

Mercury Emissions During Steam-Curing of Cellular Concretes That Contain Fly Ash and Mercury-Loaded Powdered Activated Carbon

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ABSTRACT

To evaluate possible mercury emissions during steam curing of cellular concretes containing fly ash and mercury-loaded PAC, we collected and measured gaseous mercury released during curing for concretes that contained different quantities of fly ash and mercury-loaded PAC (HgPAC). All experiments were conducted in a laboratory and were configured to enable estimation of an upper limit on the potential gaseous mercury release from steam-curing concretes that contain fly ash and mercury sorbent material. The observed emissions of mercury for the complete curing process generally increased with rising mercury concentrations in the concrete, where fly ash and fly ash-HgPAC by-products were the principal sources of mercury. However, release of mercury from concretes that contain these byproducts was less than 0.022% of the total quantity of mercury present in the concrete from all sources. Therefore, the steam-curing of cellular concrete containing fly ash and mercury-laden PAC does not result in a significant release of the mercury captured following coal combustion.

INTRODUCTION

Approximately 48 tons of mercury emitted into the atmosphere annually come from coal-fired utility boilers. To reduce mercury emissions from these boilers, injection of powdered activated carbon (PAC) into hot flue gas streams and subsequent collection of the PAC along with fly ash is being developed. If this PAC technology is implemented, the total mercury content in the final coal combustion by-product will increase. Therefore, an understanding of the fate of mercury in re-use applications of fly ash concrete is needed to support future applications of the new fly ash-sorbent by-products.

One possible application of fly ash containing PAC is use in the production of steam-cured cellular concrete. Unlike conventional Portland cement concrete, the incorporation of fly ash containing organic carbon into cellular concretes is less problematic since the porosity of the concrete is obtained through the generation of hydrogen gas created by chemical reactions of aluminum powder in the mix. Also, instead of replacing cement, the fly ash is substituted for the fine sand component of the mix when making cellular concrete. In fact, 100% of the sand can be replaced with fly ash and as a result fly ash may represent a greater fraction of the concrete.

Given the high levels of fly ash that can be incorporated into cellular concrete, the potential for utilizing mercury-laden PAC, as well as the elevated temperatures during curing, it is important to examine the potential release of mercury from these materials. To this end, gaseous mercury emissions from cellular concretes containing fly ash and mercury-laden PAC were measured during steam curing.

METHODOLOGY

Mercury emissions from 3 fly ash cellular concrete mixes were examined including (1) a mix for which fly ash replaced 50% of the sand, (2) a mix in which fly ash replaced 100% of the sand, and (3) a 50% mix which also contained mercury-laden PAC. In addition, mercury emissions from the sampling container were collected as a “blank” and emissions from a cellular concrete not containing fly ash served as a measure of the release of mercury from other components in the mix. A single class F fly ash was used in this work. A detailed description of the methodology can be found elsewhere.¹

The cellular concrete mixtures contained Type III portland cement (Holcim), Chardon 110™ silica sand (100 to 270 mesh, 99.5% silica, Fairmount Minerals), pulverized burnt lime containing a silica flow aid (Carmeuse), coal fly ash (Class F, Headwaters Resources) and 0.6 μm aluminum powder (99.9% pure, Aldrich). Powdered activated carbon (PAC), DARCO-FGD™, used in select experiments, was obtained from Norit Americas. PAC was loaded with mercury by placing PAC in a closed gas desiccator containing mercury-saturated air obtained by heating metallic mercury. Cellular concrete ingredients were mixed and then placed in a rectangular mold within an inert sampling container.

Curing of cellular concrete samples was carried out in four steps: a) presteaming, b) temperature rise, c) maximum-temperature hold, and d) soaking. For all stages, iodated carbon (IC) traps were placed on the inlet of the sampling chamber to provide a source of mercury-free air or steam. An IC trap on the outlet of the sampling chamber collected mercury emissions from the curing concrete samples. During the temperature rise, hold and soak, the sample chamber was placed in a carbon-steel autoclave. Steam was supplied using microprocessor-controlled electric boiler at 80 psig. Temperature during steam curing varied from 21 to 80 °C. The total time of curing over which mercury emissions were sampled was approximately 21 hours for each experiment. Mercury emissions from each cellular concrete mix were determined in triplicate.

Mercury-free air passed over curing concrete samples using air sampling pumps at a flow rate of 0.350 L/min. Mercury emissions from the curing concrete samples were collected using IC traps. IC traps were heated using silicone-insulated heating tape to prevent condensation. After curing, IC traps were analyzed for mercury by cold vapor atomic fluorescence spectroscopy (CVAFS) by Studio Geochemica (Seattle Washington).

RESULTS AND DISCUSSION

Table 1 shows the mercury content of the different cellular concrete mixtures. Mercury content was determined by summing the mercury levels in the various concrete ingredients as determined by CVAFS. All concrete ingredients, including cement, sand, lime, aluminum, and fly ash contributed mercury to the mix, though fly ash (and mercury loaded PAC for HgPAC) contributed the majority of mercury to the concrete. The FA0 mix had the lowest mercury content and mercury content increased with increasing fly ash content. HgPAC had a higher mercury content than FA50 due to the addition of mercury-loaded PAC, but the mercury content of HgPAC was not as high as FA100.

Mercury collected from all the curing concrete samples during the pre-steaming phase was below the limit of detection. During steam curing, average mercury release varied from 1.9 ± 0.4 to 5.8 ± 1.3 ng/kg for cellular concrete containing fly ash and/or mercury-loaded PAC. The FA0, which did not contain fly ash or mercury-loaded PAC, had the lowest mass mercury release. The mass of mercury released generally increased with increasing fly ash content with the FA50 having the lowest mass release and FA100 having the highest. The mercury release from the HgPAC, which contained 50% fly ash plus mercury-loaded PAC, was greater than the release from FA50.

Table 1. Mercury content of cellular concrete and release during steam curing.

Concrete	Mercury Content		Hg Release	
	Total Hg (μg)	Hg Conc. ($\mu\text{g}/\text{kg}$)	ng/kg	%
FA0	27	1.4	0.4 ± 0.6	0.03 ± 0.05
FA50	283	14.2	1.9 ± 0.4	0.013 ± 0.003
HgPAC	468	23.4	2.5 ± 1.1	0.011 ± 0.005
FA100	538	26.9	5.8 ± 1.3	0.022 ± 0.005

Examining mercury release on a percent basis demonstrates that HgPAC and FA50 had the lowest percent releases of mercury. Interestingly, the FA0 had the highest percent release of mercury. However, release of mercury from all concretes containing these byproducts was less than 0.022% of the total quantity of mercury present in the concrete from all sources. Therefore, the steam-curing of cellular concrete containing fly ash and mercury-laden PAC does not result in a significant release of the mercury captured following coal combustion.

Extended sampling of mercury release from a single batch of FA100 was conducted under ambient laboratory temperatures for a period of 10 days following steam curing. During this extended sampling interval the calculated mercury release was only 0.073 ng/kg (or 0.005% of the initial mercury content). Therefore, the release over this extended time period was an order of magnitude less than the release observed during steam curing, indicating that the majority of the low mercury release occurred during the steam curing interval.

REFERENCES

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