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Short Communications

Development of an accelerated leaching method for incineration bottom ash correlated to toxicity characteristic leaching protocol

Shengxuan Lin^{a,†}, Xuedong Zhou^{a,†}, Liya Ge^{a,*}, Sum Huan Ng^b, Xiaodong Zhou^c, Wei-Chung Chang^{a,d}

^a*Residues and Resource Reclamation Centre, Nanyang Environment and Water Research Institute, Nanyang Technological University, 1 Cleantech Loop, Singapore 637141, Singapore*

^b*Singapore Institute of Manufacturing Technology, A*STAR (Agency for Science, Technology and Research), 71 Nanyang Drive, Singapore 638075, Singapore*

^c*Institute of Materials Research and Engineering, A*STAR (Agency for Science, Technology and Research), 2 Fusionopolis Way, #08-03, Innovis, Singapore 138634, Singapore*

^d*Division of Environmental and Water Resources Engineering, School of Civil and Environmental Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798, Singapore*

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[†] The authors contributed equally to this work

* Corresponding author: E-mail address: lyge@ntu.edu.sg; Tel.: +65-6790-4100; Fax: +65-6792-7319

Abstract

Heavy metals and some metalloids are the most significant inorganic contaminants specified in toxicity characteristic leaching procedure (TCLP) in determining the safety of landfills or further utilization. As a consequence, a great deal of efforts had been made on the development of miniaturized analytical devices, such as ME and μ TAS for on-site testing of heavy metals and metalloids to prevent spreading of those pollutants or decrease the re-utilization period of waste materials such as incineration bottom ash (IBA). However, the bottleneck lied in the long and tedious conventional TCLP which requires 18 h of leaching. Without accelerating the TCLP process, the on-site testing of the waste material leachates was impossible. In this study, therefore, a new accelerated leaching method (ALM) combining ultrasonic assisted leaching (UAL) with tumbling was developed to reduce the total leaching time from 18 h to 30 min. After leaching, the concentrations of heavy metals and metalloids were determined with ICP-MS or ICP-optical emission spectroscopy (ICP-OES). No statistical significance between ALM and TCLP was observed for most heavy metals (i.e. cobalt, manganese, mercury, molybdenum, nickel, silver, strontium and tin) and metalloids (i.e. arsenic and selenium). For the heavy metals with statistical significance, correlation factors derived between ALM and TCLP were 0.56, 0.20, 0.037 and 0.019 for barium, cadmium, chromium and lead, respectively. Combined with appropriate analytical techniques (e.g. ME), the ALM can be applied to rapidly prepare the IBA samples as well as other environmental samples for on-site determination of heavy metals and metalloids.

Keywords: Heavy Metal; Metalloid; Accelerated Leaching Method; Toxicity Characteristic Leaching Protocol; Incineration Bottom Ash

Due to limited land availability, more and more municipal solid wastes (MSW) had been treated by incineration [1, 2]. The IBA generated from MSW contains a relatively high content of heavy metals, certain metalloids and a low content of organic compounds [3]. Because a trace amount of heavy metals and some metalloids (*e.g.* arsenic) can be toxic and carcinogenic due to accumulation effect in human body [4, 5], rapid testing and analysis of heavy metals and metalloids in IBA and other environmental sample leachates are of crucial importance for regulating the spread of the toxins.

Nowadays, novel analytical technologies such as ME and μ TAS had been successfully developed for on-site rapid detection of heavy metals and metalloids [6-8]. However, conventional leaching procedures such as TCLP and Bureau of Reference (BCR) sequential extraction required extended period of time, which hampered the on-site detection of the environmental samples [9, 10]. Therefore, it was necessary to develop a new ALM which could correlate to conventional leaching procedure (*e.g.* TCLP).

So far, several attempts were made to accelerate conventional leaching procedures [11-16]. Among them, UAL was one of common approaches to increase the efficiency of conventional leaching. In UAL, the local heat and power generated by the ultrasonic wave effectively accelerate the leaching of the analytes from samples [16]. Due to its advantages of inexpensive and portable apparatus, UAL was selected in accelerating leaching for environmental samples such as incineration fly ash and sludge based on leaching behaviour of a few heavy metals [16, 17]. However, to the best of our knowledge, there were no ALM targeted to IBA, the overwhelming product generated from incineration treatment.

The aim of this work was to develop a novel, simple and efficient ALM to reduce the total leaching time from 18 h to 30 min for IBA samples. With the assistance of ICP-MS and ICP-OES, a comprehensive comparison of ALM and TCLP would be carried out based on leaching behaviour of 12 heavy metals and 2 metalloids. As the first attempt, a combination of UAL and tumbling was proposed to realize the similar performance as TCLP.

For the reagents used in the experiments, acetic acid (glacial 100%) and nitric acid (65%) were from Merck (Darmstadt, Germany); ICP standards of arsenic (As), barium (Ba), cadmium (Cd), chromium (Cr), cobalt (Co), lead (Pb), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), selenium (Se), silver (Ag), strontium (Sr) and tin (Sn) were from PerkinElmer (Waltham, MA, USA). The extraction fluid was prepared by adding acetic acid into the deionized water from TKA water purification system (Niederelbert, Germany) to get a pH value of 2.88 ± 0.05 , measured by a G20 Compact Titrator (Mettler Toledo, Greifensee, Switzerland).

IBA obtained from Senoko Incineration Plant (Singapore) was used as the testing sample throughout the entire study. The IBA samples were manually crushed by a homemade portable crusher into grit having a diameter of less than 1 cm aligned with TCLP. After homogenization, IBA samples were split into small portions (100 g each) and mixed with extraction fluid at a solid to liquid ratio of 1:20. Thereafter, the IBA samples were leached with three leaching techniques, namely, TCLP, UAL, and a combination of ultrasonic leaching and tumbling.

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Firstly, TCLP (USEPA SW-846 Method 1311) was used as the conventional leaching procedure in this study [9]. The extraction vessel was tumbled end-over-end for 18 h at 30 rpm (Figure S1a). Next, UAL was performed by degassing the ultrasonic bath for 10 min before commencement of leaching. The IBA samples sealed in a glass bottle was subjected to ultrasound in an ultrasonic bath (37 kHz, Elma, S180H) for a certain period of time (Figure S1b). Lastly, a combination of ultrasonic leaching and tumbling was performed on the IBA sample by subjecting it to 5 min of UAL then followed by tumbling with a rotator spinning at 30 rpm over a period of 25 min (Figure S1c). All the experiments were conducted in triplicates.

After filtered through a 0.45 μm nylon membrane filter (Whatman, UK) and recorded the pH values, the leachates were acidified by 2% (v/v) of nitric acid for further ICP-OES (Optima 8300 Cross Flow, PerkinElmer Inc., Waltham, MA, USA) and ICP-MS (7700x, Agilent, Santa Clara, CA, USA) analyses.

The concentrations of Ba, Co, Mo, Mn, Ni and Sn in the leachates were determined by ICP-OES. The r^2 values of the calibration curves of Ba, Co, Mo, Mn and Ni were greater than 0.995. The analysis of each leachate was conducted three times, and the average concentrations of the heavy metals were recorded.

The concentrations of Ag, As, Cd, Cr, Hg, Pb, Sb and Se were determined by ICP-MS. The instrument was equipped with a Micromist nebulizer and other high matrix introduction accessories. The radio frequency power was set at 1550 W, and plasma, auxiliary, and nebulizer gas flow rates are 12, 0.95, and 0.75 L min⁻¹, respectively. Calibrations were

conducted with ICP-MS standard solutions of Ag, As, Cd, Cr, Hg, Pb, Sb and Se which were prepared for a range of concentrations encompassing expected sample concentrations.

All the statistical analyses of the results were performed using Microsoft Excel 2013. Unpaired *t*-tests with unequal variance were performed to provide the *p*-values. A *p*-value less than 0.05 was considered as significant difference.

Preliminary results of 60 min UAL from ICP-MS and ICP-OES indicated that concentrations of most heavy metals (including Ag, Ba, Cd, Cr, Pb and Hg) and metalloids (*i.e.* As and Se) of interest were significantly higher than TCLP. As the further attempt, the time of UAL was reduced from 60 min to 6 min to avoid over-leaching. However, the leaching amounts for the most over-leaching analytes (including Ag, Ba, Cd, Cr, Pb, Hg and Se) only dropped slightly. Moreover, the leaching amounts for under-leaching analytes (including Co, Mn, Ni and Sr) also dropped, which made them departing even further from TCLP.

During further investigation, it was discovered that final pH values obtained from accelerated leaching with ultrasound did not match up with pH values obtained from TCLP. The average pH values of TCLP leachates were about 7.54 ± 0.12 , while the average of pH values from ultrasonic leachates only reached 4.61 ± 0.35 . As shown in Figure 1, the pH values increased with prolonged tumbling of TCLP and UAL. However, the gradient of increase was significantly lower in UAL. As the pH value is a critical factor that significantly influences some metals leaching behavior [18], a better leaching method to match the pH level with TCLP was desired.

TCLP used mechanical washing by end-over-end tumbling for a period of 18 h, which indicated that tumbling could play a critical role for changing the pH value. As shown in Figure 1, the pH value increased considerably during the first 0.5 h, and slightly from 0.5 to 18 h. This phenomenon inspired us to develop a combination approach of both UAL and tumbling, which could have a synergetic effect to better fit pH and concentration of heavy metals and metalloids in the leachates of accelerated leaching to TCLP.

Therefore, a new ALM combined UAL with tumbling of the sample was developed as follows: After the 5 min UAL procedures, tumbling of the sample was performed with a rotator spinning at 30 rpm with a period of 25 min. The average pH of this ALM was 6.15 ± 0.03 , which is much closer to pH level of TCLP at 7.54 ± 0.12 . More importantly, there was no statistical significance ($p > 0.05$) between ALM and TCLP for the concentrations of As, Co, Mn, Mo, Ni and Sr. On the other hand, the concentrations of Ag, Hg, Se and Sn from both TCLP and ALM were below limit of detection (LOD) of ICP-MS or ICP-OES. Therefore, leaching amounts of Ag, Hg, Se and Sn from ALM were considered to be constant with results from TCLP. The concentrations of heavy metals and metalloids in IBA leachates obtained from different leaching methods were shown in Table 1.

As shown in Table 1, the concentration of As leached by UAL is higher than both ALM and TCLP. Al-Abed *et al.* found similar leaching pattern for leaching of As. The higher concentration of As in the leachates obtained from UAL might due to exchangeable or acid extractable carbonate phases which contributed to release As species from iron oxides or dissolution of carbonates driven by cation exchange [19]. On the other hand, at relatively neutral pH, leaching of As is limited by co-precipitation of As and iron hydroxide onto the

solid matrix [19-21]. This explained the relatively lower concentrations of As for TCLP and ALM.

Similar to the findings from Komomweeraket *et al.*, very low levels of Cr and Pb were measured from the TCLP leachates which have nearly neutral pH. This phenomenon could be attributed to the dissolution of oxide or hydroxide materials which greatly influences the solubility of these amphoteric elements [22]. Amphoteric elements could leach out from the solid matrix for pH levels below 6 or above 8.

Studies performed by Jing *et al.* found that desorption of Hg peaks at both acidic end and alkaline end of the pH spectrum [23]. According to previous findings, the desorption rate decreased steadily between pH of 3-5, while minimum desorption occurred between pH 5-7 [23]. This finding matched with the results shown in Table 1, in which level of Hg was below the LOD of ICP-MS of $0.05 \mu\text{gL}^{-1}$ for both TCLP and ALM.

The encouraging results from ALM indicated that the method is a very promising alternative for TCLP. Although the main aim of this study was to develop an ALM as close to TCLP as we can, over-leaching was generally more desirable than under-leaching due to the safety reasons. The alarm potential is raised when concentrations of any targeted analyte from ALM exceeds TCLP's criteria. Meanwhile, false alarm from over-leaching analytes by ALM are mitigated with correlation factors. The statistical tool of *t*-test was performed on each analyte to determine the relationship of ALM and TCLP. For the heavy metals (*i.e.* Ba, Cd, Cr and Pb) with statistical significance ($p < 0.05$), correlation factors in magnitude were provided

between ALM and TCLP. Correlation factors of Ba, Cd, Cr and Pb are 0.56, 0.20, 0.037 and 0.019, respectively.

In conclusion, a novel ALM was successfully developed for leaching out the heavy metals and metalloids in IBA, which significantly reduced in the total leaching time from 18 h to 30 min while offered comparable leaching amounts of most heavy metals and metalloids. In general, the testing results showed that there was no statistical significance between ALM and TCLP for most heavy metals (*i.e.* Ag, Co, Hg, Mn, Mo, Ni, Sn and Sr) and metalloids (*i.e.* As and Se). For the heavy metals (*i.e.* Ba, Cd, Cr and Pb) with statistical significance, correlation factors were derived between ALM and TCLP. The successfully developed ALM would be applied to rapidly leaching of IBA and other environmental samples for on-site of heavy metal and metalloid detection.

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Conflict of Interest Statement

The authors declared that they have no conflicts of interest.

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Table 1 Concentrations of heavy metals and metalloids in IBA leachates obtained from different leaching methods

Elements	Concentration (μgL^{-1})			
	TCLP	6 min UAL	60 min UAL	Final ALM
Arsenic (As)	4.0 ± 0.4	22 ± 8	8 ± 3	3.00 ± 0.05
Barium (Ba)	282 ± 4	449 ± 28	616 ± 294	501 ± 7
Cadmium (Cd)	21.6 ± 0.4	51 ± 12	73 ± 18	106.9 ± 0.6
Chromium (Cr)	6 ± 3	259 ± 96	278 ± 71	160 ± 2
Cobalt (Co)	185 ± 45	9 ± 3	17 ± 7	152 ± 6
Lead (Pb)	4 ± 3	1311 ± 324	1404 ± 314	331 ± 6
Manganese (Mn)	1922 ± 423	213 ± 70	974 ± 402	2280 ± 52
Mercury (Hg)	< 0.05	0.6 ± 0.1	1.3 ± 0.5	< 0.05
Molybdenum (Mo)	107 ± 3	< 0.5	< 0.5	102 ± 3
Nickel (Ni)	167 ± 24	9 ± 5	18 ± 8	162 ± 1
Selenium (Se)	< 1	5 ± 1	9 ± 4	< 1
Silver (Ag)	< 0.05	0.3 ± 0.3	0.8 ± 0.5	< 0.05
Strontium (Sr)	3410 ± 255	299 ± 76	419 ± 117	3225 ± 44
Tin (Sn)	< 2	< 2	< 2	< 2

Figure Caption:

Fig. 1. pH profile of 18 h TCLP and 60 min UAL.

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Figure 1