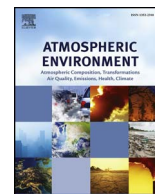




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Mercury release from fly ashes and hydrated fly ash cement pastes

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ABSTRACT

The large-scale usage of fly ash in cement and concrete introduces mercury (Hg) into concrete structures and a risk of secondary emission of Hg from the structures during long-term service was evaluated. Three fly ashes were collected from coal-fired power plants and three blend cements were prepared by mixing Ordinary Portland cement (OPC) with the same amount of fly ash. The releasing behaviors of Hg⁰ from the fly ash and the powdered hydrated cement pastes (HCP) were measured by a self-developed Hg measurement system, where an air-blowing part and Hg collection part were involved. The Hg release of fly ashes at room temperature varied from 25.84 to 39.69 ng/g fly ash during 90-days period of air-blowing experiment. In contrast, the Hg release of the HCPs were in a range of 8.51–18.48 ng/g HCP. It is found that the Hg release ratios of HCPs were almost the same as those of the pure fly ashes, suggesting that the hydration products of the HCP have little immobilization effect on Hg⁰. Increasing temperature and moisture content markedly promote the Hg release.

1. Introduction

Mercury (Hg) is highly toxic and its chemical compounds usually have strong biological toxicities (Pavlish et al., 2003). Hg may cause global air pollutions due to its long-term persistence and long-range mobility (Pavlish et al., 2003; Dastoor and Larocpue, 2004). Moreover Hg contamination is seriously concerned due to its cumulative effect in the body of animals and human being and the strong toxicity to the nervous system and vital organs (Mergler et al., 2007). A large amount of Hg is released to ambient atmosphere every year by fossil fuels combustion (Naylor, 2009). During coal combustion process a part of Hg contained in the raw coal is transferred to fly ash. Hg remaining in the coal-fired flue gas exists in the forms of elemental mercury (Hg⁰), oxidized mercury (Hg²⁺), and particulate-bound mercury (Hg^p) respectively (Pavlish et al., 2003). Hg⁰ is highly volatile and insoluble in water causing global air pollution by long-distance transportation. Hg²⁺ is soluble in water and can be removed by wet flue gas desulfurization (WFGD). Hg^p is collected by the particulate control systems in coal combustion process. The proportion of the three forms of Hg is determined by many factors including coal type, elemental compositions in the coal and fly ash (Laudal et al., 2000; Galbreath et al., 2004), the existing air pollution control devices (APCDs) and Hg sorbent in the power plants (Srivastava et al., 2006; Meij and Winkel,

2006; Pudasainee et al., 2009) etc. As the widespread use of APCDs and Hg sorbent, an increasing amount of Hg is transferred to fly ash.

Cement and concrete are the most important construction materials in the world. Mineral admixtures including fly ash, slag etc are widely used as raw materials in modern cement and concrete production for purposes of consuming a large quantity of industrial wastes, and improving the properties of concrete, particularly durability of concrete, additionally (Berndt, 2009; Zhang et al., 2009). Fly ash is popularly utilized in the production of construction materials and a majority of China's annual output of fly ash is used as a supplementary cementitious material in cement and/or a mineral admixture of concrete. The proper incorporation of fly ash in concrete formulation may effectively reduce the temperature rise of massive concrete by reducing the early hydration heat of concrete and hence mitigate the cracking risk induced by temperature change during hardening of concrete (Bendapudi and Saha, 2011; Uysal and Akyuncu, 2012; Bae et al., 2014). The long-term pozzolanic reaction of fly ash in cementitious system leads to denser micro-structure of concrete and hence improves durability of the resulted concrete (Zeng et al., 2012; Hanehara et al., 2001). However, the large-scale usage of fly ash in cement and concrete introduces a non-negligible amount of heavy metals to concrete structures, such as mercury, lead, arsenic, etc, which poses a risk of releasing such heavy metals to the environment during the long-term service of the concrete

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structures. Nonvolatile heavy metals could be stabilized or immobilized by the cement matrix by precipitation, sorption and substitution reactions (Batchelor, 2006; Chen et al., 2009). Hg is a highly volatile element, the risk of secondary pollution by Hg release from concrete structures is of concern.

It has been demonstrated that temperature, light exposure, Hg concentration in air, air chemistry, Hg concentration in soil have effects on Hg emissions from soils (Gustin et al., 1997, 2002; Zehner and Gustin, 2002; Engle et al., 2001, 2004). The release of mercury from coal combustion byproducts such as fly ash, FGD gypsum, and wall-board made from FGD gypsum were previously tested under both dark and illuminated conditions, with or without the addition of water to the samples by N.J. Pekney (Pekney et al., 2009). Previous study has also shown that detectable amounts of Hg were constantly emitted from flue gas desulfurization gypsum samples (Zhu et al., 2017). Considering the vast amount of fly ash in the world, systematic study on the Hg release from fly ash is needed. When fly ash is used in cementitious materials, it is well known that due to the cement hydration and the pozzolanic reaction of fly ash with hydration product of cement, heavy metals may be solidified (Batchelor, 2006; Chen et al., 2009). However, as Hg is a highly volatile heavy metal, whether Hg can be stabilized by the cement hydration product is a question to consider. Closer look is needed at the Hg release behavior from hydrated fly ash cement pastes.

The aim of this study lies on experimentally disclosing Hg release from fly ash as well as hydrated fly ash cement pastes using laboratory techniques. Three fly ash samples were selected from coal-fired power plants and were mixed with ordinary Portland cement (OPC) in a fixed proportion. Hg release behaviors from the fly ash samples and from the powdered hydrated cement pastes (HCPs) were measured by self-developed experimental system which includes a solid phase mercury analyzer. The effects of temperature, moisture content in the powdered samples were investigated. The Hg release amounts from fly ashes and HCPs of fly ash cement were tested. The physico-chemical solidification processes affecting the Hg release were discussed.

2. Materials and methods

2.1. Materials

Samples of fly ash (FA) were obtained from three different coal-fired power plants in China, labeled as FA1, FA2 and FA3. All of the fly ash samples were dry ash. Ordinary Portland cement (OPC) classified by P.I.42.5, complying with the Chinese National Standard GB8076-1997 was used to prepare fly ash cement paste samples. Average chemical and mineralogical compositions of Ordinary Portland cement are listed in Table 1. The chemical compositions of the fly ash samples are shown in Table 2. As shown in Table 2, considerable amounts of SiO₂, Al₂O₃, CaO were detected in the fly ash samples.

Table 1
Chemical compositions of the Ordinary Portland cement.

Composition	Concentration (%)	Standard deviation
SiO ₂	20.60	0.03
Al ₂ O ₃	4.61	0.02
Fe ₂ O ₃	3.23	0.02
CaO	62.59	0.04
MgO	2.61	0.03
SO ₃	2.92	0.01
R ₂ O	0.53	0.01
fCaO	0.75	0.07
LOI	2.04	0.02
Cl	0.011	0.001
C ₃ S	54.23	0.26
C ₂ S	18.23	0.28
C ₃ A	6.76	0.02
C ₄ AF	9.82	0.06

Table 2
Chemical compositions of the fly ash samples.

	FA1 (wt.%)	FA2 (wt.%)	FA3 (wt.%)
SiO ₂	47.00	49.53	49.87
Al ₂ O ₃	39.87	25.92	24.93
CaO	5.03	8.95	9.73
Fe ₂ O ₃	3.00	8.78	8.33
TiO ₂	1.64	1.73	1.76
K ₂ O	0.77	1.39	1.53
SO ₃	0.71	0.71	1.15
MgO	0.63	0.71	0.87
Na ₂ O	0.37	0.74	0.56
P ₂ O ₅	0.37	0.19	0.28
MnO	0.05	0.17	0.19
BaO	0.04	0.18	0.10
SrO	0.20	0.53	0.39
ZrO ₂	0.13	0.10	0.10
ZnO	0.02	0.04	0.04
PbO	0.02	0.03	0.02
Ga ₂ O ₃	0.01	0.01	0.02
CuO	0.01	0.02	0.02
Cr ₂ O ₃	0.01	0.04	0.03
NiO	0.01	0.03	0.02
Y ₂ O ₃	0.01	0.01	0.01
SUM	99.90	99.81	99.95

2.2. Sample preparation

The fly ash cement (FAC) pastes were prepared by mixing OPC, fly ash and deionized water. The mass ratio of fly ash to OPC was 1.0 and water to the blend cement ratio was 0.4. Mixing procedure complied with ASTM C1738-11a. The fresh cement pastes were filled in standard moulds with dimension of 40 mm × 40 mm × 160 mm. The filled moulds were kept for setting for 24 h in sealed condition by plastic membrane and the hardened samples were then demoulded. The hydrated FAC, which is called here hydrated cement paste (HCP) was obtained by curing the FAC pastes at 20 ± 2 °C and 95% relative humidity for 28 days. After curing, the HCPs were air dried for two days. The dried samples of HCPs were triturated into fine powders labeled as HCP1, HCP2 and HCP3 for the following Hg release measurements. Hydrated OPC paste was made by mixing deionized water and OPC in the weight ratio of 0.4 following the same procedure and the powdered hydrated OPC paste was labelled as HCP0.

2.3. Characterization methods

The Hg concentrations in the fly ash (FA) samples, OPC and HCP samples were quantified by Lumex RA915M + PYRO915 (Lumex, Russia) which was calibrated with the Standard Reference Material 1632d (provided by the U.S. National Institute of Standards and Technology) within a ± 5% error range. Lumex 915M can directly measure Hg concentration in solid matrix. The concentrations of other elements in the samples were analyzed by X-ray Fluorescence Spectrometer (XRF-1800, Shimadzu, Japan). The surface area of the samples was measured using the Brunauer-Emmett-Teller (BET) method at 150 °C. X-ray diffraction (XRD) was performed to determine the crystalline phases in the air-dried samples and the experiments were carried out with a diffractometer (Bruker D8, Karlsruhe, Germany) using Cu K α radiation. The microstructure and morphology of each sample were analyzed by scanning electron microscopy (SEM Merlin, ZEISS, Germany).

2.4. Hg releasing measurement

The Hg release from FA samples and HCP samples was tested in the self-developed Hg measurement system shown in Fig. 1. 20 g of sample was placed in the bottom of a cubic quartz glass chamber. Ambient dry

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