Journal of Hazardous Materials 180 (2010) 449–455

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Destruction of acenaphthene, fluorene, anthracene and pyrene by a dc gliding arc plasma reactor

Liang Yu [a], Xin Tu [b], Xiaodong Li [a], Yu Wang [a], Yong Chi [a], Jianhua Yan [a,*]

[a] State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, 310027, China
[b] School of Chemistry, The University of Manchester, Oxford Road, Manchester, M13 9PL, UK

ARTICLE INFO

Article history:
Received 5 January 2010
Received in revised form 22 March 2010
Accepted 13 April 2010
Available online 18 April 2010

Keywords:
Polycyclic aromatic hydrocarbons
Destruction
Gliding arc
Non-thermal plasma

ABSTRACT

In this study, four kinds of PAHs (polycyclic aromatic hydrocarbons) i.e. acenaphthene, fluorene, anthracene and pyrene are used as targets for investigation of PAHs treatment process assisted by dc gliding arc discharge. The effects of carrier gas and external resistance on the PAHs decomposition process are discussed. The results indicate that the destruction rate can be achieved to the highest with the carrier gas of oxygen and the external resistance of 50 kΩ independent of type of PAHs. Furthermore, experimental results suggest that destruction energy efficiency of gliding arc plasma would be improved by treating higher concentration pollutants. Based on the analysis of experimental results, possible destruction mechanisms in different gas discharge are discussed.

1. Introduction

Incomplete combustion of fossil fuel or municipal solid waste (MSW) may lead to release many kinds of persistent organic pollutants (POPs) such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzo-p-dioxines (PCDDs), polychlorinated dibenzofuran (PCDFs) [1]. PAHs and PCDD/Fs have been designated as priority pollutants by US EPA and attracted increasing attention due to their contribution to environmental and health problems [2]. Many countries have legislated strict laws on these kind of POPs emission. Therefore, it is necessary to develop friendly technologies to destruct POPs. In this study, we focus our attention on the PAHs destruction technology.

In recent years, several technologies have been utilized to remove PAHs from the sewage sludge, contaminated soils and wastewater. Bioremediation has received wide acceptance as a viable and low-cost treatment method for remediation soils and water polluted by PAHs for many years. However, biodegradation-based technologies are usually long-term processes which cost several weeks [3]. In addition, bioremediation often has limited applicability when soils or sewage sludge are contaminated with complex mixtures of highly PAHs. In order to overcome these disadvantages, scientists have developed kinds of new technologies to destruct PAHs. Nam et al. developed a new method, which combined biodegradation and Fenton reaction, to enhance the PAHs degradation process in soils [4]. Photocatalytic degradation of PAHs on soils surface using TiO_2 under UV light was studied by Zhang et al., which concluded that catalyst TiO_2 can accelerate the PAHs photodegradation speed for 3–4 times [5]. Owing to having large number of energetic electrons and reactive radicals such as OH, O and H, electron beam irradiation has been applied to PAHs destruction in sewage sludge and the destruction rate of PAHs was reported to be about 90% at absorbed dose 5 kGy [6]. The aforementioned technologies are mainly used for destruction of PAHs in solid or liquid phase and a few publications are concerning with PAHs removal in gas flow. We have ever studied the degradation of PAHs and soot particles, released from polyethylene (PE) and polyvinyl chloride (PVC) combustion in a laboratory-scale drop tube furnace, using an ac gliding arc reactor [7]. The results of this research indicated that the PAHs removal rate can be achieved up to 74.4%. However, degradation products and destruction mechanisms were not discussed in that work. Recently, we have developed a dc gliding arc system for the decomposition of PAHs. The specific PAHs decomposition processes [8], dc gliding arc reactor performance and destruction mechanisms are investigated in our further research. In this study, four kinds of PAHs destruction mechanisms are discussed based on a qualitative and quantitative analysis of products. Additionally, a comparative assessment of the gliding arc reactor performance on destruction energy efficiency under different working conditions is done.

As an alternative approach, non-equilibrium plasma technologies have been utilized for volatile organic compounds (VOCs) abatement, wastewater treatment and energy conversion for many years and received increasing attentions [9–10]. Because of the

* Corresponding author. Tel.: +86 571 87952443; fax: +86 571 87952438.
E-mail addresses: yanjh@zju.edu.cn, yuliang@zju.edu.cn (J. Yan).

0304-3894/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2010.04.051

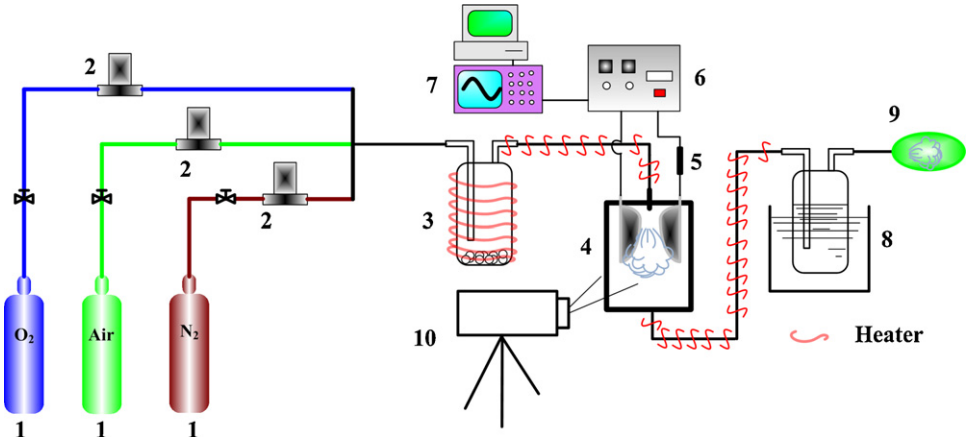

Fig. 1. Experimental setup: 1. gas source, 2. mass flow controller, 3. PAHs source, 4. gliding arc reactor, 5. external resistor, 6. power source, 7. oscilloscope, 8. absorption bottle containing hexane, 9. gas bag; 10. high speed camera.

large number of high energy electrons and radicals and relative low gas temperature, non-equilibrium plasma is regarding as a promising technology for gas phase pollutants destruction. In this study, we study the four kinds of PAHs decomposition processes by one of non-thermal plasmas, namely gliding arc discharge, which has been utilized for abatement of some other gas pollutants and considered as an efficient kind of non-thermal plasma by many scientists.

2. Experimental

2.1. Experimental setup

The experimental setup shown in Fig. 1 mainly consists of PAHs-containing gas generation system, a dc gliding arc plasma reactor and an analysis system. PAHs powders are placed in glass bottle and be heated to set temperature. The temperature of transfer pipe in the experiment is kept at 130 °C. The vaporized PAHs are carried by gas flow at 6 l/min and sprayed into gliding arc plasma system which consists of a dc power supply (0–6150 V, 900 W), an external resistance bank (50–93 kΩ) and a quartz gliding arc reactor. Compared with the PE and PVC combustion system [11], this gas phase PAHs-generations system are more stable. The arc voltage and current are measured by using a current probe (Tektronix, TCP303) and a high voltage probe (Tektronix, P6015A), respectively. The electrical signals are sampled synchronously through a digital oscilloscope (Tektronix, TDS2024). The spatial evolution of the gliding arc column is recorded by an HG-100K high speed camera. Emission spectra of the gliding arc discharge are measured by an optical fiber connected with a SBP300 spectrometer. To collect the reaction products, Post-destructed gas flows pass through the absorption bottle containing hexane. We carried out each experiment for 3 min. Every experiment is started after 20 min stabilization without discharge.

2.2. Materials and methods

Four kinds of PAHs as test pollutants, namely acenaphthene (AcP), fluorene (Flu), anthracene (AnT) and pyrene (Pyr), are purchased from Sigma–Aldrich (HPLC grade), Japan, and are used without further purification. Hexane (analytical grade) is purchased form Sinophram Chemical Reagent Co. Ltd., China. In these experiments, nitrogen, air and oxygen are used as carrier gas and external resistance varies from 50 kΩ to 93 kΩ. Post-destructed products in hexane are analyzed by gas chromatography with mass spectrometry detection (GC/MS). The off-gas is analyzed by a FTIR spectrometer.

2.3. Gliding arc gas discharge

Fig. 2 presents the photographs of gliding arc gas discharge recorded by common speed camera and high speed camera (5000 frames/s). The gliding arc gas discharge produces a plasma region characterized by periodic arc movement between at least two electrodes. This type of plasma starts at the shortest gap between electrodes by an initial breakdown of introduced gas when the power source provides high enough voltage between electrodes. Then the arc is pushed downstream by the gas flow and glides along the electrode surface until it quenches. After the decay of discharge, there is a new breakdown at the narrowest gap and the cycle repeats. This periodic discharge evolves during a cycle from arc to transitional discharge with a relative high level of electron density [12]. Electrical signals and an emission spectrum of a nitrogen discharge are shown in Figs. 3 and 4, respectively. An opposite variation tendency can be observed from arc voltage and arc current signals in one period presented in Fig. 3. During the arc evolution, the arc voltage increases and arc current decreases due to increase of discharge length. In Fig. 4, it is obvious that the 300–400 nm scans are predominated by the $N_2(C^3\Pi_u \rightarrow B^3\Pi_u)$ 2nd positive system, which indicates that electronic excited state of nitrogen molecules abound in a nitrogen gliding discharge. Nowadays, gliding arc plasma has attracted considerable interest on its

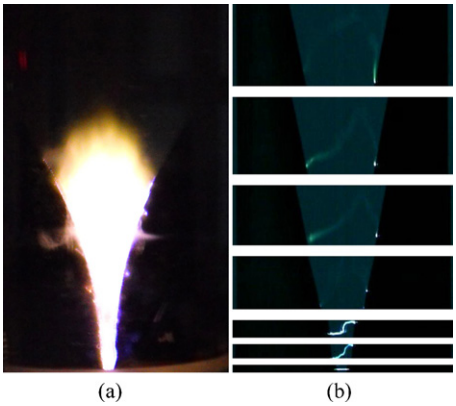

Fig. 2. Photographs of gliding arc discharge recorded by common (a) and high speed (b) cameras.