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Stabilization of fly ash using cementing bacteria. Assessment of cementation and trace element mobilization

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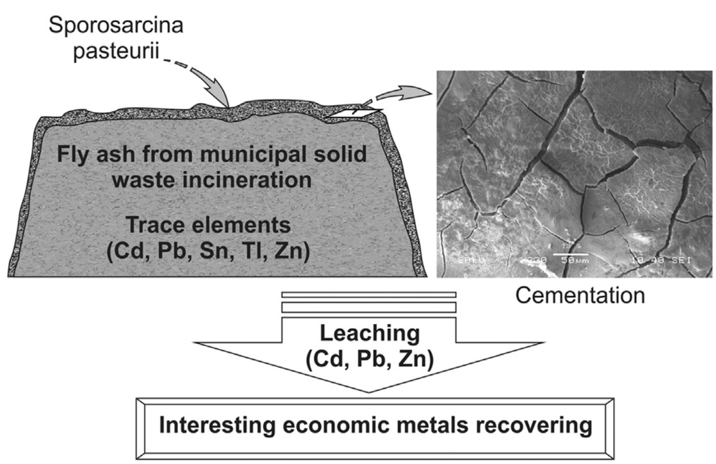
HIGHLIGHTS

- Fly ash from municipal solid waste incineration has a high trace element content.
- Cd, Pb, and Zn are primarily associated with carbonates and mobile under acid pH.
- Microbial treatment promotes the cementation and stabilization of fly ash.
- The treatment enhances the leachability of metals.
- It can be applied to recover metals of economic interest.

GRAPHICAL ABSTRACT



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ABSTRACT

Fly ash from municipal solid waste incineration (MSWI) was treated with microorganisms (*Sporosarcina pasteurii* and *Myxococcus xanthus*) to assess their capacity for cementing this waste material. Leaching tests on the samples treated with bacteria were also performed to assess the possibility of recovering and recycling trace elements from the fly ash. Sequential extractions combined with mineralogical studies demonstrated that Pb is mobile in water when associated with portlandite. Also, Cd, Pb, and Zn are primarily associated with carbonates and are mobile in acidic environments (up to 4.8, 13.9 and 248 mg/l of Cd, Pb and Zn, respectively, extracted with acetic acid). Microbial treatment of the fly ash, especially with *Sporosarcina pasteurii*, led to its cementation and stabilization, preventing its dispersion into the environment. But samples treated with bacteria exhibited a higher capacity for trace element leaching than did untreated fly ash. The ability of these bacteria to mobilize metals can be applied to recover those of economic interest. The use of low cost biotechnologies can be an alternative to chemical treatments currently utilized for the recovery and reuse of these wastes.

1. Introduction and objectives

The primary sources of fly ash are coal combustion power plants, biomass combustion, and incineration of municipal solid waste [1].

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The generation and management of municipal solid waste (MSW) is a topic of great interest due to the large amount of waste produced and the limited space available in landfills for deposition. MSW incineration (MSWI) is one of the most effective management techniques because it reduces the volume and mass of waste by more than 70%–90%. However, two basic types of solid wastes are generated during this process: bottom ash, or slag, and fly ash. Due to high trace element content, these types of wastes are classified as hazardous wastes.

Fly ash is typically stored in landfills, but due to the environmental problems resulting from accumulation [2], they are being reused in multiple industrial sectors. In recent years, the reuse of fly ash has been focused in four main recovery routes: cement/concrete, ceramic/glass, paving, and absorbents. For all uses it is necessary to treat materials to reduce the total concentration of pollutants and/or stabilize them to diminish leaching of contaminants. The primary treatments consist of separation processes, stabilization/solidification, and heat treatment [1,3–6]. In most cases stabilization of some heavy metals has been obtained at laboratory scales; however, the costs are often too high for practical implementation.

In addition to these treatments, new technologies using microorganisms, primarily bacteria, have been developed in recent decades for the decontamination of water and soil [7]. This process is known as *bioremediation* [8–10]. Microorganisms capable of this process can alter physical and chemical characteristics, thus changing their mobility [11]. Bioremediation has been effective for wastewater treatment [12].

Bioleaching is another potential option that is a natural dissolution process carried out by bacteria, such as *Thiobacillus ferrooxidans*, *Leptospirillum ferrooxidans*, *Acidithiobacillus ferrooxidans*. This is used to recover heavy metals from sludge drying, such as Zn, Cu, Ni, Pb, Cd, and Cr. Bioleaching has been shown to be more effective for metal recovery than chemical leaching, but it is more time intensive [13].

In addition to trace element leaching, the particles are very small and easily suspended, which can lead to air pollution. This can lead to adverse health consequences depending on the concentration and duration of exposure. In recent decades, cementing bacteria have been used for engineering, soil stabilization, and heritage restoration through injection of ureolytic bacteria [14–18]. Therefore, we hypothesized that this methodology could also be successfully employed for fly ash; previous geochemical and mineralogical studies have been conducted to assess the risks from trace element leaching and fly ash mobility [19].

The objectives of this study were a) to assess the hazards derived from MSWI fly ash and b) to perform cementation and bioleaching tests with bacteria, stabilize the fly ash, and study the possibility of metal recovery.

2. Materials and methods

Two samples of different MSWI fly ash were studied (MO and COR). The MO sample is passed through an electrofilter and a reactor with sodium bicarbonate and activated carbon at the MSWI plant. The COR sample is subject, in addition, to pre-treatment with lime and active carbon. To stabilize these raw fly ash samples, two types of non-pathogenic ureolytic bacteria were used: *Sporosarcina pasteurii* and *Myxococcus xantus*.

2.1. Characterization of fly ash

The particle size distribution of fly ash was analyzed with a Malvern Instruments Ltd. Mastersizer 2000 UK. pH was measured in a solid:liquid mixture 1:100 after stirring for 1 h with a micropH 2002 pH meter (Crisson) calibrated with pH 4 and 7 solutions.

The mineralogy was determined by X-ray diffraction (XRD) with a Bruker D8 Advance diffractometer with Cu-Ka radiation at 40 kV and 30 mA at the central research facilities of the University of Seville (CITIUS, http://investigacion.us.es/scisi/sgi/servicios/area-de-rayosx). The samples were prepared on side-loaded sample holders to guarantee random powders and were scanned from 3 to 70° 2θ at a rate of 0.03° 2θ step intervals and 1-s counting time per step. The assessment of mineral content was estimated with DIFFRACplus software and the powder diffraction file (Pdf2) from the International Centre for Diffraction data (ICDD).

In order to complete the mineralogical study, samples were observed by scanning electron microscopy (SEM) with a JEOL 6460LV microscope equipped with a secondary electron detector (SE), backscattered electron detector (BSE), and microanalysis INCA X-sight by energy dispersive X-ray spectroscopy (EDS).

Chemical analyses of major (i.e., Al, Ca, Cl, F, Fe, K, Mg, Mn, Na, O, P, S, Si, Ti) and trace elements (i.e., As, Cd, Cr, Cu, Ni, Pb, Sn, Tl, Zn) were performed by X-ray fluorescence (XRF) with an Axios-Panalytical-X-Ray fluorescence spectrometer at CITIUS. The samples were prepared in pressed powder pellets using wax as an agglomerating agent. Quality control of the analyses was carried out by calibration curves with several standards. The errors for of major elements were below 10% and the results were compared against the Fischer pattern 603–683. The results of trace elements were compared against several certified materials, such as GEOPT 22, 24, 26, 28 (http://www.geoanalyst.org/). (The detection and quantification limits and relative error of trace elements are detailed in Table 1).

2.2. Mobility tests

Sequential extractions assays were performed by the BCR method, which was modified according to the sample characteristics [20]. In the first extraction, the mobility of water soluble compounds was studied (solid-liquid ratio 1:100). In the second extraction, the labile fraction associated with carbonates was studied by attacking the residue of the first step with acetic acid at pH 3 with the same solid-liquid ratio. Both extractions were obtained via 1 h of agitation. They were then centrifuged at 10,000 rpm for 10 min to separate the slurry phase. The supernatant was filtered at 0.2 mμ and acidified with 2% HNO3 for conservation. Chemical analyses of the most abundant and dangerous trace elements (according to the US Environmental Protection Agency) were performed by inductively coupled plasm optical emission spectroscopy (ICP-OES) at the CITIUS. Calibration curves were constructed with patterns for each element. The relative standard deviation (RSD) was below 10%.

After each extraction, a sample of the residue was analyzed by XRD to control dissolved mineral phases during the attack. The assessment of the dissolved mineral phases was performed by differential XRD (DRXD) by subtracting the diffraction patterns of the treated samples from the untreated samples [21,22].

2.3. Bacteria cementation assays

Fly ash was treated with non-pathogenic bacteria. Both bacteria were cultivated in Tryptic Soy Broth (TSB) and 2% agar. In addition, 2% urea was added for *Sporosarcina pasteurii*, which requires an ureolytic environment for survival. The pH was kept above 8 with NaOH. Bacteria were collected with an inoculation loop and a culture broth was prepared in TSB.

The cementitious solution for *Sporosarcina pasteurii* contained NH_4Cl (150 mM), $NaHCO_3$ (25 mM), and $CaCl_2$ (50 mM). The solution for *Myxococcus xanthus* contained NH_4Cl (50 mM), $NaHCO_3$