operation. Over 99% of the SF<sub>6</sub> was recovered from the used gas and the purity of the purified gas was over 99.7 vol%. The operation cost, which is the cost of refrigerant (liquid nitrogen), electricity and labor, per kilogram of purified SF<sub>6</sub> was 6,526 KRW. Considering the price of new SF<sub>6</sub> in Korea is about 15,000 KRW per kilogram this year, about 56% of the purchase cost can be saved.

*Keywords: SF<sub>6</sub>, SF<sub>6</sub> Recovery, SF<sub>6</sub> Purification, Cryogenic Solidification, Liquid Nitrogen, Demonstration Scale*

## I. Introduction

KEPCO uses SF<sub>6</sub> in electrical equipment such as 22.9 kV gas insulated switches (GIS) and main bus. The lifetime of electrical equipment using SF<sub>6</sub> is about 22 years and they are regularly replaced [1]-[5]. To minimize the emission of SF<sub>6</sub> into the atmosphere and save purchasing cost, KEPCO is developing SF<sub>6</sub> recovery/separation/purification technology [5][6]. There exist several SF<sub>6</sub> purification technologies in commercial use [7][9]. One of the technologies is liquefaction of SF<sub>6</sub>. It liquefies SF<sub>6</sub> and separates SF<sub>6</sub> from gaseous impurities. The physical properties of SF<sub>6</sub> is shown in TABLE 1. However, due to its relatively higher operating temperature than cryogenic solidification, the emission of SF<sub>6</sub> with impurities is larger [6][10]. A membrane separation technology is good for purifying low concentration SF<sub>6</sub> gases but not suitable for high concentration SF<sub>6</sub> gases from electrical equipment. There is a report that 0.1% SF<sub>6</sub> from electronic industry was concentrated to 1.4% with recovery rate of over 96% [11]-[15]. Another technology is SF<sub>6</sub> hydrate technology which forms solid SF<sub>6</sub> hydrates [16]-[19]. It can operate under mild conditions (near room temperature and ambient pressure) and use less energy to operate, but is limited to purify SF<sub>6</sub> up to relatively low concentration of 70-80%. The most suitable technology to purify high concentration SF<sub>6</sub> gases from electrical equipment seems to be cryogenic solidification technology even though it uses more energy than other technologies due to cryogenic operation. It utilizes relatively high freezing point (melting point) of SF<sub>6</sub> as shown in TABLE 1. By cooling SF<sub>6</sub> below -64°C, SF<sub>6</sub> solidifies on the surface of chiller and can be separated from impurity gases such as nitrogen

and oxygen which are still in gas phase [10]. This technology can purify SF<sub>6</sub> to very high purity of over 99.9% and it is easy to scale up to commercial scale. KEPCO has built a prototype SF<sub>6</sub> purification system using cryogenic solidification technology in demonstration scale [6]-[10]. It used liquid nitrogen as refrigerant.

In June of 2018, a construction branch of KEPCO requested us to process 5.4 tons of used SF<sub>6</sub> gases recovered from a 345 kV switchyard replacement project of a substation. It was a good opportunity to evaluate the prototype system for the feasibility of commercialization through processing large amount of SF<sub>6</sub> in long term operation. In this paper, the modification of the prototype system for processing large amount of the SF<sub>6</sub> at a time, the operation of the system to process 5.4 tons of used SF<sub>6</sub> from power transmission equipment for 2 months from July to September in this year, and the evaluation of the operation results are presented.

## II. Experimental

### A. Materials for purification

Used SF<sub>6</sub> gases subject to purification were supplied from a 345 kV switchyard replacement project of a substation. Fig. 1 shows the project site (a), a main bus containing used SF<sub>6</sub> gas (b), and a gas recovering system from the equipment (c). The 30-year old SF<sub>6</sub> gases were supplied in 47-L and 600-L cylinders: 5,150 kg in 10 cylinders of 600-L capacity and 327 kg in 6 cylinders of 47-L capacity - about 5.4 tons in total.

## Article Information

Manuscript Received July 26, 2019, Accepted November 13, 2019, Published online March 30, 2020

H. Seo is with KEPCO Research Institute, Korea Electric Power Corporation, 105 Munji-ro Yuseong-gu, Daejeon 34056, Republic of Korea.

J. E. Lee, K. S. Kim and K. Kim was with KEPCO Research Institute, Korea Electric Power Corporation, 105 Munji-ro Yuseong-gu, Daejeon 34056, Republic of Korea.

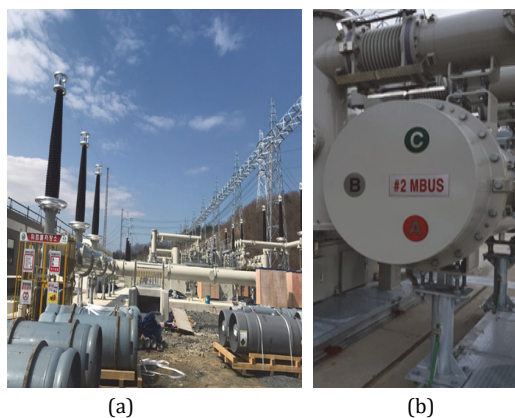
Correspondence Author: Kyeongsook Kim (kskim9451@gmail.com)



This paper is an open access article licensed under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International Public License.

To view a copy of this license, visit <http://creativecommons.org/licenses/by-nc-nd/4.0>

This paper, color print of one or more figures in this paper, and/or supplementary information are available at <http://journal.kepco.co.kr>.



(a) (b)



(c)

Fig. 1. (a) Replacement site. (b) Main bus. (c) Recovering system with a recovery system.

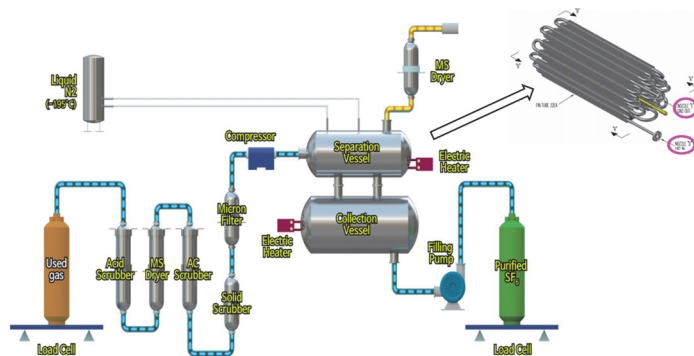


Fig. 2. Schematic diagram of purification system and the shape of chiller.

**B. Analysis procedure**

On-site analysis was performed with a portable SF<sub>6</sub> analyzer. It can also analyze moisture, SO<sub>2</sub>, and SF<sub>6</sub>. The analyzer was connected to the cylinder directly with a pressure regulator. Off-site samples were sampled in Tedlar bags or 80-L SF<sub>6</sub> sampling bags. In off-site analysis procedure, SF<sub>6</sub> was analyzed with GC-TCD first. Then the gas was analyzed with GC-DID. However, GC-DID analysis was not performed when concentrations of O<sub>2</sub> and N<sub>2</sub> were over 1,000 ppm to prevent from damaging DID sensor, which is for microanalysis. The analysis up to this point was ‘the simple analysis’. The following analysis procedure after the simple analysis was ‘the precise analysis’. Substances which were hard to be analyzed with GC-DID or there were not in standard samples were analyzed with

**TABLE 1**  
Physical Properties of SF<sub>6</sub> Gas and Major Impurities

	SF <sub>6</sub>	CF <sub>4</sub>	N <sub>2</sub>	O <sub>2</sub>
Boiling point (°C)	-50.8	-128	-196	-183
Melting point (°C)	-64	-184	-210	-219

**TABLE 2**  
Major Specification of the Prototype Purification System

	Separation temperature	Purification capacity	SF <sub>6</sub> assay	Storage capacity	Operation pressure
Specification	-60°C	80 kg/batch	99.9 vol%	0.27 m <sup>3</sup>	20 bar

**TABLE 3**  
Analysis of Used SF<sub>6</sub> Before Purification

Gas Bombay No	Portable SF <sub>6</sub> Analyzer				Simple Analysis (GC-TCD)		
	SF <sub>6</sub> (vol%)	H <sub>2</sub> O (ppm)	SO <sub>2</sub> (ppm)	HF (ppm)	SF <sub>6</sub> (vol%)	O <sub>2</sub> (vol%)	N <sub>2</sub> (vol%)
1	99.29	5.4	0	0	91.24	1.92	6.84
2	95.48	11.5	0	0	95.72	1.10	3.18
3	98.04	7.8	0	0	94.78	1.18	4.04
4	98.99	4.3	0	0	95.43	1.01	3.55
5	99.15	12.6	0	0	96.09	0.90	3.01

GC-MSD and FT-IR equipped with 10 m cell for very low concentration impurities. The FT-IR was very effective and had very high sensitivity since it used liquid nitrogen in the analysis. The moisture analyzer used diode laser adsorption spectroscopy. To analyze ppm level moisture, sufficient purging was required to stabilize the peak. The analyzers used were: GC-TCD (GC-Thermal Conductivity Detector, Clarus 680, Perkin-Elmer), GC-DID (Gas Chromatograph-Discharge Ionization Detector, Novachrom 1000, AGC), GC-MSD (Gas Chromatograph-Mass Selective Detector, Clarus SQ 8T, Perkin-Elmer), FT-IR (Fourier Transform-Infrared Spectrometer, MIDAC I1801, MIDAC Corp.), and Moisture Analyzer (DF-745SGMAX, SERVOMEX). A portable analyzer (SF<sub>6</sub>-6100 pump back analyzer, Rapidox) was used for on-site analysis of SF<sub>6</sub> and moisture [20]-[27].

**C. The purification system**

The prototype cryogenic SF<sub>6</sub> purification system was comprised of three subsystems: a pre-cleaning, a purification / separation, and a collection/refilling system (Fig. 2) [6][10]. The system was cooled with liquid nitrogen. The pre-cleaning system retrieved used SF<sub>6</sub> gases from supply vessels. And in this system, impurities such as moisture, solid particles, acidic gases, and others which could not be separated in the following cryogenic purification / separation system were removed with scrubbers and filters. A compressor and a vacuum pump were used to retrieve SF<sub>6</sub> from 47-L cylinders. A 47-L cylinder can store 40 to 50 kg of SF<sub>6</sub>. Since the system was designed to process 80 to 100 kg of SF<sub>6</sub> in a batch as shown in TABLE 2, two cylinders were evacuated at a time. Each cylinder was evacuated to -0.3 bar<sub>g</sub>.

The purification / separation system cooled the gas below the freezing point of SF<sub>6</sub> with liquid nitrogen in the purification / separation vessel. The tube coil of the chiller in the purification / separation vessel, on whose surface SF<sub>6</sub> froze is shown on the right side of Fig. 2. At the operation temperature of -60°C, the major impurities N<sub>2</sub> and O<sub>2</sub> still remained in gaseous state and the gas containing impurities was vented to the atmosphere. It was important to remove impurities such as moisture with higher

freezing point than SF<sub>6</sub> in the previous pre-cleaning system to prevent blocking the pipeline in the purification / separation system.

After venting gas with high impurity contents, SF<sub>6</sub> was melted into liquid and transferred to the collection vessel of the collection / refilling system located below the purification / separation vessel by gravity. The collection / refilling system stored purified SF<sub>6</sub> as liquid and refilled purified SF<sub>6</sub> into the cylinders for transfer. The major specification of the system is summarized in TABLE 2.

This system was designed to process 80 kg of SF<sub>6</sub> in two 47-L cylinders (40 kg per cylinder) in two hours. However, some modifications of the system and the operation procedure were required to process gases stored in 600-L cylinders. The modification is described in the next section.

### III. Results and Discussion

#### A. Supply of used SF<sub>6</sub>

Since the purification system was designed to retrieve SF<sub>6</sub> from 47-L cylinders, modifications of the system and operation procedure were required when SF<sub>6</sub> gases were supplied from 600-L cylinders. Residual amount of SF<sub>6</sub> from 600-L cylinders were retrieved to the recovery system, and SF<sub>6</sub> stored in the recovery system was processed when enough SF<sub>6</sub> were collected in the recovery system.

##### 1) Retrieval from 47-L cylinders

Since the system was designed to process 47-L cylinders, no modification was required in the operation. The cylinder was turned upside down so that liquid SF<sub>6</sub> was retrieved first. This operation saved retrieval time and energy. If gaseous SF<sub>6</sub> was retrieved first, liquid SF<sub>6</sub> had to be vaporized and this required time and heat for vaporization. The operation is shown in Fig. 3(a). At first, the pressure in the cylinder was higher than that of the system which was low due to pre-cooling, SF<sub>6</sub> was retrieved spontaneously until the pressures were equalized. This stage was defined as 'liquid retrieval' stage. When the pressure difference decreased to 6 bar<sub>g</sub>, a compressor and a vacuum pump were operated to retrieve the remaining SF<sub>6</sub> until the pressure went down to -0.3 bar<sub>g</sub>. This stage was defined as 'gas retrieval' stage. This operation was repeated twice before purification / separation operation started.

##### 2) Retrieval from 600-L cylinders

Since the system was not designed to process these cylinders, modification was required in the system and the operation. The cylinders can store up to 600 to 700 kg of SF<sub>6</sub>. Considering the capacity of the system, the batch size was decided to be 80 kg. For that purpose, a platform scale was used to measure the weight of the cylinder. And a longer connecting hose was required. When the cylinder weight decreased by 80 kg, the valve was closed. The 'liquid retrieval' stage ended when the pressure difference between the connecting hose and the system reached 6 bar<sub>g</sub>. And SF<sub>6</sub> in the connecting hose was retrieved until -0.3 bar<sub>g</sub> in the 'gas retrieval' stage. The cylinder was placed such that liquid SF<sub>6</sub> could be retrieved first. Due to their size and weight, 600-L cylinders are used in lying down position unlike 47-L cylinders. In this position, the cylinder is designed such that gaseous SF<sub>6</sub> can come out first for the analysis. After that, by rolling the cylinder 180°, we made liquid SF<sub>6</sub> come out to process. The operation is shown in Fig. 3(b).



(a)

(b)



(c)

Fig. 3. Retrieving SF<sub>6</sub> (a) from 47-L cylinder, (b) from 600-L cylinder, and (c) from the recovery system.

##### 3) Retrieval from SF<sub>6</sub> recovery system

KEPCO uses SF<sub>6</sub> recovery systems to recover SF<sub>6</sub> from electrical equipment such as 22.9 kV GIS and main bus. The recovery system is comprised of a compressor, a vacuum pump, a storage vessel, a cooler, a heater, and control system. The operation pressure is 29.4 bar and the operation temperature is -20~40°C. This recovery system was also used in the SF<sub>6</sub> purification operation. It was better to transfer residual SF<sub>6</sub> (less than 80 kg) remaining in the 600-L cylinders to the recovery system than to retrieve to the purification system since evacuation of 600-L cylinder took a long time. The operation time could be saved by transferring residual SF<sub>6</sub> to the recovery system while another 600-L cylinder was connected to the purification system for retrieval. The operation procedure was similar to that of 600-L cylinder. However, since the weight of the SF<sub>6</sub> retrieved from the recovery system could not be measured, the mass flow of SF<sub>6</sub> was measured with a mass flowmeter (Coriolis type). Since SF<sub>6</sub> came out from the recovery system as gas, the retrieval from the recovery system took longer than from cylinders. The operation is shown in Fig. 3(c).

#### B. Purification performance

The on-site and the simple analyses were performed prior to processing used SF<sub>6</sub> gases. As shown in TABLE 3, all samples from the cylinders had SF<sub>6</sub> purity over 90%, while SF<sub>6</sub> recovered from a substation had been purified stably over 30 years. Even though values of on-site analysis were higher than those of the simple analysis, they were less reliable than those from GC-TCD. The on-site analysis was to check the condition of the supplied gas, not for quantitative analysis [20].

The result of the simple analysis of purified SF<sub>6</sub> is shown in TABLE 4. TABLE 5 shows a sample of the precise analysis. All of

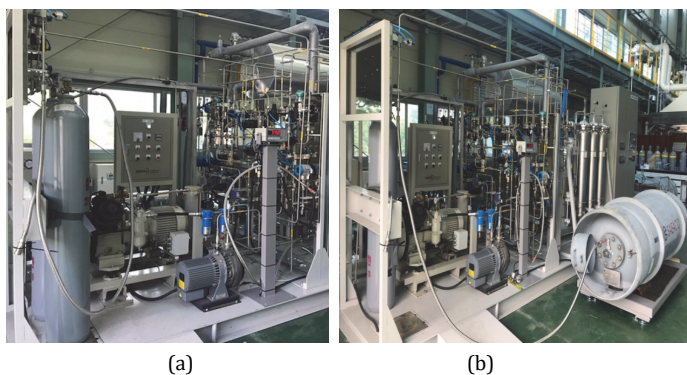


Fig. 4. (a) 47-L cylinder refilling. (b) 600-L cylinder refilling.

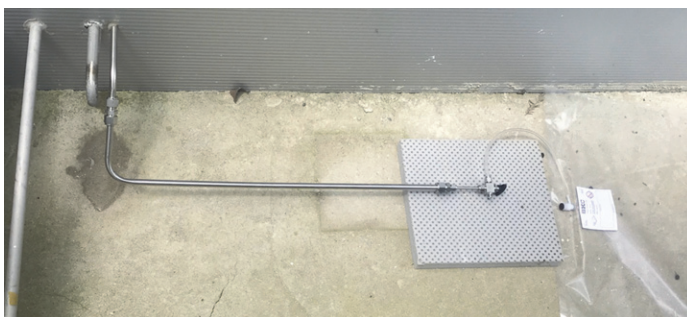


Fig. 5. Sampling the vented gas.

the purified SF<sub>6</sub> satisfied the SF<sub>6</sub> reuse guide line of KEPCO (99.7 vol%). Other than major impurities O<sub>2</sub> and N<sub>2</sub>, minute amounts of CO<sub>2</sub>, CF<sub>4</sub>, and H<sub>2</sub>O, were detected in the precise analysis.

### C. Refilling of the purified SF<sub>6</sub>

Purified SF<sub>6</sub> gases were transferred back to the original cylinders. For 47-L cylinders, the scale built into the purification system was used to transfer SF<sub>6</sub> to a cylinder. The refilling operation was stopped automatically by the system at the pre-set value, which was 40 kg in this operation.

For 600-L cylinders, the new platform scale was used to transfer SF<sub>6</sub> to a cylinder. In this case, the refilling operation was stopped manually when the transfer reached desired value, which was 300 kg in this operation, since the new platform scale could not be connected to the control program of the purification system. Fig. 4 shows the refilling operation. 5,188.6 kg of purified SF<sub>6</sub> was refilled and returned.

### D. Discharge of waste gas

The separation temperature was set to near the freezing point of SF<sub>6</sub> at 1 bar. Thus, the vapor pressure of SF<sub>6</sub> in the purification / separation vessel was near 1 bar during the separation stage. Considering the surface temperature of the chiller was much lower (near -100°C), actual vapor pressure in the vessel was expected to be lower than the equilibrium pressure. If the pressure of the vessel was lower than 0 bar<sub>g</sub> after the separation, the outside gases could be introduced during venting. Thus, the remaining gas after the separation was vented until the pressure decreased to 0.3 bar<sub>g</sub>. The margin of 0.3 bar was to minimize the emission of SF<sub>6</sub> into

TABLE 4  
The Simple Analysis of Purified SF<sub>6</sub>

Cylinder No	(GC-DID) & (GC-TCD)		
	SF <sub>6</sub> (vol%)	O <sub>2</sub> (ppm)	N <sub>2</sub> (ppm)
#1	99.99	13.30	18.23
#2	99.99	36.87	62.59
#3	99.99	21.61	27.80
#4	99.99	33.08	55.72
#5	99.99	18.13	28.66

TABLE 5

A Sample of the Precise Analysis of Purified SF<sub>6</sub> (Cylinder #5)

Component	Concentration (μmol/mol)	Analyzer
H <sub>2</sub>	N.D	GC-DID
O <sub>2</sub>	22.10	GC-DID
N <sub>2</sub>	32.95	GC-DID
CH <sub>4</sub>	N.D	GC-DID
CO	N.D	GC-DID
CO <sub>2</sub>	3.60	GC-DID
CF <sub>4</sub>	6.67	GC-DID
C <sub>2</sub> F <sub>6</sub>	N.D	FT-IR
H <sub>2</sub> O	7.8	Moisture analyzer
HF	N.D (Detection limit <0.1)	FT-IR
SO <sub>2</sub>	N.D (Detection limit <0.2)	FT-IR
SOF <sub>2</sub>	N.D (Detection limit <0.1)	FT-IR
SO <sub>2</sub> F <sub>2</sub>	N.D (Detection limit <0.1)	FT-IR
SOF <sub>4</sub>	N.D (Detection limit <0.1)	FT-IR
SF <sub>6</sub>	99.99% (mole fraction)	

\* Assay is by difference, after impurity content is determined.

the atmosphere and prevent outside gases from being introduced to the separation vessel.

The discharge of the waste gas was measured twice as can see in Fig. 5. In the first measurement, the vent started at 0.39 bar<sub>g</sub> and 361.7 L of waste gas was vented in 3 minutes. As shown in TABLE 6, the main component of the waste gas was air (N<sub>2</sub> and O<sub>2</sub>, 95.97 vol%). Besides minute amount of other impurities, concentration of SF<sub>6</sub> was 4.03 vol%, which is about 87 g of SF<sub>6</sub>. This means that only 0.1 vol% of SF<sub>6</sub> was lost while one batch (80 kg) of SF<sub>6</sub> was purified. In other words, the recovery rate was 99.9 wt%. In the second measurement, the vent started at 1.47 bar<sub>g</sub> and 374.7 L of waste gas was vented in 10 minutes. As shown in TABLE 7, the gas was analyzed twice. The concentration of the air was 90.7 vol% and 90.2 vol%. And the concentration of SF<sub>6</sub> was 9.25 vol% and 9.79 vol%, which translated to about 215 g, 227 g of SF<sub>6</sub> loss. The loss of SF<sub>6</sub> was slightly higher but the recovery rate was about 99.7 vol%, which was good enough for commercial operation.

### E. Operation cost evaluation

#### 1) Operation time

This operation demonstrated that the prototype purification system was capable of processing large amount of SF<sub>6</sub> at a time. The system could receive SF<sub>6</sub> from large vessels (600-L cylinders) as well as from SF<sub>6</sub> recovery systems directly, which retrieve SF<sub>6</sub> from electrical equipment.

Examples of operation times are summarized in TABLE 8. The retrieval time was shortest when 600-L cylinders were used. It was due to the short gas retrieval time since only gas remained in the connecting pipe was retrieved in 600-L cylinder operation while the whole cylinder was evacuated in 47-L operation. In 600-L cylinder operation, residual gas less than 80 kg in the cylinder was retrieved to the recovery system and purified later. This retrieval operation could be performed while another cylinder was connected

TABLE 6

Composition of the Waste Gas in the First Measurement

Component	Concentration ( $\mu\text{mol/mol}$ )	Analyzer
H <sub>2</sub>	71.40	GC-DID
O <sub>2</sub>	22.56%	GC-TCD
N <sub>2</sub>	73.41%	GC-TCD
CH <sub>4</sub>	32.34	GC-DID
CO	51.87	GC-DID
CO <sub>2</sub>	2.97	GC-DID
CF <sub>4</sub>	5.53	GC-DID
C <sub>2</sub> F <sub>6</sub>	4.92	FT-IR
HF	N.D (Detection limit <0.1)	FT-IR
SO <sub>2</sub>	N.D (Detection limit <0.2)	FT-IR
SOF <sub>2</sub>	N.D (Detection limit <0.1)	FT-IR
SO <sub>2</sub> F <sub>2</sub>	N.D (Detection limit <0.1)	FT-IR
SOF <sub>4</sub>	N.D (Detection limit <0.1)	FT-IR
SF <sub>6</sub>	4.03% (mole fraction)	

\* Assay is by difference, after impurity content is determined.

TABLE 7

Composition of the Waste Gas in the Second Measurement

Component	Concentration ( $\mu\text{mol/mol}$ )		Analyzer
	1st	2nd	
H <sub>2</sub>	76.19	65.46	GC-DID
O <sub>2</sub>	23.19%	19.79%	GC-TCD
N <sub>2</sub>	67.55%	70.42%	GC-TCD
CH <sub>4</sub>	20.43	28.94	GC-DID
CO	28.54	50.76	GC-DID
CO <sub>2</sub>	1.74	2.28	GC-DID
CF <sub>4</sub>	3.23	3.78	GC-DID
C <sub>2</sub> F <sub>6</sub>	0.40	0.83	FT-IR
HF	N.D (Detection limit <0.1)		FT-IR
SO <sub>2</sub>	N.D (Detection limit <0.2)		FT-IR
SOF <sub>2</sub>	N.D (Detection limit <0.1)		FT-IR
SO <sub>2</sub> F <sub>2</sub>	N.D (Detection limit <0.1)		FT-IR
SOF <sub>4</sub>	N.D (Detection limit <0.1)		FT-IR
SF <sub>6</sub>	9.25%	9.79%	(mole fraction)

\* Assay is by difference, after impurity content is determined.

to the purification system, and overall operation time could be saved. The retrieval time from the recovery system could not be measured precisely because the platform scale could not be used as explained above. However, it took more time because SF<sub>6</sub> was retrieved in gas phase from the recovery system. Since same amount of SF<sub>6</sub> was purified in a batch, the purification time was same in all cases. The refilling time was shorter in 47-L cylinder operation. However, the refilling rate (kg/min) was faster in 600-L cylinder operation (9.375 kg/min. for 600-L cylinders, 6.67 kg/min. for 47-L cylinder). The overall operation time per kg of SF<sub>6</sub> was 1.3 and 0.54 minutes for 47-L cylinder operation and 600-L cylinder operation respectively. Thus, processing large supply was better in operation time. These operation times did not include handling times such as weighing, transferring SF<sub>6</sub>, and connecting the cylinders. Since large vessels need less handling, the real operation time will be much shorter in 600-L cylinder operation.

## 2) Operation cost and economic evaluation

The major operation cost came from electricity, liquid nitrogen [28][29] and personnel labor. The total amount of electricity and liquid nitrogen consumed in the operation and labor cost are summarized in TABLE 9. The price of liquid nitrogen was 400 KRW per kg. The liquid nitrogen was used to drive pneumatic valves and freeze the SF<sub>6</sub> gas. It was also used to maintain the pressure of the system at safe level while it was not in operation (idling). The electricity was used mainly to operate compressor,

TABLE 8

Operation Times by Operation Stages (unit: minutes)

	Retrieval	Purification	Refilling	Total time per kg SF <sub>6</sub>
47-L cylinder (80 kg SF <sub>6</sub> )	Liq : 31 Gas : 33	28	12	1.3
600-L cylinder (300 kg SF <sub>6</sub> )	Liq : 34	90	32	0.54

TABLE 9

Total Amount of Liquid Nitrogen (LN<sub>2</sub>), Electricity for the Operation, and Labor Cost

	LN <sub>2</sub> amount (kg)	Electricity (kWh)	Labor cost (KRW)
Operation	9,442	33.19	28,690,000
Operation + Idling	12,904	42.31	28,690,000

\* the monetary of Korea, 1 USD = 1,290 KRW (based on the exchange rate of Nov. 12, 2018))

TABLE 10

Operating Cost of the System

	LN <sub>2</sub> cost (KRW)	Electricity cost (KRW/kg SF <sub>6</sub> )	Labor cost (KRW)	Total (KRW)
Operation	3,776,800	6,381	28,690,000	32,473,181
Operation + idling	995	2	5,529	6,526

vacuum pump, and liquid pump. The electricity billing system in Korea is so complicated that the electricity cost of the operation could not be easily isolated from the whole electricity expense of the research institute. However, it was calculated as about 192 KRW per kWh, which included the basic charge of electricity, the cost of power consumption, value added tax (VAT) and the foundation fund of the electrical industry.

The operation cost is summarized in TABLE 10. The total amount of refilled SF<sub>6</sub> was 5,188.6 kg. Thus, dividing the cost by the refilled SF<sub>6</sub> gives the cost per kg of SF<sub>6</sub>. As shown in TABLE 10, the major cost came from labor cost and then liquid nitrogen use. Except for labor cost, the major operation cost of cryogenic purification technology depends mainly on cooling of the system. Therefore, optimizing the cooling temperature is most important. And considering the consumption of nitrogen during idling period, insulation of the system is also important. The system was operated during summer season so that the cost during the idling would be near maximum. Less idling cost would be expected during cold seasons.

In summary, the operation cost was 6,259 KRW per kg of SF<sub>6</sub>. And it was 6,526 KRW per kg of SF<sub>6</sub> if the idling cost was included. Since the price of new SF<sub>6</sub> is 15,000 KRW per kg in Korea this year, the reuse of SF<sub>6</sub> can save 8,474 KRW per kg of SF<sub>6</sub>, in other words 56% of cost saving. Thus, this system is viable for commercial operation.

## IV. Conclusion

A prototype SF<sub>6</sub> purification system using cryogenic solidification technology has been evaluated for the feasibility of commercialization of the system through a long-term operation. From the evaluation of the operation results, we can conclude as follows:

- 1) 2-month operation of the system demonstrated the long-term operation stability of the system enough for commercial operation. During this period, about 5.4 tons of used SF<sub>6</sub> from power transmission equipment were purified and stored.
- 2) Two types of cylinders with different storage capacity were used to store SF<sub>6</sub> gases before and after purification: 47-L cylinders and 600-L cylinders. The purification time including refilling was 99-104 minutes to process two 47-L cylinders of SF<sub>6</sub> (80 kg), which is similar to that of a comparable commercial system. For 600-L cylinders, the time was even shorter: 102 minutes to process 300 kg of SF<sub>6</sub>. This demonstrated that the process time of the system was at commercial level.
- 3) Considering the amount and the SF<sub>6</sub> concentration of the vent gas, the recovery rate was over 99 vol%—good enough for commercial operation.
- 4) Various transfer procedures were developed to suit the various SF<sub>6</sub> supply vessel. Considering the additional time and labor, transferring SF<sub>6</sub> from and to the large vessels was better than from small vessels.
- 5) The purity of the purified SF<sub>6</sub> was over 99.99 vol%, which is higher than 99.7 vol%, the SF<sub>6</sub> reuse guideline of KEPCO-99.7 vol%. And it will be reused in KEPCO equipment.
- 6) The operation cost, which is the cost of refrigerant (liquid nitrogen), electricity and labor, per kilogram of purified SF<sub>6</sub> was 6,526 KRW. Considering the price of new SF<sub>6</sub> in Korea is about 15,000 KRW per kilogram this year, about 56% of the purchase cost can be saved.

## Acknowledgment

This study has been proceeded as a self-funded internal research project (No. R16VA06) of Korea Electric Power Corporation (KEPCO) since 2016. The research team deeply appreciate the help of Daesung Industrial Gases Co., Ltd. for detail design and manufacturing of the purification system.

## References

- [1] W-T. Tsai, "The decomposition products of sulfur hexafluoride (SF<sub>6</sub>): Reviews of environmental and health risk analysis," *Journal of Fluorine Chemistry*, 128, pp.1345-1352, 2007.
- [2] M. Rabie, "C.M. Franck. Assessment of eco-friendly insulation to replace the most industrial greenhouse gases SF<sub>6</sub>," *Environmental Science Technology*, 52, pp.369-380, 2018.
- [3] M. Maiss, C.A.M. Brenninkmeijer, "Atmospheric SF<sub>6</sub>: trends, sources, and prospects," *Environmental Science Technology*, 32, pp.3077-3086, 1988.
- [4] International Electrotechnical Commission, IEC 60376. Specification of technical grade sulfur hexafluoride (SF<sub>6</sub>) for use in electrical equipment. Second edition, 2005. International Electrotechnical Commission, CIGRE 234, SF<sub>6</sub> recycling guide, 2003.
- [5] IEC 60480. Guidelines for the checking and treatment of sulfur hexafluoride (SF<sub>6</sub>) taken from electrical equipment and specification for its reuse. Second edition, 2004.
- [6] K. Kim, K.S. Kim, J.E. Lee, S. Park, C.K. Ahn, G.H. Kim, "Status of SF<sub>6</sub> separation /refining technology development for electric industry in Korea," *Separation and Purification Technology*, 200, pp.29-35, 2018.
- [7] C.K. Ahn, G.H. Kim, J.E. Lee, K.S. Kim, K. Kim, "An SF<sub>6</sub> purification process utilizing gas hydrate formation developed for electric power industry," *Separation and Purification Technology*, 217, pp. 221-228, 2019.
- [8] W.S. Cho, K.H. Lee, H.J. Chang, W. Huh, H.H. Kwon, "Evaluation of pressure-temperature swing adsorption for sulfur hexafluoride (SF<sub>6</sub>) recovery from SF<sub>6</sub> and N<sub>2</sub> gas mixture," *Korean Journal of Chemical Engineering*, 28, pp.2196-2201, 2011.
- [9] M.B. Kim, K.M. kim, T.H. Kim, T.U. Yoon, E.J. Kim, J.H. Kim, Y.S. Bae, "High selective adsorption of SF<sub>6</sub> over N<sub>2</sub> in a bromine-functionalized zirconium-based metal-organic framework," *Chemical Engineering Journal*, 339, pp.223-229, 2018.
- [10] A plant and method for recovering sulphur hexafluoride for reuse, WO 2012/142672, PCT/AU2012/000417.
- [11] S. Lee, J.W. Choi, S.H. Lee, "Separation of greenhouse gases (SF<sub>6</sub>, CF<sub>4</sub> and CO<sub>2</sub>) in an industrial flue gas using pilot scale membrane," *Separation and Purification Technology*, 148, pp.15-24, 2015.
- [12] Y. Dai, Q. Li, X. Ruan, Y. Hou, X. Jiang, X. Yan, G. He, F. Meng, Z. Wang, "Fabrication of defect-free Matrimid asymmetric membranes and the elevated temperature application for N<sub>2</sub>/SF<sub>6</sub> separation," *Journal of Membrane Science*, 577, pp.258-265, 2019.
- [13] S. Lee, J.W. Choi, S.H. Lee, "Separation of greenhouse gases (SF<sub>6</sub>, CF<sub>4</sub> and CO<sub>2</sub>) in an industrial flue gas using pilot scale membrane," *Separation and Purification Technology*, 148, pp.15-24, 2015.
- [14] S.J. Kim, Y.I. Park, S.E. Nam, H. Park, P.S. Lee, "Separation of F-gases from nitrogen through thin carbon membranes," *Separation and Purification Technology*, 158, pp. 108-114, 2016.
- [15] K. Golzar, H. Modarress, S.A. Iranagh, "Separation of gases by using pristine composite and nanocomposite polymeric membranes: a molecular dynamics simulation study," *Journal of Membrane Science*, 539, pp.238-256, 2017.
- [16] I. Cha, S. lee, J.D. Lee, G.W. Lee. Y. Seo, "Separation of SF<sub>6</sub> from mixtures using gas hydrate formation," *Environmental Science Technology*, 44, pp.6117-6122, 2010.
- [17] H. Lee, J.D. Lee, Y. Kim, "Effect of nonionic surfactants on F-gases (HFC-134a and SF<sub>6</sub>) Hydrate formation," *Industrial & Engineering Chemistry Research*, 57, pp. 12980-12986, 2018.
- [18] V. M Vorotyntsev, V. M. Malyshev, P. G. Taraburov, G. M. Mochalov, "Separation of gas mixtures by continuous gas hydrate crystallization," *Theoretical Foundations of Chemical Engineering*, 35, pp. 513-515, 2001.
- [19] V. M Vorotyntsev, V. M. Malyshev, G. M. Mochalov, P. G. Taraburov, "Separation of gas mixtures by the gas hydrate crystallization method," *Theoretical Foundations of Chemical Engineering*, 35, pp. 119-123, 2001.
- [20] J. E. Lee, K.S. Kim, A.R. Kim, S. Park, K. Kim, "Installation and test run of comprehensive analysis system for SF<sub>6</sub> in power equipment," *KEPCO Journal on Electric Power and Energy*, vol. 3, no. 1, pp. 41-47, 2017.
- [21] Standard test method for air and carbon tetrafluoride in sulfur hexafluoride by gas chromatograph, ASTM D2685-11 (2011).
- [22] Standard test method for acidity of sulfur hexafluoride, ASTM D2284-11, (2011)
- [23] Standard specification for sulfur hexafluoride, ASTM D2472-15 (2015)
- [24] Standard test methods for water vapor content of electrical insulating gases by measurement of dew point, D2029-97 (2008).
- [25] S. van der Laan, R.E.M. Neubert, H.A.J. Meijer, "A single gas chromatograph for accurate atmospheric mixing ratio measurements of CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub> and CO," *Atmospheric Measurement Techniques*, 2, pp.549-559, 2009.
- [26] X. Zhang, H. Liu, J. Ren, J. Li, X. Li, "Fourier transform infrared spectroscopy quantitative analysis of SF<sub>6</sub> partial discharge decomposition components," *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 136, 884-889, 2015.
- [27] R. Kurte, H.M. Heise, D. Klockow, "Quantitative infrared spectroscopic analysis of SF<sub>6</sub> decomposition products obtained by electrical partial discharges and sparks using PLS-calibration," *Journal of Molecular Structure*, 565-566, pp. 505-513, 2001.
- [28] H. Cho, D. Woo, Y. Choi, M. Han, "Characteristics of SF<sub>6</sub> gas recycling processes," *Clean Technology*, 17, pp. 329-335, 2011.
- [29] H. Tajima, A. Yamasaki, F. Kiyono, "Energy consumption estimation for greenhouse gas separation processes by clathrate hydrate formation," *Energy*, 29, pp.1713-1729, 2004