Journal of Hazardous Materials 325 (2017) 230–238

Contents lists available at ScienceDirect

Journal of Hazardous Materials

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

Application of microwave energy in the destruction of dioxins in the froth product after flotation of hospital solid waste incinerator fly ash

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HIGHLIGHTS

- Microwave treatment is applied for the destruction of dioxins in the froth.
- Combination of microwave irradiation and carbon constituents favors decomposition of dioxins.
- Recovery potential of PAC in the treated froths for reuse was found.

ARTICLE INFO

Article history:
Received 22 August 2016
Received in revised form
23 November 2016
Accepted 30 November 2016
Available online 2 December 2016

Keywords:
Microwave treatment
Dioxins
Fly ash
Powder activated carbon
Froths

ABSTRACT

Most of polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs) and powder-activated carbon (PAC) in hospital solid waste incinerator fly ash are enriched in the froths produced through flotation. Because PAC is an excellent microwave absorber, microwave treatment was performed on the froths in this study to decompose PCDD/Fs. The results showed that the destruction efficiency of PCDD/Fs increased with increasing microwave incident power and processing time, particularly for highly chlorinated PCDD/Fs. With a microwave incident power of 2100 W at 7 min, the total mass destruction efficiency of the PCDD/Fs in the froths reached 99.6 wt.% and the total toxic equivalent (TEQ) of PCDD/F was substantially reduced from 29.0 to 0.08 ng–I-TEQ/g. PCDD/Fs in the froths were mostly decomposed and evaporated very little into exhaust gas under microwave treatment, especially at 2100 W. The treated froths displayed good porous structures, enabling the potential recovery of PAC for reuse. Microwave treatment of the froths could promote the rapid decomposition of PCDD/Fs and the recovery of a typical waste resource; also it could present a viable alternative to combustion treatment for the froths.

1. Introduction

Powder-activated carbon (PAC) has high adsorption capacity because of its extensive surface area and microporous structure [1]. During hospital solid waste incineration, a large amount of PAC is sprayed into exhaust gas to meet the new Chinese emission standards of less than 0.1 TEQ ng/m^{-3} for polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDD/Fs). PCDD/Fs tend to enrich on hospital solid waste incinerator (HSWI) fly ash by the presence of PAC which adsorbs PCDD/Fs. On the basis of the lipophilicity and hydrophobic nature of PCDD/Fs in fly ash, a previous study established an effective flotation process for HSWI fly ash [2]. After flotation, two types of solid products, froths and tailings, are produced. Tailings are deposited directly into landfills because their pollutant concentrations are below the upper limit specified in Chinese landfill site regulation [3]. However, most of PCDD/Fs in HSWI fly ash would be enriched and then separated as float products by flotation. In our previous study, the froths were found to contain a high concentration of PCDD/Fs (29.0 ng–TEQ/g) and a high content of carbon constituents (including PAC and unburned carbon), with a loss of 56.4 wt.% on ignition (LOI). This indicated an urgent need for the development of an effective technology to properly treat and dispose of the froths.

The detoxification of PCDD/Fs is exceptionally difficult because of their stable chemical structure [4,5]. In recent years, numerous methods for decomposing PCDD/Fs have been developed, including mechano–chemical treatment, reductive dechlorination, hydrothermal treatment with supercritical water oxidation (SCWO), and thermal treatment with microwave peroxide oxi-

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http://dx.doi.org/10.1016/j.jhazmat.2016.11.078
0304-3894/

dation (MPO). For example, Yan [6] and Mitoma [7] have concluded that PCDD/Fs in fly ash can be directly degraded using mechano–chemical treatment. Mechano–chemical treatment is a non–combustion technology that requires no heating process or exhaust gas treatment, but ball milling generally must be maintained for approximately 2–12 h to achieve the desired detoxification. Zhang indicated that the removal efficiency of TEQ reached almost 98% through catalytic degradation of PCDD/Fs with Pd/C in an ethanol–water solution [8]. However, catalysts can easily be poisoned when they make contact with fly ash and most are quite expensive [9]. Hu found that the hydrothermal treatment of fly ash exhibited considerably high destruction efficiency for PCDD/Fs and the use of a mixture of ferric and ferrous sulphate as an auxiliary chemical could enhance decomposition [9]. Okajima indicated that SCWO could decompose the PCDD/Fs adsorbed in the activated carbon and that the destruction efficiency of PCDD/Fs was higher than 99% under conditions of 500 °C and 20 MPa [10]. Zou found that SCWO enabled the simultaneous detoxification of organic pollutants in municipal solid waste (MSW) leachate and PCDD/Fs in fly ash [11]. However, these methods, which require high-pressure conditions, are not optimal for the recovery of vaporized dioxins and have comparatively high operating costs [8]. The melting process of fly ash can easily decompose PCDD/Fs by exposing them to very high temperature (above 1300 °C), but the process requires extremely high energy consumption [12].

Thermal treatment at low temperatures is therefore a promising technology because its energy requirements are much lower [13]. The most common method is to heat fly ash at approximately 300 °C under inert gas conditions to detoxify the dioxins through dechlorination and hydrogenation [14]. Wu found that 99.2% of PCDD/Fs in the HSWI fly ash were removed and desorbed under thermal treatment at 350 °C in a nitrogen atmosphere, and dechlorination and destruction reactions were critical in the removal of PCDD/Fs [15]. Hung indicated that the total PCDD/F destruction efficiency was greater than 90% when the fly ash was pyrolyzed at 300 °C for 1 h [16]. However, longer periods of operation, at least 0.5 h, are the most common limitations of low temperature treatments [15–17]. The reburning of the froths in the combustion chamber of an incinerator might be a feasible procedure for destroying PCDD/Fs, but resource waste is inevitable because PAC is burned concurrently [18]. The exhausted PAC in the froths should be effectively treated because PAC is relatively expensive.

Microwave heat possesses the capabilities of interior heating, rapid heating and selective and volumetric heating, which have notable advantages over conventional heating processes [19–26]. In recent years, a growing number of studies have focused on the applications of microwave processing in the stabilization of heavy metal in industrial sludge as well as the treatment of fly ash using incinerators [27,28]. The heavy metal in industrial sludge could be stabilized with the assistance of microwave absorbing materials such as aluminum powder, iron powder, chitosan, and alumina (α-alumina and γ-alumina). Studies on the microwave sintering of fly ash for heavy metal stabilization have demonstrated reductions in leaching concentrations, which are caused by the solidification of heavy metals into glass ceramics [29]. Chou et al. further evaluated the effects of four different additives (Al_2O_3 powder, c-Al_2O_3, SiO_2, and kaolin) on heavy metal immobilization through pelletization and the subsequent microwave sintering of fly ash [19]. However, few studies have investigated the decomposition of PCDD/Fs in fly ash under a microwave field. Chang et al. verified that the reduction of PCDD/Fs in MSW incinerator fly ash was feasible by using MPO in a H_2SO_4/HNO_3 solution, and found that nearly all PCDD/Fs in fly ash could be reduced in 120 min at 150 °C. However, the MPO process raises problems related to the application of treatment using acid liquid wastewater in additional engineering applications [30].

Carbonaceous materials demonstrate favorable microwave-absorbing characteristics and act as microwave heating accelerators [31]. Activated carbon combined with microwave radiation has been widely used for the removal of organic pollutants such as chloramphenicol, chlorobenzene, and polychlorinated biphenyl in soil [32–38]. Granular activated carbon (GAC) is usually added to the soil as a microwave-absorbing medium to rapidly increase the temperature of the reaction system [33]. Zhang found that Congo red could be degraded fully by increasing the amount of PAC added, because PAC can absorb a large amount of microwave energy [39]. Microwave heating energy was also used effectively for the treatment of volatile organic compounds adsorbed onto GAC [40]. Yuen found that exhausted carbon could be regenerated in addition to destroying the adsorbed organic matter [41]. Liu adopted an integrated granular active carbon (GAC) adsorption/microwave regeneration process for pentachlorophenol (PCP) decomposition [42].

The froths displayed high PAC content, and microwave treatment of the froths did not require an additional microwave-absorbing medium, meaning that the froths could be directly treated with microwave energy. However, decomposing PCDD/Fs in the froths or in incinerator fly ash through microwave heating with carbonaceous materials has not yet been reported in the literature. The objectives of this paper are to evaluate the feasibility of microwave treatment of the froths using PAC as a microwave absorbent and to investigate the destructive effect of PCDD/F congeners in the froths. The effects of microwave incident power and processing time on the decomposition of PCDD/Fs in the froths are also assessed.

2. Materials and methods

2.1. Materials

The fly ash sample used in the study was obtained from a 20 t/d gyration kiln incinerator at a HSWI center in northern China. The incinerator was equipped with a PAC sprayer device and bag filters as air pollution control devices for managing PCDD/Fs. The fresh, dry fly ash sample was collected from the hopper of a bag filter; it therefore contained PAC, which was injected into a flue gas duct before the bag filters. The ash sample was collected and dried at 383 K for 24 h.

Fly ash was treated with column flotation, and the experimental flotation apparatus and operating conditions were the same as those in our previous study [2]. According to our previous report, suitable flotation conditions were 0.05 kg/l of concentrated slurry, 12 kg/t of kerosene dosage, 3 kg/t of frother dosage, and a 0.06 m^3/h air flow rate. After flotation, PCDD/Fs and carbon constituents in fly ash were separated and enriched into a froth product, with chloride simultaneously washed out. The froths were carefully vacuum-filtered, dried, weighed and stored. This was followed by analysis of chemical composition and the carbon content, namely LOI in the fly ash and the froth product, respectively, as shown in Table 1. A PAC sample injected into the air pollution control devices of the incinerator was obtained from the PAC storage tank of the waste incineration plant. The parent PAC is made from a coconut shell, and 96.5% of the PAC particles are less than 106 μm [43].

2.2. Methods of microwave treatment

The froths and the PAC sample were pelleted using a hydraulic press operated for 5 s at a maximum pressure of 50 MPa. Pelletization pretreatment was beneficial for reducing the gap between the particles, increasing the contact area between them and shortening the processing time to reduce energy consumption [44]. A water