Non-thermal plasma assisted VOCs decomposition: Influence of humidity, residence time, pellet materials

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The emission of volatile organic compounds (VOCs) from industrial processes, vehicular exhaust, paints, and coatings has become a significant environmental concern in recent years. Many VOCs are carcinogenic and pose severe risks to living organisms upon prolonged exposure [1]. Although various studies have focused on the removal of VOCs using plasma, there remains a need to explore the effects of different reactor designs, carrier gases, and material parameters, as well as the underlying reaction mechanisms involved in the process.

This study investigates toluene oxidation in a dielectric barrier discharge (DBD) reactor packed with catalysts. The reactor was filled with commercially available pellets (glass, Al_2O_3, TiO_2, and $BaTiO_3$) and its performance was evaluated for materials of distinct catalytic properties. The effects of toluene residence time and relative humidity on decomposition efficiency were examined. Additionally, experiments were conducted with varying N_2/O_2 gas compositions, revealing that higher N_2 concentrations increase power consumption while reducing the formation of primary reactive species (e.g., O_3 and N_2O). Temperature profiling over a 1-hour operational period confirmed that the reactor maintains stable thermal conditions (< 45°C), ensuring suitability for prolonged use. During extended operation, the discharge power decreased from 2.39 W to 1.77 W, likely due to surface charge accumulation on the dielectric, which suppresses microdischarges over time. Increased relative humidity was found to reduce discharge power due to water vapor lowering the dielectric surface resistance and decreasing discharge current. However, elevated humidity also promoted the generation of additional reactive species, such as HNO_3, alongside O_3 and N_2O. The mechanism of toluene abatement via non-thermal plasma (NTP) coupled with catalysis was explored, with particular attention to the formation of O_3, N_2O, and organic byproducts. A comparative analysis of energy efficiency between standalone NTP and NTP-catalyst systems demonstrated that catalyst integration enhances process sustainability. Notably, the NTP system facilitates catalyst regeneration, and when combined with catalysts, it achieves faster reaction kinetics and higher reactive species yields than conventional advanced oxidation processes (AOPs). Compared to the non-catalytic reactor, the catalyst-loaded system exhibited superior toluene removal efficiency, as decomposition occurred not only homogeneously in the gas phase but also heterogeneously on the catalyst surface [2].

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[1] C. He, J. Cheng, X. Zhang, M. Douthwaite, S. Pattisson, Z. P. Hao, Chem. Rev., **119** (2019), pp. 4471-4568
[2] X. X. Feng, H. X. Liu, C. He, Z. X. Shen, T. B. Wang, Catal. Sci. Technol., **8** (2018), pp. 936-954

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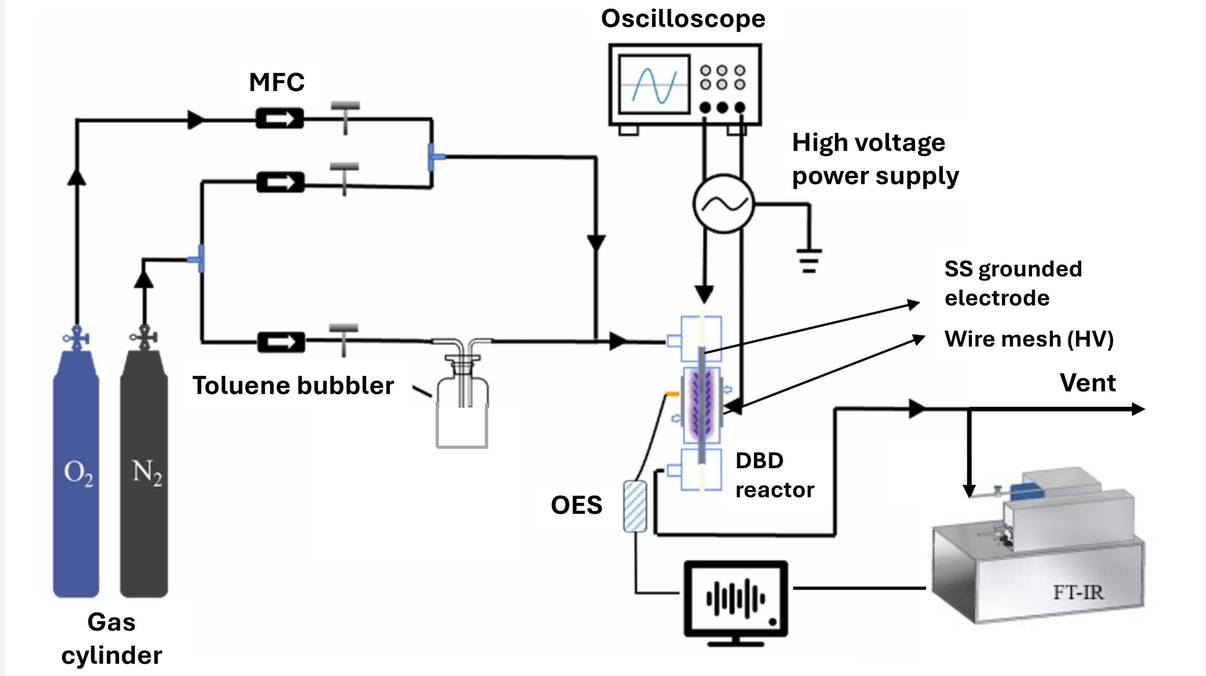
Introduction and Experimental Setup

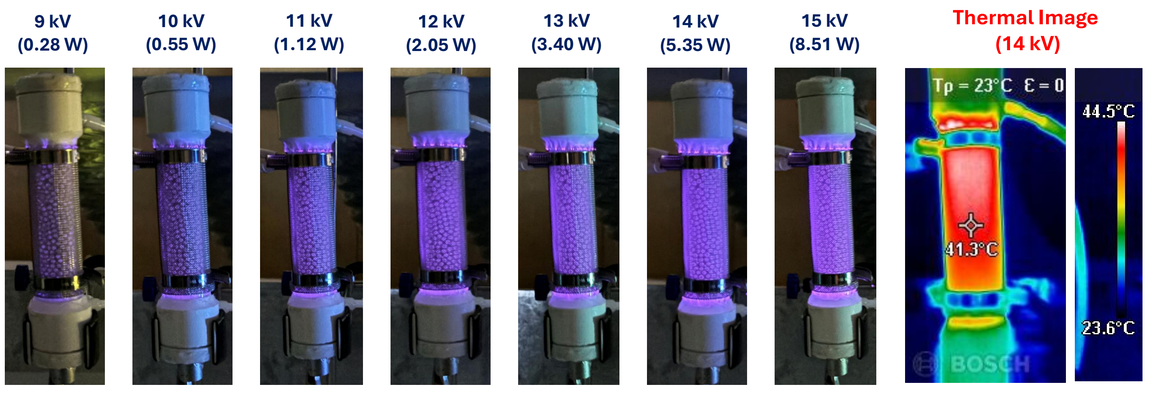
- **Volatile organic compounds (VOCs)** emissions from industries, vehicles, and consumer products are a major source of **air pollution**, contributing to smog, secondary organic aerosols, and **ozone formation**. Many VOCs (e.g., benzene, toluene, formaldehyde) are **toxic or carcinogenic**, so their abatement is critical to protect public health and the environment.
- **Non-thermal plasma** can generate highly **reactive species** (·OH, O·, O_3, NOx radicals) under mild conditions, enabling VOC degradation at **ambient temperature and pressure**. This makes plasma particularly attractive for treating intermittent VOC emissions where conventional methods (thermal incineration, adsorption, and biofiltration) are inefficient.
- Despite numerous plasma studies, comparative evaluations of different **reactor configurations** (DBD, packed-bed, corona, gliding arc, etc.), carrier gases (air, O_2, N_2), and **catalyst materials** (TiO_2, $BaTiO_3$, MnO_2, etc.) remain limited. These factors strongly influence **energy efficiency**, mineralization rate, and **by-product suppression**, highlighting the need for systematic understanding.
- The **plasma–catalyst–VOC interaction mechanisms** (radical generation, adsorption–desorption, surface reactions, intermediate decomposition pathways) are still not fully resolved. Better mechanistic insights will guide the design of next-generation plasma-catalyst systems with improved selectivity, **lower energy consumption, and scalable operation** for real-world VOC abatement.
- **Here, we** conducted experiments using **different plasma-catalysts combinations** to investigate their influence on **VOC degradation**. The **reactive species** generated in each reactor configuration were analyzed, and the **toluene removal efficiency** was compared across different plasma–catalyst combinations. Additionally, the **energy efficiency** of each configuration was evaluated to identify the most effective and sustainable strategy for **VOC abatement**.

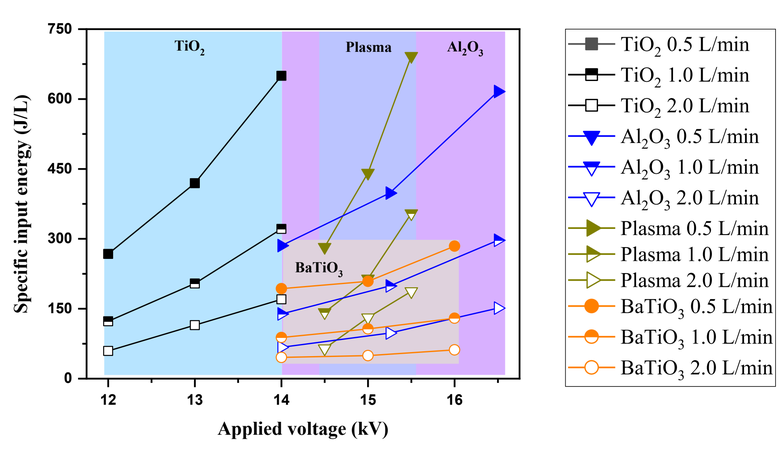


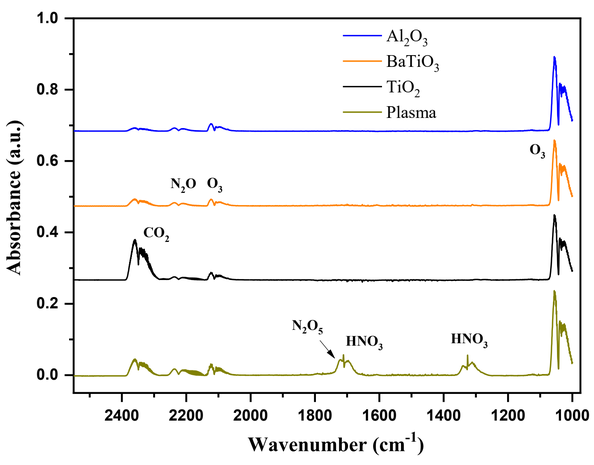
Plasma Reactor Characteristics

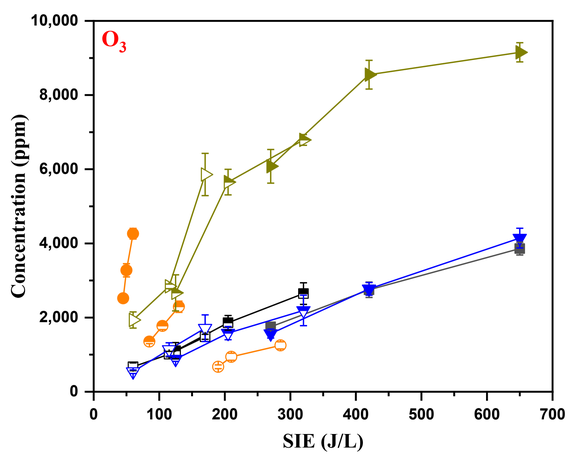
Experimental Conditions

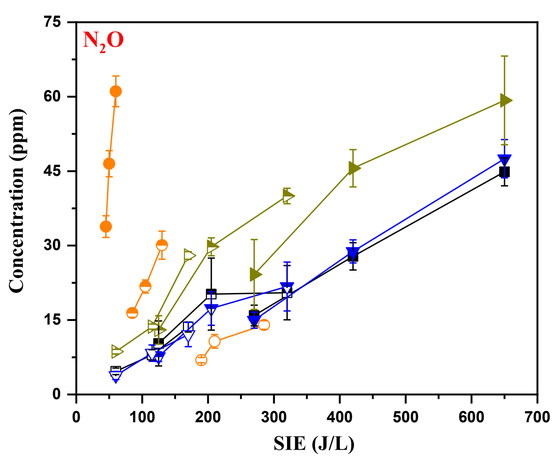
- Discharge gap: 3.5 mm
- Effective reactor length: 120 mm
- Catalytic materials: γ-Al_2O_3, TiO_2, $BaTiO_3$
- Carrier gas: N_2+O_2 (50:50)
- Gas flow rate: 0.5, 1.0, 2.0 L/min
- Applied voltage: 12 – 16.5 kV @ 1500 Hz
- Discharge power: 1 – 6 W





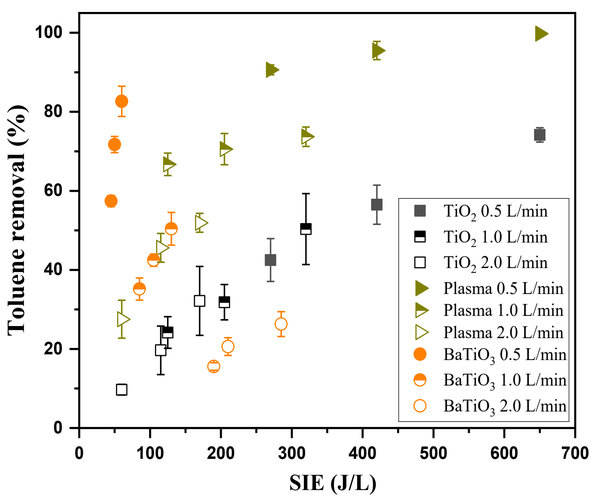


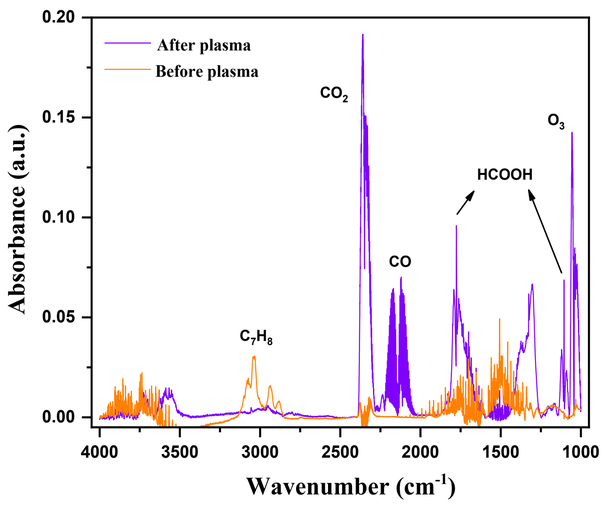


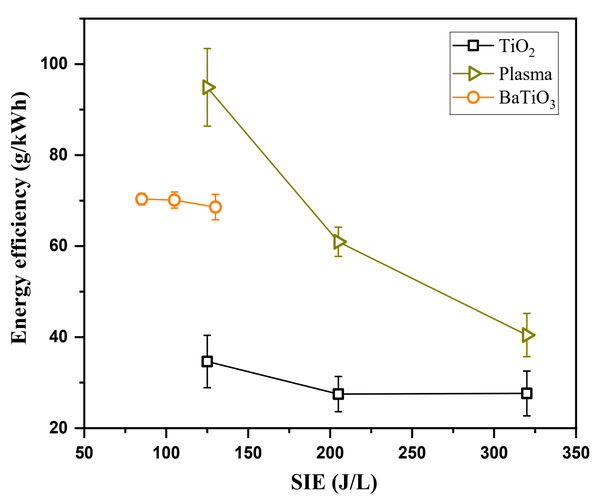


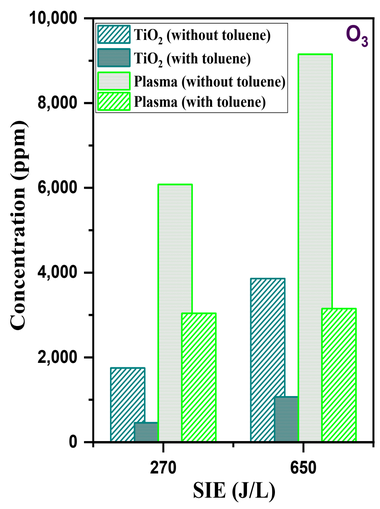
- **At higher dielectric constant**, a voltage drop across the dielectrics is bigger than across the gas gap. This reduces the discharge power delivered into the plasma, so the **SIE becomes lower.**
- At higher dielectric constant, more of applied voltage drops across the dielectric instead of the gas. This weakens the electric field in the plasma gap, reduces electron impact dissociation of O_2, and therefore **O_3 production decreases**.
- When the reactor is empty, the plasma volume is large and gas-plasma interactions dominate. Due to the various series reactions in the **empty reactor we detect HNO_3 and N_2O_5**.

Toluene Degradation and Associated Parameters

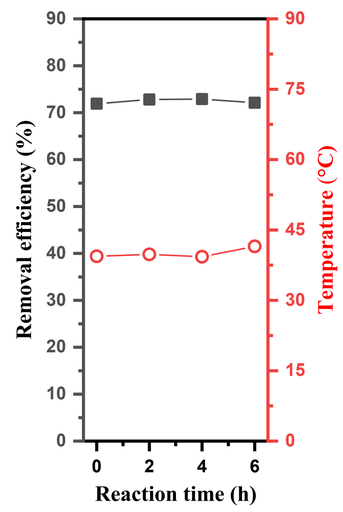




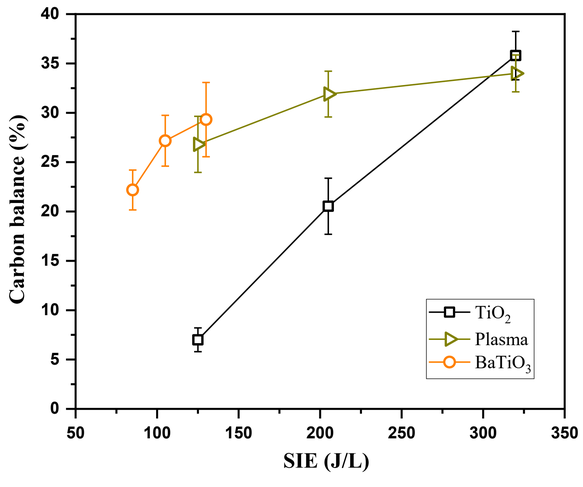


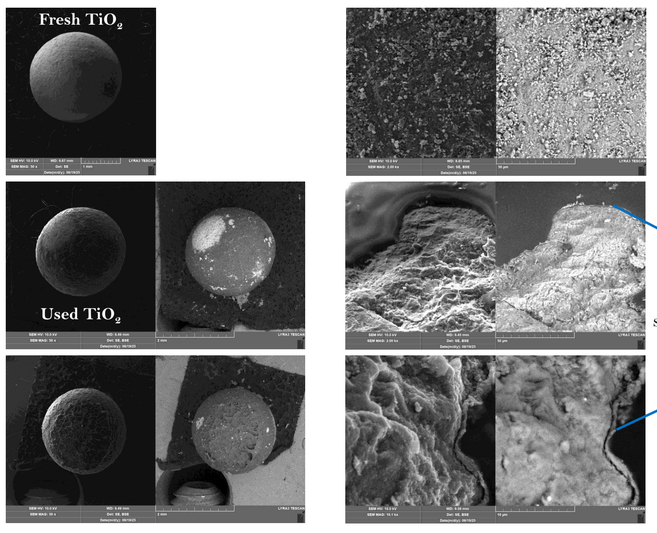


Catalyst stability experiment

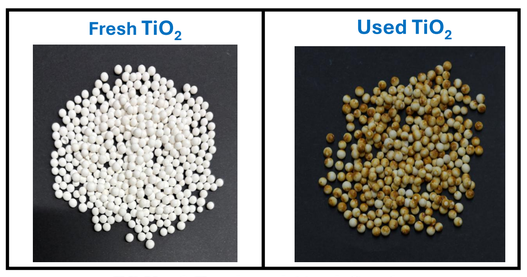


FESEM Images of Fresh and Used Catalyst

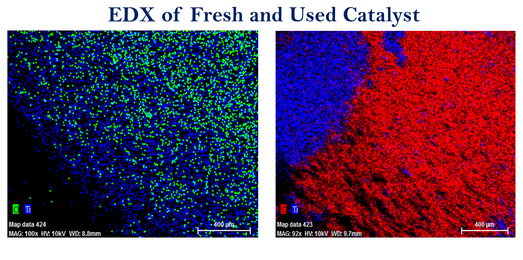




Solid carbon deposits on surface of TiO_2 after VOC treatment



EDX of Fresh and Used Catalyst



- ✓ Catalyst-assisted plasma enhances toluene removal efficiency, especially **$BaTiO_3$ at lower SIE**, highlighting the role of catalysts in **improving VOC degradation**.
- ✓ The presence of CO_2, CO, HCOOH, and O_3 peaks after plasma treatment indicates **partial oxidation of toluene**. Complete mineralization was not achieved, showing the need for optimized plasma–catalyst conditions.
- ✓ **Plasma alone** showed **higher energy efficiency at lower SIE**, but its efficiency decreased as SIE increased. In contrast, **TiO_2 and $BaTiO_3$ catalysts maintained more stable performance**, indicating their suitability for energy-efficient VOC abatement.
- ✓ Despite surface fouling, the catalyst maintained relatively **stable removal efficiency** and **low-temperature operation** over extended reaction times, suggesting good potential for practical applications with periodic regeneration.

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