Waste Management xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

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

Characterization of heavy metals and PCDD/Fs from water-washing pretreatment and a cement kiln co-processing municipal solid waste incinerator fly ash

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ARTICLE INFO

Article history:
Received 9 October 2017
Revised 2 March 2018
Accepted 3 March 2018
Available online xxxx

Keywords:
Heavy metals
Dioxins
Water washing
Cement kiln co-processing
Fly ash

ABSTRACT

A disposal method for fly ash from a municipal solid waste incinerator (MSWI-FA) that involved a water washing pretreatment and co-processing in a cement kiln was tested. The mass flows of toxic heavy metals (HMs), including volatile HM (Hg), semi-volatile HMs (Pb, Cd, Tl, and As), and low-volatility HMs, and polychlorinated dibenzo-p-dioxin/polychlorinated dibenzofuran (PCDD/Fs) in the input, intermediate, and output materials were characterized. The flue gas Hg concentrations from tests 0, 1, and 2, fed with 0, 3.1, and 1.7 t/h of dried-washed FA (DWFA), were 28.60, 61.95, and 35.40 μg N m^{-3}, respectively. Co-processing of DWFA did not significantly affect the metal concentration in clinker as most of the major input metals, with the exception of Cd, Pb, and Sb (which came from DWFA), were from raw materials and coal. Co-processing of DWFA did not influence on the release of PCDD/Fs; baseline and co-processing values ranged from 0.022 to 0.039 ng-TEQ/N m^3, and from 0.01 to 0.031 ng-TEQ/N m^3, respectively. The total destruction efficiency for PCDD/Fs in MSWI fly was 82.6%. This technology seems to be an environmentally sound option for the disposal of MSWI-FA.

1. Introduction

The amount of municipal solid waste (MSW) generated in China has increased rapidly with economic growth. Over recent years, MSW has increasingly been disposed of by incineration in China, and 40 million tons of MSW were incinerated in 2015, generating approximately 4 million tons of municipal solid waste incinerator fly ash (MSWI-FA) (Big data of MSW dispose, 2016). As it contains various toxic metals and PCDD/Fs and soluble salts that can contaminate soil and groundwater if not disposed of properly, MSWI-FA should be treated as hazardous waste. At present, MSWI-FA is mostly disposed of in MSW landfills in China (Tang et al., 2013), but this is not considered environmentally sound as PCDD/Fs and other organic pollutants cannot be destroyed and toxic metals may leach from the MSWI-FA into the environment around the landfill site (Ham et al., 2008).

The use of solid wastes in cement manufacturing, either as a supplementary fuel or substitute for other raw materials, provides an option for eliminating waste and recovering energy and materials at the same time; this concept has been developed and used for co-processing problematic waste such as MSW, sewage sludge, POPs waste, contaminated soil, and MSWI-FA (Yan et al., 2009; Li et al., 2012a, 2012b, 2012c; Yan et al., 2014; Chen et al., 2016; Liu et al., 2015; Zhao et al., 2017a, 2017b). However, the public is increasingly concerned about whether these processes will release pollutants into the environment or have detrimental effects on the cement quality. The materials contained in MSWI-FA are simultaneously beneficial and detrimental for cement clinker production and concrete use. The beneficial part, which includes CaO, SiO_2, Al_2O_3, and Fe_2O_3, can be used as raw material and typically accounts for less than 50% of the composition. The detrimental substances, including Cl, Na, and K salts, and toxic trace heavy metals (HM) such as Hg, Cd, Pb, As, Cr, and Tl have negative impacts. Large amounts of sodium chloride (NaCl) and potassium chloride (KCl) make the concrete porous and decrease its strength; the deposition of chloride compounds on ducts and induction fans may cause corrosion and clogging in the kiln system (Quina et al., 2008; Wang et al., 2010; Kikuchi, 2001; Pan et al., 2008). During co-processing of MSWI-FA, it is thought that all non-volatile and semi-volatile HMs might be transferred into clinker during clinker formation, which would significantly increase the HM

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https://doi.org/10.1016/j.wasman.2018.03.006
0956-053X/

levels in clinker. To date, the risks of HM contamination in clinker and cement products have not been addressed properly (Huang et al., 2012).

At present, there are two main methods for co-processing MSWI-FA, namely water washing pretreatment and subsequent co-processing and eco-cement technology. The water washing pretreatment and co-processing technology is used to produce ordinary cement clinker in which part of the raw material is substituted with dechlorinated MSWI-FA. There are two steps to the technology. The first step involves a water-washing pretreatment to remove alkali chlorides such as NaCl, KCl, and $CaCl_2$, other soluble salts, and the amphoteric HM in the fly ash, such as Pb and Zn. The HMs are precipitated from washing effluents by reacting with CO_2 or other chemicals and then are reintroduced and mixed with washed MSWI-FA, ready to be fed into the cement kiln. The second step involves co-processing by continuously feeding the dechlorinated MSWI-FA into the flue gas chamber at the kiln inlet to produce ordinary clinker.

The second method, the eco-cement technology, was developed in Japan and uses MSWI-FA and sewage sludge to produce specialized cement called eco-cement, which is defined as having a mass of at least 500 kg or more of MSWI-FA or sewage sludge for every 1 t of clinker produced. The chlorine content of eco-cement is higher than that of ordinary cement and is classified into two types, namely ordinary eco-cement with a Cl less than 0.1% of the cement mass and rapid hardening eco-cement with a Cl content of between 0.5% and 1.5% of the cement mass. Even though it seems a successful product, there are many limitations associated with the use of eco-cement, and situations should be assessed individually to determine the suitability of eco-cement before application (Ecocement JIS R 5214, 2009; Yen et al., 2011).

Previous studies of the effects of co-processing MSWI-FA to clinker have reported that the compression strength and setting time of the clinker produced from MSWI-FA were similar to those of normal clinker and have concluded that the quality might satisfy the requirements for general use (Wang et al., 2010; Kikuchi, 2001; Pan et al., 2008; Yen et al., 2011). In other major studies, the emissions, distribution, and destruction of POPs, i.e. PCDD/Fs, PCBs and PCN (Liu et al., 2015, 2016a, 2016b; Zhao et al., 2017a, 2017b), during co-processing of MSWI fly ash were characterized, and the stack emissions of different pollutants, including PCDD/Fs, PAHs, HMs and conventional pollutants, were examined when alternative fuels and materials were used in cement production (Yang et al., 2012; Conesa et al., 2011, 2008; Zemba et al., 2011; Rivera-Austrui et al., 2014; Ames et al., 2012). In a limited study of the HM content in cement that produced by using alternative fuels and materials, the concentration of some HMs, such as Sb, Cd, Zn, Pb, and Co in the cement increased, because the input materials included some waste (Achternbosch et al., 2005; Gleis, 2003); however, the increases were small and their subsequent impacts on the cement quality have not been examined. To the best of our knowledge, the distribution of HMs and PCDD/Fs in the water washing and co-processing method and the impacts of pretreatment on the destruction of PCDD/Fs and transfer of HMs into cement clinker have not yet been examined.

In this study, toxic HMs, comprising 3 groups, namely volatile HMs (Hg), semi-volatile HMs (Pb, Cd, Tl, and As) and non-volatile HMs, and PCDD/Fs in the input (original MSWI-FA (FA), coal, and raw meal (RM)) (RM refers mixture of raw materials as calcareous, clay material and a small amount of correction materials according to the proportion, grinding to certain fineness), intermediate (washed MSWI-FA (WFA), washing water, HM sludge, dried-washed MSWI-FA (DWFA), and salt), and output materials (clinker (CK), cement kiln dust (CKD) and flue gas (Flue)) produced during pretreatment and co-processing in a cement kiln, were systematically measured. The transfer of HMs and destruction of PCDD/Fs during the pretreatment and co-processing of MSWI-FA are discussed separately. The results obtained in this study might provide important knowledge for the control and reduction of heavy metal and PCDD/Fs in cement kilns co-processing MSWI-FA.

2. Materials and methods

2.1. Characterization of MSWI-FA and pretreated MSWI-FA

MSWI-FA from a municipal solid waste incinerator with grate-based technology and a daily disposal capacity of 1000 tons was used in this field study. The chemical compositions of the original and pretreated MSWI-FAs were analyzed, as shown in Table 1. The major useful components of the original fly ash (FA), which accounted for 49.52% of the total, were CaO, SiO_2, Fe_2O_3, and Al_2O_3, while Cl, Na, and K accounted for 20.32% of the total. After water washing and drying process, the major useful components of the dried washed fly ash (DWFA) accounted for 65.19%, and the detrimental components accounted for 2.36%, of the total content.

2.2. The washing pre-treatment and co-processing system

The system consists of a pre-treatment facility and a preheating–precalcining cement kiln. The plant is equipped with state-of-the-art air pollution control devices, and has a daily clinker production capacity of 2700 t. The pretreatment is designed to remove the NaCl and KCl dissolved salts without extracting the dissolvable HMs contained in the MSWI-FA. The pretreatment process and the cement kiln system are shown in Fig. 1. The process involves three rounds of washing-dewatering to maximize the removal of soluble salts from the FA. After the washing process, the HMs dissolved in washing water are precipitated in the chemical depositing tank, and the clean salt solution on the surface goes through the mechanical vapor recompression (MVR) tower, in which the industrial salt is crystalized and the condensate water is recirculated into the washing tank. The deposited HM sludge is pumped into the 3rd washing tank, and then join with washed FA in the 3rd mechanical drying process to obtain WFA. The WFA is further dried by hot air from the clinker cooler to form DWFA. The DWFA is stored in a silo until it is fed into the flue gas chamber at the inlet of the rotary kiln.

2.3. Sampling scheme

The testing comprised one blank test (test 0) and two tests (tests 1 and 2) under co-processing conditions. Test 0 was conducted on the first day with no DWFA co-processing to determine

Table 1
Composition of the FA, WFA, and DWFA.

Compositions	FA	WFA	DWFA	Salt
Moisture (%)	6.63	37.77	0.72	0.66
LoI (%)	23.00	57.63	23.45	5.84
Ash (%)	79.12	42.34	76.99	95.68
Cl (%)	14.47	0.50	1.04	59.91
Na (%)	2.46	0.32	0.82	33.58
K (%)	3.32	0.21	0.5	0.37
SO_3 (%)	4.64	1.80	3.92	0.11
Al_2O_3 (%)	3.64	2.36	3.88	–
Fe_2O_3 (%)	1.19	0.89	1.2	–
MgO (%)	3.06	1.98	3.19	–
CaO (%)	34.87	29.25	48.84	–
SiO_2 (%)	9.82	6.60	11.27	–

1. – No detectable; 2. LOI and Ash are given in dry-basis, and the other variables are given in wet basis.

Please cite this article in press as: Yan, D., et al. Characterization of heavy metals and PCDD/Fs from water-washing pretreatment and a cement kiln co-processing municipal solid waste incinerator fly ash. Waste Management (2018), https://doi.org/10.1016/j.wasman.2018.03.006