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Plasma Based Methods, New Air Cleaner Technologies for VOCs Removal in Non-Industrial Buildings

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Abstract

Development of plasma based methods, and their remarkable capabilities for removal of volatile organic components (VOCs) from indoor air have motivated designers to employ these methods for indoor air treatment. This paper addresses the outcome of an extensive literature review on plasma based air cleaner technologies and their application for VOCs removal in non-industrial buildings. Furthermore, it reports the role of the catalyst on the performance of the systems as well as the reduction of greenhouse gas emission.

Keywords: plasma, air cleaner, VOCs, non-industrial building

Résumé

Le développement de méthodes utilisant un plasma, dont la capacité d'élimination des composés organiques volatils (COV) de l'air intérieur est remarquable, a conduit les concepteurs à les employer en traitement de l'air intérieur. Ce document présente le résultat d'une revue approfondie de la documentation sur les technologies de purification de l'air au plasma et leur application à l'élimination des COV dans les bâtiments non industriels. Par ailleurs, il fait état du rôle du catalyseur dans la performance des systèmes et dans la réduction des émissions de gaz à effet de serre.

Mots-clés : plasma, purificateur d'air, COV, bâtiment non industriel

1. Introduction

Modern building ventilation design must take into account the health and comfort of the occupants as well as energy consumption and the environment. The system needs to protect occupants against chemical contaminants from numerous internal sources, as well as bio-contaminants from the occupants themselves. To reduce exposure to internal contaminants outdoor air is brought in to dilute the contaminants and exhaust a portion to outdoors. However, the quantity of the outdoor air brought into the building can have a direct negative effect on the energy cost of ventilation. There is a cost to heat, cool, humidify or dehumidify the outdoor air depending on the location and season. The required energy for these processes contributes up to 35% of greenhouse gas emissions.

A number of factors must be taken into consideration in selecting the most effective air cleaning system. They include long-term performance, minimum energy consumption, and minimum amount of unwanted by-products formation. Furthermore, the capability of the system to work in indoor conditions (ambient temperature, relative humidity and variety of VOCs) is an important parameter in selecting that system [1].

VOCs are a group of chemical components that can be emitted from several indoor sources. The proven role of these components in several health problems, has motivates researchers to eliminate the amount of VOCs in indoor air by means of air cleaning systems.

Plasma based methods refer to a series of processes in which a high voltage discharge (HV) is used for the destruction of pollutants [2]. In fact, in these processes, oxidation reaction of gaseous pollutants takes place in the destruction of pollutants. Development of plasma methods in recent decades has improved the performance of this technology and made it more cost-effective and applicable for applying as an air treatment method in non-industrial buildings. The paper reports the outcomes of an extensive literature review of developing plasma techniques, from thermal plasma, to plasma catalyst, and the effect of the catalyst on VOCs decomposition and reduction of greenhouse gas emission.

2. Thermal Plasma

In thermal plasma, a high electrical voltage (1kW-50MW) is applied to generate a plasma flame in an active zone at atmospheric pressure [3]. This high voltage discharge causes an initial excitation in the working gas, followed by ionization and generating of large amounts of reactive species, including ionized and dissociated radicals (>99% ionization). In this state, the temperature of the reactive species in the field gas can be reached at 1000K while the peak of the temperature may reach up to 10,000 – 20,000 K ($T_{peak}=1-2$ eV) depending on the voltage, gas flow rate, and the source of plasma [4].

Thermal plasma is one of the suggested processes for the removal of noxious compounds from gaseous wastes, especially VOCs. Using high energy density and consequently high temperature, results in high fluxes of reactant species, swift reaction rate and shorter response time for the reactor to start up. As a result, a smaller reactor could be used in thermal plasma technology [5]. Obtaining the full fragmentation of molecules and high specific activation are other advantages of thermal plasma [6]. However, using electricity, as the most expensive source of energy, is a major consideration against applying this method [5]. On the other hand, in thermal plasma all particles including electrons, ions, atoms, and background gas molecules are at thermal equilibrium ($T_e \approx T_{ion} \approx T_{gas}$). Therefore, overheating of the reaction media is inevitable [3]. Furthermore, thermal plasma is more applicable for wastes containing high concentration of organic components. As a result, the application of thermal plasma for indoor air treatment and its integration in the buildings mechanical ventilation system is practically impossible.

3. Non-thermal Plasma (NTP)

NTP technique, works based on creating a quasi-neutral environment including, ions, radicals, electrons, neutrals and UV photons [7]. Because of the lower mass, electrons are accelerated selectively in the field. The temperature of these accelerated electrons can reach to about 10,000–250,000 K (1–25 eV) while the background gas remains at ambient temperature ($T_e \gg T_{ion} \sim T_{gas}$). Therefore, the ionization of the molecules is significantly lower than thermal plasma (around 1%). The collision of the electrons with background molecules including oxygen, nitrogen and water vapour, results in producing excited bulk gas molecules (N_2^* and

O_2^* , etc.). These excited molecules emit photon or heat to lose their energy. This energy forms reactive ions and radicals (for instance OH^* and O^*) which are unstable and contribute to the next oxidation reactions [8].

NTP systems commonly work in low pressure to prevent the system to reach arcing. Working in atmospheric pressure causes instability in the system due to increase in the number of collisions and consequently heat generation, which leads the system to spark and arc. Nevertheless, preventing the reaction media temperature from increasing is a very significant advantage of this method. This advantage has motivated scientists to develop this field for indoor air treatment applications including removal of VOCs as well as acid gases (NO_x , SO_x , etc.), particulate matters, and bacteria.

3.1 NTP Reactors

Generally, classification of NTP reactors is based on the type of the employed discharge mode in them [8], [9]. Figure 1 illustrates the configuration of the most commonly used NTP reactor configurations.

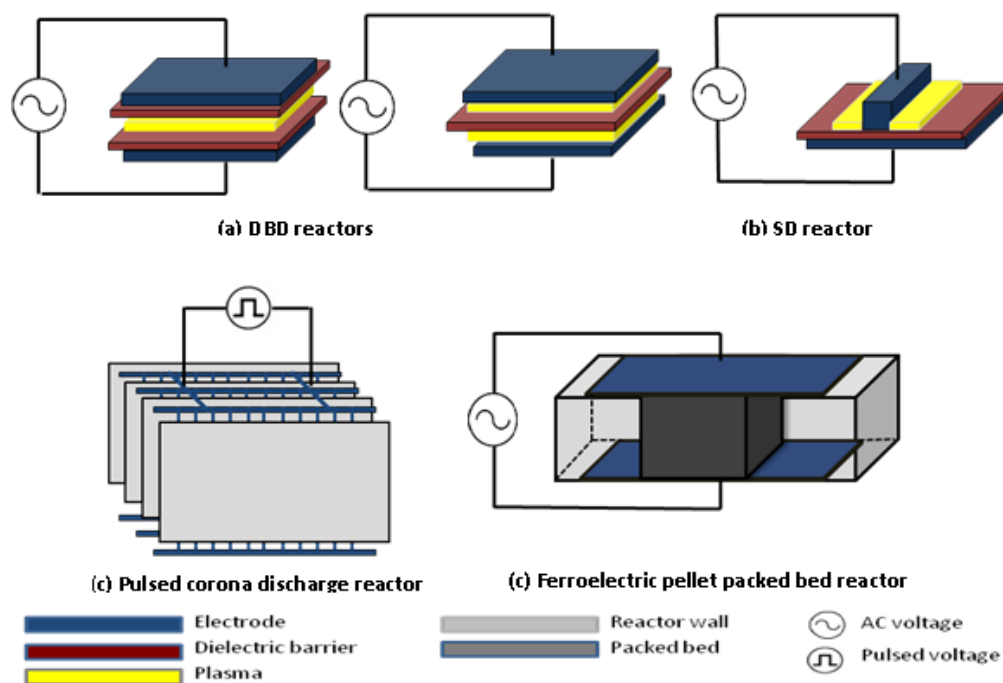


Figure 1. Configuration of some common NTP reactors (a) Dielectric barrier discharge (DBD), (b) Surface discharge, (c) Pulsed corona discharge, (d) Ferroelectric pellet packed bed

Dielectric barrier discharge (DBD): DBD or silent discharge is one of the most common types of NTP reactors [10]. This reactor consists of at least one dielectric, such as glass, quartz, ceramic, alumina, teflon or mica between electrodes which plays a key role in NTP stabilizing [11]. Easy application and non-complicated reactor scale up, lower power consumption and higher energy efficiency are the most important advantages of this discharge mode compared to the other modes [12].

Surface discharge (SD): SD has a similar configuration to DBD. In this type of reactor, a dielectric barrier discharge is used to generate plasma. Ceramic-based alumina can be used in this reactor. This reactor can be used for decomposition of VOCs; though, ozone accumulation without the possibility of its destruction restricts the applicability of this method for indoor air treatment [9].

Pulsed corona discharge: In this reactor applying voltage pulses between electrodes produces plasma. The spark formation in this way can be prevented which means a higher efficiency and longer lifetime of the reactor [8]. Because of reducing ozone formation, pulse corona discharge can be applied for indoor air treatment[13].

Ferroelectric (dielectric) pellet packed-bed reactor: In this type of reactor, ferroelectric pellets (BaTiO_3 , NaNNO_2 , MgTiO_4 , CaTiO_3 , SrTiO_3 , PbTiO_3 and $\text{PbZrO}_3\text{-PbTiO}_3$) are used as packed-bed [14]. The efficiency of a dielectric to store more charge depends on the dielectric of used materials. The presence of packed bed makes a uniform distribution of gas and electrical discharge. However, pressure drop increase is a side effect that should be considered [8].

3.2 Limitations in NTP Technology

NTP shows capability of VOCs abatement in a wide range of concentrations (1-10,000 ppm) in ambient temperature. The lower requirement of energy compared to thermal plasma and high efficiency in removing particulate matters [15] are the advantages of this method. However, despite the advantages of NTP, this method has poor energy efficiency especially for low concentrations of VOCs. Relative humidity has a strong impact on the system performance [16]. Besides, incomplete oxidation of target pollutants results in formation of several by-products including CO , NO_x , O_3 and organic by-products (i.e. formaldehyde) that may be more hazardous compared to the target pollutants [17]. Consequently, these have made this method impractical for indoor air treatment applications.

4. Plasma Catalyst

To overcome the deficiencies of NTP, combination of this method and catalyst, which is named plasma catalyst has been proposed. In plasma catalyst, the presence of the catalyst and the synergic effect of plasma and catalyst enhances the removal efficiency and total oxidation of the components. Catalyst and plasma can be combined in two different ways in a plasma catalyst reactor:

In plasma catalyst (IPC): In this single-stage method, catalyst is introduced to the NTP in the discharge zone of the reactor (fig 2.a). The catalyst can be used either as a packed bed or honey comb monolith form or be coated on the electrode or reactor wall [19]. In industrial applications, due to the pollutant high concentration, high level of energy is needed for destruction of pollutants. Increasing the SIE amount causes an increase in the rate of NO_x and ozone concentration. The ozone coverage of the catalyst surface diminishes the catalyst activation and leads to the formation of CO due to non-complete oxidation of pollutants. In this case, IPC reactors are more efficient as they need lower SIE and have higher energy performance compared to PPC reactors [20].

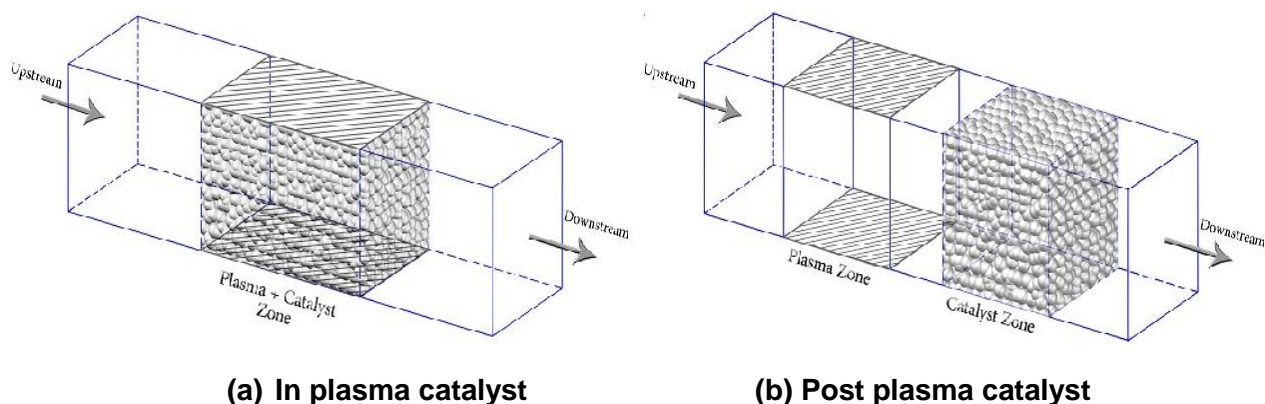


Fig.2 Schematic diagram of plasma reactors: (a) in plasma catalyst (IPC) reactor, and (b) post plasma catalyst

Post plasma catalyst (PPC): In a two-stage plasma catalyst method, the catalyst can be placed either upstream or downstream of the discharge zone. However, the installation of the catalyst downstream of the plasma (PPC) is more efficient (Fig. 2.b) since the generated reactive species in the plasma zone participate in oxidation reactions in the catalyst zone and complete the mineralization reactions. The result is an increase in CO₂ selectivity rather than CO formation as well as abatement of ozone concentration in the effluent gas [21]. In indoor air applications, because of the low concentration of the VOCs there is less demand to SIE. Moreover, based on the nature of the pollutants, generation of NO_x is negligible, and CO and ozone formation is more of a concern. Therefore, PPC is a more appropriate method for indoor air application [20].

4.1 Catalyst

Selecting an appropriate catalyst is an important challenge for enhancing the VOCs removal efficiency in plasma catalyst. The presence of the catalyst increases the residence time of reactants in the reactor. Consequently, the probability of surface reactions between the reactant molecules and reactive species is increased, leading to more selective reactions and higher efficiency of the reactor. The textural properties of the catalyst including specific surface area, pore volume, pore size and size distribution, as well as particle size and shape have significant role in oxidation of target components in a plasma catalyst process [22]. The efficiency of the catalytic plasma process is strongly related to the adsorption capacity of the catalyst. To have a high efficiency of VOC removal from air, VOC component should form a strong bond with the surface of the catalyst as adsorbent. This effect is more significant in low levels of VOCs concentration [23]. Therefore, the higher the storage capacity of the catalyst, the more efficient is the catalyst. Using an appropriate dual functional adsorbent/catalyst with high capacity of adsorbing the pollutants increases the concentration and retention time of components on the catalyst surface as well as the probability of collision between the component and reactive species [20]. The results of VOCs removal and CO₂ selectivity as well as the energy efficiency of plasma catalyst reactors are represented in Table 1. In this table, specific input energy (SIE, J L⁻¹) and energy efficiency (EE, g kWh⁻¹) are calculated as:

$$SIE = \frac{60U I}{Q} \quad (1)$$

$$EE_i = \frac{3.6 C_{in} \cdot \eta \cdot M}{24.4 SIE} \quad (2)$$

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where, U is the applied voltage (kV); I is the discharge current (mA); Q is the reaction gas flow rate (l/min); EE_i is the energy efficiency for conversion of component (gkWh/h); C_{in} is the inlet concentration of component (ppm); η is the conversion of component; M indicates the molar mass (g/mol); 24.4 shows the molar volume of gas (L/mol) under the defined condition and finally 60 and 3.6 are the units conversion.

Results show significant increase in VOCs removal efficiency and CO_2 selectivity as well as decrease in energy efficiency in plasma catalyst reactors compared to NTP.

Table 1. VOCs removal, CO_2 selectivity and the energy efficiency of plasma catalyst reactors

Ref	Reactor Type	Catalyst	SSA (m ² /g)	Target pollutant	Carrier Gas	Conc. (ppm)	Flow Rate (L/min)	Removal efficiency (%)	CO ₂ yeild (%)	SIE (J/L)	EE _i (g/kWh)
[24]	DBD/ PPC	MnO ₂ -Fe ₂ O ₃	219	Toluene	20%O ₂ + 80%N ₂	240	0.315	76	23.5	172	15.7 ^c
		MnO ₂ /□- Al ₂ O ₃	169					88	18		18.2 ^c
		MnO/AC	1024					99.7	30.2		20.6 ^c
[25]	SD /PPC	NaY HY	750 520	Toluene	80% N ₂ + 20% O ₂ + 0.5% H ₂ O	200	0.5	78 87	60 38	5W 600 ^b	3.85 ^c 4.3 ^c
[26]	DBD/IPC	γ -Al ₂ O ₃ α- Al ₂ O ₃	133 0.26	Carbowax	Air	200	0.1	77 100	96 61	4W, 2400 ^b	0.64 ^c 0.83 ^c
	DBD/PPC	γ - Al ₂ O ₃ α- Al ₂ O ₃ + γ- Al ₂ O ₃		Carbowax	Air	200	0.1	69 45	98 92	4W 2400 ^b	0.57 ^c 0.37 ^c
[27]	DBD/IPC	Pt/ Al ₂ O ₃	---	2-heptanone	Dry air	180	0.42	98	64	200	16.2 ^c
[28]	AC DBD/	3 wt% MnO _x /SMF	---	Isopropanol	Air	100	0.5	100	100	200	4.8 ^c
[29]	DBD/IPC	Co ₃ O ₄ / Al ₂ O ₃ /nickel	---	Toluene	N ₂ + 5%O ₂	50	0.1	96	75	500	1.42 ^c
[30]	DC corona /PPC	MnO _x / Al ₂ O ₃	---	Formaldehyde	Air+ 30%RH	2.2	6	87	---	20	0.46 ^c
[31]	DC tooth wheel cylinder / PPC	MnO _x / Al ₂ O ₃	200	Benzene toluene p-xylene	Air +25% RH	1.5 1.4 1.2	6	94	100	10	1.8 ^c
								97			2.0 ^c
								95			1.95 ^c
[32]	DC Corona/P PC	CuOMnO ₂ /TiO ₂	50	Toluene	Dry air	0.5	10.8	78	---	2.5	2.3 ^c
	DC Corona/IP C	TiO ₂	49					82 ± 2			
[33]	DC tooth wheel cylinder /PPC	MnO _x / Al ₂ O ₃ 5 wt.% Mn	---	Benzen Toluene p-Xylene	Air+ 30%RH	0.47 0.81 0.73	6	100 100 95	~35 ^a	10	0.6 ^a 1.2 ^a 1.2 ^a
[16]	DC corona / PPC	Pd/ Al ₂ O ₃	230-280	Toluene	Air	0.5	10 L	94	---	10	0.7 ^c

SSA: specific surface area

^a Approximate amounts extracted from graphs

^b Calculated by equation (1) data extracted from reference.

^c Calculated by equation (2) data extracted from reference.

SMF: sintered metal fiber

5. Cycled Storage–Discharge (CSD) Plasma Catalytic Process

Energy consumption is an important concern in employing plasma-based methods for indoor air treatment applications. Although, catalyst implementation in plasma reactors has made this method more cost-effective, optimization of these systems to decrease the expenses is still a goal in developing plasma processes. One promised technique to decrease the energy consumption is using CSD plasma catalytic reactors. These reactors work based on a cycle in which in the first step (storage stage) the target component is adsorbed on the catalyst while plasma is off. In the second step (discharge stage), plasma is applied in the reactor to oxidize the stored components [23]. Figure 3 shows a schematic diagram of this method. In this figure,

Q_1 and Q_2 are the flow rate of the polluted air in the storage state and discharge stage, respectively; t_1 is the storage period, and t_2 is the discharge period of the CSD process. Energy cost (kWh/m³) in a CSD plasma catalyst reactor is calculated by:

$$E_c^{CSD} = \frac{P_{discharge} \times t_2}{Q_1 \times t_1} \quad (3)$$

In this equation, Q_1 is the flow rate in the storage step (m³/h) and t_1 and t_2 are the storage and discharge period (h), respectively.

Employing this method for indoor air quality application, in which the concentration of pollutants is very low, can optimize the energy consumption of the plasma systems. In this situation large surface area and storage capacity are two important factors for the employed catalyst to increase the storage time (t_1) and consequently the energy cost. Furthermore, in high levels of relative humidity, the existing water molecules compete with the target components for adsorbing on the surface of the catalyst and limit the access of these molecules to the surface of the catalyst. The result is VOCs removal efficiency diminishing. Thereby, an appropriate catalyst should provide an excellent adsorbent characteristics as well as hydrophobic properties. Table 2 represents an overview of the studies on the CSD plasma process.

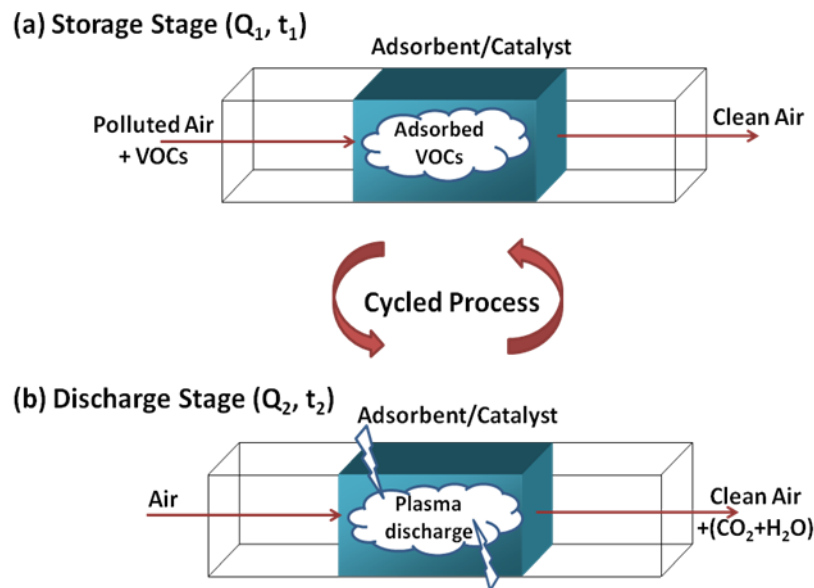


Fig. 3 Schematic diagram for a CSD plasma catalytic reactor

CSD plasma process is an affordable method for VOCs removal for IAQ improvement. However, selecting an appropriate catalyst with high oxidation capacity in the presence of plasma and a high capacity of VOCs storage is the most important challenge in developing the catalyst.

7. Conclusion

The importance of improving IAQ in sustainable buildings reveals the need of a comprehensive study for selecting the best method to reach this aim. In this study, the development of an appropriate approach for VOC treatment using plasma technique and the feasibility of their implementation in indoor environment was reviewed. The disadvantage of the temperature

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rising in the field gas and non-applicability of work in low concentration levels make thermal plasma method inappropriate approach for indoor air quality application. NTP overcomes these limitations and makes plasma technology an applicable method to work in low concentration and ambient temperature. However, poor energy efficiency and formation of greenhouse gases including O₃, CO, NO_x and other hazardous organic by-products are disadvantages of this method that make it improper for indoor air applications.

Table 2 Overview of studies on the CSD plasma process

Ref	plasma Type	Catalyst	SSA (m ² /g)	Target pollutant	Carrier Gas	Conc (ppm)	Flow Rate (ml/min)	t(min)	Removal efficiency (%)	CO ₂ yield (%)	P _{dis} (W) SIE(J/L)	Ec (kWh/m ³)
[34]	DBD	Ag/HZSM-5	334	Benzene	Air+ 50%RH	4.7	Q1=600 Q2=60	t ₁ = 840 t ₂ = 24	100	99.8	4.7W	3.7e ⁻³
[23]	DBD	Ag-Cu/HZSM-5		Formaldehyde	Air+ 50%RH	5.3	Q1=300 Q2=60	t ₁ = 780 t ₂ = 14	100	> 99 ^a	2.3W	10 ⁻⁵ – 10 ⁻⁴
[35]	DBD	1%Ag/TiO ₂	≤68	Benzene	Air (50%PP)	200	Q=4000-5000	---	100 ^a	---	169J/L	---
		4%Ag/TiO ₂	≤68						75 ^a	80 ^a	136 J/L	
		0.5%Ag/γ-Al ₂ O ₃	≤210						90 ^a	75 ^a	160 J/L	
		H-Y	520						100	>70 ^a	160 J/L	
		2%Ag/H-Y	520						100	65 ^a	140 J/L	
[36]	DBD	0.8 wt%Ag/HZSM-5	334	Benzene	Air+ 50% RH	4.7	Q1=600 Q2=60	t ₁ = 60 t ₂ = 9	100	100	4.7 W	---
		2 wt%Ag/HZSM-5	329						92 ^a	100		
		4.2wt%Ag/HZSM-5	306						63	100		
		HZSM-5	328						89 ^a	100		
		2 wt%Ce/HZSM-5	---						100	91 ^a		
		2 wt%Cu/HZSM-5	---						97 ^a	93 ^a		
		2 wt%Ni/HZSM-5	---						75 ^a	94 ^a		
									t ₁ = 60 t ₂ = 15			

^a Approximate amounts extracted from graphs
PP: O₂ partial pressure

Plasma catalyst, on the other hand, is capable of increasing the VOCs removal efficiency with the lower amounts of hazardous by-product formation. Energy consuming is improved in this method; even though, more considerations should be given for optimization of the energy consumption in plasma catalyst methods.

Finally, employing a CSD plasma catalytic reactor is an effective method for completely oxidizing of VOCs as well as optimizing the energy consumption in non-industrial buildings. Significant decrease in the formation of hazardous greenhouse gases, especially CO and O₃, and organic by-products, is the other advantage of this method that has made it a good candidate for non-industrial buildings application. Employing an appropriate catalyst still is a challenge to achieve the best performance of this method.

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EIC Climate Change Technology Conference 2013

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