Simulation study of a hybrid cryogenic and membrane separation system for SF6 recovery from aged gas mixture in electrical power apparatus

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Abstract

Despite its wide use in the electrical industry, sulphur hexafluoride (SF6) is a potent greenhouse gas with high global warming potential (GWP) and atmospheric lifetime. Therefore, there is a growing interest in recycling, reconditioning, and reusing aged SF6. In addition, the upcoming F-gas regulation with increasing restrictions imposed on SF6 could result in banning the production of virgin SF6, consequently putting pressure on end-users to recycle aged SF6 for further reuse. Currently, the cryogenic separation process is often utilised to recondition aged SF­6 in electrical power apparatus. However, this method is not suitable to treat mixtures with SF6 content lower than 40 mol.% and produces a waste gas containing up to 15 mol.% of SF6. This study aims to tackle the aforementioned limitations of the existing SF6 reconditioning process by implementing membrane units. For this purpose, a mathematical model of the membrane gas separation process is developed and integrated into an Aspen Plus simulation via CAPE-OPEN interface. A set of cost model equations collected from the open literature is also used to study the cost of the SF6 ­recovery process. The simulation for a feed mixture of 20 mol.% SF6 and 80 mol.% N2 with a flow rate of 10,000 kg/day demonstrated the system’s capability to recover SF6 efficiently, achieving the product purity of 98.3 mol.% and SF6 recovery of 97.5 %. Further optimisation reduced the specific recovery cost and specific energy consumption by 3.1 % and 10.5 %, respectively. The SF6 content in the waste gases was also reduced to less than 1 mol.%.

**Keywords**: sulphur hexafluoride; gas separation; membrane; simulation; Aspen Plus.

* 1. Introduction

Sulphur hexafluoride (SF6) is highly stable due to its chemical inertness that is comparable to nitrogen (N2) gas and incombustible nature. Two prominent factors for the use of SF6 in the electrical industry are its excellent dielectric property, almost 2.5-3 times higher than air, under similar test conditions, and its low dissociation temperature and high dissociation energy, which makes it an excellent insulating and arc-quenching gas medium. However, SF6 is a potent greenhouse gas with a global warming potential (GWP) 24,300 times higher than carbon dioxide (CO2) and an atmospheric lifetime of 3,200 years.

Despite these facts, demand for SF6 in medium- and high-voltage switchgear continues to rise, driven by increasing electricity demand and electrification of transportation. The installed base of medium-voltage switchgear units is expected to increase between 40 to 90 per cent by 2050 (Zhai et al., 2021). In the UK, National Grid is the largest user of SF6 with an inventory of 920 tonnes with SF6 presently accounting for 92 % of their Scope 1 emissions (National Grid, 2022). The continued phase-out of SF6 is paramount to ensure the wider electricity industry in the UK and Europe as a whole can realise the ambition of Net Zero by 2050. Despite decades of research, there is no perfect like-for-like replacement for SF­6 across different power equipment. For example, some SF6 alternatives, such as fluoronitrile and perfluoroketone, despite having been commercialised and successfully operational in gas-insulated equipment (Hyrenbach et al., 2017; Kieffel and Biquez, 2014), belong to the group of per- and polyfluoroalkyl substances (PFAS), which will potentially be banned according to the EU PFAS Restriction Proposal (Zeinoun and Reisser, 2023). This leaves non-greenhouse gases including N2 and air to be the only viable option to substitute SF6. However, due to their low dielectric strength, it would dramatically increase the equipment size and/or the filling pressure of the device by a factor of at least 2.5 (Kieffel et al., 2016). This would not only impact enclosure design, safety and cost but also go against the need to develop more compact electrical equipment in metropolitan areas.

The new proposed F-gas regulation draft prohibits the sale of new SF6 switchgear starting in 2026 (European Commission, 2022). However, it does not ban the use of existing SF6 switchgear which has a typical asset lifespan of 20 to 30 years for medium-voltage and >40 years for high-voltage equipment. Hence, there is a growing need to recondition aged SF6 for further use. The conventional method of reconditioning used SF6 is a commercialised cryogenic process to recover SF6 from aged mixtures of SF6 with other components mainly including N2 or air (DILO, 2022). The recovered product contains 99 mol.% SF6 and approximately 85 % recovery of SF6 is achieved. As shown in Figure 1, the process consists of two separation columns: one operates at relatively lower pressures of approximately 8-14 bara, and the other operates at a higher pressure of up to 40 bara. The refrigeration units are used to cool the gas mixture to the optimal temperature between -40 ℃ to -30 ℃. Note that the operating pressure and temperature of the two separation columns are dependent on the composition of the gas mixture. The main limitation of this process is that it is only suitable for treating insulating gas mixtures with a high content of SF6 (>60 mol.%), while the SF6 content in a binary mixture with a buffer gas such as N2 typically ranges from 5 to 20 mol.% (Hama et al., 2018). In addition, a certain amount of waste gas is produced and cannot be directly released into the atmosphere, which requires a further stage of incineration that can be energy intensive.



Figure 1: Conventional SF6 cryogenic process (simplified from (DILO, 2022))

In gas separation applications, there is a growing interest in membrane-based hybrid systems due to their inherent benefits. These include straightforward installation and operation, minimal energy requirements, continuous operation with the option of partial or complete recycling, and the potential for integration with other separation units. This study aims to address the current shortcomings of the SF6 reconditioning process by introducing a design of a membrane-cryogenic hybrid system.

* 1. Methodology
		1. Aspen Plus simulation

Aspen Plus was used to model a plant capable of treating approximately 10,000 kg/day of SF6 mixture, a representative scale for a small gas treatment plant. The SF6 content in the feed mixture was set at 20 mol.% with the remaining gas being N2. Two membrane units were incorporated: the first one was used to enhance the SF6 content in the feed for the cryogenic cycle, while the second unit was employed to minimise the SF6 concentration in the residual gas. A process flow diagram of the process is included in Figure 2.

Since a membrane module is not available in Aspen Plus and in its Model Library, the membrane model was solved numerically in MATLAB and integrated into Aspen Plus as a CAPE-OPEN COM unit operation with a license provided by AmsterCHEM. Additionally, the Peng-Robinson property package was chosen for the simulation.



Figure 2: Membrane-cryogenic process

* + 1. Model assumption

The model was assumed to operate for 358 days per year (7 maintenance days) for a period of 5 years. The membrane is assumed to be in constant operation during this period with no decrease in productivity. The process operated at steady state, which means no replacements or adjustments to the production rate were considered and any replacements during the 5-year period were accounted for in the contingency and fee costs (15 % and 3 % of the bare module cost, respectively) (Turton et al., 2018). For simplification, other costs including operational labour, waste treatment and administration were neglected.

The membrane module was simulated as a hollow fibre module using a mathematical model that describes the counter-current flow pattern with bore-side feed, based on a number of assumptions. The most important ones are as follows:

1. The permeation rate obeys Fick’s law;
2. Permeance of gas components through the membrane is independent of pressure on both sides;
3. Plug flow on the feed side with changes in velocity and pressure are assumed to be negligible on the feed side;
4. Laminar flow on the permeate side.
5. Gas properties including viscosity and specific heat capacity are independent of pressure and temperature.

Before being utilised to examine the plant performance under various operating conditions, the membrane model was validated using experimental data of SF6/N2 separation using a commercial membrane from Yamamoto et al. (2002). The analysis revealed that the membrane model aligned well with experiment data for high feed-side pressures of 4 and 5 bara but deviated significantly at a lower pressure of 3 bara. To address this, a corrective factor of 1.12 was applied to the membrane area variable, reducing the model’s average percentage error to 8 %. Turton et al. (2018) reported the correlations of cost estimation in USD back in 2001. Hence, all the costs were updated to 2023 price using cost indexes and converted to GBP adopting the current exchange rate of 1 USD to 0.8 GBP.

* + 1. Simulation study of a hybrid SF6 recovery system

The simulation of the process was carried out to estimate the key performance indicators including the specific SF6 recovery cost and energy consumption. Table 1 presents a set of important equations of the membrane and economic models used in this study.

Table 1: Membrane and economic models and equations for key performance indicators

|  |  |  |
| --- | --- | --- |
| **Equation** | **Description** | **Unit** |
| **Membrane model** |
|  $A\_{m}=$ $N\_{T}πD\_{i}L$ | (1) | Effective membrane area | m2 |
|  $\frac{dN\_{t}x\_{i}}{dz}=-N\_{T}πD\_{i}J\_{i}$ | (2) | Molar flow rate profile on the feed side | mol/m·s |
| $\frac{dN\_{s}y\_{i}}{dz}=-N\_{T}πD\_{i}J\_{i}$  | (3) | Molar flow rate profile on the permeate side | mol/m·s |
| $\frac{dp\_{s}}{dz}=\frac{192μRT\_{s}ND\_{o}(D+ND\_{o})}{π(D^{2}-ND\_{o}^{2})^{3}P\_{s}}\sum\_{i=1}^{nc}N\_{s}y\_{i}$  | (4) | Pressure profile on the permeate side | Pa/m |
| $\frac{dT\_{t}}{dz}=-N\_{T}πD\_{i}\left[\frac{\sum\_{i=1}^{nc}J\_{i}c\_{pi}(T\_{t}-T\_{s})}{\sum\_{i=1}^{nc}N\_{t}x\_{i}c\_{pi}}\right]$  | (5) | Temperature profile on the feed side | K/s |
| $\frac{dT\_{s}}{dz}=-N\_{T}πD\_{i}\left[\frac{\sum\_{i=1}^{nc}J\_{i}c\_{pi}(T\_{t}-T\_{s})}{\sum\_{i=1}^{nc}N\_{s}y\_{i}c\_{pi}}\right]$  | (6) | Temperature profile on the permeate side | K/s |
| **Capital cost** |
| $log\_{10}C\_{P}^{0}=K\_{1}+K\_{2}log\_{10}\left(A\right)+K\_{3}\left[log\_{10}(A)\right]^{2}$  | (7) | General equipment cost | £ |
| $C\_{BM}=C\_{P}^{0}F\_{BM}$  | (8) | Bare module equipment cost  | £ |
| $C\_{TM}=\sum\_{i=1}^{n}C\_{TM,i}=1.18\sum\_{i=1}^{n}C\_{BM,i}$  | (9) | Total module cost | £ |
| $C\_{M}=1.12A\_{m}C\_{m}$  | (10) | Membrane cost | £ |
| $C\_{TM}=0.2(C\_{TM}+C\_{M})$  | (11) | Annual capital related cost | £/y |
| **Operational cost** |
| $C\_{Elec}=C\_{E}∙P\_{C}$  | (12) | Annual electricity cost | £/y |
| $C\_{UT}=C\_{R}∙Q\_{C}+C\_{S}∙Q\_{H}$  | (13) | Annual utilities cost | £/y |
| $C\_{OPEX}=(C\_{Elec}+C\_{UT})∙ τ$  | (14) | Annual operating cost | £/y |
| **Performance indicator** |
| $(C\_{TM}+C\_{OPEX})/F\_{SF\_{6}}$  | (15) | Specific recovery cost | £/kg SF6 |
| $(P\_{C}+Q\_{C}+Q\_{H})/F\_{SF\_{6}}$  | (16) | Specific energy consumption | kWh/kg SF6 |

* + 1. Optimisation of a hybrid SF6 recovery system

The operation of the SF6 recovery system was optimised to ensure the purity of the liquid product is at 99 mol.% SF6. The feed conditions were fixed at 10,000 kg/day at 25 $℃$ and 4 bara. The nonlinear single-objective optimisation framework can be described as:

**Objective function:** Minimise specific recovery cost (Equation 15)

**Constraint:** Minimum purity of 99 mol.% SF6 in the liquid product

**Decision variables:** The initial values, lower and upper bounds for the decision variable in the optimisation problem are listed in Table 2

Table 2: Initial values, lower and upper bounds for the decision variables in the optimisation problem

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **Decision variable** | **Unit** | **Initial** | **Minimum** | **Maximum** |
| C-1 operating pressure | bara | 9 | 7 | 14 |
| C-1 operating temperature | $$℃$$ | -30 | -40 | -30 |
| C-2 operating pressure.  | bara | 38 | 30 | 40 |
| C-2 operating temperature  | $$℃$$ | -38 | -40 | -30 |
| M-1 area  | m2 | 145 | 140 | 160 |
| M-2 area | m2 | 45 | 30 | 60 |

* 1. Results and discussion

The simulation was first carried out using initial operating conditions as listed in Table 2 and the results obtained are shown in Table 3. Results from the initial simulation show that the hybrid process is capable of treating feed gas with a low concentration of SF6 of 20 mol.%, while achieving a high SF6 recovery of 97.5 % and product purity of 98.3 mol.%. The SF6 content in the waste gas streams was 0.555 and 0.844 mol.%, respectively, which are significantly lower in comparison to that of the conventional cryogenic process.

Table 3: Simulation results using initial operating conditions

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **SF6 recovery productivity** | **Product purity** | **SF6 content in Waste Gas 1** | **SF6 content in Waste Gas 2** | **SF6 recovery** | **Total capital cost** | **Total operational cost** | **SF6 recovery cost**  | **Specific energy consumption** |
| **kg/h** | **mol.%** | **mol.%** | **mol.%** | **%** | **106 £/y** | **106 £/y** | **£/kg** | **kWh/kg** |
| 230 | 98.3 | 0.555 | 0.844 | 97.5 | 1.058 | 0.649 | 0.580 | 0.171 |

Table 4 shows the optimisation results of several performance indicators. In general, there was no significant change in the product purity and SF6 recovery in comparison to the initial simulation. Notably, the purity of the liquid product was only influenced by the operational parameters of the low-pressure separation column C-1, with a purity of 99 mol.% attainable at operating pressures below 8 bara. The SF6 content in the waste gas streams was kept below 1 mol.% at 0.539 and 0.549 mol.%, respectively. This could potentially eliminate the requirement of the further destruction of the waste gas. Furthermore, increasing the membrane area in both stages led to a reduction in the total energy consumption, indicating a trade-off between capital and operational costs. Nevertheless, the associated increase in capital costs was negligible, resulting in a lower specific recovery cost by 3.1 %.

Table 4: Optimisation results

|  |  |
| --- | --- |
| **Optimised decision variable** | **Optimised results** |
| **Parameter** | **Unit** | **Value** | **Parameter** | **Unit** | **Value** |
| C-1 operating pressure | bara | 8 | SF6 recovery productivity | kg/h | 230 |
| C-1 operating temperature | $$℃$$ | -33 | Product purity | mol.% | 99.0 |
| C-2 operating pressure | bara | 32 | SF6 content in Waste Gas 1  | mol.% | 0.539 |
| C-2 operating temperature | $$℃$$ | -40 | SF6 content in Waste Gas 2 | mol.% | 0.549 |
| M-1 area | m2 | 150 | SF6 recovery | % | 97.8 |
| M-2 area | m2 | 50 | Total capital cost | 106 £/y | 1.058 |
|  |  |  | Total operational cost | 106 £/y | 0.531 |
|  |  |  | SF6 recovery cost | £/kg | 0.562 |
|  |  |  | Specific energy consumption | kWh/kg | 0.153 |

* 1. Conclusion

The combination of Aspen Plus simulations with a techno-economic analysis provided an overview of the feasibility of a hybrid membrane-cryogenic plant to recover SF6 from the aged gas mixture. Firstly, the initial simulation showed success in addressing the two main drawbacks of the current SF6 reconditioning process: i) capable of treating SF6/N2 insulation mixture with a low SF6 content below 20 mol.%, and ii) achieve high SF6 recovery of 97.5 % and low SF6 content of less than 1 mol.% in the waste gas streams. This degree of recovery was significantly higher, as opposed to the 85 % recovery of SF6 achieved by the conventional cryogenic process. The content of SF6 in the waste gas streams produced of less than 1 mol.% could potentially obviate the need for additional disposal process of the waste gas. The cost analysis reveals that the compressor cost constitutes the primary component of the overall capital expenditure, whereas the operational cost is predominantly influenced by the combined expenses of electricity required for the compressors and the cost of refrigerants used in the refrigeration unit. Therefore, the incorporation of membrane units does not substantially affect the overall process cost. The determination and optimisation of various process variables has attained the reduction in both SF6 recovery cost and SF6 content in waste gas. The specific SF6 cost, and energy consumption were reduced by 3.1 % and 10.5 % respectively.

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