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Accepted manuscript

A review of Non-thermal plasma -catalysis: The mutual influence and sources of synergetic effect for boosting volatile organic compounds removal

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Abstract:

This review is aimed at researchers in air pollution control seeking to understand the latest advancements in volatile organic compound (VOC) removal. Implementing of plasma-catalysis technology for the removal of volatile organic compounds (VOCs) led to a significant boost in terms of degradation yield and mineralization rate with low by-product formation. The plasma-catalysis combination can be used in two distinct ways: (I) the catalyst is positioned downstream of the plasma discharge, known as the "post plasma catalysis configuration" (PPC), and (II) the catalyst is located in the plasma zone and exposed directly to the discharge, called "in plasma catalysis configuration" (IPC). Coupling these two technologies, especially for VOCs elimination has attracted the interest of many researchers in recent years. The term "synergy" is widely reported in their works and associated with the positive effect of the plasma catalysis combination. This review paper investigates the state of the art of newly published papers about catalysis, photocatalysis, non-thermal plasma, and their combination for VOC removal application. The focus is on understanding different synergy sources operating mutually between plasma and catalysis discussed and classified into two main parts: the effect

of the plasma discharge on the catalyst and the effect of the catalyst on plasma discharge. This approach has the potential for application in air purification systems for industrial processes or indoor environments.

Keywords:

Plasma, Catalysis, Synergy, VOC, Reactor, catalyst, Conversion, Mineralization

1 . Introduction

Air pollution is an urgent issue worldwide, and it causes about seven million premature deaths annually around the world as reported by the World Health Organization (WHO). Faced with this urgent situation, the international community is increasingly focused on curbing air pollution and its associated consequences.

Volatile organic compounds (VOCs) occupy a significant portion of the pollutants released into the atmosphere. They are defined as organic substances that readily evaporate at room temperature and atmospheric pressure, constitute a substantial portion of airborne pollutants (i.e., carbon, hydrogen, and heteroatoms) et al., 2016). Human activities across various industries, including petroleum (Rajabi et al., 2020), paints and varnishes (Bauer & Buettner, 2018), cement production (C. Wang et al., 2018), wood-based furniture (Yuanzheng Wang et al., 2021), pharmaceutical formulation and pesticide manufacturing (D. C. He et al., 2022) are the primary sources of VOCs emissions. The United States Environmental Protection Agency (U.S. EPA) carried out a study called "Total Exposure Assessment Methodology (TEAM)," which reported that indoor VOCs concentration is 2 to 5 times higher than outdoor levels. Additionally, VOCs can react with carbon monoxide or nitrogen oxides molecules in the presence of sunlight to generate ground-level ozone (Xiong & Du, 2020). The high toxicity and the rapid spreading of this kind of chemical pollution can cause several serious problems for human health, including cancers, allergies, cardiovascular anomalies (Leachi et al. 2020), brain diseases, eyes and skin irritations, and severe pneumonia (Ratiu et al. 2021) as

presented in the **Fig.1** . Recognizing these dangers, environmental regulations are increasingly addressing VOC emissions. In March 1999, a European directive was issued, aimed to reduce the industrial VOC emissions from organic solvents (Journal, 2017). Between 1970 and 2021, the USA successfully reduced their VOCs emissions by approximately 50%. Moreover, for the best execution of the government's directives, several technologies have been adopted to limit the emission of VOCs into the atmosphere. They are divided into two approaches: recovery and elimination approach. Destructive techniques comprise biofiltration, ozonation, thermal oxidation, catalytical combustion, catalytic and photocatalytic degradation, and non-thermal plasma. Among these technologies, catalytic oxidation is mainly investigated in the literature. It allows higher VOCs conversion and mineralization. However, it presents several failures, including high-temperature requirements, catalytic deactivation after a series of reactions, and the ineffectiveness of some chemical compounds. Recently, NTP has been approved as an effective process for the remediation of air contaminants at low-temperatures and atmospheric pressure. It is considered a green technology that does not require toxic chemicals; it operates with a simple setup, and can destroy a large variety of VOCs at low or high concentrations (Assadi et al.,2016). Nevertheless, NTP is limited by three significant drawbacks: (I) usually, the discharge occurs at high voltage, which presents a direct risk of electric shock (II), the production of toxic and undesired by-products such as ozone and NO_x; and (III) low energy efficiency due to the high applied electric potential.

Hence, hybrid systems combining plasma and catalysis draw special attention to overcome problems previously highlighted. The presence of the catalyst in the plasma discharge (In Plasma Catalysis) led to a series of benefits as enhancement of the conversion efficiency and mineralization, prevention of catalyst deactivation, reduction of harmful by-products such as ozone, lowering energy consumption thanks to low temperatures and optimization of discharge voltage applied. These qualities result from the synergetic effect occurring between the two processes. In the last decade, several researchers reported the presence of this behavior in NTP-Catalysis/Photocatalysis systems, and a number of hypotheses have been suggested

highlighting the mutual effect between the catalyst and the discharge (Palau et al., 2015). Precedent reviews on VOCs remediation are published with a focus on the application of catalysis and plasma separately (C. Yang et al., 2019) (C. He et al., 2019) (Mu & Williams, 2022). However, recently, some papers have reviewed coupling plasma and catalysis without exploring in depth the nature of the interaction discharge - catalyst and potential synergy sources. This review paper addresses this gap by summarizing recent research on VOC remediation using catalysis/photocatalysis alone, NTP alone, and their combined application. We delve into the mutual interactions between the discharge and the catalyst, exploring the mechanisms underlying the observed synergistic effect.

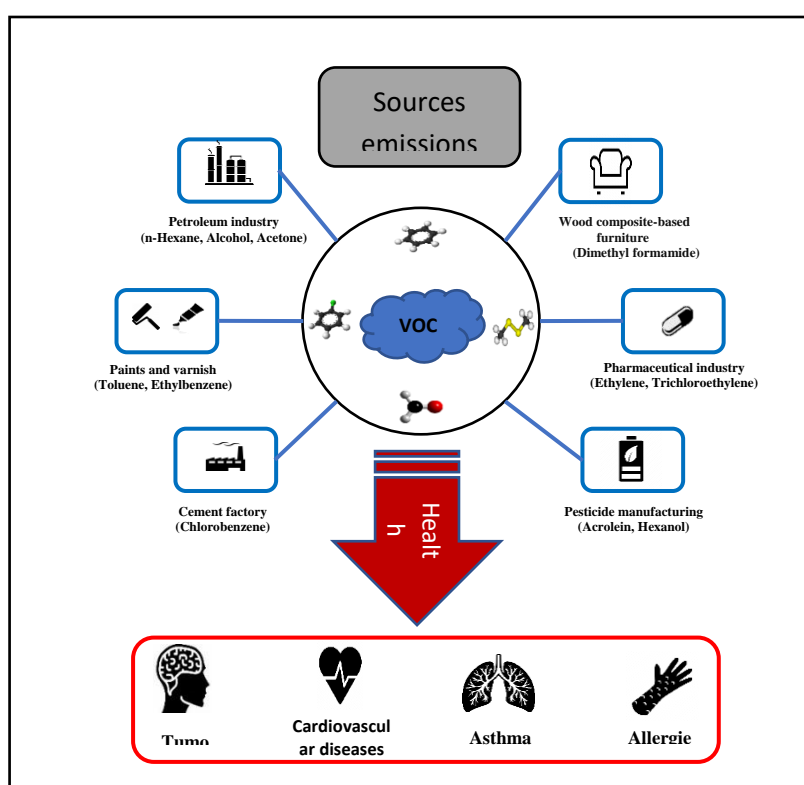


Figure 1. VOCs emission sources and effects on human health.

2. VOCs removal technologies

Various techniques were developed to reduce and control VOCs emissions into the atmosphere. They are classified into two categories, as defined in **Fig. 2**: recovery technologies and elimination technologies (Parmar & Rao, 2009). Recovery methods consist

of the trapping and reconversion of gaseous pollutants to beneficial compounds as solvents by adsorption/absorption (W. Zhang et al., 2022) (Pirola & Mattia, 2021), membrane separation (W. Yang et al., 2018) and condensation (H. Wang et al., 2020). The destruction methods include, thermal catalysis (Yuxin Wang et al., 2021)(Weng et al. 2017a), photocatalysis (Abou Saoud et al., 2021) (Yanagida et al. 2019), non-thermal plasma (Zadi et al., 2020) (Abdelaziz et al., 2013), combustion (W. B. Liet al., 2010), which are based on the oxidation process of the target VOC by highly oxidants species , such as hydroxyl radicals and reactive oxygen species (ROS). Except for biofiltration (Marycz et al. 2022), which is based on using microorganisms to convert the pollutant and to produce energy. Herein, special attention will be given to the destruction processes, particularly plasma, catalysis, and their combination.

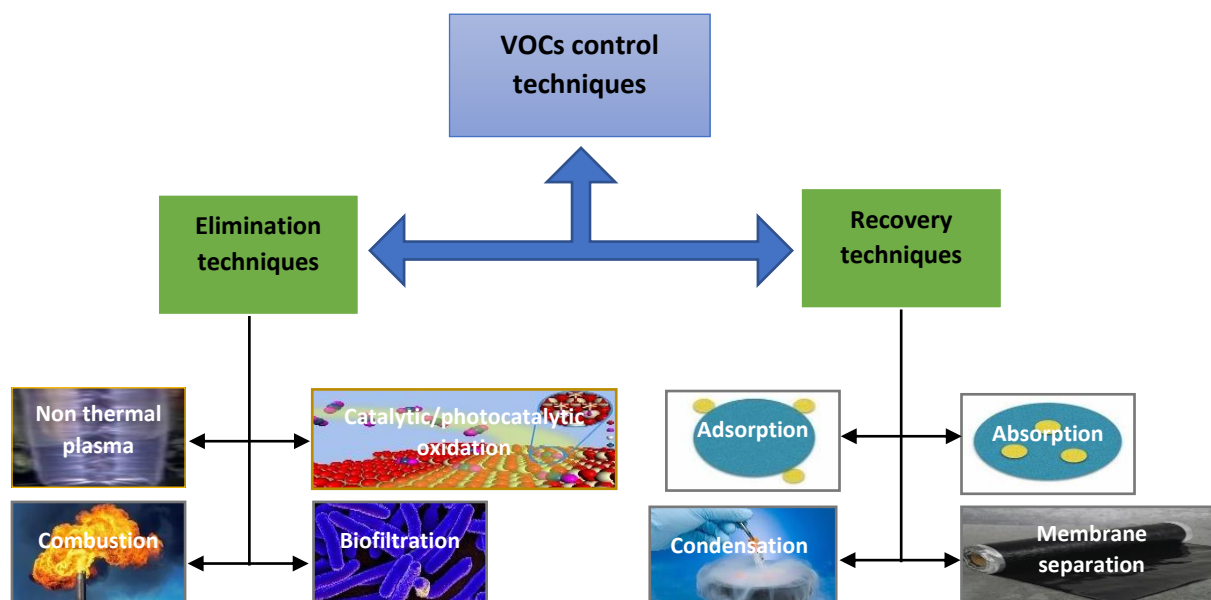


Figure 2. Flowchart of VOCs controlling methods.

2.1. Heterogeneous catalysis

Catalysis is the heart of chemical processes; since its discovery, it has been the foundation of diverse industrial processes since its discovery, including petroleum refinement (Armor, 2011), chemical manufacturing, and environmental technologies for water and air depollution (Junhua Li et al., 2013) . It is also one of the twelve principles of green chemistry (Sheldon, Arends, and Hanefeld, n.d.).

The catalyst, a key player in chemical reactions, significantly reduces the activation energy required for these chemical reactions to occur on its surface-active sites. The catalysts' physicochemical properties and elemental composition dictate the types of active sites. Three well-known sites come into play in the case of heterogeneous catalysis for VOC elimination: Basic sites, acidic sites, and metal sites, as illustrated in **Fig. 3**.

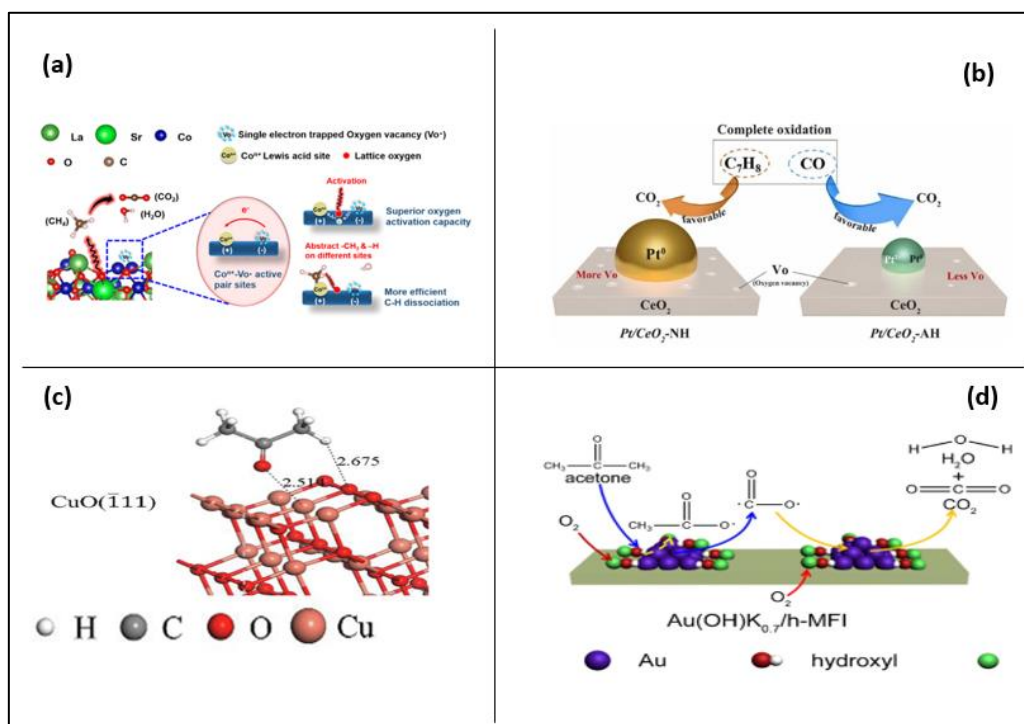


Figure 3. (a) Lewis's acid sites over Perovskite Oxides catalyst for CH_4 combustion (Reprinted with permission from (J. Yang et al. 2021). Copyright 2021, American Chemical Society), (b) Pt Metallic site for toluene oxidation (Reprinted with permission from (Q. Yang et al. 2022a). Copyright 2021, Elsevier), (c) Acetone adsorption on acid-base Lewis pair (Reprinted with permission from (K. Zhou et al. 2019). Copyright 2019, Elsevier), (d) Acetone decomposition over Au metallic site (Reprinted with permission from (Z. Jiang et al. 2022). Copyright 2021, Elsevier).

2.1.1. Catalysts used for VOCs oxidation

Catalytic oxidation of gaseous organic pollutants has been widely reported in the literature; it involves a large catalyst family with variable compositions and structures, such as mixed and single noble metal-based catalysts and non-noble and metal oxide-based catalysts.

Table 1. lists some recent works dealing with catalytic VOC removal.

- Noble metal-based catalyst

Despite their elevated cost, noble metal-based catalysts are the most used materials for VOCs oxidation due to their high activity, low operating temperature, and best resistance against poisoning (Liotta, 2010) (Z. Zhang et al., 2016).

The chemical elements Ag, Pd, Au, Pt, Ru, and Rh supported on carbon nanotubes (CNT), zeolites, or metal oxides compose the core of this catalyst type. Many studies have been performed to oxidize VOCs with different noble metal-based catalysts. The authors started with the synthesis of the catalyst material, and then they tested its catalytic activity. Zhiwei Wang and collaborators prepared a TiO₂-supported Pt catalyst via the molten salt method. Furthermore, they tested the performance of the catalyst for toluene removal. A total 1000 ppm of toluene conversion and high selectivity were achieved at 190°C over a Pt/TiO₂ catalyst (H. Huang & Deng, 2020). Pd nanoparticles loaded on the surface of silicalite material exhibited the best performance toward toluene oxidation at 160°C thanks to the intrinsic properties of the catalyst, which combine high adsorption capacity, high Pd⁰ composition, and suitable acidity (L. Wu et al., 2022). Gold-based catalysts are known as efficient materials for VOC oxidation, and the presence of Au atoms makes the catalyst less exposed to deactivation problems such as coke deposition and sulfur-containing compound poisoning (Assadi et al., 2014 a, b,c). In a recent paper reporting the synthesis of gold supported on Co₃O₄-CeO₂-Al₂O₃ mixed metal oxide supports, the authors indicated that the presence of gold nanoparticles led to a remarkable conversion efficiency of formaldehyde even at room temperature (Ilieva et al. 2022).

X. Zhang et al. demonstrated the excellent effect of loading Ag nanoparticles on a zirconium-based metal-organic framework (MOF) structure toward toluene conversion. The authors expected the high lattice oxygen and Ag content on the surface to be the reason for the enhanced activity (Xiaodong Zhang et al., 2020).

According to Sun et al., the incorporation of sub-nanometer Rh on TiO₂ support led to a complete oxidation of formaldehyde at ambient temperature, in contrast with the TiO₂ support alone, for which no activity was recorded. The different characterization methods showed the

Rh species' high dispersion, allowing the dissociative adsorption of O_2 and, therefore, the reactivity toward HCOOH conversion (X. Sun et al., 2018).

- **Mixed noble metal catalyst**

Mixing noble metals in an optimal ratio improves their catalytic activity and eliminates the high-cost problem of certain metals.

He et al. synthesized monometallic Pt, Pd, and bimetallic Pt-Pd nanoparticles loaded on MnO_2 nanospheres to test their catalytic activity toward toluene removal. They concluded that the catalyst with a Pt/Pd molar ratio of 7:3 showed the best catalytic performance compared to the Pt and Pd monometallic catalysts with 100, 85, and 80% of toluene conversion at 175 °C, respectively. They associated the obtained result with the O_{ads}/O_{latt} optimal molar ratio reached due to mixing two metals (Jiaqin He et al., 2021).

A recent study elucidated a synergy in oxidation of formaldehyde by combining platinum and gold nanoparticles on the surface of TiO_2 . They confirmed from different characterization techniques the role of Au and Pt nanoparticles in enhancing the reducibility and the mobility of oxygen species, which is the key to catalytic surface reactions (Ye et al., 2022).

Ag-Pd/ MnO_2 catalyst prepared by the galvanic replacement method was tested for toluene oxidation. The researchers stated the effect of mixing the two metals on improving the lattice oxygen at the surface and, therefore, the oxidation of toluene at low temperatures compared to a Pd/ MnO_2 catalyst (Y. Li et al., 2019).

- **Non-noble metal-based catalyst**

A considerable number of studies on the use of catalysts such as transition metals or metal oxides as a good alternative toward VOC conversion were carried out. Non-noble metal elements have the advantage of being less expensive, more durable, and resistant to deactivation by coke formation. The synthesis method of the catalyst material is crucial for its catalytic activity. Aggett et al. prepared CeO_2 catalysts from two Ce precursors with different oxidation states (Ce^{4+} or Ce^{2+}) using the precipitation method. Then, they evaluated the

performance of the corresponding catalysts toward propane oxidation. The Ce^{4+} exhibited a high conversion percentage of propane due to its morphology and high surface area (Aggett et al., 2021). Despite the efficiency of CeO_2 as a catalyst, its nanostructure played an essential role in preventing coke formation. Y. Zhang et al. investigated the effect of the CeO_2 structure on coke deposition. The order of coke accumulation was established as follows: $\text{CeO}_2\text{-C}$ structure (Cubic shaped) > $\text{CeO}_2\text{-O}$ structure (Octahedral-shaped) > $\text{CeO}_2\text{-R}$ structure (Rod-shaped) > $\text{CeO}_2\text{-S}$ structure (Sphere-shaped); they assigned this trend to the high concentration of oxygen vacancy on spherical CeO_2 catalyst (Ying Zhang et al., 2022)

Manganese oxides MnO_x are among the materials investigated in the literature for organic pollutant elimination. Its catalytic activity is commonly associated with diverse structures and oxidation states (S. Mo et al., 2020). L. Chen et al. reported the impact of redox reaction temperature during the synthesis of $\alpha\text{-MnO}_2$ catalyst on the toluene oxidation. They found that the catalyst prepared at 60 °C showed a high toluene conversion yield, which was probably assigned to the surface defects observed for this material (L. Chen et al., 2021).

The ABO_3 perovskites (**Fig. 4**) have gained recognition for their catalytic activity in VOCs oxidation. A group of researchers has specifically highlighted the relationship between LaCO_3 perovskite structure and its performance in toluene oxidation. They provided evidence that the sample contains abundant surface reactive species, including adsorbed oxygen and Co^{3+} (Xuejun Zhang et al., 2019).

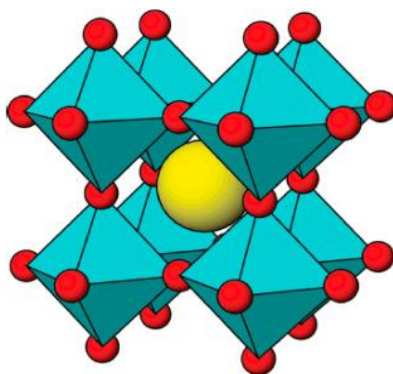


Figure 4. ABO_3 Perovskite structure (yellow = A, cyan = BO_6 and red = O) (Reprinted with permission from(Mamba et al. 2022). Copyright 2022, Elsevier)

Furthermore, Weng et al. prepared a stable and performant mixed metal oxide catalyst supported on acidic zeolite ($Mn_xCe_{1-x}O_2/HZSM-5$) to overcome the catalyst deactivation during the removal of chlorinated volatile organic compounds (CVOCs). They explained the synergetic impact between the acidic zeolite sites and the oxide ions supported on the surface to convert chlorobenzene and remove the deposited chlorinated byproduct (Weng et al., 2017b).

Noble metal-based catalyst							
Catalyst	Support	Reactor	VOCs	Temperature	Flow rate	Conversion	Reference
Pd	Silicalite-1	Continuous- flow fixed-bed quartz tubular microreactor	Toluene (1000 ppm)	190 °C	16.7 ml/min	90 %	(L. Wu et al. 2022)
Pd	NC/BN ^a	Continuous flow fixed-bed quartz microreactor	o-xylene (1000 ppm)	210°C	30 ml/min	80%	(H. Chen et al. 2022)
Pd	Al ₂ O ₃ CoMn ₂ O ₄	U-shaped quartz tubular reactor	Toluene (50 ppm)	165°C	30 ml/min	100 %	(Jiaqin He et al. 2022)
Pd	UiO-66 ^b	Single-pass flow reactor	Toluene (1000 ppm)	235°C	50 ml/min	90 %	(Bi et al. 2022)
Pd	Al ₂ O ₃	Con- tinuous flow fixed bed reactor with	Ethyl acetate (500 ppm)	300°C	250 ml/min	100 %	(M. Ma et al. 2021)
Pd	Ti-SBA-15 ^c	Single-pass flow reactor	Styrene (50 ppm)	225 °C	50 ml/min	100 %	(Wen et al. 2021)
Pt	CeO ₂ -TiO ₂	Fixed- bed quartz micro- reactor under	Benzene (1000 ppm)	160 °C	75 mL /min	100 %	(Yijun Shi et al. 2022)
Pt	TiO ₂	Fixed-bed quartz reactor	Benzene (100 ppm)	200 °C	200 ml/min	100 %	(J. Kim et al. 2022)
Pt	Y-zeolite-	Continuous flow fixed bed reactor	Toluene (1000 ppm)	160°C	200 ml/min	100 %	(H. Chen et al. 2022)
Pt	CeO ₂ -NH	Fixed-bed quartz tube reactor	Toluene (1000 ppm)	220°C	80 ml/min	100 %	(Q. Yang et al. 2022b)
Pt	ZrO ₂	Tubular flow quartz microreactor	Toluene (1000 ppm)	250 °C	100 ml/min	97%	(Drozdek et al. 2022)
Au	Co ₃ O ₄ -CeO ₂ -Al ₂ O ₃	Continuous flow equipment with a four-channel isothermal stainless- steel reactor	Formaldehyde (120 ppm)	25°C	ND	95%	(Ilieva et al. 2022)
Au	Ami-SiO ₂	Fixed bed tubular reactor	Toluene (178 ppm)	175°C	40 ml/min	53%	(Sanghuaypai, Sirikanjanawanit, and Sukpirom 2022)
Ag	Co ₃ O ₄	Fixed-bed quartz reactor using	Benzene (500 ppm)	204 °C	100 ml/min	100%	(Fang et al. 2022)
Ag	CeO ₂	Fixed-bed reactor equipped with an	Toluene (1000 ppm)	240 °C	50 ml/min	100%	(Yuxin Wang et al. 2021)

open tube furnace was							
Ag	TiO ₂	Quartz tube	n-butanol (1000 ppm)	225 °C	200 mL/min	100%	(Bhat et al. 2021)
Ag	UiO-66 derivative	Fixed bed microreactor	Toluene (1000 ppm)	295 °C	50 ml /min	100%	(Xiaodong Zhang et al. 2020)
Rh	TiO ₂	fixed-bed microreactor made of quartz tube	Chlorobenzene (500 ppm)	350 °C	100 ml/min	100%	(Liu et al. 2019a)
Rh	TiO ₂	Continuous-flow fixed bed reactor	Formaldehyde (140 ppm)	20 °C	50 ml /min	100 %	(X. Sun et al. 2018)
Mixed noble metal-based catalyst							
Pt-Pd	MnO ₂	U-tubular reactor	Toluene (50 ppm)	175 °C	30 ml/min	100 %	(Jiaqin He et al. 2021)
Pt -Au	TiO ₂	Batch reactor	Formaldehyde (200 ppm)	25 °C	-	100 %	(Ye et al. 2022)
Pt-Ru	ZrO ₂	Continuous-flow fixed-bed reactor	Toluene (50 ppm)	160 °C	30 mL/min	100 %	(M. Wang et al. 2020)
Pd-Au	CeO ₂ -Y ₂ O ₃	Continuous flow fixed bed microreactor	Benzene (13 ppm)	150° C	ND	100 %	(Ilieva et al. 2018)
Ag-Pd	MnO ₂	Fixed bed continuous-flow reactor	Toluene (260 ppm)	185 °C	ND	100 %	(Y. Li et al. 2019)
Non-Noble metal-based catalyst							
CeO₂	No support	Contin-uous flow fixed bed microreactor	Propane (5000 ppm)	600 °C	50 mL /min	100 %	(Aggett et al. 2021)
CeO₂-S	No support	Quartz tube	Styrene (200 ppm)	150 °C	50 mL /min	100 %	(Ying Zhang et al. 2022)
MnO₂	No support	Fixed-bed quartz reactor	Toluene (1000 ppm)	210 °C	500 ml/min	97%	(L. Chen et al. 2021)
LaCoO₃	No support	Fixed bed reactor	Toluene (500 ppm)	260 °C	100 ml/min	100 %	(Xuejun Zhang et al. 2019)
CoMn₂O₄	Carbon nanotubes	Quartz reactor with	Formaldehyde (100 ppm)	133 °C	100 ml /min	90%	(Y. Zeng et al. 2021)
CrO_x	Al ₂ O ₃		Methanethiol (nd)	375° C		100 %	(J. Lu et al. 2020)
Mn_xCe_{1-x}O₂	HZSM-5	Quartz fixed-bed reactor	Chlorobenzene (1000 ppm)	250 °c	ND	90 %	(Weng et al. 2017b)

Table 1. Experimental conditions and VOCs conversion performances at the different kinds of catalyst materials.

^a NC/BN: Nitrogen doped carbon/Boron nitride, ^b UiO-66: Zirconium-based metal organic framework (MOF), ^c Ti-SBA-15: Titanium ordered mesoporous silica, ND: not defined.

From this part of the review and **Table 3**, Noble metal and mixed noble metal-based catalysts were extensively investigated in the removal of aromatic VOCs such as toluene, benzene, and xylene. This can be explained by the high efficiency of this kind of catalyst and its resistance to the deactivation problems (Q. Li et al., 2019).

The mechanism of VOCs degradation varies from one catalyst to another depending on several parameters, including, catalyst composition, affinity of VOCs molecules with the catalyst and the conditions of the reactions. Thus, it is crucial to elucidate the reaction mechanism involved.

2.1.2. Catalytic oxidation mechanism of VOCs

2.1.2.1. Chemical reaction step:

The elucidation of the mechanisms involved in VOCs oxidation is of the great interest in preparing highly active and selective catalysts. Nevertheless, the reactivity of the catalyst surface with VOCs molecules depends on many parameters, such as the chemical composition of the catalyst and the pollutants, the design and morphology of the catalyst surface, and the support. Zou et al. reported the oxidation mechanism of toluene over PtCo₃-C catalyst (**Fig. 5**). It was proposed that the first steps consist of the simultaneous adsorption of toluene and oxygen molecule on PtCo₃-C surface. Under heating of the catalyst, the surface adsorbed oxygen species were generated and the conversion of the adsorbed toluene to several intermediates, including benzaldehyde, benzoic acid and maleic anhydride was initiated. These intermediates completely oxidize into H₂O and CO₂ (Zou et al., 2022). Tingyu Zhu and coworkers carried out a study on the benzene oxidation mechanism over a MnO_x/TiO₂ catalyst. From in situ FTIR analysis, a possible reaction mechanism was proposed and shown in **Fig. 6**. Firstly, Benzene reacted with the Mn center, leading to the formation of two conjugated phenolate structures. The oxygenated group is considered as an electron donor and ortho-para positioned director; the phenolate intermediates could easily undergo an oxidation to form Para and Ortho-benzoquinone. The catalyst and the active oxygen promoted the ring opening, allowing further the generation of small molecules such as CO, CO₂, and H₂O (J. Zeng et al., 2015). In another study, Haghghi and collaborators investigated the mechanism of P-xylene abatement over Pd/ceria-clinoptilolite material. In this mechanism, it was supposed that p-xylene could be adsorbed on both Pd and Ceria inside the zeolite pores (Imessaoudene et al., 2022). At the same time, Pd is oxidized by oxygen molecules adsorbed

on its surface to form Pd-O that can be further reduced by xylene. **Fig.7** shows clearly the role of surface oxygen species on xylene oxidation. The lattice bulk oxygen diffused to the surface of the ceria and reacted with the adsorbed xylene. The dissociative adsorption of the molecular oxygen promoted the provisioning of bulk oxygen in order to keep the cycle continuity (Asgarie et al., 2013).

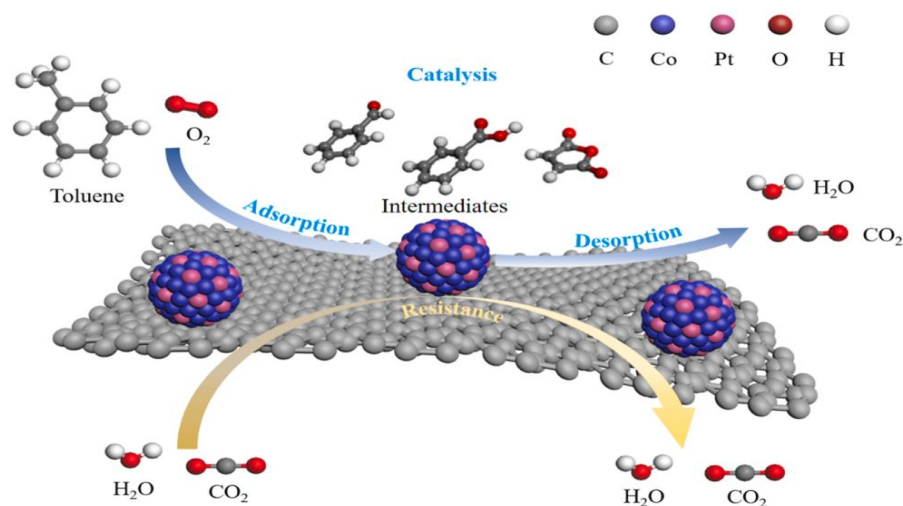


Figure 5. Plausible catalytic mechanism for the catalytic oxidation of toluene over the PtCo_3 catalyst (Reprinted with permission from (Zou et al. 2022) Copyright 2021, Elsevier)

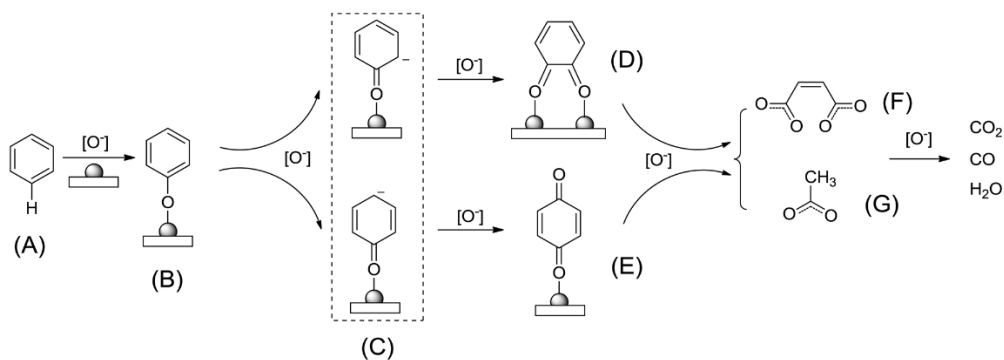


Figure 6. Possible mechanism for Benzene decomposition over $\text{MnO}_x/\text{TiO}_2$ catalyst (Reprinted with permission from (J. Zeng et al. 2015) Copyright 2015 Elsevier)

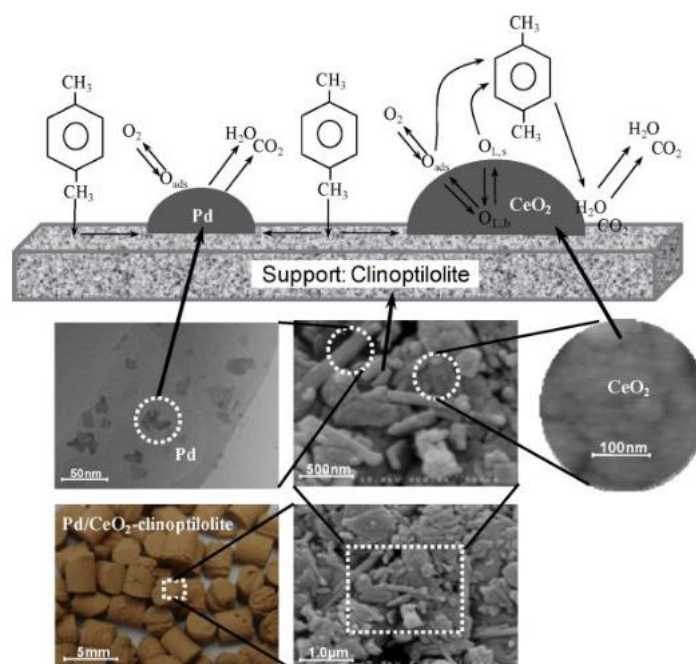


Figure 7. Reaction channel for xylene oxidation over nanostructured Pd/CeO₂(30%)–clinoptilolite (Reprinted with permission from (Asgari, Haghighi, and Shafiei 2013) Copyright 2013 Society of Chemical Industry)

Acetaldehyde is an oxygenated VOC released typically from biomass combustion, and its catalytic degradation has been reported by many researchers. Zeyu Zhao experimented with the degradation of Acetaldehyde over a MnO₂ catalyst (**Fig. 8**); the first step consisted of the adsorption of Acetaldehyde on MnO₂, and under dry conditions, the hydrogen atom of the aldehyde interacted with the oxygen atom of MnO₂ surface, leading to the formation of acetate and formate intermediates, which decomposed after to CO₂ and H₂O. The presence of water vapor enhanced the oxidation of Acetaldehyde at moderate temperatures due to the generation of hydroxyl active bonds, which reacted efficiently with the pollutant (Zhao et al., 2022).

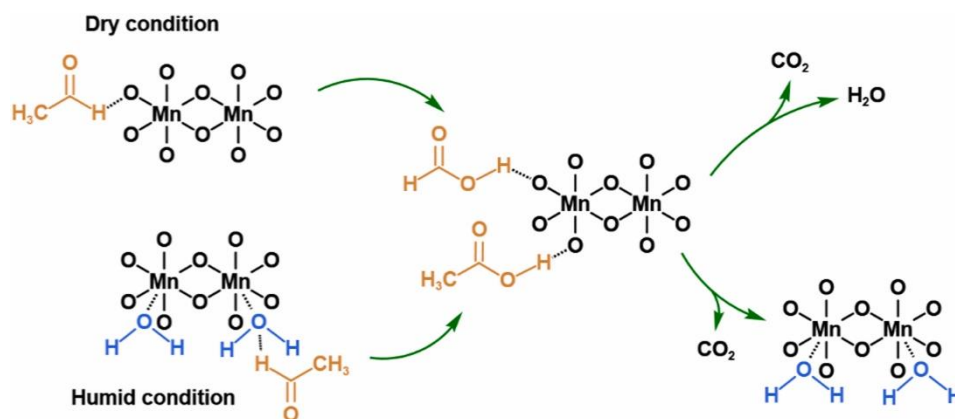


Figure 8. Mechanism of Acetaldehyde molecule over MnO_2 catalyst under dry and humid conditions (Reprinted with permission from (Zhao et al. 2022) Copyright 2021, Elsevier)

The catalytic abatement of chlorinated VOC has been widely discussed in the literature. Zhu and coworkers investigated the oxidation mechanism of chlorobenzene over nobles' metals (Pd, Pt, Ru, Rh). They supposed the adsorption of CB on the catalyst surface with the breaking of C-Cl bonds, and the formation of surface phenolate species would be the first step. Then, o-benzoquinone or p-benzoquinone was formed after phenolate oxidation. A nucleophilic attack occurred through oxygen or chlorine species and promoted the maleate or maleic anhydride formation. Finally, the above species were eventually oxidized to CO_2 , CO, H_2O , HCl, and Cl_2 (Liu et al., 2019b). Kai Shen and coauthors elucidated trichloroethylene (TCE) oxidation over the MnO_x - CeO_2 catalyst. . The rate-determining step was the dissociative adsorption of the C-Cl bond over CeO_2 , then the total oxidation of TCE fragments compounds by active oxygen species present on the surface of MnO_x and CeO_2 , as shown in the **Fig. 9** (K. Shen et al., 2022).

In addition to the conventional catalytic oxidation of VOCs, photocatalysis has demonstrated potential for odor and volatile organic compound removal, which will be discussed deeply in the next paragraph.

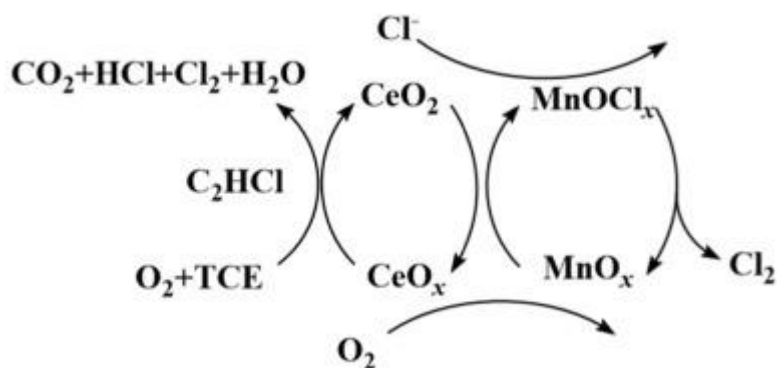


Figure 9. Catalytic cycle of TCE degradation over $\text{MnO}_x/\text{CeO}_2$ (Reprinted with permission from (K. Shen et al. 2022) Copyright 2022, Elsevier)

2.2. Photocatalysis

2.2.1. Principle

Photocatalysis, or photocatalytic oxidation process (PCO), is one of the effective and economical technologies used for air treatment. It can easily decompose VOCs in the gas phase into less toxic compounds such as H_2O and CO_2 (Belkessa et al., 2023) (Y. Shen et al., 2022). Despite the advantages cited above, photocatalysis has disadvantages, such as catalyst deactivation and limitations on active sites (Ahmad et al., 2016). The Photocatalysis process takes place in the following steps: Firstly, the valence band (VB) electrons absorb UV photons. Therefore, the excited electrons cross the energy gap and migrate to the conduction band (CB). Thus, electron-hole pairs in CB and VB are generated, respectively (Lacombe & Keller 2012). The second step considers the reaction of the generated electron and hole with adsorbed O_2 and water molecules to produce the superoxide radical $\text{O}_2^{\cdot-}$ and the hydroxyl radical OH^\bullet (Kenfoud et al., 2021; Baaloudj et al., 2021). Finally, the target molecules undergo an oxidation reaction on the catalyst surface by the produced oxidizing agents (Shayegan, et al., 2018) (**Fig. 10**).

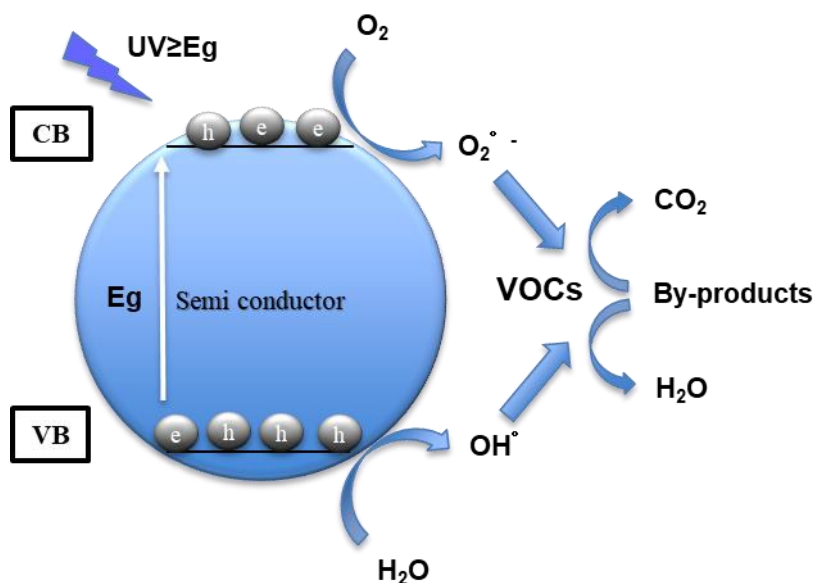


Figure 10. Photocatalysis principle.

2.2.2. Photocatalysts for VOCs removal

A significant amount of research is currently being conducted on developing photoactive catalysts for VOCs degradation. As summarized in **Table 2**, semiconductor-based catalysts are widely investigated, such as (TiO_2 , ZrO_2 , WO_3 , and ZnO) (Bellè et al., 2022) (Rangkooy et al., 2019) (Rangkooy et al., 2021), which combine several captivating characteristics such as availability, suitable electronic properties, high adsorption ability, enhanced photoactivity, and stability (Baaloudj et al., 2021b). However, they have some limitations, primarily due to electron-hole recombination and inactivity in visible light. Therefore, many researchers are focused on developing highly photoactive materials in a broad UV-Vis spectrum. The common method primarily involves doping the existing metal oxide catalyst with transition metals, noble metals, and metal oxide. TiO_2 remains the pioneer of semiconductor catalysts because it is chemically stable and durable, has a high oxidation power, and is inexpensive. Recently, Bellè et al. investigated the degradation of gaseous toluene on TiO_2 photocatalyst in a plug flow reactor. According to the result, 90% of toluene conversion was reached under UVA irradiation (Bellè et al., 2022).

Qiu et al. reported an efficient elimination of low toluene concentration under simulated sunlight irradiations on Fluorine ion doped TiO_2-SiO_2 (Qiu et al., 2018). The fluorine doping led to the

formation of oxygen vacancies and Ti^{3+} , which act as charge-trapping agents, resulting in the reduction of the hole– e^- recombination. Further, the electrons of doped material located energetically below the CB of TiO_2 reduce the energetic gap of the combined catalyst, allowing excitation even under visible light. Other doping atoms and oxides are explored in the literature, including noble metals (T. Xu et al., 2020), transition metals, and metal oxide dopants (Rangkooy et al., 2019).

Nonthermal plasma (NTP) is an emerging technology for VOCs destruction, and numerous studies have been conducted on it for over twenty years.

Metal / Mixed metal oxide photocatalyst							
Catalyst	Support	Reactor configuration	VOCs	Flow rates	Light source	Conversion (%)	Reference
TiO_2	Mesh	Annular PFR reactor	Toluene (10 ppm)	1250 ml/min	UVA	90	(Bellè et al. 2022)
TiO_2	Porous Glass Fiber Cloth	Batch reactor Glass vial	2-Propanol (300 ppm)	-	UVA	100	(Yanagida et al. 2019)
$\text{TiO}_2/\text{MnO}_2$	ND	Con- tinuous-flow fixed-bed reactor as	Toluene (30 ppm)	1 L/min	VUV	96	(Yingguang Zhang et al. 2019)
TiO_2/WO_3	ZSM-5 zeolite	Continuous cylindrical photocatalytic	Xylene (50 ppm)	0.5 L/min	UVA	41.46	(Rangkooy, Ghaedi, and Jahani 2019)
$\text{TiO}_2/\text{Bi}_2\text{O}_3$	ND	Continuous planar reactor	Formaldehyde (4 ppm)	6.6 l/min	UVC	96	(Y. Sun et al. 2022)
ZnO/WO_3	Y-zeolite	Cylindrical quartz glass reactor	Styrene (20 ppm)	0.5 l / min	UVA	75	(Rangkooy et al. 2021)
Doped metal oxide photocatalyst							
Ir-TiO_2	ND	Packed-bed reactor filled	Toluene (1900 ppm)	350 ml/min	UVC	97	(Ho et al. 2022)
$\text{F-TiO}_2/\text{SiO}_2$	Glass	Flat interlaid reactor	Toluene (15 ppm)	80 mL/ min	Visible	90	(Qiu et al. 2018)
N-TiO_2	carbonaceous microsphere	Continuous flow reactor	Acetaldehyde (500 ppm)	20 mL/min	Visible	25	(G. Lu et al. 2021)
Ag-TiO_2	ND	Airtight reactor	Formaldehyde (0.5ppm)	-	Visible	91.3	(X. Jiang, Xu, and Yu 2019)
Pt - TiO_2	ND	Stainless steel cylinder	Toluene (0.2 ppm)	2 l/min	VUV	45.8	(T. Xu, Zheng, and Zhang 2020)
Mn-TiO_2	ND	Four openings PIREX glass reactor	Ethanol (400 ppm)	-	UVA	35	(Stucchi et al. 2018)
Cu-TiO_2	Optical fiber tissue	Continuous planar reactor	Butane-2,3-dione (5.6 ppm)	33 l /min	UVA	4	(Abou Saoud et al. 2021)
Au-TiO_2	ND	Batch sealed quartz reactor	Toluene (80 ppm)	-	Visible	57.3	(Yunyang Wang et al. 2019)

Table 2. *Experimental conditions and VOCs conversion performances at the different kinds of photocatalysts material.*

ND: not defined.

2.3. Non-thermal plasma discharge for VOC removal

Nonthermal plasma has drawn particular attention in the last decade among the technologies adopted for VOC remediation. . Many studies have highlighted the interest of this technology due to its higher energy efficiency, operation under normal temperature and pressure conditions, suitability for low VOCs concentration, and simple implementation process (S. Li et al., 2020a).

Plasma discharge can be defined as the fourth state of matter. Applying the potential difference between two electrodes in gas generates a plasma discharge. In other words, it is an ionized gas with various species, including electrons, photons, neutral, ions, metastable, and molecules, which are neutral electrically (Usman Hassan Dahiru and Kingdom 2023). The average electron temperature in non-thermal plasma is around 10^4 - 10^5 K (1-10eV), much higher than the gas temperature usually kept at room temperature (Schiavon et al., 2017).

2.3.1. NTP discharge types

For VOCs abatement, several kinds of NTP are developed, including dielectric barrier discharge (DBD) (Abou Saoud et al. 2020a); (Xiaohong Yao et al., 2018);(S. Li et al., 2022a) ; corona discharge (CD) (Tao et al., 2022) (Du et al., gliding arc (GA) (Kong et al., 2019), glow discharge (GD)(B. Chen et al., 2022) and microwave discharge (WD)(Q. Feng et al., 2021).

- Corona Discharge (CD)

A corona discharge appears when the action of the critical inhomogeneous electric field ionizes the gas surrounding a conductor. It is a self-sustaining discharge characterized by its luminous "corona effect," which depends on the geometry of the electrode (edges, wires, corners) (**Fig. 11a**).

Several CDs exist depending on the power source (pulsed corona discharge) and on the electrode polarity (positive and negative CD). Also, sometimes it is named according to the

shape of electrodes (pin to plate, multi-needle-plane, wire to cylinder ...) et al., 1991) (Wan et al., 2011)

- **Gliding arc discharge**

Gliding arc plasma consists of two symmetric electrodes placed perpendicularly to the centric gas carrier source (**Fig.11b**) (Fridman et al., 1998). The high voltage applied between the two electrodes generates an arc that glides along the electrodes in the presence of dynamic flow (L. Lin et al. 2006).

- **Glow discharge (GD)**

GD, commonly known as low-pressure discharge (<10milibar), is generated between two electrodes after applying the potential difference (**Fig.11c**). The atoms of the gas are ionized, giving the corresponding cations and a free electron. Furthermore, the free electrons are accelerated under the electric field, resulting in more and more ionization and excitation reactions. The excitation collisions and the radiative deexcitation from higher energetic levels result in the apparition of plasma light "glow" (Moon et al., 2004).

- **Dielectric barrier discharge (DBD)**

The dielectric barrier discharge (DBD) is the most popular NTP technology for VOCs abatement reported in the literature. Its success is due to its stability and the operating conditions (typically under normal atmospheric pressure and temperature) (Y. Zhu et al., 2023). The general concept of DBD plasma consists of dielectric material such as ceramic, glass, plastic, or mica sandwiched between a high voltage (HV) electrode and a ground electrode (GE) (**Fig.11d**). (Juan He et al., 2022). When the high voltage and frequency are applied, the gas inside is brought to its breakdown voltage, forming a discharge with a typical gap of 0.1 to 10 mm (Dou et al., 2008) (Subrahmanyam et al., 2006).

DBD reactors can have several configurations depending on the electrode configuration, including surface DBD, planar DBD, and cylindrical DBD, as demonstrated in **Fig. 12**.

Now that the various discharges configuration have been discussed, it is necessary to discuss how they play a role in VOCs degradation, focusing on the mechanism involved and the recent studies on the subject.

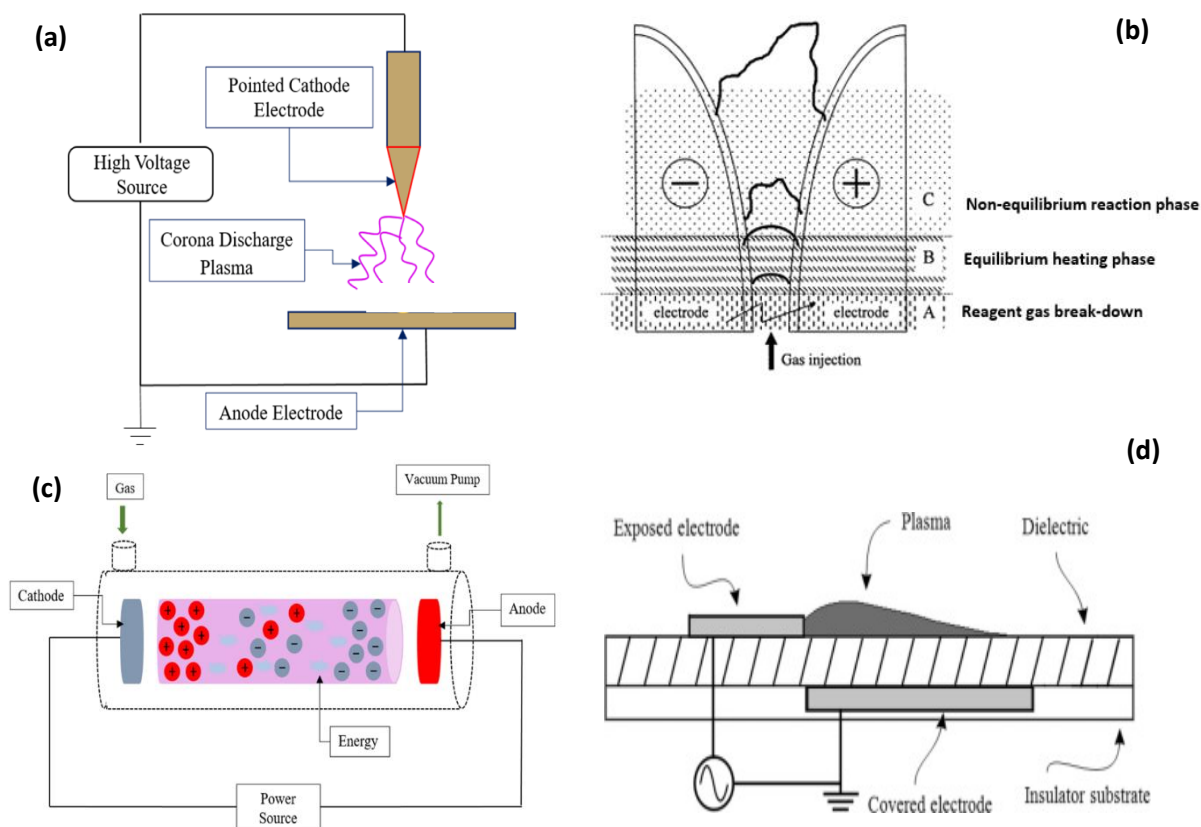


Figure 11. non thermal plasma discharges types: (a) Corona discharge (Reprinted with permission from (Birania et al. 2022) Copyright 2022 , Wiley), (b) Gliding arc discharge (Reprinted with permission from (Fridman et al. 1998) Copyright 1999, Elsevier), (c) Glow discharge (Reprinted with permission from (Birania et al. 2022) Copyright 2022 , Wiley) , (d) DBD discharge (Reprinted with permission from (Dalvand E., Ebrahimi, and Pouryoussefi 2018) Copyright 2017, Elsevier)

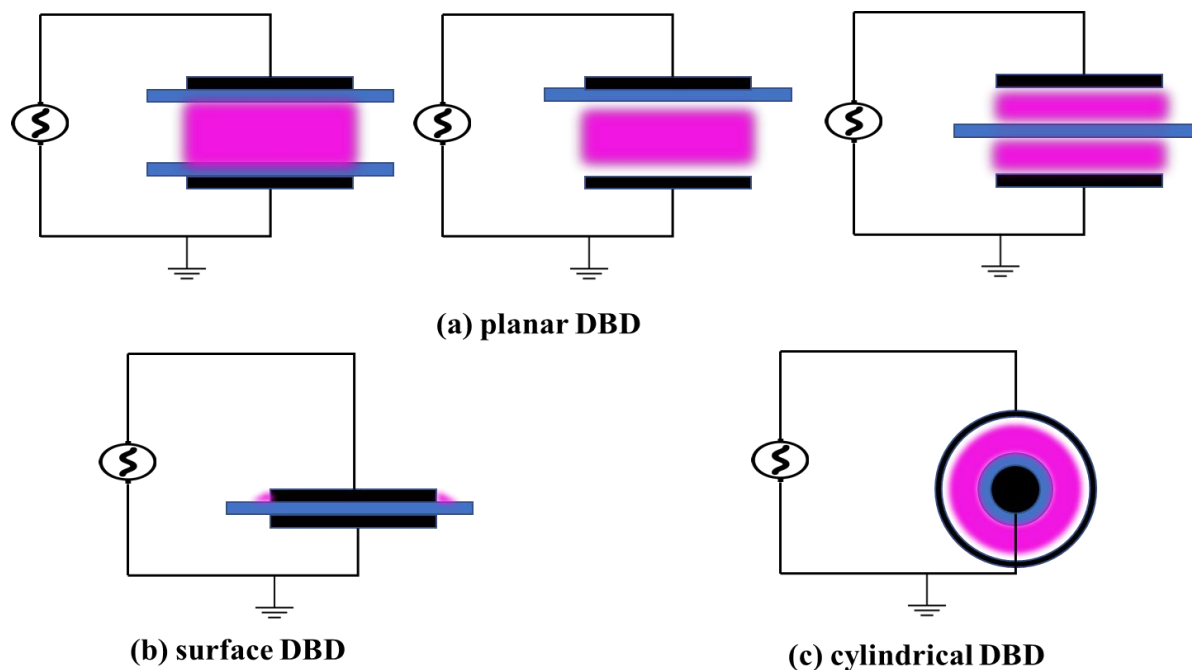


Figure 12. DBD plasma configurations

2.3.2. Mechanism of VOCs degradation under Plasma discharge

The degradation mechanism of several kinds of VOCs molecules in NTP plasma has been further studied. As in the case of the catalysis, the chemical composition of the target pollutant plays a crucial role in the degradation pathway and the reactions involved. From the summary table presented below (**Table 3**), aromatic hydrocarbons and chlorinated VOCs were the common molecules investigated (W. Zhou et al., 2021) (Saleem et al., 2021). The degradation of VOCs via plasma discharge occurs mainly through electron impact, ion collisions, and the interaction with the generated reactive species (as O, OH[°] and nitrogen metastable N₂ (A³Σ⁺_u)) in the discharge zone (Sultana., 2015). The mean energy of electrons in NTP plasma discharge can reach 10 eV (T. Zhu et al, 2017). It can easily break some molecule bonds as methyl group in the case of toluene (C–C: bond energy 4.4 eV, C–H: bond energy 3.7 eV, C=C: bond energy 5.4 eV, C–H: bond energy 4.6 eV in the ring) (**Fig.13**), leading to the formation of free radicals or initiates the ring opening, which subsequently triggers a series of reactions to produce small

inorganic molecules (NO_x , CO_x , and H_2O) or Macromolecules due to the recombination of actives components in the plasma phase (Fenglei et al., 2020). (S. Liet al., 2020).

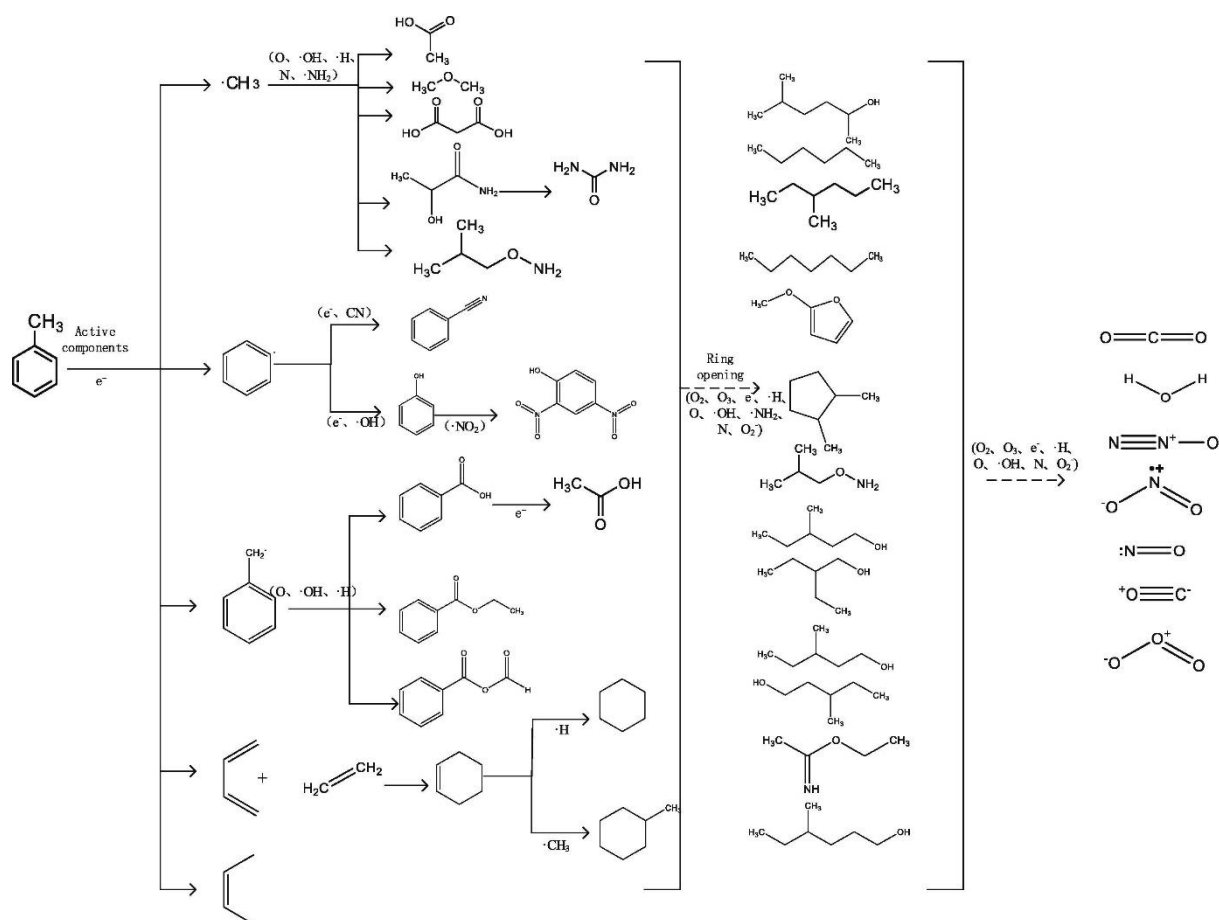
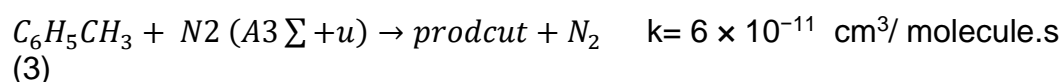


Figure 13. Possible mechanism of toluene degradation (Reprinted with permission from (Fenglei et al. 2020) Copyright 2020, Elsevier)

Michel Ondarts and coworkers report the role played by the reactive species generated in plasma discharge (i.e., O° , OH° , nitrogen metastable) on the degradation of toluene in corona plasma discharge. The following reactions were mostly expected in the case of toluene destruction (S. Li et al., 2022a):



The ions impact pathway was expected to be negligible (Ondarts et al. 2017)

Chlorinated compounds including Trichloroethylene (TCE) and chlorobenzene were also widely studied and decomposed by NTP plasma systems. The vast number of intermediates generated and the interaction with plasma species multiply the reaction channels. For TCE, the degradation mechanism starts with the abstraction of Cl under the action of energetic electrons and reactive species. The dissociation energy of the C–Cl bond (3.5 eV) is lower than that of the C = C bond (6.3 eV) and C–H bond (4.3 eV). The intermediates generated were further converted to CO₂, CO, HCl, Cl₂, and H₂O by the oxygenated species (O and OH) (T. Chang, Ma, et al., 2022).

The combination of plasma and catalyst exhibits high efficiency in the removal of VOCs with a synergetic effect. For this reason, a large number of studies have focused on this configuration, and the succeeding section will present a body of knowledge on the subject.

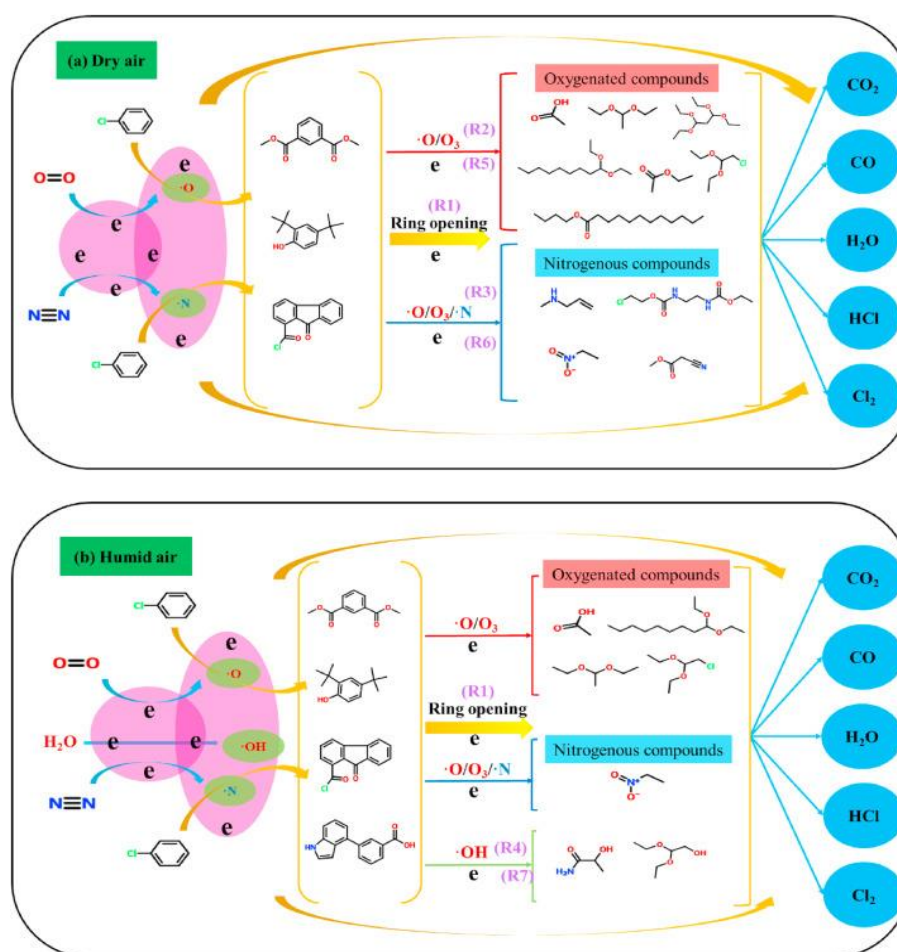


Figure 14. Degradation pathway of Chlorobenzene in DBD discharge under dry and humid conditions (Reprinted with permission from (W. Zhou et al. 2021) Copyright 2020, Elsevier)

DBD Plasma discharge							
VOCs	Discharge Type	Back ground gas	Reactor	Conversion (%)	By-products	O ₃ (ppm)	Reference
Cyclohexane (220 ppm)	Volumic DBD Dielectric: quartz	Dry air, Humified air, N ₂	two coaxial quartz tubes	98.2 % 3kJ/L	H ₂ , CO, CO ₂ , C1-C4	< 10ppm	(Usman H. Dahiru et al. 2021)
Chlorobenzene (750 ppm)	DDBD Dielectric: quartz	Dry air	Multi stack reactor	82 % 4.5kJ/L	NO ₂ , Carbone BP	160 ppm	(W. Zhou et al. 2021)
Toluene (8.6 ppm)	Volumic DBD: Dielectric: quartz	CH ₄	coaxial DBD	85.9 % 60kJ/L	H ₂ , LHC	ND	(Saleem et al. 2021)
Hexane (350 ppm)	Volumic DBD Dielectric: Glass	Air, N ₂	coaxial quartz tubes reactor	94.4 % 6kJ/L	H ₂ O, CO ₂	7 ppm In dry air	(Usman H. Dahiru, Saleem, Al-sudani, et al. 2022)
Methanol (260 ppm)	Volumic DBD Dielectric: Glass	Dry air, Humid air, N ₂	coaxial DBD	96.7 % 8.5kJ/L Dry air	CO ₂ , CO, O ₃ , CH ₄ , H ₂ , LHC	10 ppm	(Usman H. Dahiru, Saleem, Zhang, et al. 2022)
Benzene (350 ppm)	Volumic DBD Dielectric: Glass	Dry air, Humid air	two coaxial quartz tubes	93.7 % 6kJ/L	CO ₂ , CO, O ₃ , CH ₄ , H ₂ , LHC	7 ppm	(Usman H. Dahiru, Saleem, Al-Sudani, et al. 2022)
Formaldehyde (100 ppm)	DBD Dielectric: alumina	Air	Rectangular box reactor	95.12 % 2.5kJ/L	NO _x , O ₃ , CO ₂	2253	(Asilevi et al. 2020)
Acetone (80 ppm)	DBD Dielectric: Aluminum powder	Air	Cylindrical reactor	90% 24.3kJ/L	Formaldehyde, Methanol, Ketene, Acetaldehyde, Formic acid, Acetic acid, CO ₂	ND	(X. Li et al. 2019)
Naphthalene (20 ppm)	SDBD Dielectric: Mica sheet	Humid air	Cylindrical reactor	94% 6kV	NO _x , O ₃	50 ppm	(Abdelaziz, Ishijima, and Seto 2018)
Toluene (100 ppm)	Pulse modulated DBD Dielectric: quartz	Air	multistage rod-type reactor	62.9 % 2.3kJ/L	benzyl alcohol, 1,4-benzenediol, 2-methyl-, phenol, 3-methyl-4-nitro-, and 3-methyl-5-nitrophenol, CO _x , O ₃ , HCOOH	1160 ppm	(N. Jiang et al. 2020)
n-Hexane (50 ppm)	DBD discharge Dielectric: quartz	Air, N ₂ , He	Twelves parallel dielectrics quartz	98 % 1.629kJ/L air	H ₂ O, CO ₂ , NO _x , O ₃	120 ppm In dry air	(Son et al. 2021)
Toluene (300 ppm)	DDBD Discharge Dielectric: quartz	Air	coaxial cylindrical double dielectric barrier discharge	100% 0.9kJ/L	CO ₂ , CO	480 ppm	(S. Li et al. 2022b)
Corona plasma discharge							
Toluene (400 ppm)	Negative DC corona discharge	Air	Wire-plate rectangular box reactor	54.2 % ND	CO ₂ , CO	ND	(Guan et al. 2022)
Toluene (150 ppm)	negative DC corona discharge	Air	Wire to cylinder discharge chamber	91.5 % 2500 J/l	formic acid, acetic acid, maleic anhydride and benzaldehyde	2000 ppm	(T. Ma et al. 2018)
TCE (1000 ppm)	pulsed corona discharge	Air	Wire to cylinder	100 % 0.15 kJ/l	ND	ND	(Kuznetsov, Filatov, and Uvarin 2019)
TCE (58 ppm)	corona discharge	Humid air	Wire to cylinder	90.7% ND	2,2-Dichloroacetyl chloride (CHCl ₂ COCl), carbonyl chloride (COCl ₂), hydrogen chloride (HCl) and CO ₂	ND	(Z. G. Li and Zhao 2019)
Toluene (100 ppm)	DC corona discharge	Dry air	multi-wire-plate Rectangular box	52% 8.1kJ/L	CO, CO ₂	ND	(Xiaomei Yao et al. 2020)
Gliding arc plasma							
Benzene (70 ppm)	Gliding arc	Air	plate gliding arc (PGA) reactor	73 % 7.5kJ/l	CH ₄ , CO ₂	ND	(Rad et al. 2019)

Naphthalene (0.8 ppm)	Gliding arc	N ₂	AC gliding- arc reactors	93% ND	ND	ND	(Sasujit, Dussadee, and Tippayawong 2019)
Toluene (1400 ppm)	Gliding arc	Ar, O ₂	Three electrode reactor	87 % ND	Benzene Butadiene Acetone Benzaldehyde CO CO ₂	ND	(Lestinsky, Grycova, and Pryszcz 2018)

Table 3. Recent works on VOCs removal by NTP technologies.

2.4. Plasma-catalysis coupling for VOCs removal

2.4.1. Plasma-catalysis configuration

- PPC and IPC mode

The combination of plasma and catalysis processes operates in two different modes: (i) post-plasma catalysis (PPC), in which the catalytic reactor is located downstream of the NTP reactor. (ii) In plasma catalysis (IPC), the catalyst is introduced in the discharge zone, and the reaction occurs in the same reactor (X. Feng et al., 2018). (**Figs. 15a and b**).

The catalyst's presence downstream of the plasma reactor improved process performance. Firstly, generated species in the discharge area (first stage) such as ozone, radicals, electrons, ions, and excited species, induce a fundamental change in the background gas composition reaching the catalyst vessel (second stage) and contribute to the elimination of the residual pollutant at the catalyst surface (Yu et al., 2020; Baaloudj et al., 2022). Secondly, the catalyst located after the discharge plays a vital role in eliminating the by-products. For instance, the ozone generated in plasma can be eliminated and converted by the catalyst to atomic oxygen, further interested in VOC oxidation. Chang et al. highlighted the effect of the MnCo/HZ-5 catalyst placed post-NTP discharge on lowering the ozone concentration from 733.60 ppm to 226.33 ppm (T. Chang et al., 2019). Furthermore, Norsic et al. eliminate all gaseous ethanol in the PPC system, and the calculated carbon balance approaches 100 percent, which explains why the presence of the catalyst downstream of the NTP discharge is important for by-product inhibition (Norsic et al., 2018).

Tian Chang and coworkers treated trichloroethylene (TCE) in a PPC system over an Mn-Ce/HZSM-5 catalyst. The plasma reactor consists of a corona discharge vessel with a multi-pin-to-plate configuration. The catalyst was placed downstream of the discharge reactor at a temperature of 400°C. Almost total degradation of TCE was reached with CO₂ and CO

selectivities of 52.84% and 22.74%, respectively. Furthermore, Ozone depletion and utilization in the catalyst stage were also confirmed. Indeed, ozone produced by the discharge can be decomposed into oxygen species by an Mn-based catalyst, which is involved in the oxidation of TCE and intermediates (T. Chang et al., 2021).

Qianqian Yan et al. were able to convert 98 % of toluene and reach 50 % of CO_x selectivity in a DBD plasma reactor followed by α -MnO₂ /Cordierite honeycomb monolithic catalyst (CHM)(Yan et al., 2024).

The DBD reactor comprises a quartz tube with an aluminum foil wrapped around it as a grounding electrode, and a stainless-steel rod placed inside the tube as a high-voltage electrode. The catalyst is introduced into the catalytic reactor and positioned downstream of the DBD reactor. The authors noticed ozone reduction and particulate matter, confirming the catalytic treatment's benefit coupled with the plasma discharge process (Yan et al., 2024).

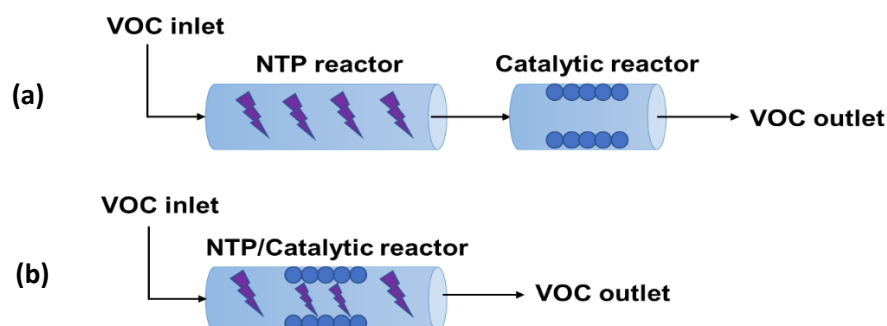


Figure 15. NTP-catalysis configuration

The combination of plasma and catalysis processes led to a beneficial effect, a phenomenon widely reported widely in the literature as a synergetic effect (X. Feng et al., 2018), holds significant promise. The term 'synergy', derived from the Greek word for 'teamwork' (Younus et al., 2019), in this context, signifies that the performance achieved by the plasma-catalysis combination surpasses the sum of plasma and catalysis performed individually, thereby piquing our curiosity about its potential.

Assadi et al. have made significant contribution to our understanding of the synergetic effect. Their investigation into the degradation of the gaseous trimethylamine by DBD plasma-coupled photocatalysis in one cylindrical reactor vessel revealed a synergetic effect for all experimental conditions studied. They attributed this behavior to several hypotheses, including plausible TiO_2 activation by the plasma UV and acceleration of by-product desorption by the reactive species generated in the plasma phase (Assadi et al. 2015).

Furthermore, Zhu et al. and coworkers demonstrated the positive effect of incorporating $\text{Au/CeO}_2/\text{Al}_2\text{O}_3$ nanocatalyst in the plasma discharge zone toward toluene removal. The removal efficiency of plasma alone in the condition considered is only 5 % and 67 % by the catalyst alone, while the combination of the plasma and catalysis in a hybrid reactor led to a complete removal of toluene with a high mineralization percentage approaching 90%. Additionally, the formation of secondary intermediates was inhibited, and catalyst poisoning was avoided (B. Zhu et al., 2020). As demonstrated in the summary Table 4, several researchers confirm the diminution of the ozone concentration after the incorporation of the catalyst in the plasma zone (Zadi et al., 2020) (Abou et al., 2018) (Assadi et al., 2015).

For instance, Pan and Chang reported the beneficial effect of integrating $\text{La}_2\text{CoMnO}_6$ perovskite in a DBD reactor (packed bed DBD). It has been observed that the $\text{La}_2\text{CoMnO}_6$ catalyst is totally ineffective for toluene removal at temperatures below 150 °C. Otherwise, the catalyst's exposure to the discharge led to a real boost in the degradation efficiency, with 100% toluene removal and 96.8% mineralization efficiency (Pan and Chang 2019).

Catalyst	Discharge type	VOCs	Reactor type	Flow rates	RE plasma alone	RE catalysis alone	RE combined	Ozone (ppm)	Synergetic factor ^c	Reference
SrTiO ₃ /rGO UV	DBD	Toluene (100 ppm)	Cylindrical quartz tube	3 L/min	90.9	3	100	Nd	1.06	(Mohammadi et al. 2020)
SiO ₂ -TiO ₂ UVA	DBD	Propanoic acid (3.24 ppm)	Co-axial Cylindrical quartz tube reactor	33 L/min	35	50	96	21 ^a 6 ^b	1.12	(Zadi et al. 2020)
Au/CeO ₂ /Al ₂ O ₃	DBD	Toluene (100 ppm)	Co-axial cylindrical reactor	0.2 L/min	67	5	100	Nd	1.38	(B. Zhu et al. 2020)
La ₂ CoMnO ₆	DBD	Toluene (150 ppm)	fixed-bed quartz reactor	1.2 L/min	82	0	100	Nd	1.21	(Pan and Chang 2019)
1 wt% Au/Al ₂ O ₃	DBD	Toluene (800 ppm)	Cylindrical reactor	0.06 L/min	60	0	96	Nd	1.6	(Quoc An et al. 2011)
TiO ₂ /GFT UVA	DBD	Trimethylamine (57 ppm)	Co-axial cylindrical reactor	33 to 166 L/min	39	34	91	215 ^a 150 ^b	1.24	(Assadi et al. 2015)
TiO ₂ /GFT UVA	DBD	Butyraldehyde (50ppm)	Co-axial cylindrical reactor	33 L/min	7	28	40	30 ^a 23 ^b	1.14	(Abou et al. 2018)
TiO ₂ /GFT UVA	DBD	Ammonia (50ppm)	Co-axial cylindrical reactor	33 L/min	20	51	80	30 ^a 23 ^b	1.12	(Abou et al. 2018)
WO ₃ /TiO ₂ UVA	DBD	Acetaldehyde (1425 ppm)	Quartz cylindrical reactor	Nd	1.75	14.03	19.29	Nd	1.22	(D. Li et al. 2022)
CoMn/TiO ₂	DBD	Chlorobenzene (300 ppm)	Cylindrical packed- bed reactor	2 L/min	2.3	78.3	97.1	Nd	1.20	(Song et al. 2018)
Pt/Al ₂ O ₃	Corona discharge	butyl-acetate (120 ppm)	Wire cylinder packed reactor	20 L/min	10	18	42	Nd	1.5	(Demidiouk, Moon, and Chae 2003)
1 wt.% Pd/Al ₂ O ₃	DBD	Propene (1000 ppm)	Cylindrical reactor	1 L/min	1	90	100	Nd	1.09	(Pham, Bui, and Khacef 2018)

Table 4. Plasma-catalysis combination for VOCs removal

^a Ozone concentration in plasma alone; ^b Ozone concentration in the presence of the catalyst; ^c Synergetic factor: the ratio of the removal efficiency of the combined system on the sum of the removal efficiency of plasma alone and catalysis alone

2.4.2. Plasma-catalysis hybrid reactor for VOC degradation

The reactor geometry plays a crucial role in the degradation performance of VOCs; in this section, special attention will be given to the plasma-catalyst hybrid reactor working simultaneously to abate gaseous contaminants.

Assadi and coworkers published several works on the degradation of VOCs using a variety of reactor configurations, among them the cylindrical pilot reactor (Assadi et al., 2014) (**Fig.16a**) which can be used to perform photocatalysis alone due to the presence of the UV lamp, contained in the reactor allowing the illumination of the catalyst wrapped against the internal reactor walls. This configuration also allows a facile generation of DBD surface plasma with an outer electrode covering the outside cylindrical tube and an inner grid High voltage electrode (Abou Saoud et al., 2020a)(Abou Saoud et al., 2020b). Coupling photocatalysis and DBD plasma simultaneously in the same reactor vessel was tested several times and high mineralization and degradation rates were achieved. The planar configuration can also be used as a photocatalytic reactor and as a plasma DBD–photocatalytic reactor was used to treat Butyraldehyde pollutants in the airstream. DBD/photocatalyst mode exhibited good CO₂ selectivity and low ozone concentration with an enhanced decomposition rate, leading to a synergetic effect (Gharib-Abou Ghaida et al., 2016).

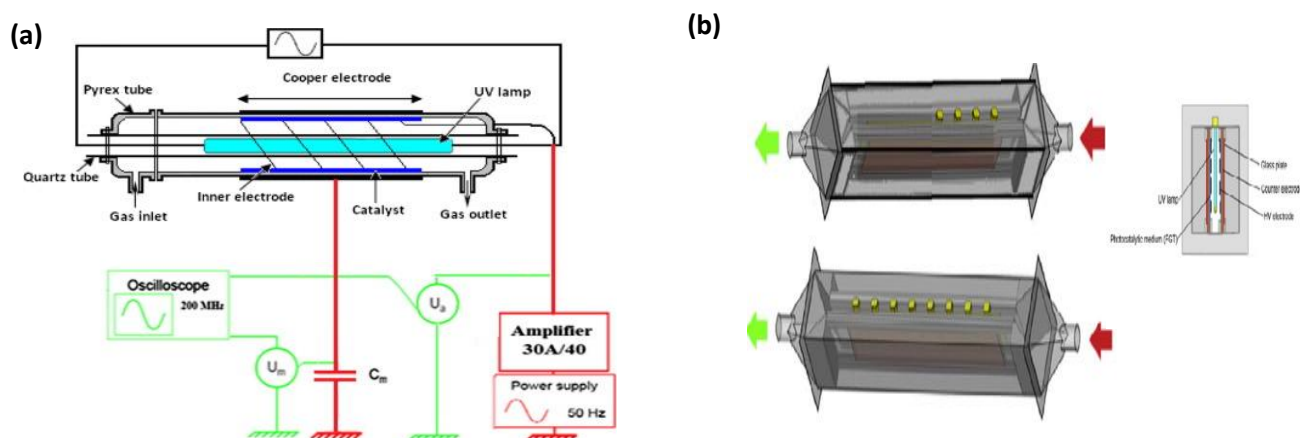


Figure16.(a) Cylindrical hybrid plasma – photocatalysis reactor, (b) Planar hybrid plasma – photocatalysis reactor

Antoine Rousseau and collaborators reported the performance of a U-shaped reactor used as an in-situ mode coupling DBD plasma and catalysis for the degradation of toluene. The reactor consists of a Pyrex tube U-shaped containing a tungsten wire electrode as the working electrode and a hollow brass cylinder as the counter electrode enrolled along the catalyst bed zone, as demonstrated in the **figure.17** (Jia et al., 2018)

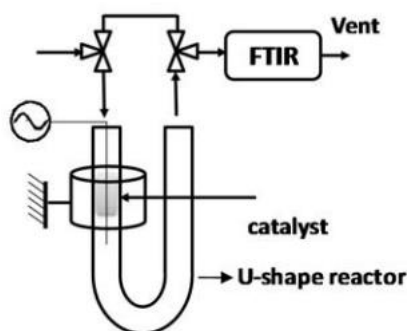


Figure17. U shape hybrid plasma- catalysis reactor , reprinted from (Jia et al. 2018)

It was reported in the literature that Packing DBD reactors with porous (Al_2O_3 , SiO_2) or non-porous material (ZrO_2) enhances significantly the characteristics of the discharge and improves the performance of the reactor in point of view of the degradation efficiency and mineralization (Veerapandian et al., 2017). The packing material possesses different shapes and sizes, primarily added in the form of spheres or cylinders. L. Sivachandiran designed a Mn_xO_y -coated glass beads packed bed non-thermal plasma reactor to convert isopropanol. It consists on a lab scale-reactor made of Pyrex glass tube. As described in the **Fig.18**, copper wire fixed inside the glass tube used as an HV electrode, and an external copper grid electrode wrapped outside the principal tube acting as a ground electrode leading to a discharge gap of 5.3 mm. This discharge zone was filled with Pyrex glass beads coated with MnO_2 (Sivachandiran et al., 2015).

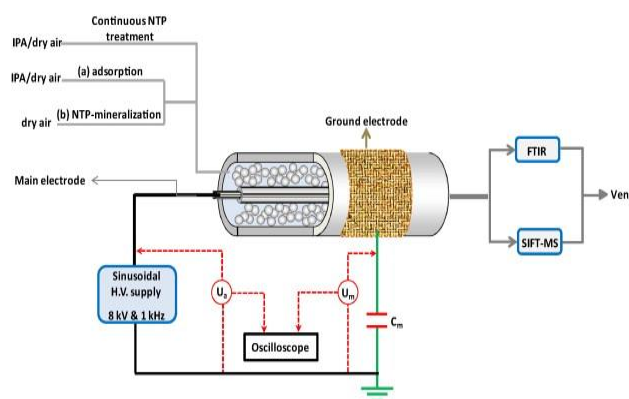


Figure18. Mn_xO_y packed bed non thermal plasma reactor for isopropanol conversion (Reprinted with permission from (Sivachandiran, Thevenet, and Rousseau 2015) Copyright 2015, Elsevier)

Recently, A great interest was given to the catalytic–plasma reactor based on a honeycomb catalyst configuration for VOCs removal (Nguyen, Yoon, et al 2022)(Nguyen, Dinh, et al 2022)(Hossain et al 2021). For instance, Shirjana Saud and coworkers investigated the degradation of gaseous diluted ethylene in 4 honeycomb PdO/ZSM-5/monolith catalysts packed in tubular polycarbonate tube and sandwiched between mesh electrodes , which act alternatively as ground or high voltage (HV) electrode (Saud et al. 2023). 97 % of Ethylene (30 ppm) was achieved at a specific discharge energy of 56 J/L and a temperature of 25 °C.

The mechanism involved in the hybrid plasma-catalyst process is still complex and based on the interactions between the catalyst and the discharge simultaneously, depending on the reactor configuration and the catalyst position. Based on a few studies, the mechanism involved will be discussed in the next paragraph.

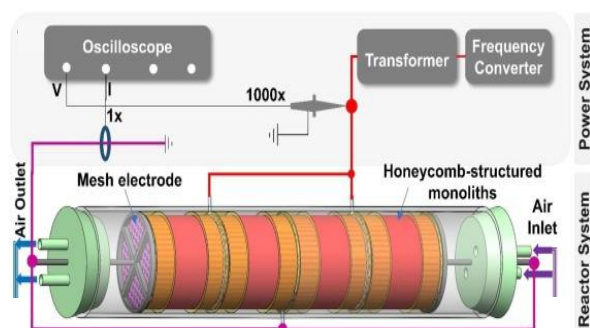


Figure19. Non thermal plasma - honeycomb PdO/ZSM-5/monolith catalysts reactor for diluted ethylene removal (Reprinted with permission from (Saud et al. 2023) Copyright 2022, Elsevier).

2.4.3. Mechanism of VOCs removal in plasma-catalytic systems

The comprehension of the mechanism and reactions involved in VOCs removal in a combined plasma-catalytic system is crucial for developing efficient and economical processes for VOCs remediation (Maxime et al., 2014). The multiple nanoseconds scale reactions of atoms and molecules involved in the plasma phase, such as dissociation, excitation, and ionization, complicate the identification of the degradation pathway (Samukawa et al., 2012). For VOCs oxidation over the catalyst surface, three well-known mechanisms are mainly evoked, like Mars-VanKrevelen (MVK) (Z. Huang et al., 2021), Langmuir-Hinshelwood (LH) (C. Feng et al., 2023), and Eley-Rideal (ER) processes. Thus, the action of plasma species and environment on the catalyst surface could impact the catalyst activity and induce change in the oxidation mechanism. Mutually, the presence of the catalyst inside the discharge zone could affect the plasma behavior.

Tian Chang and coworkers demonstrated the efficiency of NTP coupled with Mn/N-doped carbon catalyst for total toluene degradation (98%); the authors emphasized the critical role of catalyst presence under the discharge zone (T. Chang et al., 2024). It was established that toluene was firstly degraded in the gas phase by the collision with plasma active species (e.g., O_3 , O^+ , N^* , and $O\cdot$) and energetic electrons. Several by-products formed in this phase, and the unreacted toluene undergoes a catalytic oxidation into CO_2 and water over the catalyst surface. Additionally, the catalyst exhibited efficient ozone decomposition into oxygen-active species, which profits from the catalytic degradation of VOCs (Belkessa et al., 2023). Yuliang Shi investigated propane decomposition in a DBD plasma reactor combined with $Pt/CeMn_yO_x$ catalyst, and a synergetic mechanism was proposed. Through the surface analysis and characterizations of the catalyst as XPS and In-situ DRIFTS experiments, authors suggested that the first decomposition steps of propane started with the dehydrogenation reaction over the catalyst surface, which produces propylene. Then, the adsorbed oxygen species formed by the discharge promote the further degradation of propylene and other by-products, including acrolein and acrylic acid, into CO_x and water vapor (Yuliang Shi et al., 2024). Furthermore, it

was noticed that the increase in plasma discharge time reduces the number of intermediates on the catalyst surface, which is correlated with the synergy effect with the catalyst.

Most of the studies on this topic have evoked the synergetic effect exhibited by the combined process. However, its origin still needs to be better understood.

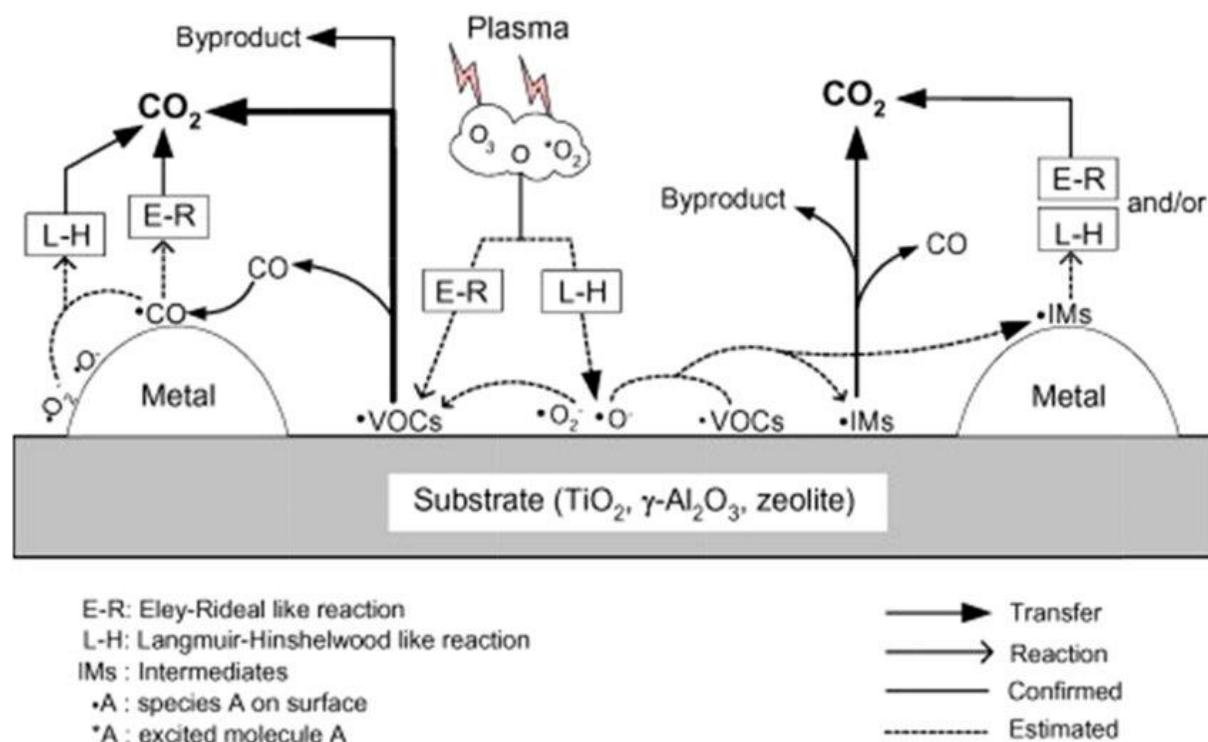


Figure 20. Mechanism of VOCs degradation under plasma -catalysis (Vandenbroucke et al. 2011) Copyright 2018, Elsevier)

3. Synergetic effect sources of plasma-catalysis

The presence of the catalyst in the plasma discharge zone improves the degradation performance of VOCs synergistically compared to the two processes operating separately. The literature announces several hypotheses to explain this trend, including the effect of the discharge on the catalyst's structure and composition (Neyts et al., 2015)..

This section of the review will highlight recent studies on the effect of plasma discharge on prepared catalysts.

3.1. Effect of the plasma on the catalyst

Conventional thermal preparation methods of heterogenous catalysts required extreme conditions, such as high temperature and pressure and toxic solvents and additives (Campanati et al., 2003). Therefore, Plasma can be used as an interesting alternative to prepare a highly active catalyst with the desired structure and composition only under mild conditions.

The impact of plasma discharge on conventionally prepared catalysts was largely highlighted in the literature. The treatment of the catalyst under discharge allows the surface modification by altering the roughness, the particle sizes, and oxygen vacancies. Plasma may also lead to surface functionalization depending on the discharge atmospheres (Neyts, 2016).

3.1.1. Catalyst surface activation

The effect of Pt/ CeO₂ catalyst treatment under DBD plasma discharge on its catalytic activity toward thermal toluene oxidation was studied by B. Wang et al. It was demonstrated that the sample exposed to the DBD discharge exhibits higher activity than the non-treated one. The characterization techniques show the modifications that occurred on the catalyst surface after DBD exposition: The surface roughness investigations provide more evidence that the catalyst is rougher after plasma bombardment. Additionally, better Pt nanoparticle dispersion and high oxygen vacancy concentrations are expected for the plasma treated catalyst, which results in lowering the activation energy and enhancing catalytic performance for toluene degradation (Bangfen Wang et al., 2018a). For instance, Shang et al. highlighted in their works the effect of the atmospheric pressure glow discharge plasma jet in enhancing the dispersion of Ni atoms in Ni/Y-Al₂O₃ catalyst and, therefore, boosting its catalytic activity (Shang et al., 2009). The characteristics of the catalyst under plasma discharge are mostly modified as demonstrated by D. Mo and Ye. After plasma treatment of the activated carbon fibers with TiO₂, more adsorbed oxygen species are observed compared to the untreated one, which can explain the enhanced photocatalytic activity towards gaseous formaldehyde. (D. Mo &Ye, 2009)

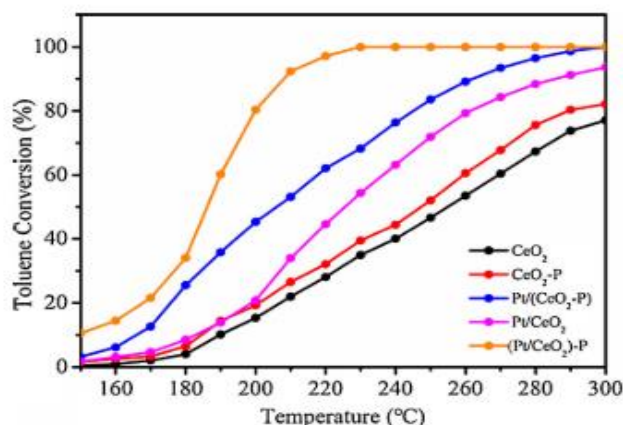


Figure 5. Toluene conversion at different catalyst (Reprinted with permission from (Bangfen Wang et al. 2018b) Copyright 2018, Elsevier)

Catalyst	OSC _{surf} ($\mu\text{mol O g}^{-1}$)	OSC _{Pt} ($\mu\text{mol O g}^{-1}$)	Ea (kJ mol^{-1})
Pt/ CeO ₂	124.87	25.46	79.1
(Pt/ CeO ₂)-P	137.86	26.48	63.8

Table 5. Characteristics of plasma treated and non-treated catalysts (Reprinted with permission from (Bangfen Wang et al. 2018b) Copyright 2018, Elsevier)

3.1.2. Catalyst regeneration

The catalyst deactivation phenomenon mostly limits catalytic reactions. This loss of activity is caused by chemical, thermal, and mechanical routes. Chemical deactivation refers to poisoning the catalyst active sites after the chemisorption accumulation of reactant compounds or by-products (Bartholomew, 2001). Furthermore, the accumulation of vapors as water vapors films leads to the blocking of active surfaces and causes severe deactivation. A Catalyst exposed for a long time to high temperatures is subject to surface degradations such as loss of catalytic surface area, pores size changes and active phase- solid support fragility (Jaison et al., 2023). The mechanical failures induced by crushing and abrasion cause irreversible loss of catalytic activity. Therefore, catalytic activity recovery is essential for the sustainable use of the catalyst. The plasma discharge process could be an exciting catalyst regeneration process (Kaliya Perumal Veerapandian et al., 2021). In a plasma-catalysis coupled system, the enhanced performances and the synergetic effect are mostly related to the in-situ regeneration of the catalyst by the reactive plasma species and the local heat provided by the discharge.

For instance, H. H. Kim et al. studied Au/TiO₂ catalyst regeneration by non-thermal plasma route. The result shows the effectiveness of the oxygen DBD plasma for removing adsorbed toluene on the catalyst surface and recovering catalyst activity (H. H. Kim et al., 2007). In other work, Zhu et al. confirmed the ability of pulsed plasma discharge to regenerate completely Au-based catalysts for CO conversion (B. Zhu et al., 2017).

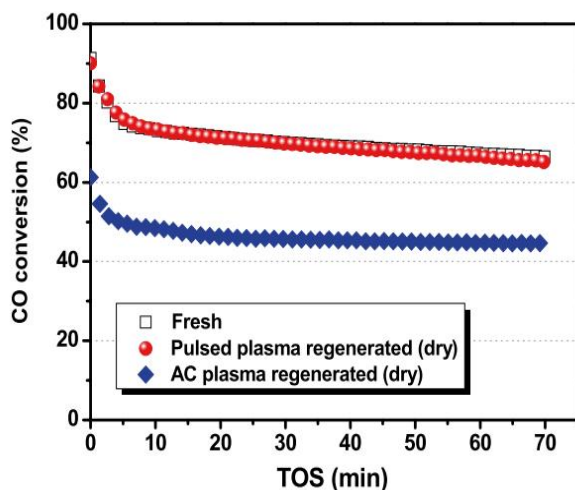


Figure 6. Catalyst regeneration by plasma discharge (Reprinted with permission from (B. Zhu et al. 2017) Copyright 2017, Springer Science)

Abou Saoud et al. studied the regeneration of TiO₂ coated on glass fiber tissue after poisoning by photocatalytic removal of DMDS and Butyraldehyde mixture. The regeneration monitored under the DBD air stream for 30 min allows the recovery of 23 % from the fresh catalyst (Abou Saoud et al., 2017).

3.1.3. Effect of plasma ionic wind on reactants adsorption

The produced ions in plasma discharge near the electrode's surface move towards the ground electrode under the action of the electric field and the momentum transfer between molecules located in the flow (Defoort et al., 2017). Thus, an electrohydrodynamic (EHD) force is produced, commonly known as the ionic wind (**Fig.23**) (Sato et al., 2019). The wind velocity depends mainly on the electrode's geometry, typically around 10 m/s (Hyun Ha Kim et al., 2016).

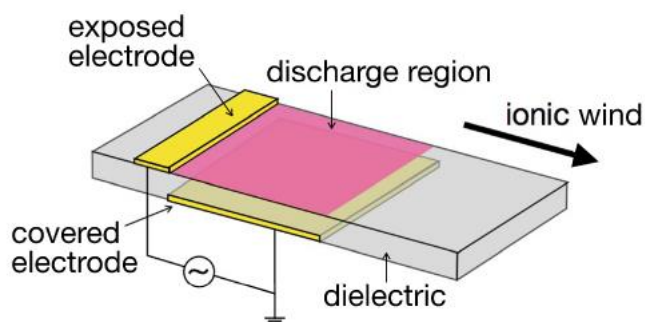


Figure 7. Ionic wind generation in surface DBD plasma (Reprinted with permission from (Sato et al. 2019) Copyright 2019, Nature)

Several works mentioned the effect of the ionic wind generated in plasma discharge on promoting gaseous molecule adsorption in a catalyst located in the plasma zone. For example, Y. Feng et al. demonstrated that mercury adsorption is enhanced by the action of the ionic wind. The EHD force causes several fluid disturbances, such as non-uniform fluid velocity and high Reynolds number, leading to the mass transfer improvement and, therefore, the best mercury adsorption was observed (Y. Feng et al., 2020). In their works, Lin et al. have reported the improvement of NO_x catalysis due to the ionic wind effect, which enhanced diffusion of gaseous NO_x towards La_{0.8}K_{0.2}MnO₃ perovskite type catalyst (H. Lin et al., 2007).

3.2. Effect of the catalyst on plasma discharge

The introduction of the catalyst inside the plasma discharge zone induces several effects on the characteristics of the discharge and enhances the performance of plasma-catalyst hybrid systems for VOCs conversion,

3.2.1. Plasma energy improvement

J. Li et al. conducted a comparative analysis of toluene degradation in DBD plasma alone and a combined plasma/CuO foam catalyst. The authors confirmed the beneficial impact of incorporating the catalyst in the discharge zone. Notably, the breakdown voltage for the DBD alone system was 18kV, while it was around 12kV for the combined system. The figure below illustrates the higher intensity and the brightness of the discharge in the plasma-catalyst reactor compared to the DBD alone (Ju Li et al., 2019).

In another study, it was conclusively demonstrated that the presence of the catalyst in the plasma volume amplified the contact points, thereby enhancing the electric field and the local electron energy (Pan & Chang, 2019). The introduction of the Ag/ZSM-5 catalyst in the plasma zone induces significant changes in plasma discharge characteristics, particularly in the current-voltage waveforms due to the surface streamers produced by the Ag/ZSM-5 catalyst (Fig.24).

Shirjana Saud demonstrated the effect of the presence of the catalyst on the enhancement of plasma energy inside the reactor. It was demonstrated through Lissajous curves with and without catalyst that the specified input energy inside the reactor was higher in the presence of the catalyst at the same applied voltage (Fig.25a-b) from (Saud et al., 2022)

Additionally, packing material has a higher dielectric constant than air, which enhances the local electric field and the electron energy (Yu et al., 2020)(Gadkari & Gu, 2018).

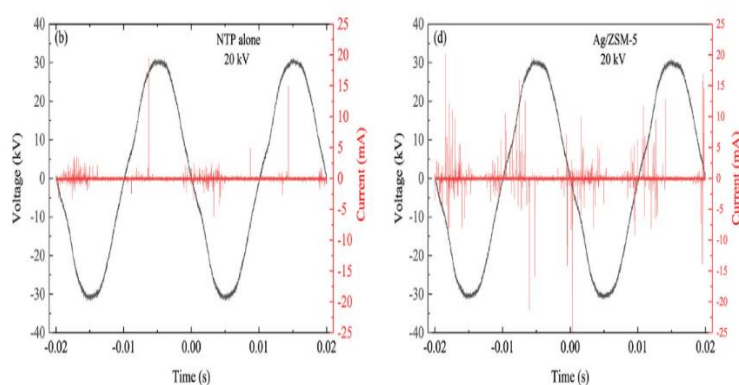


Figure 8. Voltage vs Current waveform in air alone and with the presence of the catalyst at 20 kV (Reprinted with permission from (Yu et al. 2020) Copyright 2020, Elsevier)

DBD configuration	Power density (W m^{-3})	Electron density (cm^{-3})	Electron energy (eV)
No packing	4×10^5	2.3×10^{10}	2.19
Partial packing	5.64×10^5	2.2×10^{10}	3.21
Full packing	7.13×10^5	5.8×10^6	7.33

Table 6. Effect of catalyst packing on the plasma characteristics as Power, electron density and electron energy (Reprinted with permission from (Gadkari and Gu 2018) Copyright 2018, PHYSICS OF PLASMAS)

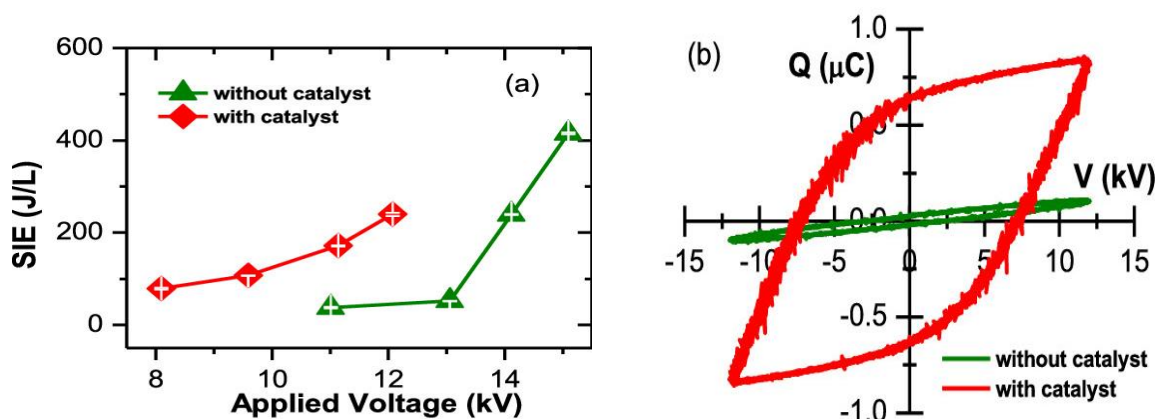


Figure 9. (a) Variation of specific input energy with and without the catalyst (b) voltage-charge Lissajous curves at 14 kV (total flow rate = 2 L/min; C_2H_4 = 250 ppm; RH = 100%; catalyst = 10 g; acquired average mode of eight samples for the oscilloscope) (Reprinted with permission from (Saud et al. 2022) Copyright 2022, American Chemical Society)

3.2.2. Formation of micro-discharge

The formation of micro-discharge in packed plasma reactors is mostly evidenced in the literature through several tools such as high-resolution camera imaging or numerical simulations (S. Li et al., 2020b)(Qu et al., 2021). Porous material inside the discharge reactor is most likely to generate micro-discharge inside the pores. The increase in the number and the intensity of micro discharge induces an increase of electron temperature and energy. Therefore, the breaking of VOCs adsorbed inside these pores is easily promoted. Yu-Ru Zhang and coworkers established a link between the dielectric constant of the catalyst and micro-discharge formation inside the pores via numerical modeling. They demonstrated that for catalysts with high dielectric constants, the plasma formation inside the pores is almost impossible, as summarized in the Fig. 26 (Y. R. Zhanget al., 2016). To support the idea of the micro discharge formation effect on the VOCs removal efficiency, Guan et al. studied the degradation of toluene in DC corona discharge with porous PTFE materials coated on the grounded electrode. Toluene removal is enhanced with an increasing the number of PTFE pores. The authors explain this trend by arguing that pores can expand the discharge zone, leading to more collisions between reactive species and toluene molecules. Additionally, micro-discharges initiate more ionization reactions, which could produce reactive entities, such as atomic oxygen and OH^\bullet radical mostly involved in VOCs abatment (Guan et al., 2022).

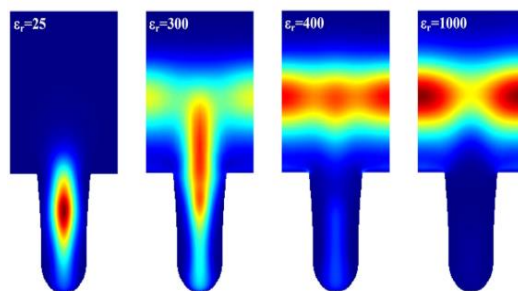


Figure 10. Effect of the dielectric constant on the micro discharges formation inside the pores (Reprinted with permission from (Y. R. Zhang, Neyts, and Bogaerts 2016) Copyright 2016, American Chemical Society)

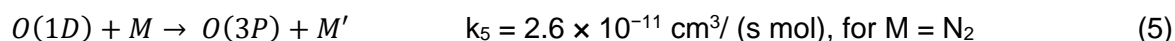
Active species generated in the gas phase discharge lose their energy by inter-species collision or with reagent molecules in the gas phase (Whitehead, 2019). For instance, OH radical lifetime is typically reduced by 5 in the presence of pollutant reagent in the plasma zone (Hibert et al., 1999). Thus, it limits the diffusion of these species to the catalyst surface and, consequently, their reactivity with the VOCs adsorbed. Hence, the generation of micro-discharge provides these strong oxidative species inside the pores near the target pollutant (Whitehead, 2019).

3.2.3. Effect of catalyst on ozone valorization

In the previous sections, the mutual effects of plasma-catalysis are discussed. Herein, the influence of plasma-catalyst combination in the same zone on the surface catalytic reactions will be detailed via the effect of the catalyst on ozone valorization.

Plasma discharge as DBD allows the generation of ozone, the most common by-product. The ozone is produced following two important steps:

- The oxygen in the carrier gas is ionized by plasma electrons leading to the formation of O (¹D) and O (³P), which corresponds to the excited-state and ground-state oxygen atoms, respectively (Holzer et al., 2002).



$$k_5 = 4.0 \times 10^{-11} \text{ cm}^3/(\text{s mol}), \text{ for } M = \text{O}_2.$$

- The ground-state oxygen atom produced via equations 4 and 5 reacts with the O_2 molecule in the presence of the third molecule to form the ozone molecule through equation 6 (Holzer et al., 2002).



Ozone is a strong oxidant agent, but the chemical reactions of VOCs with ozone are kinetically unfavorable compared to atomic oxygen (T. Changet al., 2022). For instance, the reaction rate of toluene with ozone in the gas phase at 298 K is about $1.2 \times 10^{-20} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, which is largely lower than the reactivity with an oxygen atom ($5.7 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) (Harling et al., 2009). The incorporation of some catalyst in the plasma discharge considerably reduces ozone production either by atomic oxygen adsorption in the catalyst surface and therefore limiting the production of ozone, or by the decomposition of formed ozone molecules at the surface of the catalyst (Baowei Wang et al., 2017).

J. Wu et al. performed toluene decomposition in a plasma catalysis hybrid system with Ni, Fe, Mn, Ce, and Cu oxides based catalysts. It was concluded that NiO/ γ - Al_2O_3 shows the best performance toward toluene destruction and ozone decomposition (J. Wu et al., 2013). The presence of the catalyst in the discharge zone allows a supplementary way for atomic oxygen production in addition to the classical way based on inter-species collision in plasma discharge or oxygen dissociation. In recent work, X. Zeng et al. investigated the effect of Cu-doped MnO_2 catalyst in a stage plasma-catalysis system toward KetoneVOCs degradation. The same study confirmed the effect of Cu-doped MnO_2 catalysts in ozone destruction. For example, at 630 J/L injected energy, the ozone concentration decreased by 100% over $\text{Cu}_{0.133}\text{-Mn}$ catalyst, which exhibits the best degradation performance (X. Zeng et al., 2020a).

MnO_2 is widely reported as a promising catalyst for ozone depletion, and Oyama and his coworkers first elucidated the corresponding mechanism. Three main steps are proposed:



Where * refers to the active site.

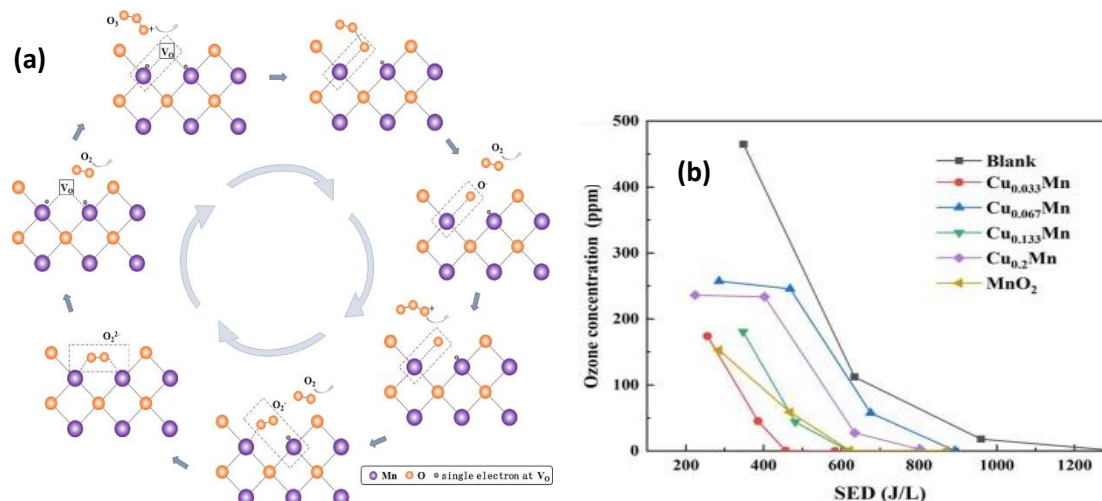


Figure 11.(a) Ozone decomposition cycle over MnO₂ catalyst (Reprinted with permission from (B. Xu et al. 2022) Copyright 2022, American Chemical Society), (b) Effect of Cu-MnO₂ based catalyst on the decomposition of ozone in plasma discharge (Reprinted with permission from (X. Zeng et al. 2020b) Copyright 2019, Elsevier)

4. Limitations and challenges:

Implementing a plasma-catalysis system for VOCs remediation has often demonstrated its performance on a laboratory scale with low or moderate concentrations of pollutants and low flow rates. However, its use at the industrial level is still a big challenge. The representative VOCs concentrations from industrial sources are around 100 to 1000 ppm, and to achieve a specific removal efficiency, high energy demand would be required, which inevitably increases the quantity of undesirable and toxic by-products such as O₃, NO_x, and CO (H. L. Chen et al., 2009). For instance, ozone concentration generated by plasma discharge is proportional to the specific energy density (SED) (Nur et al., 2016). Thus, for industrial applications, energy consumption and by-product formation are essential before implementing a plasma-based reactor.

Furthermore, the catalyst suffers from deactivation due to the accumulation of organic matter as carbonaceous, sulfur-containing, and chlorinated compounds, which drastically reducing

the process performances (T. Changet al., 2022). Therefore, industrial processes must take into account this problem. .

Finally, the design and ongoing maintenance of such industrial plants are very costly, which can be considered as a real challenge for chemical engineers.

Despite the above limitations, the plasma-catalysis process remains a very promising technology. It doesn't require the use of solvents or toxic chemicals and is suitable for a wide range of pollutants. Therefore, increasing efforts across several fields of technologies, including chemistry, physics, and mechanics, is the key to achieving a highly performant process with fewer limits.

For practical application, it is recommended that case studies be carry out under real conditions, focusing on the feasibility, overall cost, performance and safety.

5. Conclusion and outlook:

The recent works on VOCs removal by non-thermal plasma, catalysis, and their combination are discussed in this review paper. The current environmental issues facing the world led to the growth of the interest in this research area, which promises fantastic discoveries and facilitates the development of highly performant systems for air depollution. Heterogenous catalysis has always played a significant role in the chemical processes in both the laboratory and the industrial sector. But sometimes, it suffers from certain limitations, such as catalyst deactivation, the high temperature needed, and low selectivity. Non-thermal plasma alone has been integrated as a new water and air depollution technology with multiple advantages. Unfortunately, the plasma discharge is energy-consuming and generates ozone as a long-lived species. Thus, integrating the catalyst in the plasma (IPC) suppresses some limitations mentioned above and boosts the removal of VOCs.

As highlighted in numerous examples in this review, the interaction between the non-thermal plasma and catalysis demonstrated a powerful synergetic effect. This effect translates into superior process performances, including high conversion and mineralization efficiency, by-

product limitation, and catalyst conservation. Furthermore, plasma-catalysis coupling significantly reduces process costs and enhances energy efficiency, making it a viable option for domestic and industrial applications.

At the nanoscale level, the synergetic combination of plasma and catalysis is no more no less than the mutual interaction between the catalyst and plasma environment. On one side, it was demonstrated that the rich plasma environment led to several catalyst changes through the surface activation by the plasma species as electrons and reactive oxygen or nitrogen species. It is also suggested that the morphology of the catalyst can be altered during the discharge. Moreover, the catalyst deactivation issue can be resolved via in situ regeneration, which is allowed by the direct exposure of the catalyst to the discharge. Conversely, depending on the material structure and composition, the catalyst may affect the discharge entirely. For instance, the catalyst's presence enhances the electric field and the electron energy. In addition, the catalyst pores promote the formation of a micro discharge, leading to more reactive species formation. The catalyst is of great interest for reducing and valorizing the by-product, especially ozone. As is known, the hybrid systems plasma catalysis improves the mineralization efficiency. The synergy effect interaction between plasma and catalyst is crucial in optimizing existing processes regarding efficiency and cost. Therefore, it is necessary to explore in greater detail the interacting phenomena occurring at different scales using both experimental and theoretical approaches. To this end, several methods could be adopted, starting with in-situ probing of the plasma gas phase environment, catalytic reactions over the surface, and monitoring structural or chemical changes of the catalyst surface.

Furthermore, simulation and modeling are of great interest in understanding and providing design input for developing a new efficient plasma-catalytic process. Now that the synergy of the combined process has been demonstrated at the lab scale, future work under real conditions on a larger scale will be welcome to ensure its successful industrialization. To date, no study has investigated bacterial disinfection under a plasma-catalysis process. Thus, it must be a real prospect for new studies in the field.

Moreover, in the context of global warming, the control of CO₂ emission has become a hot topic. Therefore, plasma-catalytic technologies could be an interesting technology for CO₂ removal and valorizing.

Finally, this subject is a complex matrix that necessitates the collaboration of several disciplines, such as chemistry, physics, electrical, and mechanical science. This interdisciplinary approach is crucial for gaining a comprehensive understanding of the subject and for developing more efficient applications. It also underscores the broad impact of the audience's work, extending beyond their specific field and contributing to the larger goal of environmental preservation.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: