

Article

The Removal of Single and Binary Components of Hydrogen Sulfide and Dimethyl Disulfide in a Post-Plasma Catalysis Reactor: The Correlation of the Ozone Demand Factor with Specific Input Energy

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Abstract

Odorous gases such as hydrogen sulfide (H₂S) and dimethyl disulfide (DMDS) pose significant risks to human health and environmental quality. Non-thermal plasma (NTP) technology offers an effective alternative for odor treatment, but challenges remain regarding byproduct formation (e.g., ozone) and energy efficiency. In this study, a post-plasma catalysis (PPC) system combining a wire cylinder pulsed corona discharge reactor with a CuO/MnO₂-C ozone decomposition catalyst was employed to remove single and binary components of H₂S and DMDS. The effects of specific input energy (SIE), inlet concentration (C_{in}), and catalyst on removal efficiency, energy yield, and ozone emission were systematically investigated. A novel parameter, the ozone demand factor (Df), was defined to quantify the relationship between ozone consumption and pollutant removal. The results show that for single-component removal, regulating SIE/C_{in} within specific ranges (0.22–0.25 for H₂S and ~0.72 for DMDS) enables simultaneous low outlet concentrations of both pollutants and ozone. For binary H₂S-DMDS mixtures, ln(Df) exhibits a strong linear positive correlation with SIE/Σ(C_{in}) (R² = 0.968), and controlling SIE/Σ(C_{in}) at 0.28 yields ln(Df) ≈ 4, ensuring outlet H₂S < 5.0 ppm, DMDS < 3.0 ppm, and ozone < 5.0 ppm. This study demonstrates that the ozone demand factor serves as a critical control parameter for optimizing PPC systems, providing a practical tool for balancing pollutant removal and ozone emission in industrial odor treatment applications.

Keywords: post-plasma catalysis; ozone demand factor; specific input energy; non-thermal plasma



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1. Introduction

Odorous gases, which are mainly emitted from oil refineries, pulp and paper industry, sewage and wastewater treatment plants, and so on, are highly toxic and unpleasant and have a very low odor threshold value (OTV) [1–3]. It is easy for such gases to have a large negative impact on human health and environmental quality. Various approaches have been proposed to remove malodorous compounds from air streams, such as chemical scrubbing, incineration, adsorption and biofiltration [4–7]. However, there are still some technical and economic limitations due to their poor adaptability to different odors.

Non-thermal plasma (NTP) technology has been reported as a reliable alternative to conventional odor treatment methods [8–10]. NTP, which is produced by strong electric discharges and consists of electrons, radicals, neutral particles and ions, can non-selectively attack odor gas molecules. Cui et al. found that a system using a MnCe/Ti catalyst to fill a DBD reactor removes NO and SO₂ simultaneously, resulting in high removal efficiency and low energy consumption [11]. Wu et al. reported the complete elimination of ammonia and organic sulfides under an SEI lower than 4.44 J/L in the innovative multineedle-to-cylinder DBD plasma [12]. Wang and Sun studied NO_x removal in a DBD reactor and improved the NO_x removal efficiency by adding C₂H₂ [13]. However, one issue that should be raised is that NTP alone for gas purification is limited by inferior product selectivity, harmful byproducts (such as O₃, NO_x, and aerosols) and low energy efficiency [14,15]. Plasma–catalyst coupling, including in-plasma catalysis (IPC, the catalyst in the discharge zone) and post-plasma catalysis (PPC, the catalyst after the discharge zone), is considered an effective approach to overcome these drawbacks [16,17]. Of the two, the PPC system is the more widely used exhaust gas treatment system today because of its simplicity of operation, practicality and more effective byproduct control. In PPC, since high-energy electrons and short-lived active species (e.g., atomic oxygen, ions, hydroxyl groups, etc.) generated in the plasma phase have difficulty reaching the catalyst surface and reacting with the catalyst downstream of the discharge zone, the pollutant removal efficiency in many cases depends mainly on the long-lived species (ozone).

Ozone production is dependent on the energy input to the plasma reactor [18–20]. The higher the energy input, the more ozone is produced and the more complete the removal of pollutants from the catalytic zone [21]. High pollutant removal efficiency, however, is usually accompanied by excessive ozone emissions, which are a serious hazard to the environment and human health [22,23]. The trade-off between the odor gas removal efficiency and ozone emissions is a major challenge [24,25]. In addition, plasma catalysis is a complex process, and its performance is controlled by many process parameters. Previous work has focused on experimental studies evaluating the effects of individual process parameters on the performance of plasma catalysis. The lack of understanding of the overall relationship between operation parameters and between operation parameters and control objectives makes it difficult to optimize operation parameters comprehensively. Therefore, how to control and coordinate different operation parameters of the PPC system to improve ozone utilization is a key issue in achieving the simultaneous removal of pollutants and ozone.

In this paper, the PPC method was used to study the removal of odorous gases. Hydrogen sulfide (H₂S), dimethyl disulfide (DMDS) and their mixtures were selected as target contaminants. A wire cylinder pulsed corona discharge reactor was used to generate a non-thermal plasma. We investigated the relationship between operating parameters (specific energy input (SIE), inlet concentration (C_{in}), etc.) and the removal performance of binary sulfide malodor gases (H₂S + DMDS). Additionally, the ozone demand factor (Df) was defined as the key parameter of the coupling process, which determines the exit concentration of odor gases and ozone. For the first time, a relationship between ozone data and plasma-catalytic operating parameters (Df and SIE/C_{in}) was established. This relationship can be used to regulate or predict the PPC process of malodorous gases. It is hoped that this research will contribute to the removal of mixed malodor gases.

2. Results and Discussion

2.1. The Removal of a Single Component of Hydrogen Sulfide and Dimethyl Disulfide

2.1.1. H₂S Removal

In the absence or presence of downstream catalysts, the outlet H₂S concentration with different initial concentrations (198.2, 249.8 and 343.1 ppm) as a function of specific input energy is shown in Figure 1a. It is obvious that the removal of H₂S can be promoted by decreasing the inlet concentration with or without a catalyst. For example, H₂S at an inlet concentration of 198.2 ppm could be completely removed by plasma alone at 45 J/L, while the required concentrations were 53 and >78 J/L at inlet concentrations of 249.8 and 343.1 ppm. For a given initial concentration, the outlet concentration of H₂S decreased as the SIE increased. This can be explained by the fact that increasing SIE contributes to more energetic electron and reactive species generation such as $\cdot\text{O}$, OH , and excited N_2 , which in turn leads to more inelastic collisions of H₂S molecules [26–29].

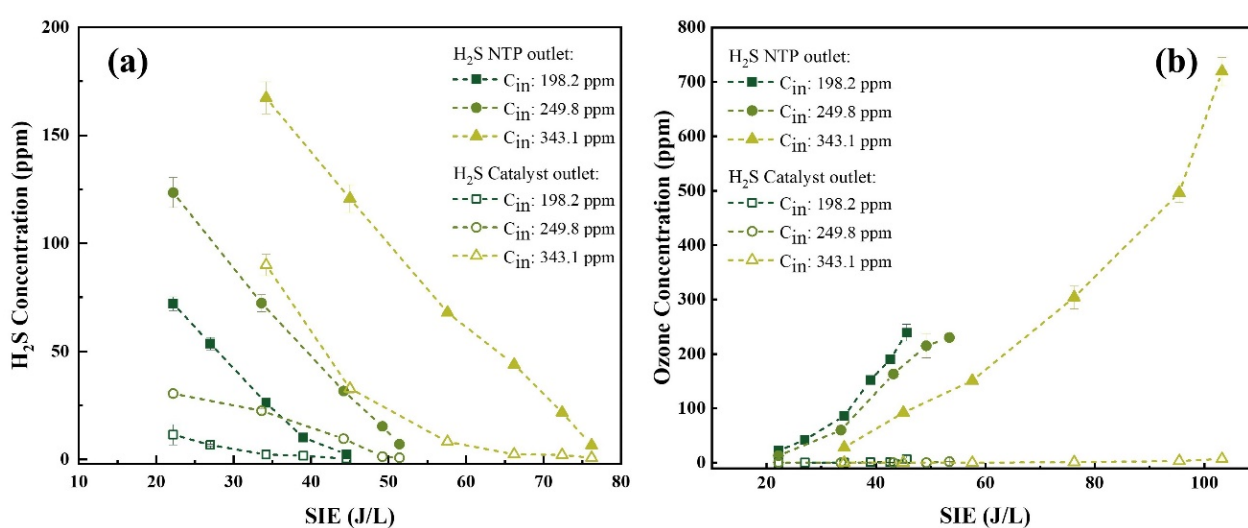


Figure 1. Under different systems, the concentrations of H₂S (a) and O₃ (b) at the outlet of the reactor vary with SIE.

In the simple NTP system, the H₂S removal efficiency increases significantly with the increase in SIE, but relative to the coupled NTP–catalyst reaction system, there is also a significant increase in the ozone concentration at the reactor outlet (as shown in Figure 1b). Compared to NTP alone, hydrogen sulfide is more easily removed in the PPC system. For >90% H₂S removal at the inlet concentration of 198.2 ppm, the SIE decreases from 44.6 J/L to 22.2 J/L. In addition, combining plasma with a downstream catalyst promotes the decomposition of ozone and the heterogeneous reaction of ozone with H₂S. The outlet ozone concentration with the catalyst was significantly lower than that without the catalyst, and the outlet ozone concentration remained almost constant at approximately 0–6 ppm regardless of the inlet concentration.

At a certain SIE, reducing the initial concentration of H₂S can improve the H₂S removal efficiency, but it also increases the concentration of ozone in the exhaust gas. For example, when the initial concentration of hydrogen sulfide is 249.8 ppm and the SIE is 53.4 J/L, the outlet concentration of hydrogen sulfide can reach a lower level (7.1 ppm, corresponding to a removal efficiency of 97.1%), but the outlet ozone concentration reaches 230 ppm. when the initial concentration of hydrogen sulfide is 198.2 ppm and the SIE is 22.2 J/L, the outlet ozone concentration was only 22 ppm, but the outlet hydrogen sulfide concentration reached 72.1 ppm (only equivalent to 63.6% of the removal efficiency). The above results show that there is a certain limitation and conflicting relationship between the pollutant

removal efficiency and ozone concentration in a simple plasma treatment system. Moreover, it is difficult to keep the outlet H_2S and ozone concentrations simultaneously at low levels.

2.1.2. DMDS Removal

Similar to the trend of H_2S removal, the DMDS removal efficiency seems to be strongly dependent on the SIE and catalyst (as shown in Figure 2a). To keep the outlet DMDS below 10 ppm, the SIE required for plasma treatment alone was 48, 86, and 100 J/L for inlet concentrations of 57.4, 89.6, and 135.1 ppm, while that for plasma-catalytic treatment was 35, 58, and 65 J/L. As seen in Figure 2b, regardless of the inlet concentration, in the plasma-catalytic system, the outlet ozone concentration remains at a low level (<10 ppm), while with increasing SIE, it reaches more than 100 ppm at >40 J/L in the plasma-only system. The presence of the catalyst reduces the energy required for DMDS degradation and increases O_3 consumption. For a given SIE, a higher inlet DMDS concentration corresponds to a lower outlet ozone concentration in a separate plasma system. For the same initial DMDS concentration, the DMDS removal efficiency, O_3 concentration and SIE were positively correlated.

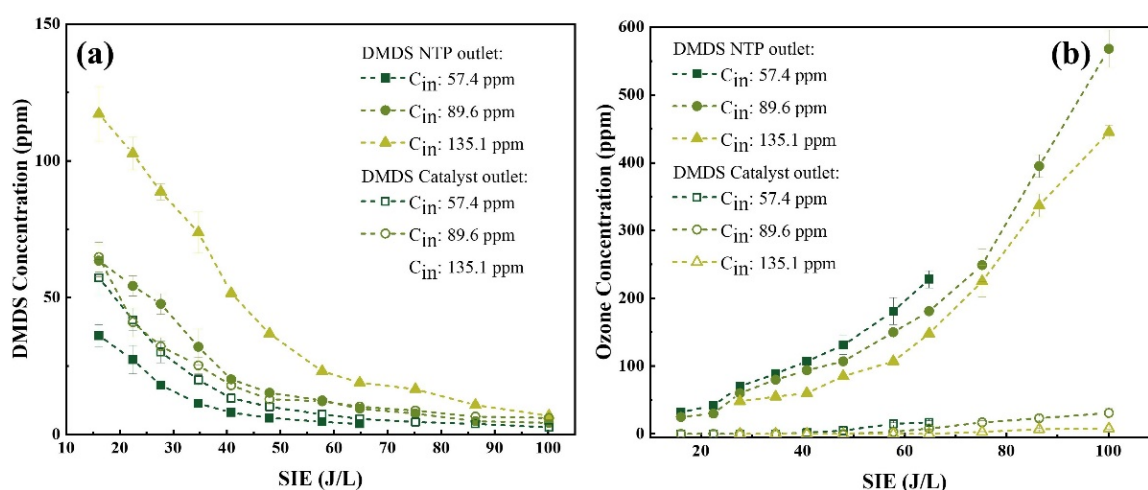


Figure 2. Under different systems, the concentrations of DMDS (a) and O_3 (b) at the outlet of the reactor vary with SIE.

2.1.3. Sulfur Balance

The SO_2 in the outlet gas was measured in a PPC system with H_2S as the contaminant. Figure 3a depicts the variation in H_2S and SO_2 concentrations for different specific input energies when the inlet concentration of H_2S is fixed at 198.2 ppm. The outlet H_2S concentration decreases with increasing SIE, while the outlet SO_2 concentration shows an increasing trend. However, compared with the inlet concentration of hydrogen sulfide, there is still an approximately 15% loss of elemental sulfur in the total outlet concentration of hydrogen sulfide and sulfur dioxide. It may be attributed to the following reasons. First, some elemental sulfur exists in the gas phase in other forms (e.g., SO_3 , H_2SO_4 , etc.). Second, elemental sulfur (elemental sulfur or other bound states) may be deposited on the surface of the catalyst.

For DMDS, the experimental results for sulfur equilibrium were similar to those for hydrogen sulfide (as shown in Figure 3b). The concentration of elemental sulfur in the DMDS removal system decreased with increasing SIE, while sulfur dioxide showed an increasing trend. However, the total concentration of elemental sulfur in the exported DMDS and sulfur dioxide is still 13.1% to 34.8% lower than the concentration of elemental sulfur in the imported DMDS. Similarly, the reasons may be summarized as follows: first,

the conversion of the product SO_2 to SO_3 , H_2SO_4 and other gaseous products; second, the attachment of some sulfur elements to the catalyst surface in some form; and third, the generation of various organic products such as CH_3SH , CH_3OH and CH_3SCH_3 during plasma degradation of DMDS (see Table S1). In addition, compared to the stable conversion of hydrogen sulfide of approximately 80%, the elemental conversion of sulfur in DMDS changed significantly. When the SIE was 22.4 J/L, the conversion of elemental sulfur was only 48.3%, while the conversion of elemental sulfur reached 80.1% at an energy density of 64.8 J/L. The results indicate that DMDS is more difficult to oxidize to SO_2 than H_2S , and more DMDS is degraded to intermediate products or deposited on the catalyst surface at low SIE.

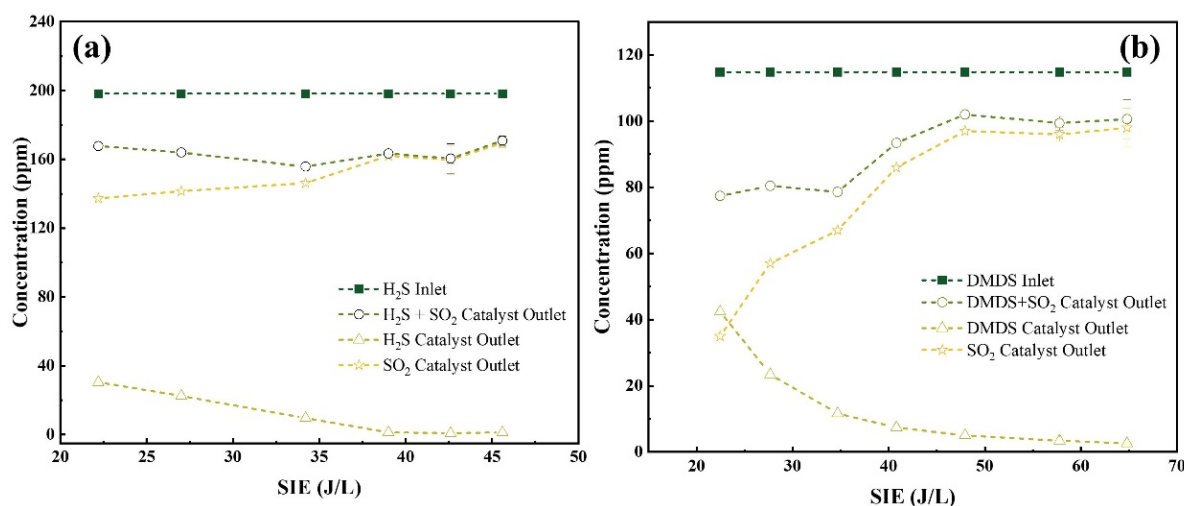


Figure 3. Sulfur balance in the H_2S removal system (a) and DMDS (b) removal system.

Although the toxicity of sulfur-containing byproducts (such as SO_2 , SO_3 , and organic sulfides) was not directly assessed in this study, it is noteworthy that SO_2 concentrations were monitored and remained below 10 ppm under optimized conditions. The formation of potentially more toxic intermediates (e.g., CH_3SH) was identified by GC-MS (Table S1), albeit at low relative abundances. A comprehensive ecotoxicological assessment of the treated gas stream would be a valuable direction for future research.

To investigate whether the S element adheres to the catalyst, the catalyst was characterized before and after the reaction in this study. The pore size distribution (Figure 4a–c) showed that the average pore size of the catalyst was 4.7 nm, and most of the sizes were in the range of 2–10 nm. Compared with the fresh catalyst, the BET specific surface area, pore volume and average pore size of the catalysts were significantly decreased after the reaction (Table 1). This may be due to the change in internal structure caused by the reaction between the contaminants and the catalyst, or the pores were blocked by the byproducts, which is consistent with the above analysis. As shown in Figure 4d–f, the SEM results showed that the catalyst surface became relatively smooth after the reaction, which is also consistent with the BET characterization results. The energy dispersive spectrometry (EDS) results of the catalyst before and after the reaction indicate the presence of active phases dominated by Cu and Mn elements on the surface of the activated carbon. In addition, the content of elemental S in the catalyst after the reaction was significantly increased (approximately 3 times) compared to that of the fresh catalyst (Figure S3 and Table S2). Combining the above characterization results and sulfur equilibrium analysis, we can conclude that sulfur elements are deposited in the catalyst through a series of catalytic reactions, blocking part of the pores of the catalyst, but the exact morphology needs to be further investigated.

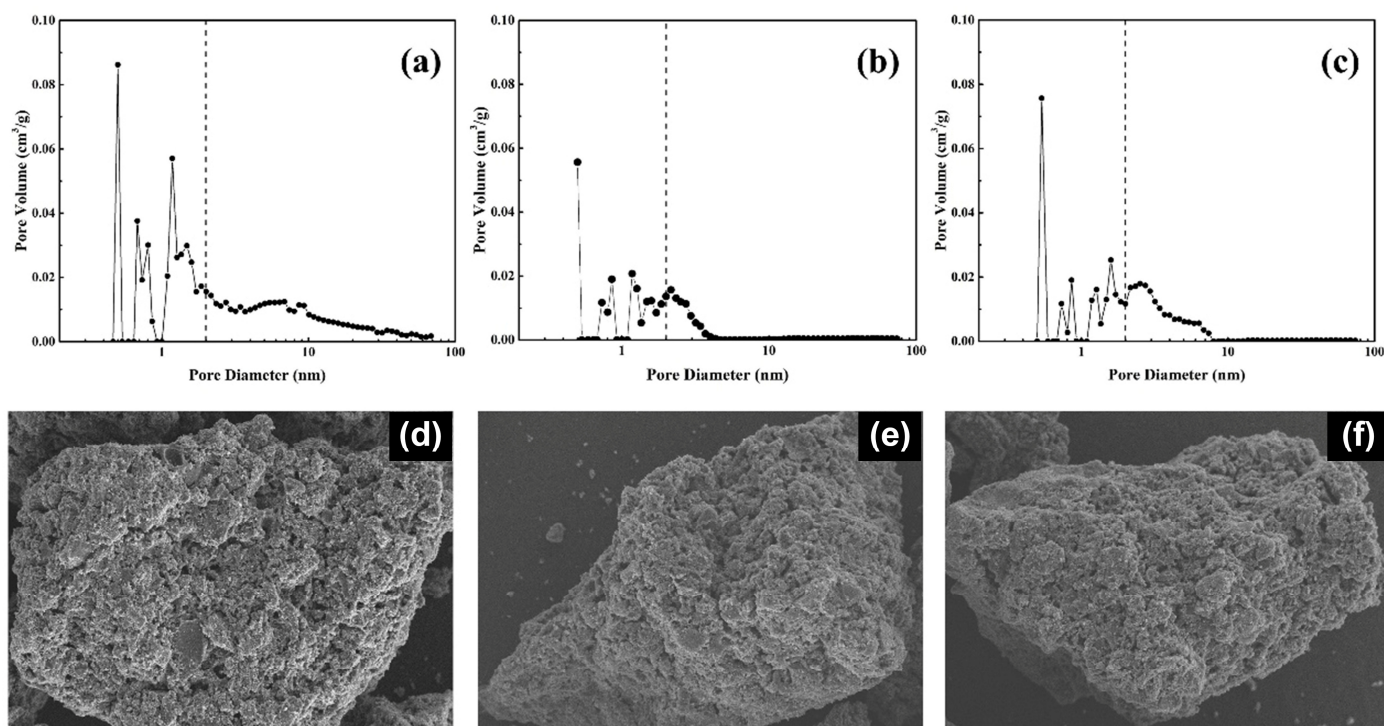


Figure 4. Characterization results of fresh and reacted CuO/MnO₂-C catalysts: pore size distribution (a–c); SEM micrographs (d–f).

Table 1. Textural properties of catalysts before and after reaction.

Catalyst	S _{BET} (m ² /g)	V _{micro} (cm ³ /g)	V _{meso} (cm ³ /g)	D _p (nm)
Before	1329.5	0.476	0.587	4.698
After (H ₂ S)	636.6	0.094	0.263	3.208
After (DMDS)	677.4	0.127	0.301	3.714

S_{BET}, V_{micro}, V_{meso} and D_p represent BET surface area, micropore volume, mesopore volume and average pore diameter.

Regarding practical applicability, the catalyst demonstrated good initial activity for ozone decomposition and pollutant removal. However, long-term stability tests are necessary for industrial implementation. Based on the BET and EDS analyses, sulfur deposition on the catalyst surface led to pore blockage and a reduction in specific surface area after prolonged use. Future work should focus on developing regeneration strategies (e.g., thermal treatment or washing) to restore catalyst activity, as well as exploring more sulfur-resistant catalyst formulations.

2.2. Simultaneous Removal H₂S and DMDS

The degradation of the binary component sulfur malodor gas (H₂S + DMDS) was carried out at an inlet concentration of 343.1 ppm H₂S and different inlet concentrations of DMDS (0, 14.5, 35.6, 57.1 ppm). As shown in Figure 5a, the outlet H₂S concentration has a slight increasing trend in the plasma-catalytic system when the inlet DMDS concentration increases. This indicates that the catalytic efficiency of hydrogen sulfide decreases slightly with increasing DMDS. The mechanisms of non-thermal plasma degradation can be divided into two categories: high-energy electron crushing of molecules and oxidation of pollutants or molecular fragments by active species (“crushing” and “oxidation”). H₂S, as a small-molecule pollutant, is more easily degraded than DMDS. For H₂S, the active species are sufficient to remove it completely, and the removal mechanism is mainly “oxidation”, while

“crushing” is auxiliary [30]. In addition, the molecular structure of DMDS is relatively complex and needs to be broken down into molecular fragments and then oxidized into small non-polluting molecules. The degradation mechanism of DMDS is mainly “oxidation” first and “oxidation” later. In summary, it can be inferred that the removal mechanisms of H_2S and DMDS are different, and there is no obvious competition between H_2S and DMDS in the plasma-catalyzed process. Therefore, the low concentration of DMDS has little effect on the removal of hydrogen sulfide.

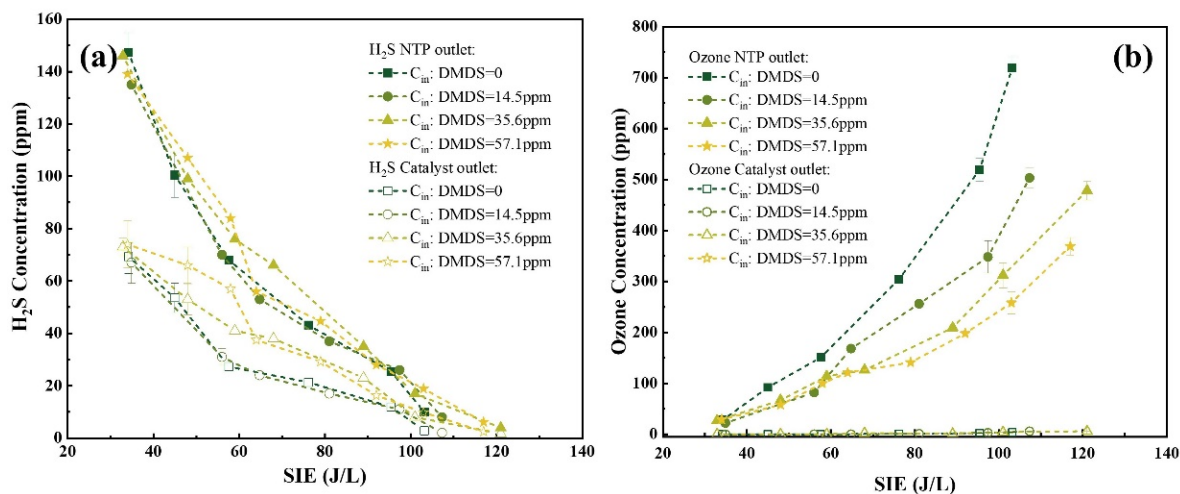


Figure 5. Variation in outlet H_2S concentration (a) and outlet ozone concentration (b) with SIE under mixed H_2S -DMDS binary system with different inlet DMDS concentrations.

As illustrated in Figure 5b, in the stand-alone plasma system, the outlet ozone concentration decreases as the inlet DMDS concentration increases. This indicates that the increase in inlet DMDS concentration leads to more depletion of ozone in the plasma. Nevertheless, in this experiment, the ozone entering the catalyst bed was more than sufficient, and the pollutant removal efficiency in the plasma-catalytic system decreased only slightly with increasing DMDS.

As shown in Figure 6, the total energy yield increases significantly with increasing DMDS concentration. This indicates that the total amount of pollutants degraded per unit of energy consumption increases significantly after mixing, and the energy use efficiency increases. It is also worth noting that the total energy production decreases as SIE increases, regardless of the DMDS concentration. Therefore, balancing the relationship between energy efficiency and removal efficiency is also a challenge for non-thermal plasma combined with catalyst technology. Table 2 shows a comparison of the energy yield of a single pollutant (H_2S or DMDS) and a mixture of them under the same conditions. The results show that when H_2S and DMDS are treated together, the energy output and their sum are reduced. This proves energetically that there is an inhibitory effect between H_2S and DMDS and does not conflict with the above discussion. Since the concentration of hydrogen sulfide was much higher than that of DMDS, the presence of DMDS did not significantly reduce the removal efficiency of hydrogen sulfide (Figure S1), but the presence of hydrogen sulfide significantly reduced the degradation efficiency of DMDS.

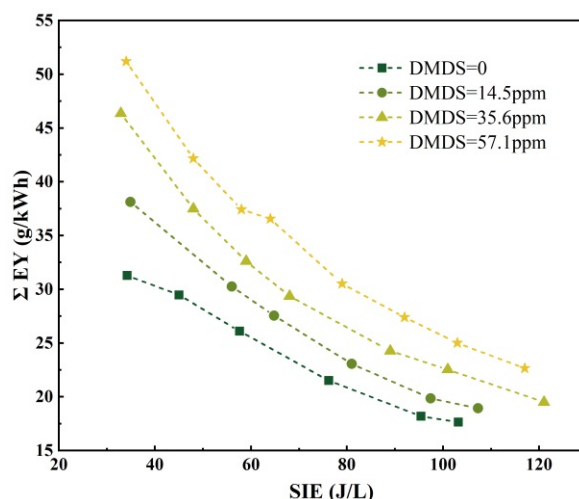


Figure 6. Total energy yield of mixed H₂S-DMDS malodorous gas as a function of SIE under different inlet DMDS concentrations.

Table 2. Comparison of energy yield between single and binary components at different SIE.

SIE (J/L)	EY _H (g/kWh)	EY _D (g/kWh)	EY _H + EY _D (g/kWh)	EY _{mixH} (g/kWh)	EY _{mixD} (g/kWh)	EY _{mix} (g/kWh)
34.7	31.28	20.08	51.36	30.80	14.76	45.56
47.9	29.47	16.20	45.67	27.87	14.39	42.26
57.8	26.09	13.80	39.89	24.41	11.02	35.43

EY_H: Single-component H₂S energy yield; EY_D: Single-component DMDS energy yield; EY_H + EY_D: Sum of energy yields; EY_{mixH}: H₂S energy yield under dual-component conditions; EY_{mixD}: DMDS energy yield under dual-component conditions; EY_{mix}: Total energy yield under dual-component conditions.

2.3. Ozone Demand Factor (Df)

2.3.1. Df for the Single Component (H₂S or DMDS) Removal

As shown in Figure 7a, for different initial concentrations of hydrogen sulfide, the outlet concentration of hydrogen sulfide decreases with increasing $\ln(Df)$, and the outlet concentration of hydrogen sulfide stabilizes at a low level (<5 ppm) when $\ln(Df) > 3$. The relationship between ozone outlet concentration and $\ln(Df)$ during hydrogen sulfide removal is shown in Figure 7b. It is obvious that the ozone outlet concentration increases with increasing $\ln(Df)$. When $\ln(Df) < 4$, it has little effect on the ozone outlet concentration, which remains below 3 ppm, because the ozone is almost completely decomposed. In summary, by adjusting $\ln(Df)$ so that it is 3~4, the outlet concentration of hydrogen sulfide and ozone can be kept at a low level to achieve the simultaneous decomposition of H₂S and O₃. In addition to the possible reasons mentioned above, it may be that the ozone concentration is always higher than the hydrogen sulfide concentration, which is relatively insufficient, resulting in a higher theoretical value of Df.

As seen in Figure 7c, the decomposition of DMDS was carried out at three different inlet concentrations of 57.4, 89.6, and 135.1 ppm. When $Df > 25$, the outlet concentration of DMDS can be kept below 10 ppm. However, at the same time, when $Df < 25$, the ozone outlet concentration can be kept below 5 ppm (Figure 7d). Therefore, it can be concluded that controlling the ozone demand factor of DMDS to approximately 25 can ensure the low outlet concentration of DMDS and ozone to achieve the best coupling effect.

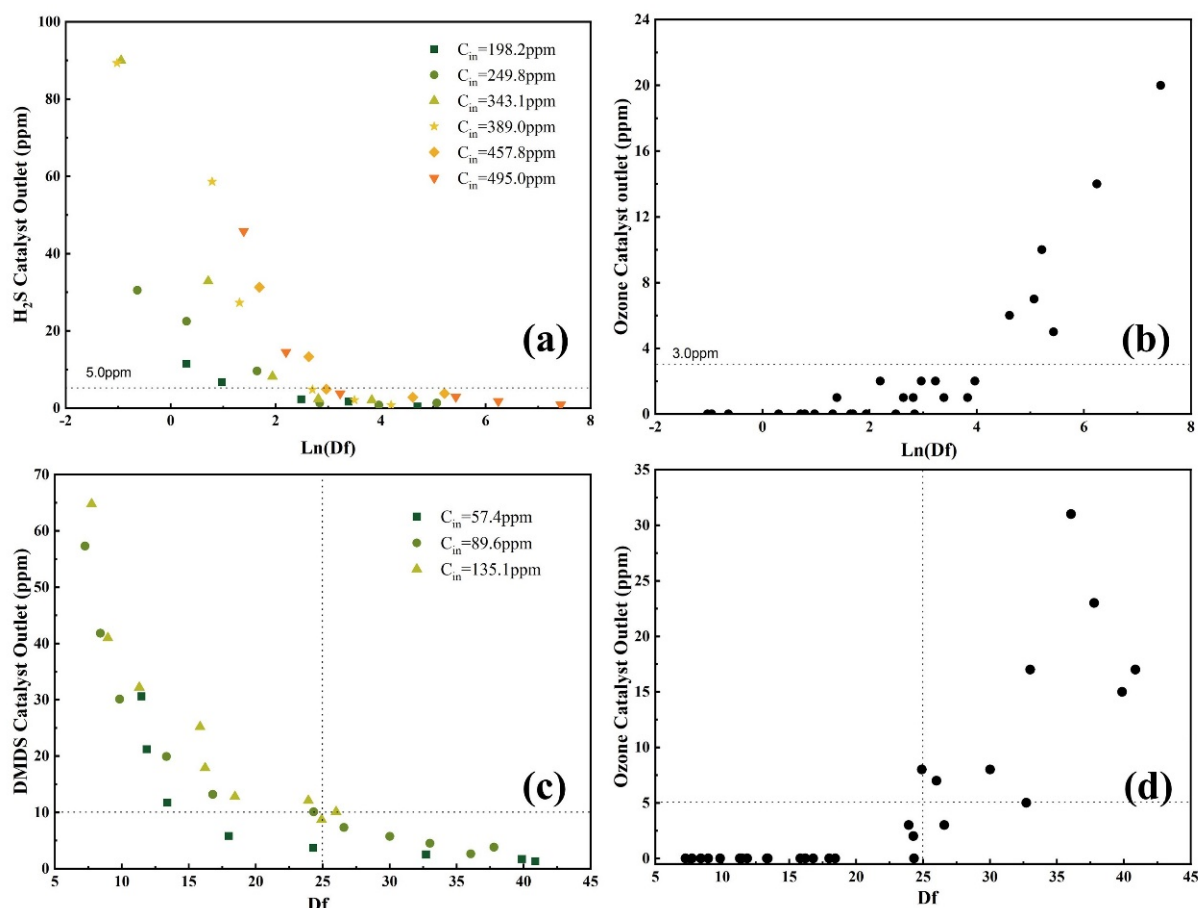


Figure 7. Relationship between Df, outlet pollutant concentration and outlet ozone concentration for single-component sulfide removal: H₂S (a,b); DMDS (c,d).

Figure 8a shows the relationship between hydrogen sulfide removal and ozone consumption in a separate plasma system. The results show a significant positive correlation between $\Delta\text{H}_2\text{S}$ and ΔO_3 ($R^2 = 0.958$) regardless of the variation in the inlet concentration (C_{in}) and the specific input energy (SIE). That is, for a specific PPC system (including catalyst, target contaminant and reactor), the value of Df is determined (the relationship between the ozone yield and SIE shown in Figure S2). The logical relationships of the different parameters in the plasma-only system are shown in Figure 8b. The inlet concentration (C_{in}) and the specific input energy (SIE) are the main factors that directly affect the O₃ and pollutant concentrations at the NTP outlet, which in turn affect the value of the ozone demand factor in the catalytic process. Therefore, it can be speculated that there is a certain functional relationship between SIE, C_{in} and Df, i.e., $Df = f(\text{SIE}, C_{\text{in}})$.

The data for H₂S and DMDS were processed and analyzed separately, and the results are shown in Figure 8c,d. For hydrogen sulfide, $\ln(Df)$ was linearly and positively correlated with SIE/C_{in} ($R^2 = 0.92$); when SIE/C_{in} was controlled at 0.22–0.25, the outlet concentrations of hydrogen sulfide and ozone could be lower than 5.0 ppm and 3.0 ppm, respectively. C_{in} also showed a linear positive correlation ($R^2 = 0.79$); when SIE/C_{in} was controlled at 0.72, the outlet concentrations of hydrogen sulfide and ozone could be lower than 10.0 ppm and 5.0 ppm, respectively.

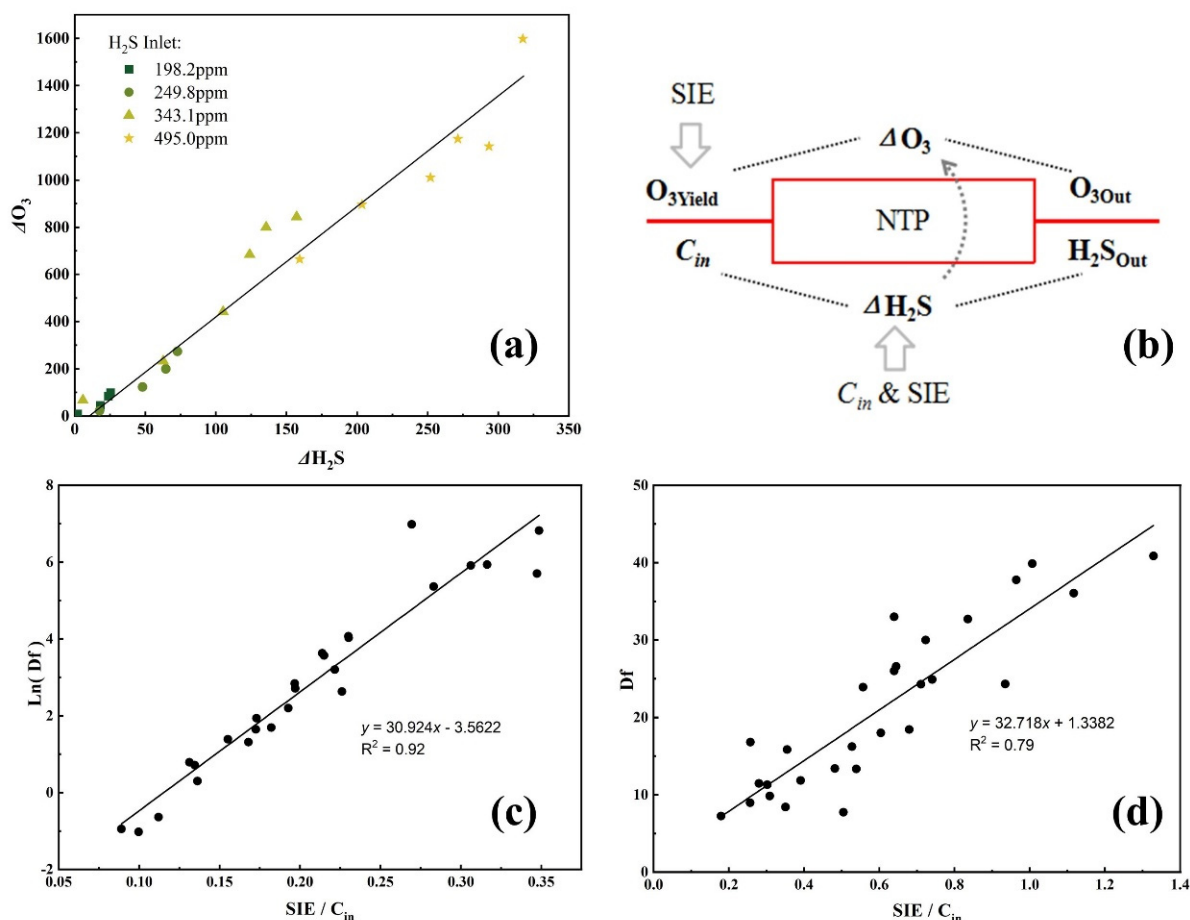


Figure 8. Correlation between ozone consumption and H_2S removal in single NTP system (a); parameter logical relationship of NTP-only system (b); linear fitting of $\ln(Df)$ versus SIE/C_{in} for single H_2S (c) and single DMDS (d).

2.3.2. Analysis of Df on Binary Component (H_2S and DMDS) Removal

The relationship between the ozone demand factor and the degradation characteristics of the binary sulfide odor gas ($H_2S + DMDS$) during the coupling process is shown in Figure 9. Similar to the single component hydrogen sulfide, the outlet H_2S and DMDS concentrations decrease as the natural logarithm of the ozone demand factor ($\ln(Df)$) increases. For different inlet DMDS concentrations, when $\ln(Df) < 4$, the outlet concentrations of hydrogen sulfide and DMDS were below 5.0 ppm and 3.0 ppm, respectively. The outlet ozone concentration remained below 5.0 ppm when $\ln(Df) > 4$. Therefore, for binary sulfide malodor gas, under the present experimental conditions (inlet H_2S concentration is 343.1 ppm and inlet DMDS concentration is below 57.1 ppm), $\ln(Df)$ should be controlled at approximately 4 to ensure that the pollutant and ozone are at a low level.

Based on the empirical relationship (positive correlation between SIE/C_{in} and Df (or $\ln(Df)$)) in Section 2.3.1, a similar relationship between $SIE/(C_{H_2S_{in}} + C_{DMDS_{in}})$ and $\ln(Df)$ is postulated for binary sulfide odor gases. The results are shown in Figure 9c. $\ln(Df)$ is linearly and positively correlated with $SIE/\Sigma(C_{in})$ ($\ln(Df) = 24.039 SIE/\Sigma(C_{in}) - 2.8342$, $R^2 = 0.968$). When $SIE/\Sigma(C_{in})$ is controlled at 0.28, $\ln(Df)$ is 4, and the outlet concentrations of hydrogen sulfide, DMDS and ozone can be lower than 5.0 ppm, 3.0 ppm and 5.0 ppm, respectively.

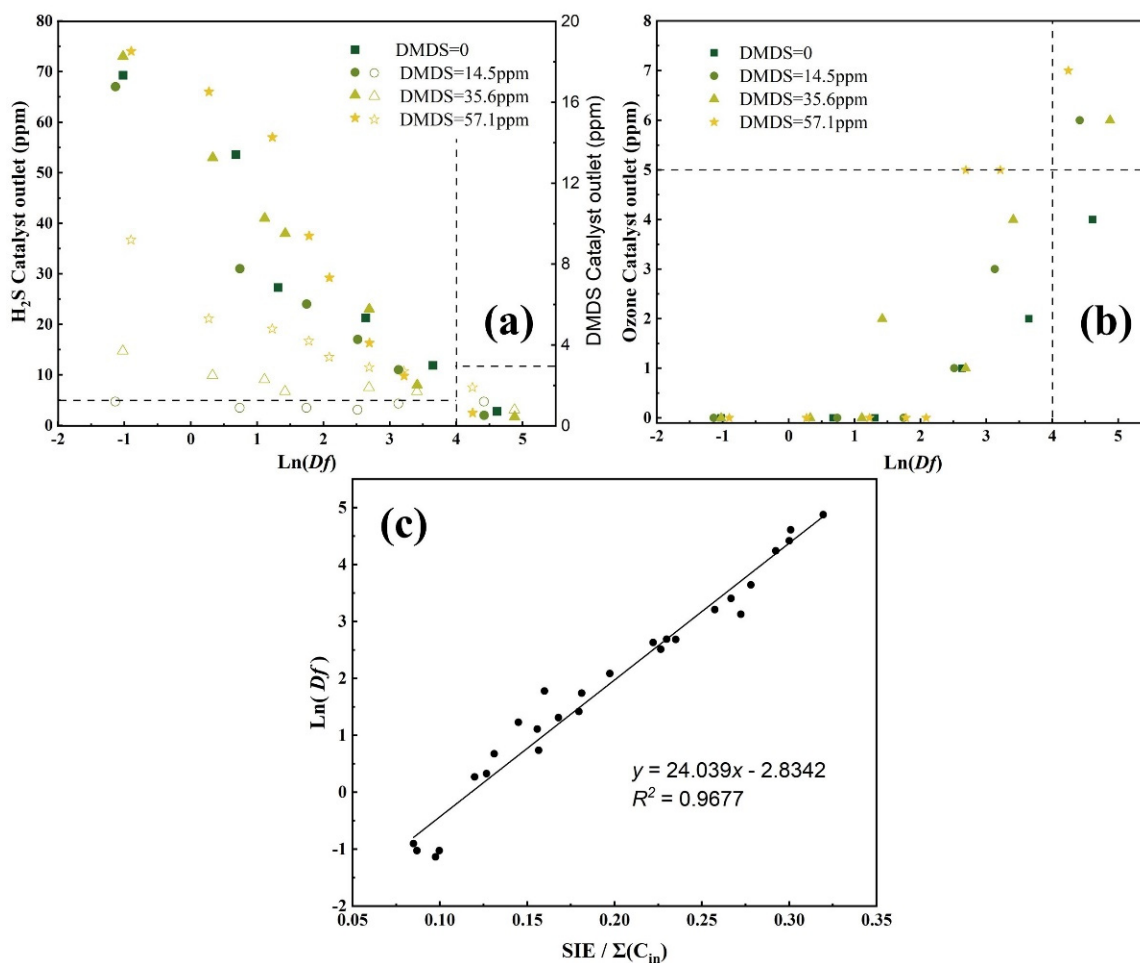


Figure 9. Variation in outlet pollutant and ozone concentration with $\ln(Df)$ (a,b); linear fitting of $\ln(Df)$ versus $SIE/\Sigma(C_{in})$ for binary H₂S-DMDS mixture (c).

Compared with the conclusion in Section 2.3.1 (hydrogen sulfide: SIE/C_{in} control between 0.22 and 0.25; DMDS: SIE/C_{in} control approximately 0.72), the $SIE/\sigma(C_{in})$ value of two-component sulfide odor gas should be controlled at 0.28, between H₂S and DMDS, in favor of hydrogen sulfide. This may be because the experiment is designed on the basis of simulated industrial exhaust gas, and the concentration of hydrogen sulfide in the two components is much higher than that of DMDS. Then, the degradation characteristics of the gas mixture are more similar to those of hydrogen sulfide.

3. Materials and Methods

3.1. Chemicals

All the chemicals used in this experiment were of analytical purity grade and supplied by Aladdin Corporation (Shanghai, China). Both nitrogen and oxygen (Nanjing Tianze Gas Co., Ltd., Nanjing, China) have a purity of 99.99%, while hydrogen sulfide has a purity of 99.5%.

A commercial ozone decomposition catalyst (CuO/MnO₂-C) was supplied by Wuxi Kaixi Catalyst Company, Wuxi, China. The catalyst was sieved to 20–40 mesh and used as received, except that it was dried at 105 °C for 12 h prior to each experiment to remove adsorbed moisture. This pretreatment minimizes the interference of adsorbed water with ozone decomposition and pollutant removal. The surface areas and pore size distributions of catalysts before and after reaction were measured by ASAP2020 (Micromeritics Instrument Corporation company, Norcross, GA, USA) using Brunauer–Emmett–Teller (BET)

and Barrett–Joyner–Halenda (DA) methods. The morphologies and microstructures of the catalysts were observed using field emission scanning electron microscopy (FESEM, S-3400NII, Hitachi, Ibaraki Prefecture, Japan). Additionally, the elements in the catalysts were measured using EDX analysis.

3.2. Experimental Setup

The flow diagram of the experimental system is shown in Figure 10. The system consists of a gas generator, NTP-catalytic reactor, power supply and gas analysis. The gas generator is composed of gas cylinders, mass flow controllers, bubble bottles and a mixing bottle. The dynamic distribution method was used to obtain the concentrations of H₂S, N₂, and O₂ required. To ensure comparability across different experimental conditions, the feed gas consisted of N₂ and O₂ mixed at a ratio of 80:20 (*v/v*) to simulate air. The total gas flow rate (*Q*) was maintained at 1.0 L/min throughout all experiments. The input concentration of DMDS and the relative humidity (RH) could be regulated by controlling the flow rate of N₂ passing through the bubblers. All gas streams were premixed before entering the reactor. Since the concentrations of H₂S and DMDS were relatively low (<400 ppm for H₂S and <140 ppm for DMDS), their effect on the bulk gas composition and thus on the discharge characteristics was minimal. The relative humidity (RH) was detected prior to the NTP reactor using a hygrometer (Testo 605-H1) with an accuracy of $\pm 3\%$ RH. A wire cylinder plasma reactor was adopted to produce a corona discharge. The effective working volume of the NTP reactor was 0.3 dm³ (65 mm inner diameter and 90 mm length). The NTP reactor body was a heat-resistant glass tube with aluminum foil attached to the inner wall as the grounding electrode. The NTP reactor was made of borosilicate glass, which is known for its high dielectric strength and thermal stability. While wall effects cannot be completely ruled out due to the relatively small reactor volume, we minimized their influence by regularly cleaning the reactor between experiments. Specifically, the reactor was rinsed with deionized water followed by ethanol, and then dried at 120 °C for 2 h. Visual inspection was performed periodically, and no significant deposits were observed on the reactor walls. The contribution of wall reactions to overall pollutant removal is expected to be negligible compared to the dominant gas-phase plasma reactions and subsequent catalytic oxidation. The discharge electrode was a 5 mm diameter stainless steel wire passing axially through the center of the NTP reactor. The NTP reactor was powered by a pulsed power supply that can generate high voltage pulses with an adjustable peak voltage of 0–30 kV and a variable frequency of 10–300 Hz. The voltage and the current were measured and recorded using a digital oscilloscope (Tektronix, TBS 1020, Beaverton, OR, USA) with a voltage probe (Tektronix P6015, Beaverton, OR, USA) and a current probe (Tektronix P6021, Beaverton, OR, USA). Representative voltage and current waveforms of the pulsed corona discharge are provided in Figure S4. The catalytic reactor was a quartz tube (7.5 mm inner diameter) packed with 10 g of CuO/MnO₂-C catalyst, corresponding to a catalyst bed volume of approximately 9 mL. The residence time was approximately 0.54 s at a total flow rate of 1.0 L/min. The heating device was located on the periphery of the catalyst reactor to maintain a constant temperature. The inlet and outlet concentrations of H₂S and DMDS were measured using a gas chromatograph (GC-9890A, Shanghai, China) equipped with a flame photometric detector (FPD). The concentration of O₃ was monitored by using an ozone analyzer (SKS-900, Guangzhou, China). All experiments were repeated at least three times, and the average values with standard deviations are reported. The organic byproducts of DMDS decomposition were analyzed using a gas chromatograph–mass spectrometer (Thermo Scientific trace GC ISQ, Waltham, MA, USA). The off-gas was purified by using an absorber containing concentrated NaOH solution.

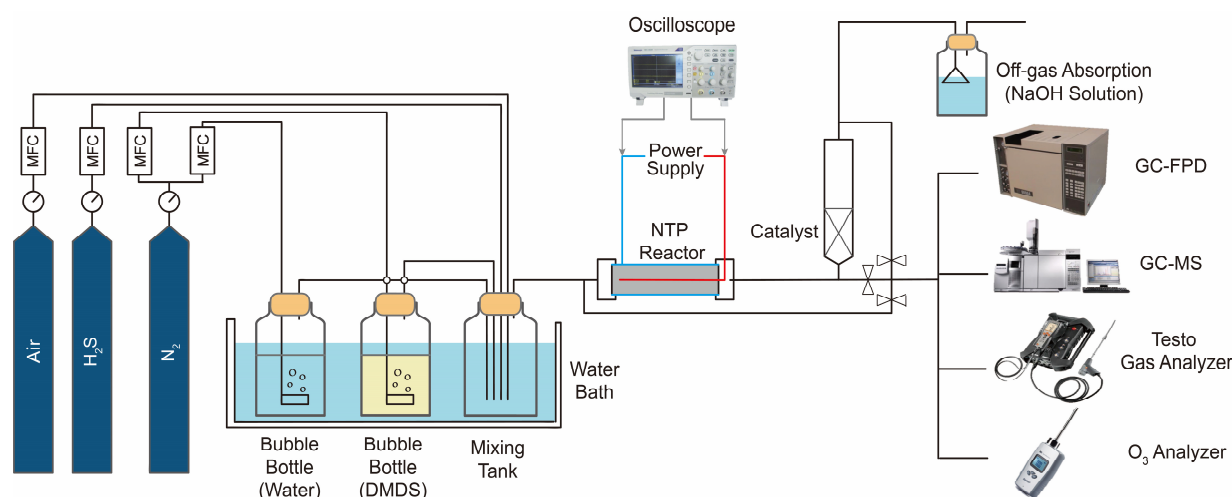


Figure 10. Schematic diagram of experimental setup.

3.3. Definition of Calculations

3.3.1. Removal Efficiency, Energy Yield and Specific Input Energy

The removal efficiency of H_2S or DMDS is expressed as follows:

$$\text{Total removal efficiency (\%)} : \eta_{\text{total}} = \frac{C_{\text{M.in}} - C_{\text{M.cata.out}}}{C_{\text{M.in}}} \times 100\% \quad (1)$$

$$\text{NTP removal efficiency (\%)} : \eta_{\text{NTP}} = \frac{C_{\text{M.in}} - C_{\text{M.NTP.out}}}{C_{\text{M.in}}} \times 100\% \quad (2)$$

$$\text{Catalytic oxidation removal efficiency (\%)} : \eta_{\text{cata}} = \frac{C_{\text{M.NTP.out}} - C_{\text{M.cata.out}}}{C_{\text{M.NTP.out}}} \times 100\% \quad (3)$$

where M is H_2S or DMDS, $C_{\text{M.in}}$ and $C_{\text{M.NTP.out}}$ are the inlet and outlet concentrations of M of the NTP reactor, and $C_{\text{M.cata.out}}$ is the outlet concentration of M of the catalytic reactor.

The instant power was calculated by multiplying the measured pulse voltage (U) and current (I), and the energy per pulse (E_p) was obtained by numerical integration of the product of the instantaneous voltage and current over one pulse period. This method is widely used for pulsed corona discharge reactors [31,32]. The specific input energy (SIE , J/L) in this study was calculated as follows:

$$E_p = \int U(t)I(t)dt \quad (4)$$

$$SIE = \frac{E_p f}{Q} \quad (5)$$

where f is the pulse frequency and Q is the gas flow rate in the NTP reactor.

The energy yield (EY , g/kW·h) was defined to characterize the energy utilization performance and was calculated as follows:

$$EY (\text{H}_2\text{S}) = \frac{C_{\text{H}_2\text{S.in}} - C_{\text{H}_2\text{S.NTP.out}}}{SIE} \quad (6)$$

$$EY (\text{DMDS}) = \frac{C_{\text{DMDS.in}} - C_{\text{DMDS.NTP.out}}}{SIE} \quad (7)$$

3.3.2. Ozone Demand Factor

Based on previous studies, the ozone concentration may have an effect on the degradation of pollutants [22,24,33]. If the O_3 concentration at the inlet (i.e., NTP outlet) is too high

or the pollutant concentration is too low, the O₃ concentration at the outlet may exceed the allowed standard; otherwise, the pollutant concentration may exceed the standard. Therefore, it is necessary to adjust the operating parameters to produce the appropriate ozone concentration to effectively remove the target pollutant and achieve a win–win effect [34,35]. In this study, we define the ozone demand factor (Df) as the ratio of ozone consumption to pollutant removal to reflect the efficiency of the catalyst in degrading pollutants, which in turn provides a reference for the adjustment of the process parameters. It can be calculated by the following equation:

$$Df = \frac{C_{O_3.NTP.out} - C_{O_3.cata.out}}{\Sigma(C_{M.NTP.out} - C_{M.cata.out})} \quad (8)$$

4. Conclusions

This study investigated the removal of single H₂S, single DMDS and their binary mixture via post-plasma catalysis (PPC) with a wire cylinder pulsed corona discharge reactor and CuO/MnO₂-C ozone decomposition catalyst. For individual H₂S and DMDS treatment, lower inlet pollutant concentrations facilitated higher removal efficiency; the downstream catalyst reduced the energy demand for pollutant degradation, improved removal performance and drastically cut tail gas ozone emission, while higher specific input energy (SIE) simultaneously raised pollutant removal efficiency and ozone output in the plasma-only system. During catalytic oxidation, sulfur-containing byproducts were generated, and elemental sulfur deposited on the catalyst surface, blocking partial catalyst pores and lowering the specific surface area and pore volume of the spent catalyst. When treating the binary H₂S-DMDS malodorous mixture, DMDS exerted negligible adverse effects on H₂S elimination, whereas coexisting H₂S significantly suppressed DMDS degradation, indicating mutual inhibition between the two sulfur compounds. The total energy yield of mixed pollutants rose with increasing inlet DMDS concentration yet declined monotonically as SIE grew, creating a trade-off between removal efficiency and energy utilization. The ozone demand factor (Df) was verified as a critical control indicator for PPC systems. For single H₂S removal, ln(Df) exhibited a strong positive linear correlation with SIE/C_{in}, and regulating SIE/C_{in} within 0.22–0.25 kept catalyst outlet H₂S below 5.0 ppm and residual ozone below 3.0 ppm. For single DMDS treatment, Df was positively correlated with SIE/C_{in}, and an SIE/C_{in} value of around 0.72 maintained outlet DMDS and ozone concentrations under 10.0 ppm and 5 ppm, respectively. For binary H₂S-DMDS waste gas, ln(Df) showed an excellent linear positive correlation with SIE divided by total inlet pollutant concentration Σ(C_{in}); controlling SIE/Σ(C_{in}) at 0.28 yielded an ln(Df) of approximately 4, synchronously restricting tail gas H₂S, DMDS and ozone concentrations below 5.0 ppm, 3.0 ppm and 5.0 ppm.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/catal16070630/s1>, Figure S1: Variation in outlet DMDS concentration (a) and outlet ozone concentration (b) with SIE under mixed H₂S-DMDS binary system with different inlet H₂S concentrations; Figure S2: The relationship between the ozone yield and SIE in the plasma reactor used in this study; Figure S3: EDS spectrum of catalysts before and after reaction. Figure S4: Representative voltage (a) and current (b) waveforms of the pulsed corona discharge. Table S1: GC-MS analysis of DMDS degradation products; Table S2: EDS analysis results of catalysts before and after reaction.

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