Scale-up analysis and development of gliding arc discharge facility for volatile organic compounds decomposition

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Abstract

The influences of inlet gas flow rate and reactor configuration on volatile organic compounds (VOCs) decomposition efficiency in a traditional gliding arc (T-GA) facility are studied based on laboratory experiments and numerical simulation. The ratio of the nozzle diameter and the shortest distance of two electrodes should be maintained in a suitable value range to guarantee the decomposition efficiency, which indicates simply enlarging the nozzle inner diameter is not a proper way to raise the T-GA treatment capacity in a fixed supply voltage condition. A developed gliding arc gas discharge (D-GA) reactor based on a modified gas feed system is proposed: small flow rate air goes through the bottom nozzle to process the GA evolution, and high flow rate contaminated gas goes frontal through the plasma region and decomposes. The performances of D-GA reactor in the decomposition of VOCs with relative high gas flow rate, including decomposition efficiency and specific energy consumption, are better than T-GA reactor. D-GA reactor also avoids the drawbacks of the increases of power system amount and electric power consumption in multi-electrode GA system.
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1. Introduction

Gliding arc gas discharge (GA) is an auto-oscillating phenomenon developing between at least two electrodes that are immersed in a laminar or turbulent gas flow. GA phenomenon starts at the shortest gap between the electrodes by an initial breakdown of the gas to be processed (see Fig. 1(a)). Then during few milliseconds the arc is pushed by the flow along the electrodes (see Fig. 1(b)) until the rupture of the ionized column (see Fig. 1(c)). This event is followed by a new breakdown at the electrode throat and the cycle repeats as long as the voltage delivered by the transformer is high enough.

The plasma region in GA is significantly in non-equilibrium state [1–4]: most of the energy consumed is used to produce relatively high energetic electrons as well as excited ions, atoms and other radicals which can stimulate the decomposition reactions directly and promote selective chemical transitions, rather than heat the bulk gas. The most attractive advantage of GA plasma is that, it successfully provides typical non-equilibrium plasmas at elevated power levels, with simultaneously keeping a high level of non-equilibrium, high electron temperature and high electron density [1,5]. This distinguish feature satisfies the requirement of both high productivity and high selectivity for plasma assisted chemical processes [1,2]. A flat two-dimensional GA facility [6] was firstly proposed as an innovative solution to the emission control and treatment of gaseous pollutant by Czernichowski in 1988. Nowadays, the GA assisted VOCs decomposition processes were widely investigated by numerous experiments [7–13] and showed promising potential.

However, industrial plasma assisted technologies for VOCs emission control require the plasma to be produced directly in the big volume of polluted gases at conditions similar to whose the gases are emitted to the atmosphere during the industrial processes [14]. Hence, one very important GA technique development issue is how to scale-up the GA facility with simultaneously satisfying the high treatment capacity and high decomposition efficiency at low energy consumption level.

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Nomenclature

D	smallest distance between electrodes (mm)
DTGs	deuterated triglycine sulfate
E	specific energy consumption (Wh/S m^3)
f	supply frequency (Hz)
F	total flow rate through plasma region (S m^3/h); $F = F_A + F_B + F_C$
F_A	net flow rate through Face A (S m^3/h)
F_B	net flow rate through Face B (S m^3/h)
F_C	net flow rate through Face C (S m^3/h)
H	vertical distance between electrode throat and nozzle (mm)
L_1	inductance in transformer primary circuit
L_2	inductance in transformer secondary circuit
MCT	mercury cadmium telluride
P	electrical power (kW)
Q	total gas flow rate (S m^3/h)
R_1	resistance in transformer primary circuit
R_2	resistance in transformer secondary circuit
RMS	root mean square
U	supply voltage (kV)
X	VOCs concentration after discharge (ppm)
X_0	VOCs initial concentration (ppm)

Greek symbols

η	VOCs decomposition efficiency (%)
φ	nozzle inner diameter (mm)

Some improvements based on traditional GA reactor (T-GA) were attempted in last decade: rotating electrical discharge GA system was realized by central rotating electrode [15] or using local external magnetic field conveniently oriented [16]; reverse vortex flow GA system [17] was proposed with pressurized gas entering the cylindrical reactor volume tangentially; multi-electrode was used and it was reported [14] that the more electrodes were put into the discharge chamber the bigger volume of polluted gas could be effectively treated. Above improvements mainly focused on the reactor structure and here we try to enlarge the treatment capacity by modifying the gas feed style.

In this paper, the scale-up analysis of T-GA was carried out based on experimental results and computational fluid dynamics calculation. A developed gliding arc gas discharge (D-GA) reactor with modified gas feed style was proposed. The comparative assessment, including decomposition efficiency, by-product and specific energy consumption, of above two style reactors was presented with tetrachloromethane (CCl$_4$), hexane (n-C$_6$H$_{14}$) and toluene (C$_7$H$_8$) were chosen as the target VOCs. In addition, the energy consumption of the D-GA system was also compared with that of the six-electrode GA reactor.

2. Materials and methods

2.1. Reactors

The basic structure and configuration of T-GA and D-GA reactors used in this study are similar (as shown in Fig. 2): the reactor mainly consists of two 96 mm long, 26 mm wide and 4 mm thick knife-shaped electrodes fixed on a teflon bed plate, a gas nozzle and a ceramic seal. The vertical distance between electrode throat and nozzle is equal to 25 mm.

The gas feed style is the main difference between two reactors: in the T-GA reactor, all gas goes into the reactor through the bottom nozzle; in the D-GA reactor, air with relative low flow rate goes through the bottom nozzle to process the GA evolution, and contaminated gas frontal goes through the plasma region via a 15 mm in inner diameter inlet (as shown in Fig. 2).

Either GA reactor is supplied with a 50 Hz high voltage transformer (220 V/10 kV) with leakage fluxes respectively (as shown in Fig. 3). The effect of leakage fluxes determines a reactance that produces a constant RMS value of current in the secondary coil.

2.2. Procedures

Experiments were carried out by using a T-GA or D-GA reactor at atmospheric pressure. Contaminated gas treated in this study results from the mixing of a VOCs flow with a dry air flow, while tetrachloromethane (CCl$_4$), hexane (n-C$_6$H$_{14}$) and toluene (C$_7$H$_8$) was chosen as the target VOCs respectively. Total gas flow rate Q was varying in the range of 0.7–1.8 S m^3/h. The ini-
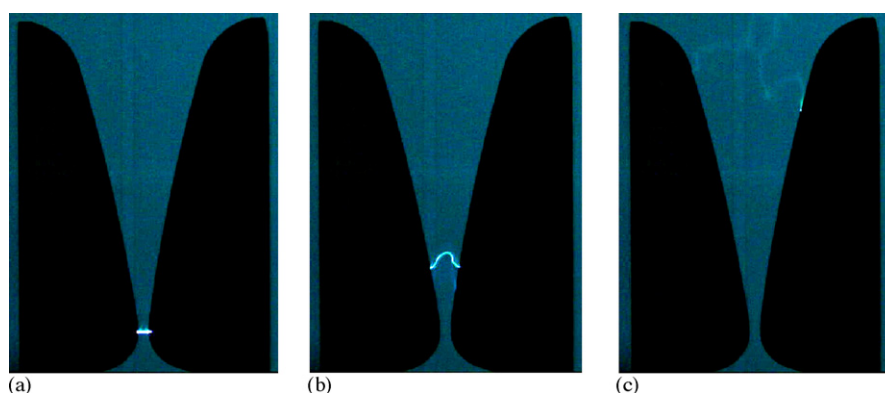

Fig. 1. Evolution of GA phenomenon (high speed camera photographs).
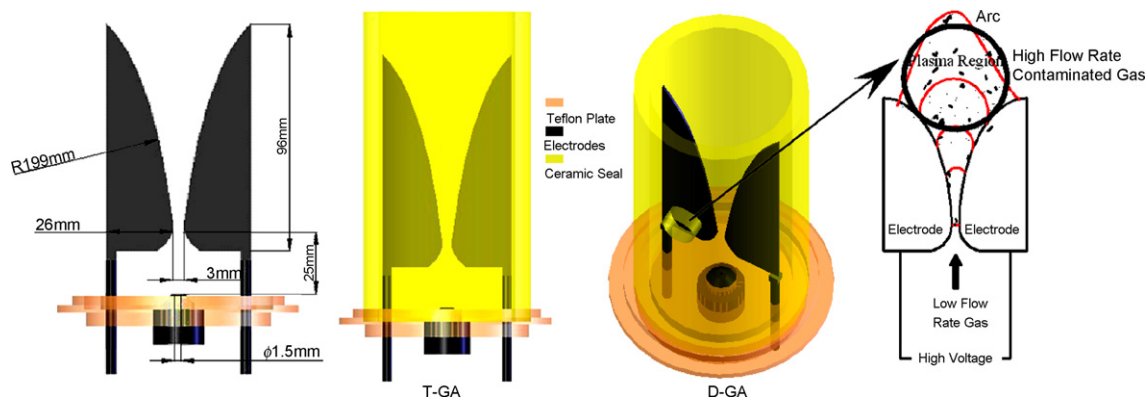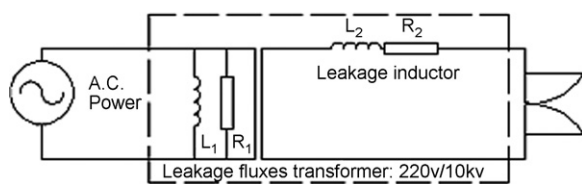
Fig. 2. Gliding arc discharge reactor.

Fig. 3. Gliding arc discharge electrical scheme.

tial concentration of VOCs was maintained at a fixed value by adjusting the mass flow controllers. Detailed experimental conditions are displayed in Table 1. The contaminated gas enters the GA reactor after passing through a drying column packed with a silica-gel desiccant to eliminate the influence from moisture.

2.3. Analytical methods

The concentrations of the target VOCs and degradation products in outlet were quantified by means of a NICOLET NEXUS 670 Fourier transform infrared (FTIR) spectroscopy equipped with a detector of deuterated triglycine sulfate (DTGs) KBr (for CCl_4, n-C_6H_{14} and decomposition by-products) or mercury cadmium telluride (MCT) (for C_7H_8 and decomposition by-products). The spectral resolution was $4\,cm^{-1}$ and every measurement was repeated four times. The temperature of sample cell was maintained at $140\,°C$. Under each set of con-

ditions, 20 min is allowed for stabilization before quantitative analysis.

The VOCs decomposition efficiency, η is defined by Eq. (1) below:

$$\eta = \frac{(X_0 - X)}{X_0} \times 100\% \tag{1}$$

where X_0 and X are its initial and final concentration respectively.

The electrical power P was measured by a DDS69# electronic single phase 2 wires Walt-hour meter installed in the electric circuit, and the pulse output constant was 1600 imp/kWh.

A Reynolds stress model (RSM), which is seen as more elaborate to GA reactor than the Spalart–Allmaras and K-ε ones [18], was selected to build an inner flow field mathematic model of the laboratory scale T-GA reactor. The 3D inner flow field was simulated with software FLUENT and the processing software GAMBIT.

3. Results and discussion

3.1. Analysis of the scale-up of T-GA facility

3.1.1. Influence of gas flow rate on VOCs decomposition efficiency

Experiments sets at different gas flow rates were studied at a T-GA reactor. The decomposition efficiencies of three target VOCs in dry air versus gas flow rate are shown in Fig. 4.

VOCs decomposition efficiency was sharply reduced with increasing the gas flow rate, irrespective of target VOCs chemical structure. This tendency is possibly due to the shortenings of both contact area and interaction time between VOCs and plasma region, which leads the reductions of energetic electrons impact dissociation and also the reactions between VOCs and reactive ions and radicals. Above limited studies indicates that some improvements to the T-GA reactor are necessary when it is applied to the decomposition of VOCs with relative high flow rate to guarantee the decomposition efficiency.

3.1.2. Influence of reactor geometry parameters on VOCs decomposition efficiency

The common sense suggests that enlarging the nozzle inner diameter is one of the most simple and direct channel to raise the
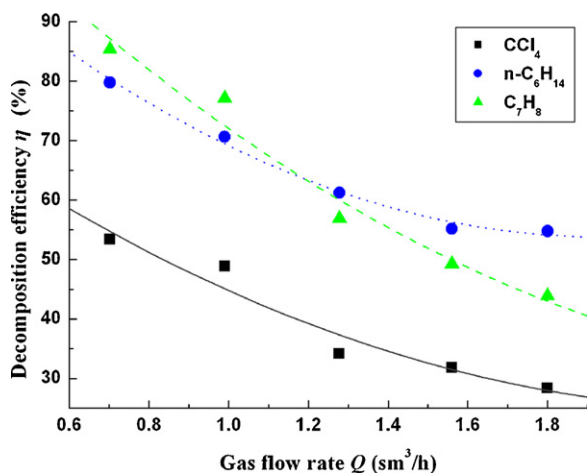
Table 1
Experimental conditions

Variable (unit)	Value	Range
Operation parameters		
Supply voltage U (kV)	10	10
Supply frequency f (Hz)	50	50
Total gas flow rate Q (S m³/h)	0.7	0.7–1.8
Smallest distance between electrodes D (mm)	3	1 & 3
Nozzle inner diameter φ (mm)	1.5	1.5 & 2.7
Vertical distance between electrode throat and nozzle H (mm)	25	25
Initial concentration X_0 (ppm)		
CCl_4	359	359
n-C_6H_{14}	968	968
C_7H_8	359	359

Fig. 4. Effect of gas flow rate on VOCs decomposition efficiency ($\varphi = 1.5$ mm, $D = 3$ mm, T-GA reactor).

Fig. 5. Geometrical model of T-GA reactor.

treatment capacity with maintaining the gas flow velocity in a proper range, when applied voltage is set in a fixed value (10 kV in present work). The feasibility of this channel was analyzed in this section.

Numerical simulation has been performed using CFD software to analyze the gas flow in the T-GA reactor in two configuration conditions: (a) $\varphi = 1.5$ mm, $D = 3$ mm, $\varphi/D = 0.5$; (b) $\varphi = 2.7$ mm, $D = 1$ mm, $\varphi/D = 2.7$. In the created geometrical model (as shown in Fig. 5), we defined the plasma region as the volume fenced by Face A, Face B, Face C and two electrodes. The gas flow rates of went through the electrodes throat and the plasma region were calculated and the results were presented in Table 2.

In $\varphi/D = 0.5$ condition, the gas flow rate went through the Face B (the electrodes throat) was 1.36 times as great as the inlet gas flow rate, while in $\varphi/D = 2.7$ condition this value was 0.17 times smaller than the inlet gas flow rate. Furthermore, the gas flow rate entered the plasma region in $\varphi/D = 0.5$ condition (2.15 S m^3/h) was obviously larger than that in $\varphi/D = 2.7$ condition (0.51 S m^3/h).

Above phenomenon is shown much clearly in the streamtraces profiles comparison between two conditions (as shown in Fig. 6). In $\varphi/D = 0.5$ condition almost all the inlet gas went through the electrode throat and some background air was also entrained by the inlet gas into the region between two electrodes. In contrast, rather big volume of the inlet gas could not enter the throat in $\varphi/D = 2.7$ condition. Hence, in theory, the contaminated
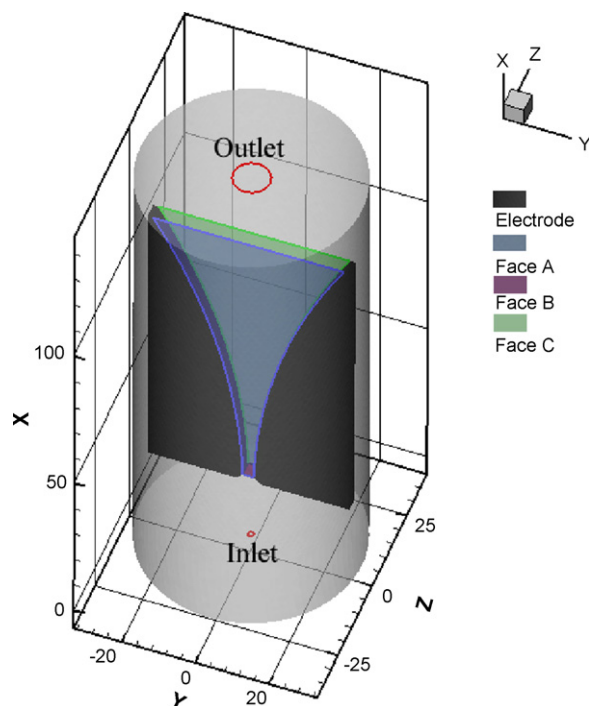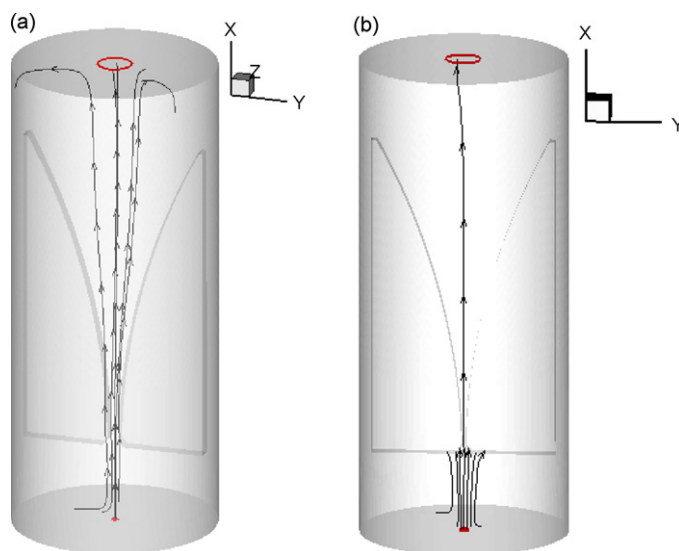

Fig. 6. Streamtrace profile in T-GA reactor ((a) $\varphi = 1.5$ mm, $D = 3$ mm, $\varphi/D = 0.5$; (b) $\varphi = 2.7$ mm, $D = 1$ mm, $\varphi/D = 2.7$).

Table 2
Flow rate calculation in proposed numerical model of T-GA reactor

Reactor configuration	Q (S m^3/h)	F_A[a] (S m^3/h)	F_B[b] (S m^3/h)	F_C[c] (S m^3/h)	F[d] (S m^3/h)	F_B/Q
$\varphi = 1.5$ mm, $D = 3$ mm, $\varphi/D = 0.5$	0.70	0.62	0.95	0.58	2.15	1.36
$\varphi = 2.7$ mm, $D = 1$ mm, $\varphi/D = 2.7$	0.70	0.21	0.12	0.18	0.51	0.17

[a] F_A: net flow rate through Face A.
[b] F_B: net flow rate through Face B.
[c] F_C: net flow rate through Face C.
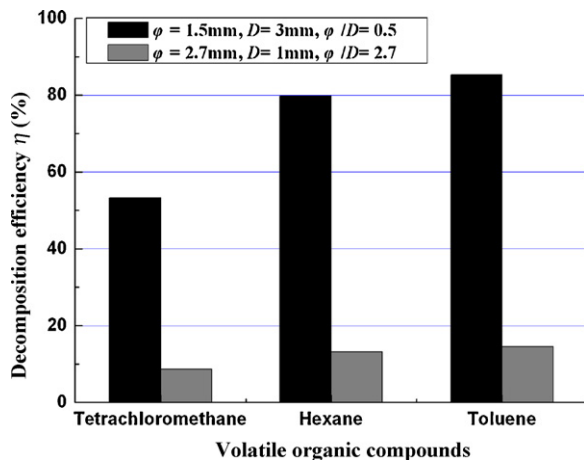[d] $F = F_A + F_B + F_C$: total flow rate through plasma region.

Fig. 7. Combined influence of nozzle inner diameter and nearest distance between electrodes on VOCs decomposition efficiency ($Q = 0.7$ S m^3/h, T-GA reactor).

Fig. 8. Comparison of VOCs decomposition efficiency in T-GA and D-GA reactors ($\varphi = 1.5$ mm, $D = 3$ mm).

gas get a greater probability to be treated in $\varphi/D = 0.5$ condition than in $\varphi/D = 2.7$ condition.

Experimental results (as shown in Fig. 7) are in good agreement with above analysis: rather higher decomposition efficiencies were obtained in $\varphi/D = 0.5$ than in $\varphi/D = 2.7$ condition for three target VOCs.

These observations suggest that the ratio of nozzle diameter φ and the shortest distance of two electrodes D should be maintained in a proper value to keep a good reaction process in the plasma region. Meanwhile, it has been reported [1,19] that for air at atmospheric pressure the shortest distance between electrodes cannot be larger than 4mm when the supply voltage is 10 kV. So the nozzle inner diameter cannot be enlarged unlimited to increase the treatment capacity in a defined power supply set.

3.2. VOCs decomposition in D-GA facility

A laboratory scale D-GA reactor was applied to the decomposition experiments of three target VOCs with relative high gas flow rate ($Q = 1.8$ S m^3/h). In the D-GA reactor, 0.4 S m^3/h dry air went through the bottom nozzle to process the GA evolution while 1.4 S m^3/h contaminated gas went frontal through the plasma region via a 15 mm in diameter inlet, as shown in Fig. 2.

VOCs decomposition efficiency comparison between T-GA and D-GA reactors is presented in Fig. 8. As explained in Section 3.1.1, an increase in the gas flow rate (from 0.7 to 1.8 S m^3/h) gives rise to a sharp decrease in the T-GA decomposition efficiency. However, by operating with flow separate (air and VOCs) and counter-flows, the mixing of the VOCs with the plasma region is strongly improved in D-GA reactor which can improve the VOCs treatment: the decomposition efficiency of 1.8 S m^3/h tetrachloromethane, hexane and toluene in D-GA reactor was 49.5, 62.4 and 59.5% respectively, which was higher than those in T-GA condition (28.2, 54.8 and 43.9%) in the same flow rate condition.
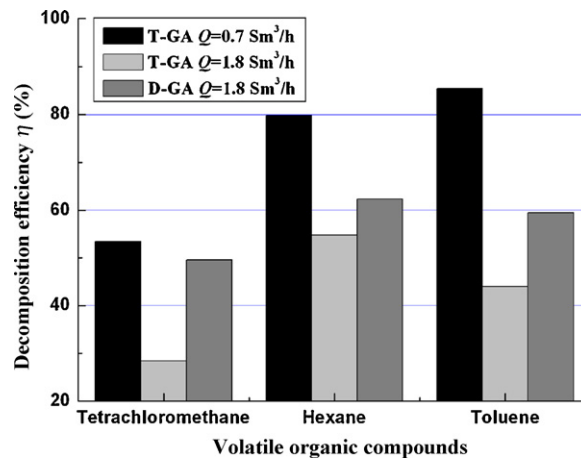
3.3. Comparative assessment between T-GA and D-GA reactors

In this section, the T-GA and D-GA reactor was applied to the decomposition experiments of 1.8 S m^3/h target VOCs respectively.

3.3.1. Decomposition by-products

Fig. 9 shows the typical FTIR adsorption spectroscopes output after T-GA and D-GA for three target VOCs. There was no significant difference in the decomposition by-product FTIR spectrum between two reactors. This observation suggests that the basic removal mechanism and decomposition process in the D-GA reactor is similar with which in T-GA condition. In other words, energetic electrons impact dissociation and reactions between VOCs molecules and ions or radicals are the main channels to VOCs decomposition, irrespective of T-GA or D-GA facility.

CO, CO_2 and NO_2 are usually the main decomposition by-products in dry air conditions [20,21], although the plasma assisted decomposition does not always completely oxidize the VOCs to above three inorganic oxides. The volumetric fractions of CO, CO_2 and NO_2 in by-product obtained in the output of D-GA reactor are higher than those in T-GA condition (see Fig. 9). This result can be mainly attributed to the larger contact area and longer interaction time between contaminated gas molecule and plasma region in the D-GA condition, which leads an enhancement to all possible reactions in the plasma region.

3.3.2. Specific energy consumption

Considering the industrial application, decomposition specific energy consumption is another important issue in the scale-up development of a plasma assisted reactor. The specific energy consumption, E is defined by Eq. (2) below:

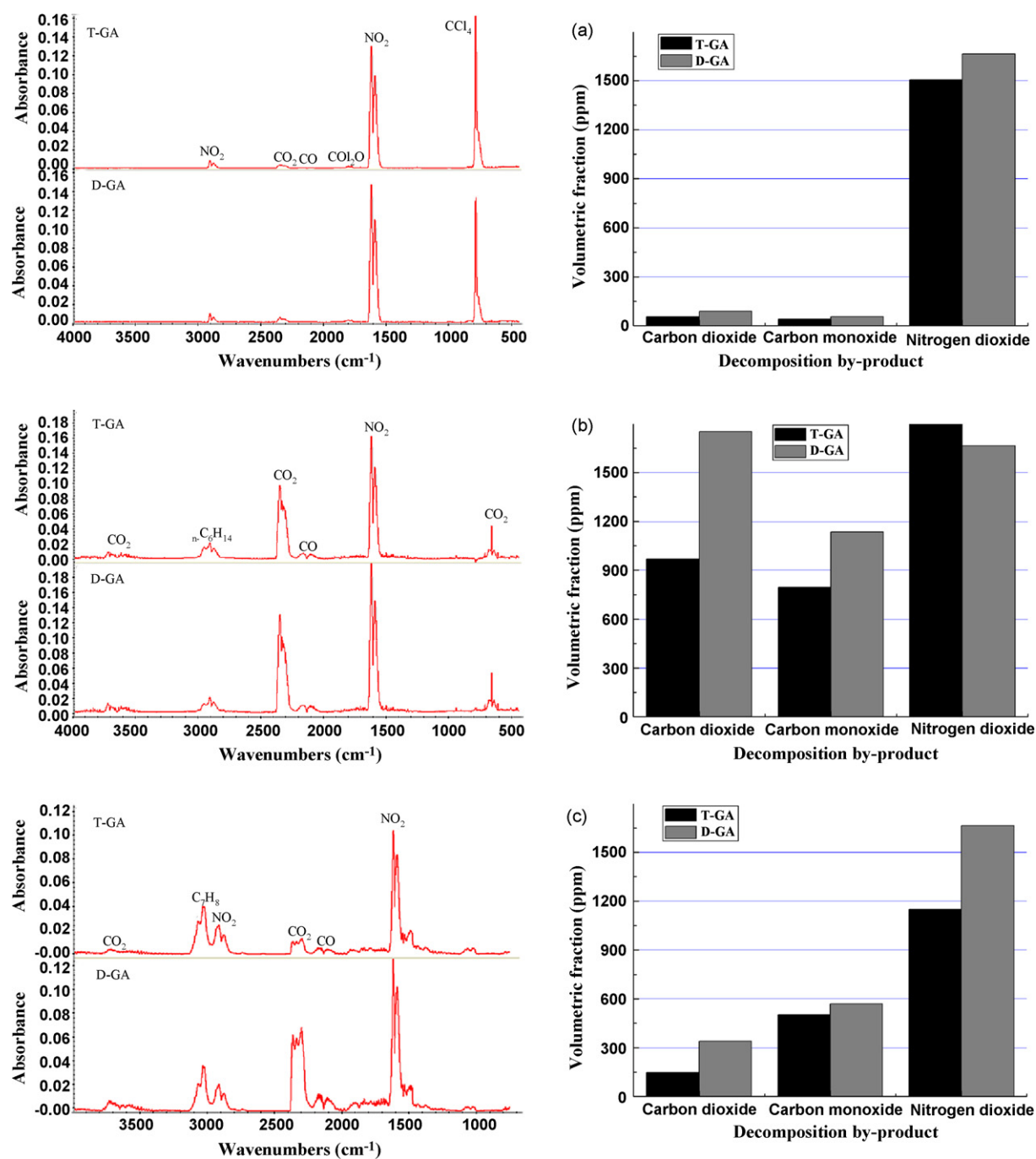$$E = \frac{P}{[Q(X_0 - X) \times 10^{-6}]} \tag{2}$$

with the unit of Wh/S m^3.

Fig. 9. VOCs decomposition by-product after GA discharge with T-GA and D-GA: (a) tetrachloromethane, (b) hexane and (c) toluene ($Q = 1.8\,S\,m^3/h$, $\varphi = 1.5$ mm, $D = 3$ mm).

Table 3
Specific energy consumption in T-GA and D-GA reactors

Reactor style	Gas flow rate Q (S m³/h)	Amount of VOCs converted $(X_0 - X)$ ($\times 10^{-4}$ S m³/h)			Specific energy consumption E ($\times 10^5$ Wh/S m³)		
		CCl_4	n-C_6H_{14}	C_7H_8	CCl_4	n-C_6H_{14}	C_7H_8
T-GA	1.8	1.8	9.5	2.8	10.5	2.0	6.8
D-GA	1.8[a]	3.2	11.0	3.8	3.4	1.0	2.8

[a] 0.4 S m³/h goes through the bottom nozzle and 1.4 S m³/h goes frontal through the plasma region.

Table 4
Comparison of power consumption for toluene decomposition between D-GA reactor and six-electrode GA reactor

Reactor style	Q (S m^3/h)	η (%)	P (kW)	E ($\times 10^5$ Wh/S m^3)
D-GA	1.8	59.5	0.11	2.8
Six-electrode GA [10]	2.0	92.0	0.84	2.5

Data in Table 3 indicate that D-GA reactor shows more effective decomposition ability with lower specific energy consumption, compared to T-GA reactor.

3.3.3. Comparison between D-GA and multi-electrode GA

Multi-electrode GA reactor is a scale-up development style based on T-GA reactor by using more than one pair of electrodes in the discharge chamber. The incontestable advantage of the D-GA compared with multi-electrode GA reactor is the application cost: multi-electrode reactor requires power systems that could ignite and sustain the discharge between many pairs of electrodes simultaneously what makes the power system rather complicated [14].

Table 4 summarizes the comparison of the power consumption for toluene decomposition between the D-GA reactor in current work and a six-electrode GA reactor [10]. The values of specific energy consumption of two styles are in the similar level, but the electric power consumption of the D-GA reactor has an obvious lower value of 0.11 kWh as compared to 0.84 kWh for the six-electrode GA system since the decrease of the power system amount. Compared to multi-electrode GA reactor, the decomposition efficiency in D-GA reactor is still in low level and can certainly be further improved. Our future work will be devoted to increase the decomposition efficiency of the D-GA reactor by optimizing the reactor configuration and operation parameters.

4. Conclusions

This study takes aim to find a proper way to scale-up the GA facility with meeting the requirements for commercial application in VOCs emission control process. Following conclusions are obtained:

(1) In the T-GA reactor, VOCs decomposition efficiency decreased sharply with increasing the gas flow rate, irrespective of target VOCs chemical structure. Some improvements should be done to the T-GA reactor when it is applied to the treatment of VOCs with relative high flow rate.
(2) The ratio of the nozzle diameter and shortest distance of two electrodes should be maintained in a suitable value range to guarantee the decomposition efficiency. In a fixed supply voltage condition, simply enlarging the nozzle inner diameter is not a proper way to raise the treatment capacity of the T-GA reactor.

(3) D-GA reactor released the treatment capacity form the limit of nozzle inner diameter by operating with flow separate (air and VOCs) and counter-flows. It showed higher decomposition efficiency with lower specific energy consumption to the treatment of relative high flow rate VOCs than T-GA reactor. D-GA reactor also avoided the drawbacks of the increases of power system amount and electric power consumption in multi-electrode GA system. This development provides an innovative option for small to large-sized GA facility.

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