

A review on application of dielectric barrier discharge plasma technology on the abatement of volatile organic compounds

Wenjing Lu (✉)¹, Yawar Abbas¹, Muhammad Farooq Mustafa², Chao Pan¹, Hongtao Wang¹

¹ School of Environment, Tsinghua University, Beijing 100084, China

² Department of Environmental Design, Health and Nutritional Sciences, Allama Iqbal Open University, Islamabad 44000, Pakistan

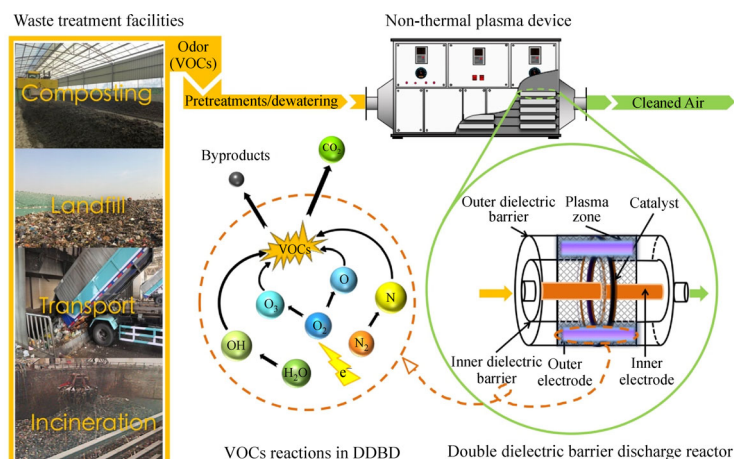
Applications of non-thermal plasma reactors for reduction of VOCs were reviewed.

Dielectric barrier discharge (DBD) plasma was considered.

Effect of process parameters was studied.

Effect of catalysts and inhibitors were evaluated.

GRAPHIC ABSTRACT



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ABSTRACT

Volatile organic compounds (VOCs) released from the waste treatment facilities have become a significant issue because they are not only causing odor nuisance but may also be hazardous to human health. Non-thermal plasma (NTP) technologies are newly developed methods and became a research trend in recent years regarding the removal of VOCs from the air stream. Due to its unique characteristics, such as rapid response at room temperature, bulk homogenized volume, high reaction efficiency, dielectric barrier discharge (DBD) plasma technology is considered one of the most promising techniques of NTP. This paper reviews recent progress of DBD plasma technology for abatement of VOCs. The principle of plasma generation in DBD and its configurations (electrode, discharge gap, dielectric barrier material, etc.) are discussed in details. Based on previously published literature, attention has been paid on the effect of DBD configuration on the removal of VOCs. Effect of various process parameters such as initial concentration, gas feeding rate, oxygen content and input power on VOCs removal are also considered. Moreover, the role of catalysis and inhibitors in VOCs removal by DBD system are presented. Finally, a modified configuration of the DBD reactor, i.e. double dielectric barrier discharge (DDBD) for the abatement of VOCs is discussed. It was suggested that the DDBD plasma reactor could be used for higher conversion efficiency as well as for avoiding solid residue deposition on the electrode. These depositions can interfere with the performance of the reactor.

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1 Introduction

Volatile organic compounds (VOCs) are emitted into the

atmosphere from numerous waste treatment facilities such as landfill sites, sludge dewatering and composting facilities, incineration and wastewater treatment plants etc. (Dobslaw et al., 2018). VOCs in the emissions of these waste treatment facilities are characterized mostly as hydrocarbons, sulfur compounds, halogenated com-

✉ Corresponding author

E-mail: luwenjing@tsinghua.edu.cn

pounds, terpenes etc. (Wu et al., 2010; Wu and Wang, 2015). Some of the VOCs are the essential precursors for the formation of particulate matter and photochemical smog (Oda, 2003; Shu et al., 2018). Some of these compounds are carcinogenic such as benzene, toluene, formaldehyde, etc. (Mustafa et al., 2017b). Therefore, the abatement of VOCs has become a serious concern to minimize their adverse effects on human health as well as to improve the air quality. Various technologies that have been developed in recent decades can be divided into three categories such as physical, chemical and biological methods.

Due to their low energy requirement, robotic and mechanical stability toward target molecules, adsorption and absorption are considered as efficient technologies for the removal of VOCs from the gas stream (Wang et al., 2014). However, these technologies have some drawbacks such as post-treatment/reactivating and disposal of used adsorbents, which may increase the overall cost.

Chemical abatement processes use chemicals to convert the VOCs into harmless or less toxic products. These processes often operate in the presence of heat as a source of energy. It primarily includes catalytic oxidation and thermal oxidation (with or without catalysts). Thermal oxidation processes operate at a temperature of 700°C to 1000°C at 0.5 to 1.0 s residence time. Catalytic oxidation proceeds in the presence of a catalyst at temperatures ranging from 250°C to 500°C. Compared to thermal oxidation processes, less NO_x is generated during the catalytic oxidation process (Carabineiro et al., 2015; Boycheva et al., 2018). However, major drawbacks of the chemical and thermal oxidation methods are catalytic deactivation, high-temperature requirement and generation of byproducts like dioxins and carbon monoxide (CO) in surplus amount (Kamal et al., 2016).

During biological technologies, organic pollutants may be converted into carbon dioxide and water; it is considered one of the cost-effective and environmental friendly technology for the VOCs abatement. Biological methods include biofilters and bioscrubbers (Schiavon et al., 2017a). Biological techniques generate less noxious by-products compared to physical and chemical technologies. However, their drawbacks include excess sludge and waste water generation, longer treatment time and low efficiency due to the slow growth of microorganisms (Padhi and Gokhale, 2014; Barbusinski et al., 2017; Xu et al., 2017a).

Non-thermal plasma (NTP) has been proved as a promising technique for the treatment of water pollution (Narengerile and Watanabe, 2012), soil pollution (Aggelopoulos et al., 2018), and air pollution (Zhang et al., 2018). Plasma the fourth state of matter and is an ionized form of gas consisting of free electrons and ions (Fridman, 2008). Plasmas possess properties such as Debye screen length, degree of ionization, electric conductivity and plasma frequency. The Debye screen length (λ_D) is a

measure of the effect of charge carriers within the specified area or certain length. The plasmas consist of both positive and negative charges. These charges exert forces on each other and collision can take place. However, these effects are limited to a certain length i.e. the Debye length (λ_D). This property of the plasma is of great importance for the interaction of charge carriers and is well explained by Keidar and Beilis (2018). The degree of ionization is also an influential factor for the plasma generation in which the atoms are ionized resulting in free electrons and ions. Mechanism of ionization in NTP is explained in detailed by Kim et al. (2017) while the calculation of the degree of ionization is well explained by Rutscher (2008). The property of electrical conductivity is significant for the plasma discharge. The electric field propagates free electrons, ions and gain drift velocities which generate an electric current density. The measurement of electrical conductivity of plasma is well explained by Crowley (2015). The electron plasma frequency is another important property of plasma since it has a significant role in the propagation of electromagnetic waves in plasmas (Rutscher, 2008).

Plasmas are classified mainly into two types according to electron temperature (T_e) and ion temperature (T_i), i.e. high temperature plasmas (HTP) and low temperature plasma (LTP). In HTP, the entire plasma particles, i.e. electrons, ions and neutral species have the same temperature, usually subjected to the high gas pressure. The temperature may reach values in the order of $T_i \approx T_e \geq 10^7$ K, it is also called hot plasma. The common example of HTP is fusion plasmas. Meanwhile, LTP can be subdivided into thermal LTP and non-thermal plasma. In the thermal LTP, all the charged and neutral particles (electrons and protons) have the same temperature in an atom and temperature of the particles may reach in order of $T_e \approx T_i \approx T \leq 2 \times 10^4$ K. Most popular example of the thermal LTP is "torch plasma" which is used in several environmental applications such as coal gasification, and other waste to energy processes etc. For NTP, the energy is only accumulated to the electrons and the temperature of electrons and ions are different, where $T_i \approx T \approx 300$ K and $T_i \leq T_e \leq 10^5$ K (Rutscher, 2008). NTP is therefore characterized as non-equilibrium plasma. Aforementioned, the energetic electrons are responsible for the propagation of reactions in NTP, resulting in dissociation of molecules and reactive species generation. The energetic electrons can degrade the molecules present in the gas environment and convert them to different transitional products (Schiavon et al., 2017b) and finally convert these into carbon dioxide (CO₂) and carbon monoxide (CO) (Ragazzi et al., 2014).

In recent decades, NTP has drawn remarkable attention for the removal of VOCs (see Table 1) from the gas stream. The merit of this technology includes rapid response at room temperature, generates high energized electrons within a very short interval of time, and at moderate

Table 1 List of typical VOCs degraded in NTP

Types of VOCs	Adverse effect on human health	Types of NTP used for abatement	Refs.
Toluene	Headache and dizziness	DBD	Guo et al. (2018b)
Benzene	Carcinogens	Corona	Ge et al. (2015)
Ethylbenzene	Eye and throat sensitivity	DBD	Hosseinzadeh et al. (2018)
Dichloromethane	Carcinogens	SPCP	Oda et al. (1995)
Trichloroethylene	Effects central nervous system	Corona discharge	Vandenbroucke et al. (2016)
Tetrachloroethylene	Skin irritation, heart and liver	DDBD	Mustafa et al. (2018)
Chlorobenzene	Lungs and urinary system	DBD	Song et al. (2018)
Xylene	Headache and dizziness	Corona discharge	Fan et al. (2009)
Styrene	Carcinogens	DDBD	Zhang et al. (2018)
Acetone	Carcinogens	DBD	Zhu et al. (2015d)
Formaldehyde	Carcinogens	DBD	Zhu et al. (2015c)
Hydrogen sulfides	Nervous system	DBD	Liang et al. (2011)
Dimethyl sulfides	Skin and eyes irritant	Corona discharge	Chen et al. (2009b)
Acetaldehyde	Probable carcinogen	Corona discharge	Klett et al. (2014)

capital cost (Raju et al., 2013; Ondarts et al., 2017). NTP can be generated through numerous ways including corona discharge (Ondarts et al., 2017), microwave discharge (Yang et al., 2017), arc/plasma torch (Choi et al., 2014), gliding discharge reactor (Młotek et al., 2015), electron beam (Kim et al., 2012), and dielectric barrier discharge (DBD) (Bahri et al., 2016). Among these various types of NTP, DBD plasma for decomposition of VOCs has attached great attention recently.

DBD, also known as barrier discharge, has the following advantages: first, a bulk homogenized volume plasma can be produced with the steady plasma area which ensures the gas to be treated uniformly. Secondly, DBD is more efficient and with lower energy consumption compared to other plasma techniques (Kogelschatz, 2003). Previous literature on treatment of air pollutants in DBD includes compounds as nitrogen oxides (NO_x) (Obradović et al., 2011), nitrous oxide (N_2O) (Trinh et al., 2016), ammonia (NH_3) (Abou Saoud et al., 2018), carbon disulfide (CS_2), airborne particles and bioaerosols (Park et al., 2011). DBD plasma already has a number of industrial uses since it has a simple configuration and can be operated under atmospheric pressure (Kogelschatz et al., 1997).

Although DBD as an end of pipe treatment, has been recommended in the literature for decomposing VOCs, it still has some disadvantages, mainly the production of undesired products, and partial mineralization, which lower the efficacy of DBD system and shorten the lifetime of the electrode (Delagrangé et al., 2006). To overcome these issues, researchers are associating the benefits of DBD by placing catalyst in the reaction zone/discharge zone or near the plasma discharge zone; the system is known as DBD-catalysis (Song et al., 2002).

This paper reviews abatement of VOCs through DBD

and DBD-catalysis system. Mechanisms occurring in DBD reactor and the effect of its configuration schemes on VOCs conversion have been evaluated. Review of the fundamental process parameters including flow rate, initial concentration, oxygen content and energy input is provided. It also gives an outline of the literature on a combined system of DBD-catalysis. Also, the effect of inhibitors (humidity, sulfur contents, chlorine, and deposition) on DBD performance is presented. Finally, a modified DBD reactor, i.e. double dielectric barrier discharge (DDBD) plasma reactor for abatement of VOCs has been reviewed and proposed for better performance.

2 Configuration of dielectric barrier discharge (DBD) reactor and their effect on VOCs abatement

DBD also called silent discharge and barrier discharge is an electrical discharge process that is created between two electrodes and a dielectric barrier. The placement of insulating material in the discharge zone which ensures the separation of gas from both sections is the major principle for the generation of plasma in the DBD reactor. As shown in Fig. 1, DBD system can be built in numerous configurations such as planer with parallel plates detached by a dielectric (a), and cylindrical type (b), in which a dielectric tube is used between the coaxial plates (Kraus et al., 2001). Planer and cylindrical are most popular configurations of the DBD reactor for pollution control, especially for water, air, and soil. Plasma discharge requires higher electrical voltage to generate a strong electric field for the degradation of targeted pollutant

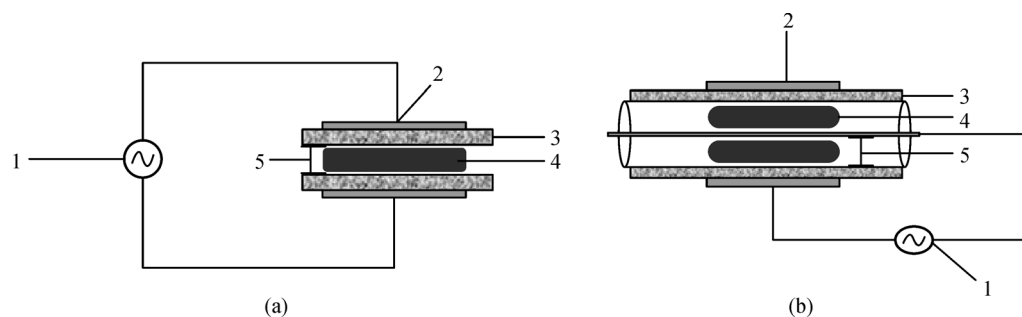


Fig. 1 Configurations of dielectric barrier discharge (DBD) reactors, where 1 is the alternating current (AC) at high voltage; 2 is the electrode; 3 is the dielectric barriers; 4 is the discharge zone; 5 is the discharge gap. (a) Planer DBD configuration; (b) Cylindrical DBD configuration.

(Kogelschatz, 2003; Yi et al., 2003). In this type of NTP techniques, plasma is produced by transferring the electrical energy into energetic electrons rather than gas heating. The gas temperature remains as low as room temperature and a voltage of 1–100 kilovolt (kV) with frequencies of 50 Hz–1 MHz is necessary to sustain plasma discharges (Wang et al., 2012; Xiao et al., 2014). Various types of discharge mode can occur in DBD system such as filamentary discharge mode, which is formed by discharges or streamers that are randomly distributed in the discharge gap between the electrodes. Another concept is the homogenous glow discharge mode, also called atmospheric pressure glow discharge mode (Ge et al., 2015).

The configuration of the DBD plasma reactor is well described in the literature (Brandenburg, 2017). The reactor configuration consists of electrodes, discharge length, volume and position of the catalyst; all these parameters have a significant effect on the conversion and energy efficiency of the system. The impact of these variables is overviewed and discussed based on published data.

2.1 Electrode

In the DBD system, electrodes play a vital role in the generation of ionized and reactive species. The physical and chemical characteristics of the electrode material affect the input power, which may enhance the energy of the ionized electrons. The intensity of plasma and discharge is related to the electrode material because different electrode materials have different electric properties (such as electric resistivity and conductivity), which can affect the plasma discharge. So far, numerous types of electrode material are used in different studies, including metallic rod (Maguraneanu et al., 2007), brass wire (Guo et al., 2010), copper (Guo et al., 2010), tungsten (Zhang et al., 2015), ferroelectric material (Subrahmanyam et al., 2010) and sintered metal fiber (SMF) (Raju et al., 2013).

Jiang et al. (2013) had examined the effect of electrode geometry on the decomposition of benzene. Three different

geometries were tested in the experiment including coil, rod, and bolt geometries. The coil electrode reactor showed better performance, i.e. 55% and 220 J/L for decomposition rate and specific input energy (SIE) respectively, than using the bolt and rod electrode. This is due to a decrease of the dielectric loss in the barrier. Iwamura and Saito (2015) studied the removal of benzene in dry air with different materials as inner electrodes, i.e. titanium, stainless steel, and oxidized titanium. They concluded that the conversion efficiency of benzene was higher with the oxidized titanium electrode due to the photo-catalytic property of the oxidized layer. The electrode with a photocatalytic property could be highly activated in the NTP at the peak voltages without extra UV light source (Chen et al., 2016). Sivachandiran et al. (2012) studied the abatement of chlorobenzene in the air. Three different electrodes including silver paste, copper wire, and aluminum foil were tested. At the SIE of 310 J/L, the conversion efficiency was 90% using silver paste electrode, which had decreased to 85% for copper wire and 80% for aluminum foil respectively at SIE 264 J/L and 236 J/L, respectively. Moreover, the CO₂ selectivity was also improved when using silver paste electrode. They concluded that the generation of abnormal corona outside the reactors with the copper wire and aluminum foil could be the reason of less conversion compared to silver paste. The corona can occur in the gaps between the dielectric barrier and ground electrode due to poor contact between them. On the other hand, in the case of silver paste, no gap between the electrodes and the dielectric barrier enhances the maximum energy consumption resulting in the generation of micro-discharges. Kim et al. (2004) also found the same results while treating benzene using various electrodes, i.e. silver paste, aluminum tape and copper mesh.

To conclude, the electrode shape and material has a noteworthy effect on the conversion efficiency of the VOCs. An appropriate electrode configuration in the DBD system could enhance the efficiency of a DBD system. For this account regarding different pollutants further investigation is needed.

2.2 Discharge gap

Discharge gap is one of the most significant factors in DBD reactor configuration. The discharge gap is the distance between the inner electrode and dielectric barrier where reactions are mostly taken place (Fig. 1). In DBD plasma reactor an appropriate discharge gap length is required to sustain stable plasma. The discharge gap in laboratory investigation for gas treatment ranges from 1 to 6 mm, and most of the studies have been conducted with the discharge gap of 2 to 3 mm (Karuppiyah et al., 2013; Mei and Tu, 2017).

Magureanu et al. (2007) studied the removal of trichloroethylene (TCE) with discharge gaps ranging from 1 to 5 mm. It was found that a shorter gap within the range of 1–3 mm appears to be more efficient for TCE abatement. The improvement of the conversion efficiency of VOCs with the shorter discharge gap may arise for two reasons: First, shorter discharge gap creates more energized and active species ($N\cdot$, $O\cdot$, $OH\cdot$, O_3 , and O_2) in discharge zone, which can increase the probability of collision between the targeted pollutant and active species. Secondly, by decreasing the discharge gap between electrodes and the dielectric layer can also increase the electric field strength that could improve the energy in the reaction zone, which ultimately enhances the conversion efficiency (Ma and Lan, 2015). Moreover, a shorter discharge gap has a positive effect on limiting the mobility of active and energized electrons in the discharge zone, which also contributes to the increased collision possibilities (Sultana et al., 2015). On the contrary, an increase in the discharge gap length requires higher input power and form non-homogenous discharge plasma and can cause a transition from the discharge to spark (Schiavon et al., 2017b). Moreover, the increase in the discharge gap decreased the power density, which is defined as discharge power/discharge volume at constant power due to the rise in plasma volume. Mei and Tu (2017) also pointed out that the increase in discharge gap could lead to a scarce or partial discharge rather than a full plasma discharge. In the latter case streamers can not cover the discharge plasma volume, which consequently reduced the conversion efficiency. To conclude that a shorter plasma discharge gap is more favorable to VOCs abatement.

2.3 Discharge length

Discharge length is linked with the discharge zone or length of high voltage electrode or reaction zone where highly ionized electrons are produced. Studies found that the discharge length has a positive link with the conversion efficiency of VOCs.

Zhang et al. (2014b) investigated the decomposition of styrene with different discharge lengths, i.e., 1.8, 3.6, 5.4, and 7.2 cm and found that the maximum conversion efficiency of styrene was attained at the most extended discharge length, i.e., 7.2 cm. It also showed that the selectivity of carbon dioxide (CO_2) and CO improved with the increase of discharge length. The highest selectivity was 84% with a discharge length of 7.2 cm and the lowest selectivity was 53% with discharge length of 1.8 cm. Similarly, Khoja et al. (2017) investigated dry reforming of methane using various DBD reactors. They found that the conversion efficiency of CH_4 could be improved with an increase in the discharge length from 10 to 30 cm. The increase in the discharge length could help in prolonging the residence time of pollutant in reaction/discharge zone and increase the probability of collision between the pollutants and high energized electrons and active species (Ashford and Tu, 2017). However, more input power is required to generate a stable plasma for an enlarged discharge plasma zone and in addition more energy is consumed in the discharge zone for the ionization and generation of energized electron and active species.

2.4 Dielectric barrier materials for DBD system

A dielectric barrier material is an electrical insulator that can be polarized by an applied electric field. Different dielectric materials have different dielectric properties as shown in Table 2. In the DBD system, the dielectric barrier is one of the important discharge parameters as it can affect the equivalent dielectric capacitance (Wang et al., 2012).

The main characteristics of the dielectric barrier become obvious, when it is placed in the electric field. In this case, the electric charges can not flow through the material and prevent spark formation. Moreover, the direct exposure of reactive species generated in the discharge zone to the metallic electrode causes its corrosion or erosion. To avoid this phenomenon, a dielectric barrier is used to cover the

Table 2 Properties of the dielectric constant of commonly used dielectric material in DBD reactor

Dielectric material	Dielectric constant
Glass	4–6
Quartz	4–7
Aluminum oxide	~10
Ceramics	Ten-10 ks
Ferroelectrics	Hundred-10 ks

metallic electrodes (Brandenburg, 2017). In most of the studies, quartz tubes (Huang et al., 2008; Raju et al., 2013; Chen et al., 2016), silica glass, alumina (Byeon et al., 2010; Kim et al., 2011) and ceramics (Gandhi and Mok, 2012; Guo et al., 2017) are used as dielectric barriers. Among them, due to its moderate cost and commercial availability, quartz is the most popular dielectric barrier for VOCs conversion in DBD.

Kundu et al. (2012) compared quartz and alumina as a dielectric barrier material for the decomposition of methane. The alumina dielectric barrier showed a 5% higher conversion of CH₄ than the quartz. Recently Khoja et al. (2017) confirmed a higher conversion efficiency of CH₄ using an alumina dielectric barrier reactor. The dielectric constant of aluminum (9.8) is higher than the one for quartz (3.7). Alumina contributed stronger to an electric field and more energized electrons, which lead to higher conversion efficiency. Moreover, the higher conversion efficiency of CH₄ in the alumina dielectric barrier may be due to porosity and roughness of the material, which provides more residence time. This was confirmed by (Kraus et al., 2001; Roland et al., 2005) who found that porous material has strong capability to adsorb the gas molecules and extend the residence time.

In general, conversion efficiency can be increased with the use of the appropriate dielectric barrier in the DBD system. The high dielectric constant can generate a strong electric field and thus positively affect the ionization of the gas. The materials with high dielectric constant have also the advantage to reduce the breakdown of voltages. The advantage of high dielectric constant materials such as ferroelectric materials can be used in industrial applications for the abatement of VOCs. Barium titanate as a ferroelectric material with a dielectric constant of 2000 to 10000 is considered a critical material when applied in the environmental pollution control. Further research regarding dielectric barrier is needed for higher energy efficiency and better performance of the DBD reactor.

3 Effect of processing parameters on VOCs abatement

In this section, based on published studies, various process parameters are discussed which have significant impact on DBD systems regarding conversion and energy efficiency. The conversion efficiency of VOCs is defined as follows;

$$\text{Removal efficiency (\%)} = \frac{[\text{VOCs}]_{\text{in}} - [\text{VOCs}]_{\text{out}}}{[\text{VOCs}]_{\text{in}}} \times 100, \quad (1)$$

where $[\text{VOCs}]_{\text{in}}$ is the initial concentration of incoming VOCs and $[\text{VOCs}]_{\text{out}}$ is the final concentration of the VOCs after DBD treatment.

An important parameter is the specific energy input efficiency defined as follows;

$$\text{Specific Input Energy (SIE)} \left(\frac{\text{J}}{\text{L}} \right) = \frac{P(\text{W})}{Q(\text{L}/\text{min})}, \quad (2)$$

where the P is the discharge power in (Watt) and Q is the flow rate (L/min).

The performance of the DBD plasma reactor depends on the removal efficiency, SIE, energy efficiency and selectivity of CO₂, which entails the degree of mineralization defined as follows:

$$\begin{aligned} \text{CO}_2 \text{ selectivity (\%)} \\ = \frac{[\text{CO}_2]_{\text{out}} - [\text{CO}_2]_{\text{in}}}{\text{No. of carbon in VOC} \times [\text{VOC}]_{\text{in}} - [\text{VOC}]_{\text{out}}} \\ \times 100, \end{aligned} \quad (3)$$

where $[\text{CO}_2]_{\text{in}}$ is the concentration of CO₂ at the reactor inlet, ppm; $[\text{CO}_2]_{\text{out}}$ is the concentration of CO₂ at the reactor outlet, ppm; $[\text{VOCs}]_{\text{in}}$ is the initial concentration, ppm; $[\text{VOCs}]_{\text{out}}$ is the final concentration of VOCs after NTP treatment, ppm.

3.1 Initial concentration of VOCs

VOCs emission varies with the pollution sources, therefore, the influence of the initial concentration of VOCs on DBD performance has been extensively studied (Abedi et al., 2014; Schiavon et al., 2015; Zhu et al., 2017). When the initial concentration increases, each VOC molecule shares fewer electrons in the discharge zone. The available energy and concentration of reactive species in the discharge zone are not sufficient to collide with the increased number of targeted molecules. Zhu et al. (2015b) studied the decomposition of toluene in dry air with different initial concentrations, i.e., 159.21 ppm, 265.25 ppm, 451.1 ppm, and 663.39 ppm. They found that the conversion efficiency is reducing with increasing initial concentrations of toluene. The maximum removal efficiency of 95% was achieved among the tested lowest initial concentration (159.21 ppm). Ye et al. (2008) investigated the decomposition of benzene with initial concentrations varying from 31 to 939.06 ppm. They found that the conversion decreased with increasing initial concentrations of benzene. The maximum conversion efficiency of 70% was achieved at an initial concentration of 31 ppm. Similarly, other studies (Vandenbroucke et al., 2011; Li et al., 2014; Mustafa et al., 2017a) also confirmed that the conversion efficiency decreases with increasing initial concentrations. It can be concluded that the treatment of gases in a DBD system with high initial VOC concentrations need either a longer residence time or a higher input power.

3.2 Gas feeding rate

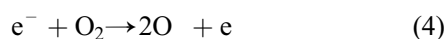
The feeding rate of the gaseous pollutants can affect the plasma discharge as well as the residence time of the active species. In laboratory scale DBD experiments, the applied gas flow ranged from 1–10 L/min, which had a significant influence on the conversion efficiency of the VOCs (Abd Allah et al., 2014; Schmidt et al., 2015).

Shahna et al. (2017) confirmed that with an increasing flow rate from 0.3 to 1.2 L/min the decomposition of chlorobenzene decreased while, at an applied voltage of 8.5 kV, the toluene conversion efficiency decreased from 50% to 30% when the gas feeding rate was increased from 1 to 5 L/min (Byeon et al., 2010). Other studies (Chang and Lin, 2005; Kwong et al., 2008; Mista and Kacprzyk, 2008; Liang et al., 2013) also found the same decreasing conversion efficiency pattern when rising the feeding rate.

As the effect of the gas feeding rate on the conversion efficiency of VOCs is related to the residence time of the substrates. Therefore high flow rates reduce the probability of collision between the targeted molecules and active species (Chavadej et al., 2007). In addition, at a high flow rate, targeted molecules can by-pass the discharge plasma without any reaction. Zhu et al. (2015a) studied the influence of the residence time of 5 to 15 s and gas feeding rates of 0.36 to 1.0 L/min on the removal efficiency of chlorobenzene. It was found that the removal efficiency is positively correlated with the residence time. The highest removal efficiency of 86.2% was achieved at a residence time of 15 s and a flow rate of 0.36 L/min. Magureanu et al. (2005) also found an improvement in conversion efficiency with an increase in residence time.

3.3 Oxygen content in the gas stream

The oxygen (O₂) concentration in the plasma is an important parameter that influences the discharge plasma and thus plays a substantial role in plasma reactions. The number and type of active species are highly influenced by the type of the carrier gas. In the case of the influence of O₂ concentrations in a NTP system, the mechanism for generation of ozone consists of two steps (Eqs. (4) and (5)) (Ma et al., 2014): oxygen molecules collide with the high energy electrons (e) and dissociate into oxygen atoms; in the second step, oxygen atoms collide with oxygen molecules and form ozone molecules.



Mok et al. (2008) studied the influence of the O₂ content on the removal efficiency of trichloromethane. They found out that limited amounts of O₂ had a positive effect on its reduction. The maximum removal efficiency was obtained at 0.5% O₂ content. Chipier et al. (2009) investigated the

removal of 2-Heptanone in N₂ and N₂/O₂ mixtures. They found out that with a low increase of the O₂ content of about 2%–3% in the N₂ a 30% higher conversion efficiency of 2-heptanone was achieved compared to a pure N₂ atmosphere. On the other hand, the removal efficiency of 2-heptanone had decreased with the further increase of the O₂ content. The highest reduction efficiency of dichloromethane (CH₂Cl₂) was achieved with the addition of 2%–4% O₂ into the nitrogen gas (Abd Allah et al., 2014). It was also discovered that at O₂ concentrations $\geq 4\%$ the reduction of chlorinated VOCs decreased.

In conclusion, a slight increase in the O₂ concentration in the DBD system can enhance the generation of active radical species, which results in a higher reduction of VOCs. On the other hand a higher concentration of O₂ in the gas stream may decrease the removal efficiency of VOCs. The reason is that excessive O₂ in the gas stream can consume reactive species generated in the discharge zone, which results in a reduction of the conversion efficiency. The formation of NO_x in the discharge zone may be responsible for a decrease of the reduction efficiency of VOCs since both the O₂ and N₂ will be consumed and by these means the active species formation such as O₃· and N₂ will be reduced.

3.4 Input power

In a DBD system input power has a significant impact on the conversion and energy efficiency of VOCs. A mass of high active species in the discharge zone is the main contributor to the decomposition of pollutants in a DBD system. These active species can be generated through a series of reactions and is directly proportional to the power supplied to the system. The effect of input power has been extensively studied (Chen et al., 2016; Karatum and Deshusses, 2016; Jiang et al., 2017) and it is obvious that increasing input power enhances the VOCs conversion efficiency. Dou et al. (2017) studied the toluene decomposition as affected by the input power, and found that toluene conversion increased from 35.4% to 78.8% when the input power increased from 26 to 65 W. Ye et al. (2008) studied the decomposition of benzene and found that its conversion efficiency increased from 5% to 40% when the input power was increased from 60 to 110 W.

The gas ionization and production of active species in NTP are well explained in the literature by Kim et al. (2017). The number of highly energized electrons and the internal electric field could be enhanced by high input power, which can lead to high removal efficiency. The high input power can also improve the pursuance for preliminary chemical reactions (Nguyen and Kim, 2015). It can be concluded that the input power has a positive correlation with the conversion efficiency of both DBD alone and DBD-catalysis system.

4 Decomposition of VOCs in plasma-catalysis systems

Plasma technology has a high conversion efficiency for most of the volatile compounds, however, for some compounds the decomposition is low combined with undesired byproduct formations (Fan et al., 2011). Despite mineralizing most of the contaminants in the air environment, the plasma process also produces oxidized pollutants such as ozone (O_3), CO, and NO_x . In addition, carbonaceous compounds are often generated during the carbonation of hydrocarbons deposit at the inner electrode of the reactor, which can affect the operation of the DBD reactor. These limitations during the operation of DBD reactors are the major hurdle for the implementation of DBD technology in the industrial sector. In recent years, many researchers are engaged with the optimization of DBD reactors in order to overcome the related problem e.g. by means of the addition of a catalyst or implementation of a ferroelectric bed reactor (Liang et al., 2010; Subrahmanyam et al., 2010; Guo et al., 2018a).

Therefore, to introduce catalysts in the NTP system not only can increase the efficiency of the decomposition of VOCs, it is also an efficient measure for controlling harmful or unwanted by-products. Plasma-catalysis combined system has been proposed and investigated by Subrahmanyam et al. (2006). The NTP-catalysis for the removal of VOCs is well described in the literature by Feng et al. (2018). Based on the placement of the catalyst, plasma catalysts can be divided into two classes, namely, in plasma catalyst (IPC) and post plasma catalyst (PPC). IPC is similar to the packed-bed reactor, in which the catalyst is packed or it is placed within the plasma discharge zone and thereby directly exposed to the plasma (Jia et al., 2013). Figure 2(a) shows the IPC in which the catalyst can be introduced in the plasma discharge zone, and the Fig. 2(b) shows the PPC where a catalyst is placed downstream of the plasma discharge.

4.1 Performance of in-plasma catalyst (IPC) on VOCs degradation

In the IPC configuration, the catalyst is directly in contact

with the discharge plasma zone. The increased performance of the IPC is quite complicated since both the plasma generation and the catalytic process take place at the same time (Whitehead, 2010). As shown in Table 3, investigations have been conducted on the conversion efficiency of VOCs using IPC; the selectivity of CO_2 (SCO_2) in the IPC system and the NTP alone system was overviewed.

Studies showed that the most common catalysts used for the abatement of VOCs are manganese, aluminum, titanium oxides combined with palladium, platinum, copper, barium, or silver (Chen et al., 2009a). Due to its low cost, large surface and high capacity for VOCs adsorption Zeolites are also commonly used for the NTP-catalysis system (Kim et al., 2015).

Xu et al. (2017b) investigated the decomposition of low concentrated toluene in air and found out that with the addition of a catalyst (i.e., cobalt incorporated in MCM-41), toluene conversion increased from 60% to >99% and CO_x selectivity increased from 25% to >80%. Dou et al. (2017) studied the toluene decomposition in a plasma-catalyst combination system (Cu-Ce-Zr supported ZSM-5/ TiO_2/Al_2O_3) and found that more oxygen vacancy, bigger pore size, and lattice oxygen in the CuCeZr/ TiO_2 system resulted in an increased conversion efficiency from 80% to 100% at an input power of 65 W. In addition higher energy efficiency, as well as higher selectivity of CO_2 were observed. Bahri et al. (2017) also compared the conversion efficiency of toluene in the DBD alone with a DBD reactor with an IPC, the latter had a twofold higher conversion efficiency of toluene than the DBD alone reactor. Recently, Wang et al. (2018) confirmed the high conversion efficiency of toluene using the DBD- TiO_2 -Ba TiO_3 catalysis system. Moreover, with the addition of a catalyst, the O_3 concentration as a by-product was also reduced from 12.3 ppb to 7.80 ppb. DBD-catalysis system has a significant influence on the electric field intensity and generation of the highly energized electrons, which can react with the O_3 molecule and decompose them in the discharge zone. Hu et al. (2016) found that the addition of the catalyst (MnO_2 - TiO_2 /zeolite) increased the conversion efficiency of benzene from 60% to 80% and the selectivity of CO_2 increased from 45% to 65%.

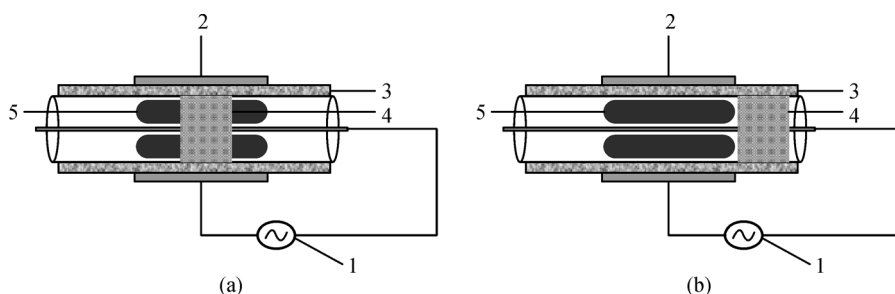


Fig. 2 Illustration of IPC (a) and PPC (b) configurations: 1. AC high voltage, 2. for the electrode, 3. dielectric barriers, 4. catalyst, and 5. discharge zone.

Table 3 An overview on comparative studies on VOCs decomposition in IPC

Type of VOC	Con (ppm)	Flow rate (L/min)	Type of catalyst	*CE (%) IPC	*CE (%) NTP alone	SCO ₂ (%) IPC	SCO ₂ (%) NTP alone	Energy density (J/L)	Refs.
Toluene	100	–	Co-MCM-41	95–100	65	80	30	292	Xu et al. (2017b)
Toluene	1	1	MIL-53 MIL-101 CPM-5	93.4 93.02 89.0	46.1	–	–	142.2	Bahri et al. (2017)
Toluene	1500	0.25	TiO ₂ -BaTiO ₃	95	70	50	35	–	Wang et al. (2018)
Benzene	60–202	4	Ag/TiO ₂ (PDC)	20–80	–	70	–	60–250	Kim et al. (2005)
Benzene (SSPBD)	400	0.5	MnO ₂ -TiO ₂ /zeolite TiO ₂ /zeolite MnO ₂ /zeolite Zeolite	83 81 75 60	60	65 62 60 46	45	10 W	Hu et al. (2016)
Benzene	400	0.5	Ag _{0.9} Ce _{0.1} /γ-Al ₂ O ₃ (PDC)	96.2	60	77.3	50	400	Jiang et al. (2016)
TCE	430	0.1	MnO _x /SMF	100	73	60	25	720	Magureanu et al. (2007)

Notes: *CE for conversion efficiency; “–” means no detection or mention.

Magureanu et al. (2007) analyzed a DBD catalytic system for the treatment of trichloroethylene (TCE) with a sintered metal fiber (SMF) interior electrode coated with transition metal oxides (MnO_x). Compared to reactors using SMF the TCE conversion and CO₂ selectivity were substantially improved from 73% to 100% and 25% to 60%, respectively.

Song et al. (2018) investigated the decomposition of chlorobenzene and found that the IPC system using CoMn/TiO₂ as a catalyst led to a higher conversion efficiency of 97% compared to plasma alone system (efficiency 81.4%); the CO₂ selectivity increased to 35%. Jiang et al. (2017) describes CeO₂/HZSM-5 as an efficient catalyst for the removal of chlorobenzene in a DBD system. Catalyst addition enhanced the mineralization of by-products. Qin et al. (2018) reported that IPC system using Mn-Ag/ZSM-5-BaTiO₃ as catalyst decreased the production of by-products such as O₃ and N₂O in the outlet of the reactor.

In conclusion, IPC shows distinctive features since it is effective on the surface and active sites of the catalyst and directly interact with the discharge plasma. Catalyst combined with a DBD as an IPC system can be various types such as γ-Al₂O₃, SMF, SBA-15, zeolite (such as MCM 41 and ZSM-5) and photocatalyst (TiO₂). Studies have proven their effectiveness in enhancing removal efficiency, energy efficiency, the selectivity of CO₂ and decrease in the by-products emission.

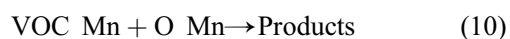
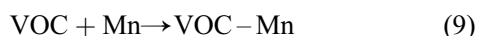
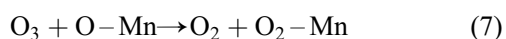
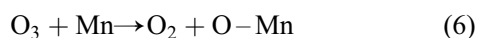
4.2 Performance of post-plasma catalyst (PPC) on VOCs degradation

In this type of NTP configuration, a catalyst can be placed downstream (after the discharge plasma zone) as shown in Fig 2(b). This type of configuration is also known as two-stage catalytic system or post plasma catalyst. Compared with the IPC configuration, PPC is characterized by a simple assembly and placement of the catalyst and is therefore more practical. Most of the active species are generated in the discharge zone where they last for a short interval of time and disappear before they reach the catalyst. However, long lifetime O₃ can be decomposed on the catalyst surface to oxygen molecules and radicals, which are more reactive than the O₃ for efficient decomposition of VOCs.

The PPC system is also verified by many studies to be effective in minimizing the toxic by-products generated in the plasma zone (Huang and Ye, 2009). The catalytic materials used in PPC system studies include Mn (Magureanu et al., 2005), Ti (Roland et al., 2002), Cu (Harling et al., 2009), Fe (Huang et al., 2008), Ni (Kim et al., 2006), Ag (Jarrige and Vervisch, 2009) and their combinations with Al₂O₃ and/or SiO₂. Among these, the placement of Mn-based catalysts in the downstream of NTP has been mostly studied. It showed a high conversion efficiency and CO₂ selectivity, as well as a reduction of ozone and other by-products (Delagrance et al., 2006; Lu

et al., 2006). Conducted studies focused more on the abatement of three VOCs, i.e., toluene, benzene and TCE (Table 4).

The reactivity for the decomposition of VOCs is higher for the atomic/molecular oxygen than for O₃ (Tang et al., 2013). Due to their capacity to oxidize the O₃ to reactive O₂ species Mn-based catalysts have been extensively used in the NTP system particularly in the PPC, they enhance the removal efficiency of VOCs (Zhu et al., 2016). The oxidation of O₃ in the presence of Mn-based catalyst can be described by the following reactions (Rezaei et al., 2013; Bo et al., 2018):



O₃ can be dissociated to molecular or atomic O₂ by three factors: 1) generated heat in discharge zone; 2) chemical reactions and catalytic decomposition; 3) especially Mn surfaces have the capacity to adsorb O₃ and convert it to O₂ species, which can react more efficiently with the VOCs adsorbed on catalyst surface than the O₃ (Bo et al., 2018).

Chang et al. (2018) studied the abatement of toluene in the PPC mode using MnO₂-Co₃O₄ as a catalyst. It showed that due to the addition of the catalyst, the conversion efficiency of toluene increased from 60% to >90% at SIE

of 455.59 J/L. Moreover, the selectivity of CO₂ also improved by 20% while the O₃ concentration was reduced from 1125 to 425 ppm at SIE of 311 J/L. Tang et al. (2013) studied the reduction of toluene using in the PPC mode as a catalyst a mixture of Ag and Mn. They reported that active species are playing a significant role in toluene decomposition in the discharge zone and also on the surface of catalyst through conversion of O₃ to O species.

Li et al. (2014) conducted a study using in the PPC system various phases of MnO₂ (α, β and γ) supported by Al₂O₃ pellets as catalyst for degradation of benzene. They reported compared to β-MnO₂ and γ-MnO₂ a higher catalytic activity of α-MnO₂ compared to β-MnO₂ and γ-MnO₂. When using α-MnO₂ as a catalyst the high conversion of benzene, the selectivity of CO₂ and the ozone suppression is achieved due to the existence of more OH· groups than other phases.

Vandenbroucke et al. (2016) characterized the performance of a PPC system for TCE decomposition. The addition of Pd/LaMnO₃ as a PPC catalyst had improved in the PPC system the conversion efficiency of TCE from 82% in a plasma alone reactor to 89% with 75% selectivity of CO_x and no ozone formation in the outlet. Sultana et al. (2018) recently confirmed the high conversion efficiency of TCE using PPC (Fe-K-OMS-2) mode and they reported an increase in conversion efficiency from 38% to 99% and CO₂ selectivity from 15% to 63%. Dinh et al. (2016) have also conducted a study on the MnO as a catalyst in DC corona and PPC system for decomposition of TCE. They confirmed that the TCE conversion could be improved from 80% to 100% and CO₂ selectivity 5% to 30% using the Mn-based catalyst in the downstream of the NTP reactor.

Table 4 An overview of decomposition of VOCs using PPC system

Type of VOC	Con (ppm)	Flow rate (L/min)	Type of catalyst	*CE (%) PPC	*CE (%) NTP	SCO ₂ (%) PPC	SCO ₂ (%) NTP	O ₃ (ppm) PPC	O ₃ (ppm) NTP	Energy density (J/L)	Refs.
Toluene	54	1.5	Mn1Co1	>90	>60	40	20	308	1125	455.59	Chang et al. (2018)
Toluene	108	6	Ag-Ce-O	75	59.8	50	–	280	640	60.5	Tang et al. (2013)
			Ag-Co-O	99				40			
			Ag-Mn-O	100				0			
Benzene	100	0.4	γ-Al ₂ O ₃	50	< 50	35	< 35	250	>250	13	Li et al. (2014)
			β-MnO ₂ /γ-Al ₂ O ₃	60		45		175			
			γ-MnO ₂ /γ-Al ₂ O ₃	70		48		125			
			α-MnO ₂ /γ-Al ₂ O ₃	80		55		< 125			
TCE	300–350	0.5	Fe-K-OMS-2	99	38	63	15	–	150	120	Sultana et al. (2018)
TEC	370	0.4	(K-OMS-2; α-MnO ₂)	>99	>80	< 30	< 5	–	–	240	Dinh et al. (2016)
TCE	500–560	2	Pd/LaMnO ₃ (100°C)	89.1	82.3	10	6	0	177	460	Vandenbroucke et al. (2016)

Notes: *CE means conversion efficiency; “–” means no detection or mention.

In general, Mn-based catalysts have better performance in the PPC system for the decomposition of VOCs (toluene, benzene and TCE) as well as suppressing O₃ concentration. Moreover, an appropriate addition of metal oxide with the zeolite can improve mineralization of reactant and reduce the production of by-products. Considering the variation in VOC composition and concentrations in the feed gas from various sources, NTP needs to operate together with catalysts either in IPC or PPC form; this is especially valid for the off-gas pollutants from industry and environmental facilities.

5 Major inhibitors of the non-thermal plasma process in DBD reactor

During the NTP process in a DBD reactor, some factors have an adverse effect on VOCs abatement, which is referred to inhibitors resulting from the reaction in the plasma. Inhibitors for both DBD and DBD-catalysis systems are discussed as followings.

5.1 Effect of chlorine

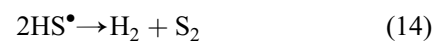
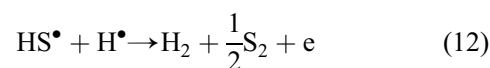
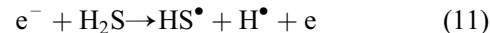
Chlorinated compounds have an extensive industrial application (Abdullah et al., 2006). However, they are considered as toxic pollutants due to their adverse effects on human health as well as on the environment. Studies on the interaction of the chlorine with the catalysts show that it could deactivate catalysts by depositing solid residues of chlorine on the active surfaces of the catalyst (Abbas et al., 2011; Aranzabal et al., 2012; Dai et al., 2008; Thevenet et al., 2014; Kamal et al., 2016). During the decomposition of chlorinated VOCs in the NTP system, in an oxygen-rich environment such as air, a significant number of various chlorinated by-products can be produced, which could be more hazardous than the parent compound (Indarto et al., 2008). The removal efficiency of chlorinated VOCs, degradation pathways and by-products generation is well explained by Vandenbroucke et al. (2011).

Shahna et al. (2017) have studied the removal of chlorobenzene with the DBD and DBD-catalysis system and found the latter had higher conversion efficiency. In the DBD alone system, the phosgene is one of the significant by-products, which can be generated by the reaction between chlorinated VOCs and active oxygen radical. Phosgene is very toxic and emitting it from the outlet is not acceptable. However, with the integration of photocatalysis in a DBD system, O₃ can be produced which has an additional oxidation effect on the conversion of phosgene to a harmless product like CO₂ and Cl₂. Presence of reactive radical species such as O·, H·, Cl· may act as auto-catalyzing agents for further decomposition reactions when this species collides with chlorinated molecules (Iijima et al., 2011). However, little attention has been paid to the effect of chlorine in the DBD plasma

reactor if the NTP-catalysis system is used; it could be shown that chlorine is also harmful for the catalyst (Abedi et al., 2015). Therefore, further investigation is needed for a better way of the abatement of chlorinated compounds in NTP systems.

5.2 Effect of sulfur

Sulfur compounds are notoriously responsible for their odour nuisance because most of them have very low odour threshold values (Chen et al., 2009b). NTP showed great potential in degrading sulfuric compounds (Sobacchi et al., 2003; Linga Reddy et al., 2012a; Zhu et al., 2018). However, there are problems, which need to be addressed before upgrading the technology for real application. While treating hydrogen sulfide (H₂S) (Dors et al., 2014) observed depositions of sulfur on the wall of the reactor, which consequently acts as a resistive layer and leads to reducing the discharge current of DBD; this could have an adverse impact on the conversion efficiency. Yan et al. (2013) found solid residues, consisting of CO₃²⁻, SO₄²⁻ and polymeric sulfur on the inner wall and electrodes of the DBD reactor. Linga Reddy et al. (2012b) proposed the possible dissociation of H₂S and formation of H₂ and S in the NTP system with Ar as a carrier gas by following reactions.



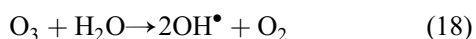
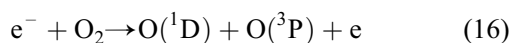
They conducted a study on the abatement of 5 vol% hydrogen sulfide diluted in Ar in a DBD reactor at the present of 500 mg of 3 wt%, 5 wt%, and 7 wt% of MnO_x/Al₂O₃. The researcher found that the presence of 5 wt% MnO_x/Al₂O₃ resulted in a better conversion efficiency (52%) than in the case of 3 wt% (45%) and 7 wt% (44%) (input power 1 W). Due to deposition of sulfur the 3 wt% catalyst was deactivated in a short time whereas the 7 wt% catalyst contained more oxides, which lead to a change in the discharge behavior. The deposited sulfur could be removed when the reactor was heated to 430 K.

5.3 Effect of relative humidity (RH)

Relative humidity is one of the influencing factors for VOCs abatement using NTP system. The problem is of great interest in DBD research and application since the water vapors always exist in the atmosphere at fluctuating concentrations (Vandenbroucke et al., 2011).

The water molecule has electronegative characteristic

and it can be converted into $\text{H}\cdot$ and $\text{OH}\cdot$ radicals in a DBD environment (Kim et al., 2003). $\text{OH}\cdot$, a short-life reactive species in the DBD reactor is considered as one of the most potential reactants for the decomposition of gaseous pollutants. The generation of $\text{OH}\cdot$ and $\text{H}\cdot$ radicals from water is described by the following reactions (Abdelaziz et al., 2018).



Lee and Chang (2003) conducted a study on the abatement of p-Xylene and found that the conversion efficiency of p-xylene increased as the humidity increased from 0.8% to 2.3%. The complete removal efficiency of p-xylene was achieved at 18 kV with 2.3% relative humidity. Improvement in conversion efficiency with the increased humidity level was also reported by others (Mfopara et al., 2009; Ni et al., 2011).

Chen et al. (2009b) investigated the decomposition of dimethyl sulfide (DMS) with varied relative humidities from 0% to 1.0% and found the highest conversion efficiency of 97% at 0.3% RH. The conversion efficiency decreased to 87% when the humidity increased further to 1.0%. Chen et al. (2016) reported that under a dry condition the conversion efficiency of toluene amounted to 27% at a voltage of 30 kV. When the RH was increased to 80%, the conversion efficiency of toluene decreased to 20%. These results show that the high humidity had an inhibitive effect on the decomposition of toluene. The trend of the decrease in conversion efficiency with increased humidity was also reported by others (Huang et al., 2008; Thevenet et al., 2008).

Nevertheless, a case of the neutral effect of the humidity on the conversion efficiency was documented by Futamura and Sugasawa (2008). They found that while increasing the humidity level from 0% to 2%, no substantial effect on toluene conversion was observed.

The humidity in the gas stream affects the conversion efficiency of VOCs in a DBD system by influencing the properties of the discharge characteristic and chemistry of the plasma. From the point of literature review, there should be optimum humidity ranges in the DBD system for the abatement of VOCs under different conditions.

5.4 Deposition problems

In the DBD system, some defective products may form during complex chemical reactions which can be either in the form of gaseous emissions or solid deposition. The

solid by-products are accumulated on the inner wall of the dielectric barrier as well as on the surface of the inner electrode. These deposits may interfere with the overall performance of the DBD reactor. Following literature shows the deposited residue in DBD reactors, which can be characterized as aromatic polymer, carbonaceous, oxygenated, and/or nitrogen-containing compounds.

During the treatment of mixtures of VOCs (toluene and ethylbenzene) in a DBD reactor, some dark brown solid deposits (petroleum or tar like-odour) were detected (Karatum and Deshusses, 2016). Magureanu et al. (2005) investigated VOCs abatement in the DBD-catalyst system and observed carbonaceous deposits on the inner wall of the dielectric barrier and the surface of the inner electrode. These by-products were estimated through the weighing method; it showed that the concentration was less than (5%) of the total carbon balance. Guo et al. (2006) observed a yellow colored solid deposit product in the DBD, which were characterized as an aromatic polymer. Ye et al. (2008) reported that some brown colored residues with the composition of 2-nitro-phenol, hydroquinone, phenol, heptanoic acid, resorcinol, 4-nitrocatechol, 3-nitro-phenol and 4-phenoxy-phenol were deposited on the inner wall of a DBD. These deposits changed the dielectric constant of the quartz and thermal energy build up consequently; mechanical failure of the dielectric barrier can also occur. Solid residues are not only deposited on the wall and electrode of the DBD reactor but may also be deposited on the surface of catalysts, which is one the main reasons for catalytic deactivation in DBD-catalysis system (Chen et al., 2009a).

Several variables such as flow rate, temperature and input power play an important role on forming depositions. Most of the published studies focused only on the qualitative analysis of deposited residues. In-depth investigations of quantitative determination of by-products, as well as their effect on the efficiency of reactors, remains a big challenge in NTP study.

6 A novel configuration of DBD reactor and its perspectives – Double dielectric barrier discharge (DDBD) reactor

The review of DBD systems demonstrates that contaminants in the environment can not be completely mineralized; undesirable by-products including chloromethane, hydrochloric acid, and greenhouse gases may be emitted from industrial activities) (Ye et al., 2016). During the decomposition of gases, solid carbon by-products may be formed and may deposit in the reactor, which may result in a further reduction of the efficacy of DBD reactors. Moreover, the direct exposure of the inner electrode to plasma and pollutants leads to corrosion or erosion, which can affect the overall performance of the DBD reactor. To overcome these drawbacks, a double dielectric barrier

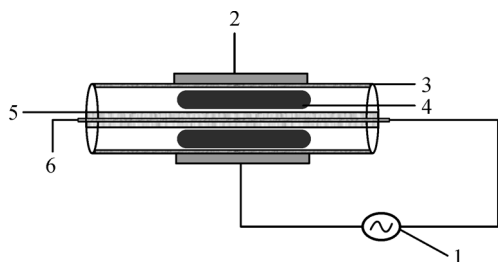


Fig. 3 Configuration of DDBD plasma reactor: 1- AC high voltage, 2- outer electrode, 3- outer dielectric barrier, 4- discharge zone, 5- inner dielectric barrier, 6- inner electrode.

discharge (DDBD) reactor has been recently used for the treatment of pollutants in air and water. In a DDBD reactor, two cylindrical tubes can be used as dielectrics; each one positioned in direct contact with the electrodes. Plasma can be generated through two dielectric barrier configurations. Figure 3 shows a DDBD reactor. The inner electrode can be placed inside the inner barrier tube and an outer electrode can be wrapped on the outer surface of the outer barrier tube. This configuration of the DDBD reactor can chemically isolate both the inner and outer electrodes from the plasma discharge zone. It is helpful for preventing arcing in the discharge zone (Florian et al., 2015), also ensures that all reactants pass through the discharge zone and are uniformly treated (Kundu et al., 2012).

Since the DDBD can actively protect both the inner and the outer electrodes from contaminants, reactive environment and by-products during the decomposing process, DDBD have higher discharge efficiency and longer lifetime of electrodes (Kundu et al., 2012).

Based on weak and robust plasma discharges respectively, Zhang et al. (2014a) developed a DDBD reactor with two discharge plasma zones including outer and inner discharge zones. They concluded that the energy utilization could be improved in the DDBD system where the low energy bond group of VOCs are first destroyed in the outer discharge, whereas the higher bond energy groups of VOCs such as aromatics are destroyed subsequently in inner discharge zone (Zhang et al., 2014a).

In the environmental pollution control field, increasing attention has been paid to DDBD for treatment of wastewater (Lu et al., 2012) and air pollutants, including exhaust gases of vehicles /NO_x (Pacheco et al., 2012), CH₄ from waste facilities (Mustafa et al., 2017a) and aromatics (Shi et al., 2016).

Zhang et al. (2014a) investigated the removal of styrene in polluted air using a DDBD reactor. They compared traditional single DBD with DDBD reactors using the same experimental conditions. They observed the production of large amounts of oil like by-products after 6 h of operation in case of DBD reactor; in addition the electrode was polluted by byproducts. In case of the utilization of DDBD reactor no such byproducts were produced. More-

over, the selectivity of CO and CO₂ was enhanced by 40% with an initial concentration of 463.50 ppm styrene at a voltage of 11 kV. The DDBD reactor concept is superior to DBD reactors not only due it has better treatment efficiency but also due to the minimization of the solid residue formation on the surface of the electrodes; this is caused through the presence of the double dielectric barrier. Recently, the authors of this paper investigated the removal of VOCs mixtures in a DDBD reactor with and without catalyst (Mustafa et al., 2018). Six different VOCs including tetrachloroethylene, toluene, trichloroethylene, benzene, ethyl acetate and carbon disulfide were mixed and investigated. The highest conversion efficiency (carbon disulfide, 83.30%, the rest of the compounds, 99.9%) was achieved at an input power of 53.7 W and a gas feeding rate of 1 L/min. In the DDBD system without addition of a catalyst, the solid residue can also form and deposit on the inner wall of the dielectric layer. However, it can be easily removed by washing or simple chemical treatment. It is reasonable to draw the conclusion that DDBD can be the future for up-grading use with all the merits it possesses.

7 Conclusions

VOCs released from the waste treatment facilities have become a significant issue and cause nuisance problems as well as health risk to humans. The VOCs abatement has been an area of interest for many researchers to meet the increasingly stringent regulations. Conventional methods to control the emission of VOCs include absorption (wet scrubbing), adsorption, incineration (either thermal or catalytic), masking, and biofiltration. However, the major disadvantages that restrict the utilization of these technologies including the requirement of post-treatment of the end products, catalytic deactivation, generation of toxic by-products and/or longer treatment time. NTP has drawn remarkable attention for the removal of VOCs from the gas stream due to its high removal efficiency and low energy consumption. Dielectric barrier discharge (DBD) is considered one of the most promising techniques of NTP due to its unique characteristics, such as bulk homogenized volume plasma with high reaction efficiency. This review addressed the key parameters of DBD including electrode configurations, dielectric barrier, discharge gap and discharge length and concluded that an appropriate configuration of the DBD reactor could be beneficial for improving the conversion and energy efficiency of the system.

Although significant amount of research has been carried out to establish decomposition processes of VOCs compounds, most of the research deals with single compounds only. Further investigations can be carried out to understand the chemical reactions and mechanism of the decomposition for multiple VOC mixtures in a DBD

system. Despite mineralization of most of the contaminants in DBD reactors, the process also produces oxidized pollutants such as O_3 , CO, NO_x , as well as solid deposits. Further research is needed to investigate the effect of by-products and eliminate deposits in a DBD system.

DBD-catalysis has been introduced and studied to overcome the issues associated with the DBD alone plasma reactor. The IPC system using catalysts such as γ - Al_2O_3 , SMF, SBA-15, zeolite (such as MCM 41 and ZSM-5) and photocatalyst (TiO_2) showed better conversion efficiency and selectivity of CO_2 . However, for the PPC system, Mn-based catalyst showed better performance in both conversion efficiency and selectivity due to its high adsorption capacity and reactivity with the O_3 molecules.

The review on the literature of DBD alone and DBD-catalysis demonstrate that several challenges need to be addressed in future research. The effect of inhibitors on the conversion efficiency has been discussed and shows that there are optimum humidity ranges in the DBD system for the abatement of the VOCs under different conditions. Catalytic poisoning can occur during the treatment of chlorinated and sulfuric VOCs. Deposition of solid carbonaceous residues can affect the overall performance of a DBD reactor. Further research of this subject could be beneficial for the large-scale application of DBD plasma.

Finally, a modified configuration of the DBD reactor, i.e. DDBD for the abatement of VOCs is discussed in details. The DDBD plasma reactor provides a better solution for the above-mentioned problems as the extra dielectric layers avoid the corrosion on electrodes. Integration of a catalyst in the DDBD configuration indicates promising results and may be a subject for future research and application of VOCs abatement.

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