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Introduction

SF₆ is instrumental in achieving space-saving electrical energy transmission and distribution equipment, such as gasinsulated switchgears (GISs) and gas-insulated lines (GILs), due to its remarkable dielectric insulation and arc-quenching performance.^{1,2} According to the report of The Intergovernmental Panel on Climate Change (IPCC) in 2013, the global warming potential (GWP) of SF₆ is approximately 23 000 times higher than that of CO₂, and SF₆ has a cumulative effect on global warming owing to its extreme chemical stability.³ The Kyoto protocol has identified SF₆ as one of the six emissionlimiting gases. Therefore, there is an urgent need to find a new environmentally friendly gas to replace SF₆.⁴

In recent years, researchers have been committed to finding environmentally friendly alternatives to replace SF_6 ; these alternatives should have low GWP, high dielectric strength and low

Synthesis and dielectric properties of the ecofriendly insulating gas thiazyl trifluoride[†]

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Sulfur hexafluoride (SF₆), which is known as a superior electrically insulating and arc-quenching medium, plays a decisive role in the modern transmission and distribution network of electric energy, especially in high-voltage power networks. However, the ever-increasing usage of SF₆ also leads to the continuous escalation of atmospheric SF₆ levels, which is considered to be the main cause of the greenhouse effect. To decrease this environmental impact, eco-friendly alternatives to SF₆ have been researched for decades. To date, no significant success has been made regarding replacement gases for transmission networks. Some potential alternatives have comparatively lower global warming potential (GWP) but involve technical trade-offs. Thiazyl trifluoride, which has some excellent chemical and electric properties, is a novel substitution candidate for SF₆. In this article, an efficient synthetic route starting from sulfur monochloride and followed by ammonization and fluorination was proposed. The structures of the intermediates and the target products were determined by X-ray diffraction (XRD), infrared spectroscopy (IR), and gas chromatography-mass spectrometry (GC-MS). The effects of some determining factors on the yield and purity, including the molar ratio of the reactants, recrystallization conditions and condensation temperature, were also investigated. The results showed that the overall yield of thiazyl trifluoride was approximately 25%, while the purity could be up to 90.6% under optimal conditions

> boiling point.^{5,6} Significant efforts have been made to study insulation gases, and many researchers have made great progress using substitutes such as perfluorocarbons (*e.g.* c-C₄F₈, C₃F₈ (ref. 7)), hydrofluorocarbons, CF₃I,⁸ C₅F₁₀O,^{9,10} C₆F₁₂O and C₄F₇N.^{11,12} These alternatives have higher insulation strength and lower GWP than SF₆, but they also have different shortcomings in terms of boiling point, toxicity, stability, and other aspects. For example, although the insulation strength of c-C₄F₈ is approximately 1.25 times higher than that of SF₆ and the GWP is far below that of SF₆, the liquidus temperature of c-C₄F₈ is high (-8 °C), and carbon deposition will occur after discharge, which will affect equipment safety and insulation strength.^{13,14} Similarly, C₄F₇N has to be mixed with CO₂ due to its high boiling point, which limits its insulation and arc-extinguishing performance.¹⁵

> Thiazyl trifluoride (NSF₃) is commonly known as a chemically stable, colorless gas that is used as an important precursor for the preparation of various sulfur–nitrogen–fluorine compounds. Recently, the results of a quantum chemical calculation based on a structure-activity relationship model show that NSF₃ is most likely to be an insulating gas with a low greenhouse effect. Baoshan Wang *et al.*¹⁶ suggested that NSF₃ has excellent performance parameters: the insulation strength is 1.35 times that of SF₆ at the same pressure, and the GWP value is 916, which is approximately one-twentieth that of SF₆, while the boiling point is -27 °C. Therefore, NSF₃ has been

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theoretically proven to be a promising alternative gas considering its liquidus temperature, insulation performance, and greenhouse effect. Research on the synthetic route of this gas is a necessary precondition for testing the comprehensive performance of NSF₃ in actual electrical equipment.

Few reports have described the synthesis of NSF₃, and these studies have seldom focused on its electrical performance. William H. Kirchhoff¹⁷ reported the general protocol for the synthesis of NSF3 by using S4N4 and AgF2. However, Oskar Glemser¹⁸ argued that the numerous side products were difficult to separate; he also reported a synthesis method involving sulfur, ammonia and silver difluoride, with carbon tetrachloride as a solvent. Moreover, he suggested that the molar ratio between the reaction materials was 1:1:6 (NH₃:S:AgF₂). However, the byproducts of the reaction contained hydrogen fluoride gas, which is extremely corrosive and harmful to the environment.19 Clifford20 proposed a method in which NSF3 was prepared by SF₂NCOF oxidation with silver difluoride. Generally, most related studies are more suitable for minor or trace synthesis rather than large-scale production.

To verify the insulation properties of NSF₃ and obtain further insight for commercial production, a comparatively economical and efficient two-step route (Fig. 1) was proposed. Ammonia and sulfur monochloride (S₂Cl₂) were used as raw materials and were fluorinated by AgF₂ to prepare NSF₃. In addition, some decisive factors were studied to maximize the yield and purity.

Experimental methods

General remarks

Some of the chemicals used in this work are potentially dangerous due to their toxicity. Thus, the detailed information on the MSDS of the materials used in this experiment were determined according to the Chemical Book, and all toxic properties are described in Table S1 in the ESI.[†]

Material and methods

All solvents used were anhydrous and analytical grade. Sulfur monochloride (greater than or equal to 98%) was purchased from Shanghai Aladdin Bio-Chem Technology Company. Silver(II) fluoride was obtained from Sinopharm Chemical Reagent Co. Ltd. NH₃ and nearly 99.99% pure nitrogen gas were obtained from Wuhan Xiangyun Industry and Trade Co. Ltd. The copper reactor and condenser were obtained from Shanghai Yanzheng Experimental Instrument Co., Ltd.

IR spectra were recorded on a Thermo FT-IR 5700 IR spectrometer (KBr), and Thermo Fisher DXR was used to record Raman spectra. GC-MS was carried out on a Varian-450 gas chromatograph coupled to a Varian-320 m selective detector equipped with EI detectors. The gas chromatograph was



Fig. 1 Two-step synthesis of NSF₃ starting from sulfur monochloride. Fig. 2 X-ray diffraction spectrum of S_4N_4 .

equipped with a 30 m and 0.250 mm, 0.25 mm df, VF-5 column. Photoelectron spectra and X-ray diffraction patterns were recorded on an X-ray photoelectron spectrometer (ESCA-LAB250Xi, Thermo Fisher Scientific, United States) and an X-ray diffractometer (XRD-6100, Shimadzu Corporation, Japan) with Cu-K α radiation ($\lambda = 0.154$ nm).

Synthesis of tetrasulfur tetranitride (S_4N_4)

 CCl_4 (250 mL) and S_2Cl_2 (40 mL, 0.5 mol) were added to a 500 mL three-necked flask at 0 °C with stirring. Afterwards, dry NH3 was introduced into the three-necked flask at a flow rate of 1200 mL min⁻¹. The whole reaction was carried out in an ice bath for 1 hour, resulting in the appearance of an orange precipitate in a red solution. The precipitate was washed with pure water with stirring for approximately 3 hours, isolated by Soxhlet extraction with dry dioxane, and then recrystallized from benzene to obtain pure S_4N_4 crystals. To generate S_4N_4 crystals, the red solution was transferred to a vacuum after evaporation treatment with dichloromethane and recrystallized with benzene.

The yield was 43%, and the crystal phase of S₄N₄ was further confirmed by XRD (as shown in Fig. 2). According to XPS, the percentage of S_4N_4 in the sample was above 99%. (N: 30.15%; S: 69.50%; C: 0.13%; O: 0.22%).

Synthesis of thiazyl trifluoride

 S_4N_4 (3.7 g, 0.02 mol) and CCl_4 (80 mL) were added to a copper reactor equipped with a copper reflux condenser under N₂ protection. The mixture was dissolved under magnetic stirring and heated to stable reflux. Then, AgF₂ (45.7 g, 0.32 mol) was quickly added to the reflux. The whole reaction was carried out at 78 °C for 2 hours. The outlet of the condenser was connected by a conduit, and the gas product was successively dried by potassium permanganate solution and anhydrous calcium chloride and finally condensed to obtain crude NSF₃ as the product. The purity of NSF3 was 90.6%, as determined by GC, while the structure was confirmed by IR and GC-MS.

GC/EI/MS (70 eV) m/z: 102.8 [M]⁻, 32 [S]⁺, 46.8 [NS]⁺, 65 $[NSF]^+$, 69.9 $[SF_2]^+$, 83.9 $[NSF_2]^+$, 88.9 $[SF_3]^+$.

IR: 815 cm⁻¹ (S–F), 945 cm⁻¹ (N \equiv S).





Fig. 3 Configuration of the high-voltage analysis platform.

The insulation performance of thiazyl trifluoride

A high-voltage analysis platform was applied to evaluate the insulation performance of NSF_3 .²¹ The configuration of the circuit is shown in Fig. 3, and the parameters are shown in Table 1. The measurement accuracy was within $\pm 1\%$.

Two tungsten-copper alloy electrodes with diameters of 10 mm were employed as the test electrodes and positioned with a 2.5 mm-long interelectrode gap. The cavity was composed of a stainless-steel base, a sealed quartz-glass body, a charging/extracting valve and an electrode spacing adjustment screw. The unit can withstand a maximum pressure of 0.7 MPa.

The gas chamber was evacuated to vacuum and then filled with inert buffer gas after stabilization for 20 min. The procedure above was repeated three times to eliminate the effects of other impurities in the test chamber. The experimental gas was injected into the chamber until the pressure was equal to the saturated vapor pressure (absolute pressure at 20 $^{\circ}$ C). An increasing voltage method was applied to the discharge chamber through a power frequency voltage to determine the instantaneous breakdown voltage. The tests were conducted at least 5 times with a gap of 4 min between every two breakdowns.

The breakdown voltages of NSF₃ and SF₆ under the same conditions are shown in Table 2, which shows that the insulation strength of NSF₃ (whose purity was approximately 87%) was 1.28 times that of SF₆.

Results and discussion

Effects of the molar ratio of reactants on the yield of tetrasulfur tetranitride

According to previous studies, the yield was greatest when ammonia was passed through at a rate of 900–1000 mL min^{-1,²²} However, the effect of ammonia on the reaction was related to not only its injection speed but also its total amount by affecting

Table 2	The dielectric	strongth o	F NISE_	and SE.
Table 2	The dielectric	strength o	I INSE3	and SF6

Gas	Pressure ^a /MPa	Purity ^b /%	Breakdown voltage/kV
SF ₆	0.3	100	35.23
NSF ₃	0.3	87	45.08

 a Isolated pressure. b The impurities of NSF3 were SF6, CCl3F, and SO2, as determined by GC-MS.

the concentration of the reactants. Therefore, the reaction of ammonia to sulfur monochloride in a molar ratio from 6 : 1 to 17 : 1 was studied, and the synthetic route and results are displayed in Table 3. The yield of 2 increased from 30.3% to 73.8% as the molar ratio increased and then remained constant when the amount of ammonia reached over 16 equivalents of sulfur monochloride. Thus, the optimal molar ratio of sulfur monochloride to ammonia should be 1 : 16.

Effects of recrystallization conditions on the yield of tetrasulfur tetranitride

Sulfur was found to be the main impurity after removal of ammonium chloride by distilled water. Recrystallization was one of the most important steps in the preparation of S_4N_4 . In addition to temperature, diverse types of solvent could also directly affect the process of crystallization since the solubility varies by material. Nevertheless, the solubilities of S_4N_4 and sulfur were extremely similar; thus, choosing an appropriate

Table 3 Effects of the various molar ratios of sulfur monochloride to ammonia on the yield of 2^a

	$\begin{array}{ccc} S_2Cl_2 & \xrightarrow{NH_3} & S_4N_4 \\ 1 & 2 \end{array}$	
Entry	$S_2Cl_2: NH_3$	$\operatorname{Yield}^{b}(\%)$
1	1:6	30.3
2	1:8	45.6
3	1:10	58.9
4	1:12	73.2
5	1:14	81.8
6	1:16	83.0
7	1:17	83.8
8	1:18	84.3

 a Reaction conditions: $S_2 Cl_2$ (0.19 mol), CCl₄ (100 mL), NH₃ at a rate of 1000 mL min^{-1}, at -4 °C for 1 hour. b Isolated yield.

Table 1	The parameters of the high-voltage test device		
		Max output	Protective

Rated voltage	Rated capacity	voltage	resistor	Voltage divider
220 V	10 kVA	100 kV	5 kΩ	1000:1



Fig. 4 (a) The solubility of S_4N_4 varies with temperature in three solvents. (b) The solubility of sulfur varies with temperature in three solvents.

solvent became key to improving the yield. Accordingly, benzene, 1,4-dioxane, and cyclohexane were selected as alternative solvents for recrystallization after taking polarity and boiling point into account; therefore, the dissolution differences in various temperature phases were tested. As shown in Fig. 4, the solubilities of S_4N_4 and sulfur in cyclohexane were relatively low, and the temperature sensitivities showed similar trends, which indicated that cyclohexane cannot be used as a solvent during the recrystallization process. By contrast, the solubilities of sulfur and S₄N₄ in 1,4-dioxane were higher than those of the other compounds. However, the solubility of sulfur was too sensitive to temperature, which resulted in the formation of a turbid liquid and eutectoid. Both S₄N₄ and sulfur can be well dissolved by benzene, which has a good thermosensitivity to S₄N₄. In general, benzene was treated as the optimal solvent for recrystallization.

Furthermore, the cooling temperature of crystallization was also a decisive factor; thus, reactions were carried out over a temperature gradient to verify the optimal temperature. Table 4 provides the relationship between recrystallization yield and cooling temperature. The results showed that the solution cooled rapidly, and the two substances were simultaneously precipitated by supersaturation at a low temperature (lower than 10 °C, entries 1–2). The yield of S₄N₄ increased obviously from 27.1% to 52.2% with the increase in temperature from 15 °C to 30 °C (entries 3–6). S₄N₄ precipitated first and separated from sulfur when the temperature ranged from 10–20 °C.

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Table 4 Effect of cooling temperature on recrystallization yield^a

Entry	Temperature/°C	Yield (%)
1	5	b
2	10	b
3	15	27.1
4	20	38.4
5	25	50.4
6	30	52.2
7	35	48.1

^{*a*} Reaction conditions: impurity-containing S_4N_4 was dissolved by stirring with benzene, and the saturated solution was heated to 80 °C before cooling. ^{*b*} No expected product was obtained.

However, sulfur crystallized after some time, resulting in a low yield. As shown in Table 4, the optimum cooling temperature was 30 °C, and several orange needle-shaped crystals of good crystal shape were obtained at this time, while the yield was up to 52.2%. At this point, the cooling rate was moderate, which contributed to the formation of the S_4N_4 crystal nucleus, thus making it easier to avoid the precipitation of sulfur. Further heating accelerated the dissolution of S_4N_4 , leading to a decrease in yield (entry 7).

Such efforts were made to promote the industrialization of the route. For instance, unpurified S_4N_4 was reacted with AgF₂, which resulted in a substantial amount of NSF₃ and effectively avoided environmental pollution by carcinogenic benzene. However, the purification process of gases remains to be further explored.

Effects of fluidizer on the yield of thiazyl trifluoride

This reaction was a combination of a S–N compound with a double-wedge cage structure and an active fluoride ion provided by silver difluoride. The reaction procedure is shown in Fig. 5. Apparently, the reactivity and mass of the active fluoride ion played key roles in the yield of NSF₃. We had already tried different kinds of fluorinating agents, such as KF (with 18crown-6 as the phase transfer catalyst), CsF and AgF₂. The target product NSF₃ could only be obtained by AgF₂. It was worth noting that AgF₂ was highly susceptible to deterioration during



Fig. 5 Pathway for the synthesis of thiazyl trifluoride from tetrasulfur tetranitride.

the loading process, which would apparently result in the loss of a considerable amount of raw material, resulting in a low yield. Therefore, we investigated the effects of AgF_2 on the yield. As shown in Table 5, the yield of NSF3 was found to increase from 21.32% to 57.38% (entries 1-5) when the $AgF_2 : S_4N_4$ molar ratio increased from 8:1 to 16:1, owing to the increase in the concentration of F⁻. After that, the yield of NSF₃ approached a steady state at approximately 58% upon further addition of AgF_2 (entry 6). Based on our findings, it could be concluded that the optimal molar ratio of AgF_2 to S_4N_4 was 16 : 1, considering economic factors and reaction efficiency.

Effects of reaction conditions on the preparation of thiazyl trifluoride

NSF₃ was synthesized from S₄N₄ and AgF₂ by ring-opening fluorination; the three S-F single bonds were formed in a stepwise manner, which meant insufficient reaction time would result in a low crude yield of the product. Consequently, it was necessary to keep the reaction stable at a constant temperature for a period of time. William H. Kirchhoff reported that the whole process was approximately 3 hours. However, it was found that the yield increased from 28.12% to 57.15% as the reaction time was extended to 2 hours (as shown in Table 6). Further extension of the reaction time had no significant effect on yield; thus, the time should be controlled for approximately 2 hours to obtain the highest yield and maximum economic benefit.

To purify the target gas, GC-MS was used to identify the impurities in the crude product, and SF₆, CCl₃F and SO₂ were confirmed to be the main impurities. The multistage condensation method was used for further purification, in conjunction with a potassium permanganate solution and anhydrous calcium chloride. In consideration of the boiling point of NSF₃ and other impurity gases, ice-salt baths with different temperature gradients for condensation were set up, and the impacts on the purity are displayed below. By comparing the results shown in Table 7, it was found that the purity increased first and then decreased as the temperature decreased. Moreover, the purity of NSF₃ reached 90% when the condensing temperature was -15 °C. It was speculated that this result was because impurities whose boiling points were higher than that of NSF₃ were easier to liquefy and separate during the temperature reduction process. However, the target gas was also liquefied, and the gas flow rate decreased as the condensing temperature

Table 5 Effects of the AgE_{2} : S_4N_4 molar ratio on the yield of NSE_4

Entry	Time ^b /h	Yield ^c /(%)
1	0.5	28.12
2	1	35.35
3	1.5	43.03
4	2	57.15
5	2.5	57.54
6	3	57.68

^a Reaction conditions: S₄N₄ (3 mmol), AgF₂ (48 mmol), and CCl₄ (waterless, 30 mL). The reaction was carried out under stirring at 78 °C. ^b Timing began after AgF₂ was added. ^c Isolated yield, measured by GC-MS.

gradually approached the boiling point of NSF₃, owing to the absence of an inert carrier gas, thereby leading to a decrease in the overall purity of the product. Therefore, it was suggested that the first-stage condensation temperature should be -15 °C to maximize the purity of NSF₃.

Hence, NSF₃ was successfully synthesized via a two-step reaction using sulfur monochloride, ammonia and silver(n)fluoride as raw materials. The total yield of NSF3 was 25% under the optimal conditions, and the purity was over 90%.

Assessment of the thermal stability of thiazyl trifluoride

To evaluate the thermal stability of thiazyl trifluoride, normal vibration mode analysis was performed, and the vibration free energy at 298 K was estimated to predict the reaction free energy of the radical decomposition. The simulation of the gas phase radical decomposition reaction was performed with Gaussian

Table 7 Effects of condensing temperature on the purity of NSF_3^{a}

Entry	Condensing temperature/°C	Purity ^b /%
1	-5	49
2	-10	60
3	-15	90
4	-20	82
5	-25	75

^{*a*} Reaction conditions: S_4N_4 (3 mmol), AgF_2 (48 mmol), and CCl_4 (waterless, 30 mL). The reaction was carried out under stirring at 78 °C for 2 hours, and the gas mixture was condensed at different temperatures. ^b GC purity.

Entry	Molar rate (AgF_2/S_4N_4)	Reaction time/h	Temperature/°C	Yield ^b /(%)	
1	8:1	2	78	21.32	
2	10:1	2	78	32.32	
3	12:1	2	78	44.79	
4	14:1	2	78	53.44	
5	16:1	2	78	57.38	
6	18:1	2	78	58.27	

^a Reaction conditions: S₄N₄ (3 mmol), CCl₄ (waterless, 30 mL), and AgF₂ were added in the specified proportions under stirring. ^b Isolated yield.

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09 software. The geometries of all molecules were optimized by B3LYP/6-31G(d,p). The convergence threshold for geometry optimization was tighter than the default value (opt = tight), and tight grid integration was applied to perform DFT simulation (int = ultrafine).

The schematic illustration of radical decomposition is shown in Fig. 6, and we consider two types of bond cleavage between N–S and S–F to assess all the decomposition patterns.

All the structures of molecules after geometry optimization are listed in Fig. 7. For the first step of radical decomposition, one of the F or N atoms is deleted, forming a single radical molecule, which results in the NSF_2 and SF_3 as shown in Fig. 7B and C.

The reaction free energy for the radical decomposition reaction of the NSF₃ molecule is shown in Table S2.[†] The bond cleavage between F and S produces the lowest reaction energy (58.22 kcal mol⁻¹) and decomposition into molecules B. The other pathway produces molecules C and nitrogen radical, resulting in a reaction energy higher than 58.22 kcal mol⁻¹. Thus, it is difficult to decompose (especially radical nitrogen atom). The decomposition from SF₃ to SF₂ (as shown in Fig. 7D) and F radical has the lowest reaction energy in this system. All the simulation results listed above suggest the formation of SF₂ radical. Furthermore, it has been confirmed that the thermal



Fig. 6 Schematic illustration of the radical decomposition of a molecule.



Fig. 7 Geometry of molecules after optimization with B3LYP/ 631G(d,p). (A) Geometry of NSF₃ molecules. (B) Geometry of NSF₂ after the first step of decomposition (losing a fluorine atom). (C) Geometry of SF₃ after the first step of decomposition (losing a nitrogen atom). (D) Geometry of SF₂ resulting from the decomposition of SF₃.

unimolecular decomposition or isomerization of NSF₃ is negligible even at extremely high temperatures (*e.g.*, \sim 3000 K whenever partial discharge or arc occurs).¹⁶

Comparison with other insulating gases

A comparison of the physical and environmental properties of NSF₃ and other mainstream potential substitute gases is shown in Table S3.[†] Although c-C₄F₈ has desirable insulating properties, the GWP remains high at 8700. Besides, c-C₄F₈ discharges at high energy may precipitate carbon, which may affect the insulation capacity. C₅F₁₀O and C₄F₇N have disadvantages of their high boiling point (26.5 °C and -4.7 °C, respectively), so they need to be mixed with buffer gases to avoid the obstacle of high boiling point to a certain extent. The gas CF₃I exhibits high dielectric strength, low boiling point and GWP. However, it has been proven that iodine precipitation can be produced with high voltage and severe discharge, which may lead to a decline of dielectric strength. It can be concluded from the comparison that the performance of NSF₃ in various aspects is relatively excellent and has certain application prospects.

Conclusions

In this paper, an efficient and convenient synthesis route of thiazyl trifluoride from sulfur monochloride was developed. It is worth noting that this compound has the potential to be a novel eco-friendly insulating gas to replace SF_6 in high-voltage transmission systems. Sulfur monochloride reacted with ammonia in an ice bath to obtain tetrasulfur tetranitride. Afterwards, tetrasulfur tetranitride was fluorinated by AgF_2 and treated with multistage condensation to produce thiazyl trifluoride. The conditions of the relevant reactions were also optimized to maximize the yield and purity. The overall yield of NSF₃ was approximately 25%, and the purity was up to 90.6%, which has not been mentioned in previous research. The dielectric strength of NSF₃ was confirmed to be 1.28 times that of SF₆ through electrical experiments, indicating that NSF₃ might be a potential alternative to SF₆.

Conflicts of interest

There are no conflicts to declare.

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