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Effect of coal combustion fly ash use in concrete on the mass transport release of constituents of potential concern

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HIGHLIGHTS

- First LEAF leaching study on US sources of concrete materials containing fly ash.
- Mass transport leaching from concrete and microconcrete with and without fly ash.
- Cumulative release dependent on liquid–solid partitioning concentration.
- Microconcretes (no coarse aggregate) can be concrete surrogates for leaching.
- Fly ash replacement causes minimal to no increases in leaching from monoliths.

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ABSTRACT

Concerns about the environmental safety of coal combustion fly ash use as a supplemental cementitious material have necessitated comprehensive evaluation of the potential for leaching concrete materials containing fly ash used as a cement replacement. Using concrete formulations representative of US residential and commercial applications, test monoliths were made without fly ash replacement (i.e., controls) and with 20% or 45% of the portland cement fraction replaced by fly ash from four coal combustion sources. In addition, microconcrete materials were created with 45% fly ash replacement based on the commercial concrete formulation but with no coarse aggregate and an increased fine aggregate fraction to maintain aggregate-paste interfacial area. All materials were cured for 3 months prior to mass transport-based leach testing of constituents of potential concern (i.e., Sb, As, B, Ba, Cd, Cr, Mo, Pb, Se, Tl and V) according to EPA Method 1315. The cumulative release results were consistent with previously tested samples of concretes and mortars from international sources. Of the 11 constituents tested, only Sb, Ba, B, Cr and V were measured in quantifiable amounts. Microconcretes without coarse aggregate were determined to be conservative surrogates for concrete in leaching assessment since cumulative release from microconcretes were only slightly greater than the associated concrete materials. Relative to control materials without fly ash, concretes and microconcretes with fly ash replacement of cement had increased 28-d and 63-d cumulative release for a limited number 10 comparison cases: 2 cases for Sb, 7 cases for Ba and 1 case for Cr. The overall results suggest minimal leaching impact from fly ash use as a replacement for up to 45% of the cement fraction in typical US concrete formulations; however, scenario-specific assessment based on this leaching evaluation should be used to determine if potential environmental impacts exist.

1. Introduction

On an annual basis, the United States (US) produces approximately 180 million cubic meters of ready mix concrete (PCA, 2013) with about 50% utilizing coal combustion fly ash as a supplemental cementitious material (Obla, 2008). Concrete materials incorporating fly ash exhibit improved handling properties as well as higher long-term strength and durability than concretes made with portland cement alone (Liu et al., 2011; Obla, 2008; Poon et al., 2000; Duran-Herrera et al., 2011). Fly ash may replace 15–40% of the portland cement fraction in Type IP cements used in read mix formulations, with higher replacement levels designed for specific applications (ACI, 1993, 2003; Poon et al., 2000).

The fly ash disposal alternatives proposed by US Environmental Protection Agency (US EPA; Federal Register, 2010) are expected to have an impact on the beneficial uses of fly ash in commercial applications. Regulatory uncertainty surrounding the disposal rule

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and the perceived effects of proposed regulation on beneficial use of fly has been attributed as a cause of the steady, but not increasing, percentage of annual fly ash used in the cement industry (Ward, 2013). The potential for impact to the beneficial use market has increased the urgency to evaluate the potential environmental impacts of fly ash used in the concrete industry. Central to this evaluation, is the understanding of the leaching behavior of those constituents of potential concern (COPCs) in fly ash identified in disposal risk evaluations and through characterization of fly ash from a wide range of sources (US EPA, 2006; Kosson et al., 2009): antimony (Sb), arsenic (As), cadmium (Cd), chromium (Cr), lead (Pb), molybdenum (Mo), selenium (Se), thallium (Tl) and vanadium (V).

Environmental assessment of concrete and cement-based materials has previously relied on the results of the Toxicity Characteristic Leaching Procedure (TCLP; EPA Method 1311) as the basis for leaching prediction (Cheng et al., 2008; Eckert and Guo, 1998; Kanare and West, 1993; PCA, 1992; Zhang et al., 2001a,b). While these studies indicate concrete and other cement-based materials are not classified as hazardous based on leaching criteria (i.e., leaching concentrations of RCRA metals are below regulatory levels), the impacts of the detectable concentrations of potentially hazardous COPCs from use of coal fly ash have been not been explored. More recently, the US EPA conducted a review available leaching data foQuantitation and detection limit values are carried r cementitious materials with and without fly ash replacement from primarily European sources (van der Sloot et al., 2012) and noted minimal impact for most species during use of fly ash-blended cements containing up to 35% coal combustion fly ash. The report cautioned that these observations were considered only indicative due to the identification of several gaps in the assembled data and underlying studies. The Electric Power Research Institute (EPRI), with partial support from the US EPA, has initiated a research project specifically to address the gaps identified in the US EPA review.

The EPRI program is the first study to date that provides comprehensive testing of the equilibrium- and mass transport-based leaching from cementitious materials representative of actual US concrete formulations containing fly ash from a range of commercial sources. The program includes leaching characterization according to new US EPA testing methods with direct comparison testing results between material component sources (e.g., fly ash, cement, aggregates), control materials made without fly ash, and fly ash blended concrete materials. In Kosson et al. (2013), the pH-dependent leaching of concrete and microconcrete materials utilizing up to 45% replacement of cement with fly ash from four US coal combustion facilities showed that the liquid–solid partitioning (LSP) as a function of pH was controlled by the hydrated cement chemistry such that only limited differences in pH-dependent leaching between concrete and microconcrete materials made with and without fly ash were observed. However, the study focused on the chemical effects of leaching through aqueous partitioning of COPCs from concrete using size reduced material and approaching liquid–solid chemical equilibrium and did not address the rate of COPC release based on the physical nature of the concrete material.

The current study investigates the impact of fly ash replacement for cement in the same materials through characterization of COPC transport from monolithic test specimens using EPA Method 1315. The objectives of this study are to (i) evaluate the impact of the use of coal combustion fly ash as a partial cement replacement for portland cement in concrete on the rate of COPC leaching from monolithic samples, (ii) compare the results of mass transport-based testing of fly ash concrete from US formulations and sources with the results of the primarily European material in the US EPA review (van der Sloot et al., 2012), and (iii) evaluate the suitability of using microconcrete as a surrogate for concrete in mass transport rate leaching testing.

2. Mass transport-based leaching and environmental assessment approaches

An overview of leaching processes and environmental assessment methodology has been provided in Kosson et al. (2002, 2013). Mass transport, the combined result of diffusion through a tortuous pore network with aqueous partitioning at the solid–liquid interface, is the primary mechanism of constituent leaching from monolithic materials (e.g., concrete and compacted soils). The driving force for mass transport is the gradient in concentration (or thermodynamic activity) between the bulk contacting solution and the pore solution at the core of the monolith. Within the pore structure, local aqueous concentrations are controlled by the same interfacial and chemical mechanisms that dominate at equilibrium (e.g., dissolution/precipitation, adsorption/desorption, complexation, interaction with dissolved organic carbon). For the purposes of leaching evaluation, these mechanisms may be approximated by the LSP as functions of porewater pH (as presented for the current project in Kosson et al., 2013) and liquid–solid ratio L/S.

2.1. Toxicity characteristic leaching procedure

Several studies have used TCLP as the basis, or a partial basis, for determining the leaching of COPCs from concretes (Cheng et al., 2008; Eckert and Guo, 1998; Kanare and West, 1993; PCA, 1992; Zhang et al., 2001a,b). TCLP is a single-batch leaching test intended to provide a leachate representative of leaching under the conditions co-disposal in a municipal solid waste landfill. However, the US EPA Science Advisory Board (SAB, 1991, 1999) and others (Eckert and Guo, 1998; Kosson et al., 2002; Thorneloe et al., 2010) have cautioned that TCLP (i) provides little relevant information for concrete assessment because the test conditions are not applicable to highly alkaline monolithic materials, (ii) the municipal solid waste landfill scenario simulated by the TCLP test condition is not indicative of actual use conditions, and (iii) single-batch tests performed on size-reduced materials do not account for the monolithic nature of concrete materials. Thus, evaluating the impacts of the fly ash source and usage rate on COPC leaching should be based on leaching approaches that provide a more fundamental understanding of the release mechanisms dominant when concrete materials are used.

2.2. Leaching environmental assessment framework

The Leaching Environmental Assessment Framework (LEAF) was developed to provide a robust approach to environmental assessment through characterization of leaching behavior of a solid material (e.g., soils, concretes, process wastes, etc.) while considering a range of potential use and disposal scenarios. The leaching methods within LEAF have been thoroughly documented (Garrabrants et al., 2010, 2012a,b; Kosson et al., 2002) and recently included in SW-846, the US EPA compendium of laboratory methods (US EPA, 2013). These procedures characterize a suite of fundamental leaching properties including (i) LSP as a function of eluate pH (EPA Method 1313), (ii) LSP as a function of L/S using an upflow percolation column (EPA Method 1314) or parallel batch extractions (EPA Method 1316), and (iii) mass transport rates from monolithic and compacted granular materials (EPA Method 1315). The LEAF methods are appropriate for beneficial use evaluations in that method selection is based on material properties, fundamental leaching mechanisms, and the conditions of the anticipated utilization or disposal scenario. For concrete materials, the most applicable LEAF tests include Method 1313 to measure partitioning between solid and liquid phases and Method 1315 to provide the