



Removal of gaseous HxCBz by gliding arc plasma in combination with a catalyst



Yong Ren, Xiaodong Li, Shasha Ji, Shengyong Lu, Alfons Buekens, Jianhua Yan *

State Key Laboratory of Clean Energy Utilization, Zhejiang University, Hangzhou, Zhejiang, China

HIGHLIGHTS

- Rotating gliding arc plasma is used in the destruction of gaseous HxCBz.
- Coupling SCR catalyst with plasma can effectively promote the destruction effects.
- Destruction mechanisms and contributions of active factors are distinguished.
- The mechanism of plasma and catalyst to HxCBz destruction are proposed.

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ABSTRACT

Hexachlorobenzene (HxCBz) owns the chemical structure of one benzene ring and six H atoms substituted by Cl atoms and it is a persistent organic pollutant present in flue gas from municipal solid waste incineration as an important precursor of dioxins. Its removal was studied using gliding arc plasma treatment, coupled downstream with a V_2O_5 – WO_3 – TiO_2 catalyst. Several parameters (input voltage, O_2 concentration, catalytic temperature and catalyst position) all influenced its removal efficiency (RE). Optimal parameter settings were tentatively determined, i.e., an input voltage of 15 kV, the temperature of the catalyst (250 °C), and the O_2 concentration (30 vol% O_2) tested at a single, fixed concentration of gaseous HxCBz (71.6 ng Nm^{-3}). A maximum RE of $76 \pm 3\%$ HxCBz was attained, with the plasma and coupled catalyst combined. Two destruction pathways, incorporating dechlorination and oxidation reactions, were recognised, both based on the detection of end- and intermediate products as well as of active species produced by the plasma. These end- and intermediate products included: low chlorinated polychlorobenzenes (mainly 1,2,4-Trichlorobenzene) as well as hydrocarbons (mainly C_2H_6), $HCOOH$, CH_4 , CO , CO_2 , etc.

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1. Introduction

Hexachlorobenzene (HxCBz) owns the chemical structure of one benzene ring and six H atoms substituted by Cl atoms and it is one of the persistent organic pollutants (POPs) present in the flue gas from all known thermal and metallurgical processes producing polychlorinated dibenzo-furans and polychlorinated dibenzo-p-dioxins, in brief dioxins (Oehme et al., 1987; Altwicker et al., 1993). To study the decomposition of dioxins, HxCBz is commonly chosen as a model compound, because it has similar characteristics and is much less toxic.

As required by the Stockholm Convention, measures have been taken to reduce HxCBz (and dioxin) emissions from various sources, e.g., adsorption on activated carbon (McKay, 2002), catalytic oxidation (Weber et al., 1999), thermal destruction (Song et al., 2008),

and (tentatively) non-thermal plasma (NTP) (Yan et al., 2007b). Although widely applied, activated carbon merely transfers semi-volatile pollutants from flue gas towards the gas cleaning residues (Chi et al., 2006). Thermal destruction faces possible dioxin formation during gas cooling considering de novo reactions in the temperature window 250–400 °C (Buekens and Huang, 1998).

NTP is an advanced technique for oxidizing pollutants (Petitpas et al., 2007; Fridman et al., 2008), e.g., volatile organic compounds (VOCs) (Indarto et al., 2007), polycyclic aromatic hydrocarbons (PAHs), polychlorinated benzenes, and dioxins (Hung et al., 2010). Plasma reactors come in various guises, including: dielectric barrier discharge, gliding arc plasma (GA), corona discharge, etc. Therein, GA plasma shows unique features (Ren et al., 2013) with regards its transitional states between thermal plasma and NTP. Abundant presence of active species (Fridman et al., 1998), appropriate application for large gas flow, and a relatively low gas temperature are all facets thereof. Therefore gaseous HxCBz removal by GA plasma was considered. In the past, attempts using GA

* Corresponding author. Tel.: +86 571 87952738.

E-mail address: yanjh@zju.edu.cn (J. Yan).

plasma to destroy VOCs, e.g., benzene, toluene, chlorobenzene, and PAHs have been investigated.

Significant conversion of VOCs, both aromatic and chlorinated VOCs, had been achieved using GA plasma and the end-products were CO, CO₂, Cl₂, and H₂O. GA plasma was regarded as an effective method for the treatment of gaseous pollutants produced on an industrial scale (Indarto et al., 2007). The destructive effects depend on the discharge characteristics of the GA plasma, including gas flow field, electric parameters, and reactor structure (Bo et al., 2007b). PAHs, with their multiple benzene ring structure, are potential candidates for treatment by GA plasma methods. High concentration PAHs could be decomposed (the highest destruction efficiency found was 92%) by DC GA discharge and the active species from the oxygen can accelerate the decomposition process. During GA plasma decomposition, some reactive radicals, i.e. O and OH, play an important role and different objective compounds perform differently because of their chemical structure and reaction channels (Bo et al., 2007a; Yan et al., 2007a).

When promoting a given treatment ability, some researchers make some specific catalyst combine with a NTP to yield a plasma/catalyst system. According to the position of the catalyst inside the plasma reactor, such a hybrid system is divided into in-plasma configuration (IPC) and post-plasma configuration (PPC) (Van Durme et al., 2008). In the NTP discharge process, gas molecules can be excited and transformed to active short living species (e.g. atomic oxygen, superoxide species (O₂⁻), hydroxyl radicals, etc.). In addition, some long living species, such as ozone, are produced from the recombination of the active species. Both of them form effective components for treatment of the target pollutant. However, IPC can more thoroughly take advantage of the short-lived active species, and long-lived species act more on the catalyst positioned in the downstream plasma reactor (PPC). Because IPC may influence the operation and cause instability in the GA plasma, the catalyst is generally positioned downstream of the GA plasma.

Considering the characteristics of HxCBz, a TiO₂-based catalyst was deemed beneficial for its decomposition. TiO₂ has significant photosensitive characteristics beneficial for VOC destruction (Lu et al., 2011). V₂O₅ is the main active component for dioxin destruction and V=O bonds are regarded as a typical active site (Ozkan et al., 1994). The combination of V₂O₅ and TiO₂ can take nucleophilic substitution reactions with chlorinated benzene (Finocchio et al., 2006). The extra loading of WO₃ plays an enhancement role in the activities of the multiple catalysts (Bertinchamps et al., 2006). Hence, a commercial V₂O₅-WO₃-TiO₂ catalyst downstream from the GA plasma was adopted for the destruction of gaseous HxCBz.

In the present work, the removal effects of HxCBz by GA plasma alone and plasma coupled with a catalyst were experimentally compared. Moreover, the effects of major parameters on the HxCBz removal efficiencies (RE, defined as the ratio of HxCBz removed and initial HxCBz expressed as a percentage), such as: input voltage, O₂ concentration, catalytic temperature, and catalyst position were considered. The destruction mechanism of HxCBz in the catalytic plasma process was inferred by the detection of the intermediate- and end-products of HxCBz and active species in the plasma region. The long-term goal was to develop a technically feasible method for the thorough destruction of dioxin-like, chlorine substituted organic compounds, and thus address the threat from municipal solid waste incineration (MSWI) effects on the environment and human health.

2. Materials and methods

2.1. Experimental system

The experimental system comprises three parts, respectively related to the generation of a steady HxCBz inflow, to the plasma

reactor system, and to the detection of residual products in the effluent, as shown in Fig. 1.

A steady continuous HxCBz generation system was established, which could be divided into three parts (HCB standard solution, a trace liquid injection pump and a stable volatile zone). A standard solution of HxCBz was mixed by pure solid HxCBz and an organic solvent with a high boiling point, and then injected by an injection pump and atomised by a quartz-made jet nozzle. The atomised solution went with a carrier gas stream and was heated in the stable volatile zone. All drops of liquid solution were fully evaporated to form a steady gas flow containing a fixed concentration of HxCBz, and the injected solvent was eliminated by oxidation and decomposition of excess O₂ under high temperature to avoid the side effect. There was no solvent detected in the outlet of the generation system. The steady and continuous injection control coupled with a thorough mixture of HxCBz and solvent could ensure the stability of HCB in gas stream. The repeatability of the HxCBz concentration was 94–103% for 8 continuous sampling periods, around an average initial value of 788 ng Nm⁻³ established for a fixed carrier gas flow rate of 1 L min⁻¹. This flow gas was called the “reactant gas flow”.

The plasma reaction sub-system comprises a rotating GA plasma, developed inside a reactor (Fig. 2), a downstream catalyst and a high voltage power supply. The structure of the rotating GA plasma reactor is shown in Fig. 2. There were two gas streams derived from the same gas bottle: a diluent gas with an adjusted flow capable of delivering between 7 and 11 L min⁻¹ (fixed at 10 L min⁻¹ in these tests) called the “drive gas flow” and the carrier gas doped with HxCBz, i.e. the “reactant gas flow”. Together they constituted the experimental gas, and were controlled by two different mass flow meters (#1 and #2). The experimental gas was a mixture of N₂ and O₂, with an O₂ concentration ranging from 0 to 30 vol% (Giugliano et al., 2001) according to practical MSWI flue gas conditions (around 10 vol%). The mixed experimental gas was used to drive the development of the electric arc, excited at the gap between the inner and outer electrodes and lead the formation of the plasma region (Fig. 2). The high voltage between the electrodes was supported by a high voltage power source (adjustable voltage of 8–17 kV, at an alternating current frequency of 50 Hz). The experimental mixed gas caused a disturbed flow field in the reactor because of its high speed and strong rotation: this was beneficial to the interactions between active species and reactant molecules. The catalyst was located behind the plasma region, i.e. in the post-plasma region, and an electric heating band (at 150–250 °C, adjusted by a separated controller and measured by a thermocouple) was also placed there so as to pack the quartz glass tube. The temperature was set according to the practical conditions of flue gas discharge behind the bag filter house of the MSWI (140–210 °C) (Fridman et al., 1998) and avoided the temperature windows of dioxin formation. The distance between the central electrode and the honeycomb catalyst was either 1.0 or 9.0 cm, defined as the two different positions at the “proximal end” and “distal end” of plasma reactor respectively: they both ensured that there was no influence on the operation of the plasma reactor. The gas hourly space velocity was set to approximately 45000 h⁻¹.

The detection sub-system consisted of the necessary sampling and detection equipment. The sampled HxCBz was collected downstream of the plasma treatment by absorption into toluene in two-stage bottles. After the pre-treatment process, the samples were then offline analysed by GC-ECD (Agilent 6890N, USA) following Chinese National Standard HJ 621-2011GB/T.

The end- and intermediate products as well as of active species from plasma are online detected by several devices in the experimental process, including monochromator (PI-Acton 2750) with ICCD (PI-MAX 2), Time Of Flight Mass Spectrometer (TOFMS, MASS-500) and Fourier Transform Infrared Spectrometer (FTIR,

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