

Geochemical modeling of contaminant transport from recycled roadbed material using recycled boiler fly ash

University of Miyazaki, Student Member, SANTIAGO J. Rodolfo
University of Miyazaki, Member, Tomoo SEKITO and Yutaka DOTE

1. Introduction

In recent years due to environmental concern and limited landfill space, several studies have focused on the potential recycling of waste materials such as incinerated ashes as a replacement of natural aggregates in civil engineering applications, in particular for road construction [5]. The major concern related to the recycling of these materials is the potential release of contaminants through leaching in the long term caused by water contact.

In this research, a multidisciplinary approach combining mineralogical analysis (X-ray diffraction and XRF) with chemical analysis, column leaching test, and pH_{stat} leaching tests was applied for the prediction of the speciation and mobility of potentially hazardous substances that may be released to the environment from the leaching of recycled ash produced from a paper mill company.

2. Methods

2.1 Recycled Boiler Fly ash sample

The Boiler Fly Ash (BFA) sample was generated from a paper mill plant that utilizes waste tires in combination with wood waste for energy production through incineration. The BFA was then used to produce the recycled material by mixing cement and sand. The mixing ratio of the sample is described in Table 1.

2.2 Recycled BFA chemical and mineralogical composition

The chemical composition of the BFA recycled material was analyzed by XRF (Shimadzu model EDX-720). To identify the major minerals present in the sample, the X-ray diffraction pattern was measured using a PANalytical X'pert PRO diffractometer.

2.3 Modified column leaching tests

The column leaching tests were performed largely according to ISO/TS 2126-8-3 with some modifications in regards to flow rate to satisfy the needs of this study [4]. Dried recycled BFA was packed in 3 acrylic columns with a height of 30cm and 5cm diameter equipped with glass wool and glass beads at the inlet and wool glass only at the outlet. The average height and weight of the packed sample were ± 20 cm and around 300g respectively. An initial eluate solution of pH 3 was pumped in an up-flow direction with an initial flow rate of 12mL/hr. The same conditions were set for another set of 3 columns using distilled water as the eluate solution. Fractions were collected at a cumulative liquid-to-solid ratio (L/S) values from 0.1 to 100. Effluent fractions were collected in PE bottles sealed to prevent carbonation processes. To evaluate further changes in the pH and element concentration, at L/S 100 the flow of the columns was gradually increased from 12mL/hr to 16 mL/h

Table 1. Composition of recycled BFA

Solid content mass ratio (%)			Moisture ratio (%)
BFA	cement	sand	
30	30	40	48.6

Table 2. Chemical composition of recycled BFA

Component	Content (%)
CaO	40.3
SiO ₂	28.0
Al ₂ O ₃	10.3
SO ₃	9.93
Fe ₂ O ₃	4.07
ZnO	2.90
K ₂ O	1.71
MgO	1.58

[4] and the pH of the eluate solution was lowered to pH 2 for both sets of columns. Shortly after collection of each effluent fractions, pH was measured and sub-samples were taken and filtered through a 0.45 μ m membrane filter and stored for element concentration analysis, by Atomic Adsorption Spectrophotometer (AAS) (Ca, Na, K, Mg, Pb, Zn, As, Cu, Cd, Cr, Ni), Ion chromatography (PO₄ and SO₄) and Total Organic Carbon.

2.4 pH_{stat} Leaching test

To evaluate the solubility of major element components and heavy metals as a function of pH, pH_{stat} leaching test were performed on the recycled BFA samples at a L/S ratio of 10 using 20g of sample and 200mL of distilled water, constantly stirred at specific pH values (pH 2, 4, 6, 8, 10, 12), which were kept constant for a duration of 24hrs using an automatic titration system (902 Metrohm). Solutions of HNO₃ and NaOH were used to maintain the pH of the suspensions. At the end of each pH_{stat} test, solutions were filtered by a 0.45 μ m membrane filter and cations (Ca, K, Na, Mg) and trace metals (Pb, Zn, As, Cu, Cd, T-Cr, Ni) were measured by AAS.

2.5 Total content analysis

To determine the total content of trace metals in the samples, the sample was digested by an acid solution mixture of HCl and HNO₃ (3:1). The concentration of trace metals (Pb, Zn, As, Cu, Cd, T-Cr, Ni) was measured by AAS.

3. Results and discussions

3.1 Chemical and mineralogical composition of recycled BFA

The chemical composition of the recycled BFA sample in the oxide form obtained from XRF analysis is given in Table 2. The main components were CaO (40.3%), silica (28.0%) and Al₂O₃ (10.25%), these results contribute to the high alkalinity of the sample (pH 12.6) due to the enrichment of calcium silicate hydrates [1].

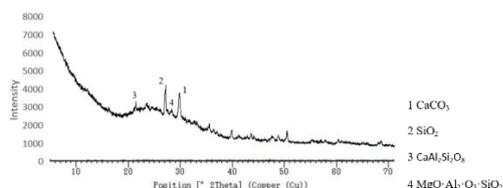


Figure 2. XRD analysis pattern and identified mineral phases

As for the mineralogy of the recycled BFA, candidate phases were assigned by XRD. It was found that quartz (SiO_2) and calcites (CaCO_3) are the main identifiable crystallized phases (Fig. 2) followed by calcium aluminum silicates ($\text{CaAl}_2\text{Si}_2\text{O}_8$) and magnesium aluminum silicates ($\text{MgO}\cdot\text{Al}_2\text{O}_3\cdot\text{SiO}_2$) to a lesser extent but still able to be identified. Based on similar studies with MSWI bottom ash as the material of study, in regards to the crystallized phases (calcite, quartz, ettringite, and anhydrate) there is a great similarity with the mineralogy composition of the recycled BFA, as described by other authors [3]. For the recycled BFA there are also amorphous shape minerals that could not be identified but based on the literature review could also be present in the sample due to the origin of the sample [3].

3.2 Column leaching tests

Figure 3 shows the pH values as a function of the applied L/S obtained from the two sets of columns. At 100 of L/S, the pH values are still high with around 11.5, due to the high buffer capacity of the components with Ca and Mg and also to the lack of carbonation processes in the system. For the major elements, the leachate is mainly attributed to solubility processes. In both sets of columns, the release was very high in the initial L/S elution describing a “first flush” effect [2] and then slowly decreasing through higher L/S ratios. Due to the pH-dependent leaching tendency of heavy metals, the release of trace metals, in particular, T-Cr were relatively low from the first L/S eluates and very minimal concentrations were detected at L/S 100 for both columns (Fig. 3).

3.3. pH_{stat} test leaching

Figure 4 shows the strong dependence on pH on the leaching capacity of trace metals by the pH_{stat} test. In particular, the highest concentrations were observed under extremely acidic conditions, at pH 2 for Cu and T-Cr. The release can be attributed to the dissolutions of minerals binding the heavy metals. The leachate concentration of As shows a low pH-dependency throughout the tested pH range. For Cu, there is a significant decrease or no detection through the pH range of 6-10 and then an increase at pH 12, this is attributed to its amphoteric nature. Comparing the total content of the trace metals (dotted line) to the concentration release at the lowest pH value (pH 2); As, Cu

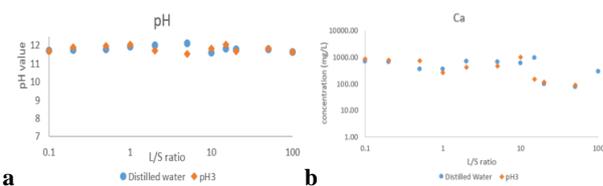


Figure 3. pH values (a) and Calcium (b) concentration release as a function of L/S ratio

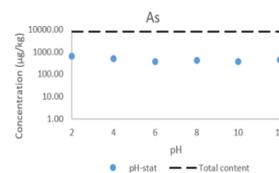


Figure 4. Leaching of Arsenic as a function of pH

and T-Cr showed a reduction in leaching capacity of 8%, 12%, and 19% respectively from the total content measured.

4. Conclusion and Recommendations

Based on the initial results, the recycled BFA has a high buffer capacity, allowing it to maintain its alkalinity and the leaching of elements is primarily attributed to solubility processes. It was implied that the leaching of heavy metals (As, Cu, and T-Cr) could not occur or be very minimal in field site applications. Further continuation of the column leaching test is needed to evaluate reactions that could affect the release of trace elements due to pH changes in the system. To interpret the current data on a long term assessment, geochemical modeling will be applied to calculate the composition of leachate based on the potential solubility controlling minerals and calculate the leaching potential of trace elements of the recycled BFA.

References

- Choi, M.J., 2019. Performance evaluation of the use of tire-derived fuel fly ash as mineral filler in hot mix asphalt concrete. *Journal of Traffic and Transportation Engineering (English Edition)*.
- Lager, T., Delay, M., Karius, V., Hamer K., Frimmel, F.H., Schulz, H.D.. 2006. Determination and quantification of the release of inorganic contaminants from municipal waste incineration ash. *Acta hydrochim. Hydrobiol*, 34, 73-85.
- Meima, J.A., Comans, R.N.J., 1997. Geochemical modeling of weathering reactions in MSWI bottom ash. *Environ. Sci. Technol.* 31. 1269-1276.
- Naka, A., Yasutaka, T., Sakanakura, H., Kalbe, U., Watanabe, Y., Inoba, S., Takeo, M., Inui, T., Takeshi, K., Fujikawa, T., Sato, K., 2016. Column percolation tests for contaminated soils: Key factors for standardization. *Journal of Hazardous Materials*. 320, 326-340.
- Zhang, Y., Soleimanbeigi, A., Likos, J.W., Edil, T.B., 2016. Geotechnical and leaching properties of municipal solid waste incineration fly ash for use as embankment fill material. *Journal of the Transportation Research Board*, 2579, 70-78.