

## Experimental Investigation on Styrene Emission with a 1000m<sup>3</sup>/h Plasma System

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**Abstract:** This paper reports experimental investigations on plasma-induced styrene emission abatement in air at room temperature (24±2°C) and pressure. Tests were performed with a 1000 m<sup>3</sup>/h plasma system under DC corona discharge. It is observed that styrene destruction in DC glow discharge is negligible in contrast to streamer corona. When relative humidity (RH) is in the range of 30%-80%, the energy consumption for styrene decomposition rises with RH value. The energy coefficient  $\beta$  value becomes larger with increasing either RH or the initial styrene concentration for RH in 30%-80% and the concentrations within 90 ppm-1000 ppm, respectively. With regards to those dependence sensitivity, the lower the concentration, the more sensitive to the RH value. Under optimal condition (100 ppm initial concentration, 35 %RH), the  $\beta$  value is 18.73 J/L.

**Keywords:** Styrene, NTPS, energy efficiency, byproducts

### 1 INTRODUCTION

Over the last 20 years, Non-thermal plasmas (NTPs) have been very active for air pollution control [1-6]. Compared with conventional techniques, NTPs may provide many advantages, such as low operation and capital costs when abating large volumes pollutants in low concentration. These NTPs are often generated by various gaseous discharges type, such as streamer corona, microwave discharge, gliding arc et al. [7].

Energy efficiency of NTPs processes has been improved by around a factor of 7-8 during the last 15 years [8], further research must be carried out to bridge the gap between industrial applications and fundamental academic studies. Worldwide research and development in recent years are engaging in large-scale tests. Yan et. al. reported a 50000 m<sup>3</sup>/h hybrid plasma system for odor treatment [10], Lee et al. reported a 42000 m<sup>3</sup>/h pulsed plasma system for flue gas emission clean [11].

In order to achieve technical and economical feasibility of industrial applications, three important issues must be considered, that are byproducts, energy efficiency and scalability [9]. Previous works have shown pollutant decomposition often depends on the initial concentration, relative humidity, corona mode and compositions of the carrier gas. However, few of authors presented their work in terms of energy efficiency.

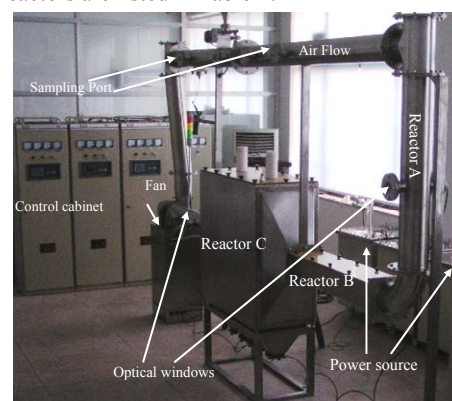
When decomposing VOCs, such as styrene, gas phase and solid phase byproducts (i.e., aerosol and polymerization) are often observed [12,13]. For high gaseous temperature, low initial concentration, and high energy density treatments, plasma induced oxidation processes mainly dominate the processes. For low gaseous temperature, high initial concentration, and low corona energy environment, however, more active electrons become directly attack organic molecules and then generate HC radicals for polymerization and solid film formation [14]. Because these gas and solid byproducts often induce secondary pollutants, it is important to know their physical and chemical properties. This study was done to develop engineering dates to illustrate the feasibility of NTPs techniques to destruct styrene at ppm level. A 1000 m<sup>3</sup>/h plasma system was developed and tested for DC corona

processes to investigate the effects of corona modes, humidity and initial concentrations on styrene decomposition in air at room temperature (24±2°C) and pressure. In addition, volatile organic byproducts were quantitatively analyzed by means of GC and GC-MS.

### 2 EXPERIMENTS

#### Reactor and Power Supply

Experiments are carried out with a 1000 m<sup>3</sup>/h closed loop flow system as shown in Fig. 1. Gases are circulated in and out of reactor in a loop driven by fan with variable flow rat (10 m<sup>3</sup>/h-1000 m<sup>3</sup>/h). The entire set-up is made of stainless steel. The total circulation volume is 780 L. It consists of four distinct parts: a wire-cylinder type corona reactor (Reactor A), a bipolar corona coagulation system (Reactor B), a five-field multipoint-plate ESP (Reactor C), and several sampling ports. Two optical windows are installed in the recirculation loop for observation of corona luminescence. Specifications of the three plasma reactors are listed in Table 1.



**Fig. 1** Schematics of the experimental setup

The system can be equipped with various power sources to achieve unique functions. Reactor A with pulsed, DC+plused or DC+AC energization induces VOCs oxidation. Aerosol produced by reactor A can be coagulated in reactor B under

positive and negative DC corona. Electrostatic precipitation is means of electrical measurements and/or optical observations.

**Table 1** Specification of the reactor configurations

Parameter	Reactor A	Reactor B	Reactor C
Height (cm)	130	15.5	100
Length (cm)	/	90	100
Plate-plate distance (cm)	/	15.5	10-350
Diameter (cm)	15	/	/
#Energized electrodes	1	2	1-32
Energized electrode distance (cm)	/	8	22-330
Energized electrode diameter (mm)	1.3	1.2	/

used to collection of coagulated aerosol. Moreover, VOCs decomposition can also be performed in reactor C with DC, pulsed, DC+plused or DC+AC power source.

This paper mainly focuses on VOCs oxidation in NTPs processes, thus reactor A and reactor C are adopted. The positive DC power source (AC/DC converter operating at 50 Hz line frequency, 50 kV maximum output) is used for energizing the plasma reactor.

A given amount of styrene is firstly injected into the closed loop, and then mixed with air under a flow rate for 30 mins–350 mins. By monitoring the concentration with GC, we can obtain a designed steady concentration. The temperature and relative humidity (RH) of system are measured with a thermometer & humidity meter (Cem DT 615). The internal flow rate is 305 Nm<sup>3</sup>/h. DC corona discharges are then run for a particular time interval (typically 60-300 Secs). After every run, Samples of the treated gas are collected with a gastight syringe for GC (Agilent GC6890N-MS5973N) and GC-MS (Fuli GC9750) analysis.

The mean value of the current is measured by a current meter in series with a 10 kΩ resistor and in parallel with a 10μF capacitor. The voltage is measured using a digital oscilloscope (Tektronix DPO 7054) and a high voltage divider (Tektronix P6015). Thus the energy density is calculated by the following equation.

$$E = \frac{V \times I \times t}{V_{STP}} = \frac{V \times I \times t \times T_{operation}}{V_{system} \times T_{STP}} \quad (1)$$

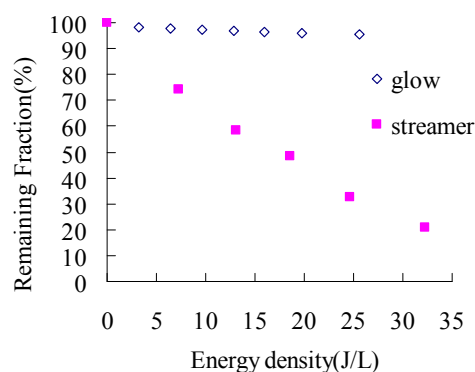
where  $E$  represents energy density (J/L),  $V$  is measured DC voltage (kV),  $I$  is mean corona current (mA),  $t$  is treatment time (s),  $V_{STP}$  is volume of the system at STP condition (L),  $V_{system}$  is system volume (L),  $T_{STP}$  is 298 K,  $T_{operation}$  is system temperature (K).

### 3 RESULTS AND DISCUSSIONS

#### 3.1 Corona Modes

Fig. 2 shows the effect of corona modes on styrene conversion. The experiment is conducted with the reactor A and the reactor C under DC energization. Because of their different electrode geometries, glow and streamer corona are produced, respectively. The two corona modes can be distinguished by

For streamer corona discharge, the streamer propagation can be observed between electrodes gap, and corona current waveforms demonstrate pulsed characteristics. However, for DC Hermstein glow, luminous sheath can only be observed around the energized electrode. GC analysis of plasma- processed gas suggests that styrene reduction in glow discharge is negligible, whereas significant amount of conversion is observed in streamer corona. This difference can be ascribed to their difference of discharge volume and radical productivity.



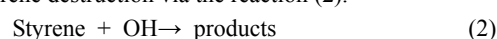
**Fig. 2** Conversion of styrene as a function of energy density with different corona modes: □ 85 ppm (RH =40%) ■ 93 ppm (RH=44 %)

#### 3.2 Humidity & Initial Concentrations

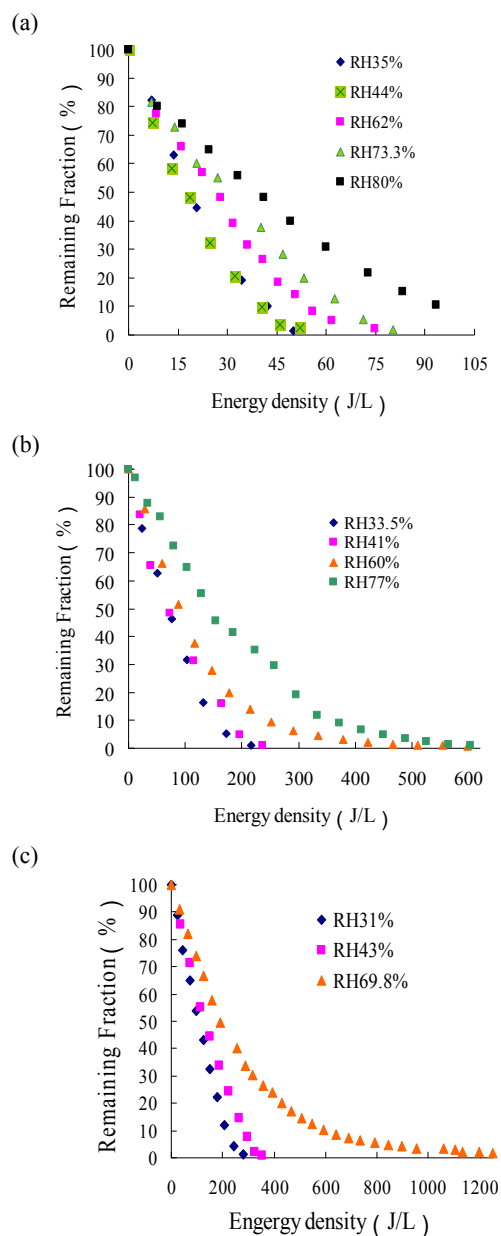
Fig. 3 shows the decomposition of 90 ppm–100 ppm (Fig. 3(a)), 667 ppm–721 ppm (Fig. 3(b)) and 942 ppm–1000 ppm (Fig. 3(c)) styrene as a function of energy density in 30%–80% relative humidity (RH). The experiment is performed with reactor C. It is found, at a given initial concentration, the energy density for styrene decomposition is increasing with RH. To convert 90% of 90 ppm–100 ppm styrene, the energy density is 42 J/L and 93 J/L, respectively when increasing RH from 35% to 80%.

The conversion becomes less sensitive for the RH in the range of 30%–45%.

In humid environment, Water molecules are collided with energetic electrons and generate OH free radicals, thus facilitate styrene destruction via the reaction (2).



Moreover, the generated active species, for example O radical, which is responsible for styrene decomposition in an oxidizing atmosphere is quenched by water vapor. It is suggested that, for high RH condition, negative effects of humidity dominate the plasma process, hence suppress styrene destruction. In low RH condition, negative and positive effects of humidity can balance with each other.



**Fig. 3** Conversion of styrene as a function of energy density with various relative humidity (RH): (a) 90 ppm–100 ppm (b) 667 ppm–721 ppm (c) 942 ppm–1000 ppm

The energy consumption for styrene decomposition is increasing with increase of its initial concentrations. When the RH is 32%, to obtain 90% of styrene conversion, the energy density for destructing 90 ppm–100 ppm and 942 ppm–1000 ppm styrene are 42 J/L and 215 J/L respectively.

The energy efficiency is evaluated with  $\beta$  value in the units of J/L. According to global corona plasma kinetic model,

the  $\beta$  value is calculated from exponential equation [9]

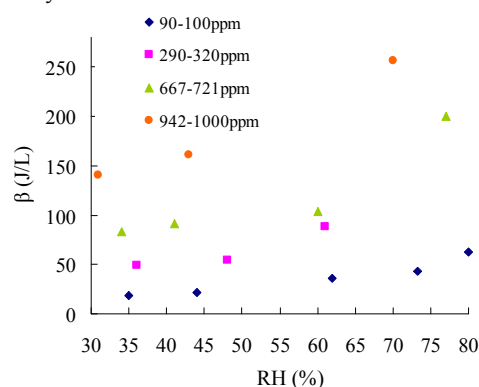
$$\frac{C}{C_0} = \exp\left(-\frac{E}{\beta}\right) \quad (3)$$

where  $C_0$  is initial styrene concentration,  $C$  is remaining concentration after applying the energy density  $E$  and  $\beta$  is the energy constant. It suggests the higher the  $\beta$  value, the lower the energy efficiency in plasma process.

According to the present work, the slopes of  $\ln\left(\frac{C}{C_0}\right)$  vs.  $E$

are not following linear trend after several decades of plotting in  $\frac{C}{C_0}$ , but the initial slopes are roughly linear. Therefore, the  $\beta$

value is still an effective concept for evaluating the energy efficiency<sup>[14]</sup>. Fig. 4 shows the  $\beta$  value as a function of RH at different styrene initial concentrations.



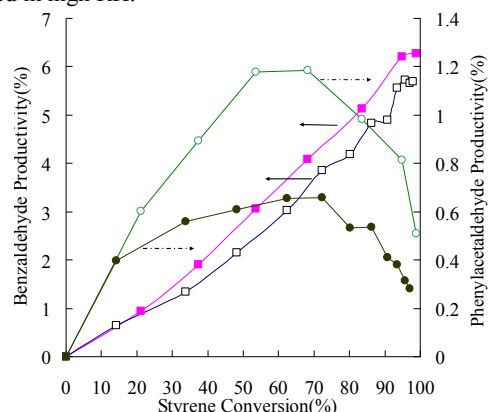
**Fig. 4** Energy constant ( $\beta$  value) for destructing various initial concentrations of styrene as a function of relative humidity (RH)

It is demonstrated that, the energy efficiency is decreasing with increase of RH and initial concentrations. The energy efficiency reduced by increase of initial concentrations can be compensated by decreasing RH. In addition, the destruction of low initial concentration of styrene is more sensitive to RH compared with high initial concentration styrene. From 35% to 73.3% RH, the  $\beta$  value for destructing 90 ppm–100 ppm styrene is increased 2.43 folds, whereas from 30% to 70% RH, the  $\beta$  value for destructing 942 ppm–1000 ppm styrene is increased 1.8 folds. This can be explained by the reaction frequency between styrene molecules and active species will be lowered in low styrene concentration at humid condition.

### 3.3 Byproducts

The by-products in present experiments can be classified into gas phase and solid phase byproducts. Common volatile organic products from oxidation of styrene in this work are benzaldehyde, phenylacetaldehyde and benzoic acid. Due to their relative high intensity of GC signals, only benzaldehyde and phenylacetaldehyde are quantitative measured. The two products are quantified by calculating the ratio of the species concentrations to the initial styrene concentrations. Fig. 5 displays the product data for treating 667 ppm–721 ppm styrene with 33% and 60% RH condition. It is seen that both the productivity of benzaldehyde and phenylacetaldehyde are

restrained in high RH.



**Fig. 5** Production of benzaldehyde and phenylacetaldehyde from the treatment of styrene (667 ppm–721 ppm in air) with DC corona, as a function of relative humidity RH:

- Benzaldehyde(RH=60%) ■ Benzaldehyde (RH=33%)
- Phenylacetaldehyde(RH=33%)
- Phenylacetaldehyde (RH=60%)

The productivity of benzaldehyde is increasing with styrene conversion. However, the productivity of phenylacetaldehyde initially increases and then decrease with styrene conversion. This difference is due to their different kinetic rate constants with active species (O, OH et. al.). The turning point of phenylacetaldehyde productivity gives some suggestions of the decomposition reactivity of the products relative to styrene<sup>[15]</sup>. Recent experiments are mainly focusing on the physical and chemical characteristics of solid phase byproduct (aerosol and polymerization), the result will be published in a separate paper.

#### 4 CONCLUSIONS

A 1000 m<sup>3</sup>/h plasma system has been developed to convert styrene in air with DC corona. The experiment is performed with RH and initial concentration ranged from 30%–80% and 100–1000ppm respectively. The following conclusions are obtained.

(1) Styrene destruction in streamer corona is significantly better in contrast to glow discharge.

(2) Energy consumption for styrene decomposition is increasing with increase of RH.

(3) Energy efficiency becomes lower with increasing either RH or initial concentration. The destruction of low concentration styrene is more sensitive to the value of RH compared with high concentration styrene.

(4) Byproduct analysis suggests benzaldehyde, phenylacetaldehyde and benzoic acid are major volatile organic products from oxidation of styrene.

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