Leachability of heavy metals from fired clay bricks incorporated with cigarette butts

(Leachability of heavy metals)

Aeslina A.K.
Senior Lecturer, Department of Environmental Engineering and Water Resources,
University Tun Hussein Onn Malaysia, (UTHM),
Batu Pahat, Malaysia.
aeslina@uthm.edu.my
Mohajerani A.
Senior Lecturer (PhD), School of Civil, Environmental and Chemical Engineering,
RMIT University,
Melbourne, Australia.
dr. abbas@rmit.edu.au

Abstract—**Although small in size, indiscriminate littering of cigarette butts (CBs) can cause serious environmental impact. Several trillion cigarettes produced worldwide annually lead to thousands of kilograms of toxic waste that will end up in the waterways. Most cigarette filters are made of cellulose acetate which is slow to biodegrade and can take up to 18 months or more to break down under normal litter conditions. CB filters release range of toxic chemicals as they deteriorate. Toxic chemicals and heavy metals trapped in the CB filters can be leached and so cause serious damage to the environment. This paper presents some of the results from a continuing study on recycling CBs into fired clay bricks. Fired clay bricks were manufactured with different percentages of CBs (0%, 2.5%, 5% and 10%) to determine the possible levels of heavy metals that can be leached from the manufactured bricks. Leaching of heavy metals from the fabricated clay bricks was tested to investigate whether the leachate values exceed the regulatory standards. Three types of leaching tests which are Australian Bottle Leaching Procedure (ABLP), the Toxicity Characteristics Leaching Procedure (TCLP) and Static Leachate Test (SLT) were carried out in this investigation to analyse eleven heavy metals: Arsenic (As), Selenium (Se), Mercury (Hg), Barium (Ba), Cadmium (Cd), Chromium (Cr), Lead (Pb), Silver (Ag), Zinc (Zn), Copper (Cu) and Nickel (Ni). The leachate results revealed trace amounts of heavy metals.**

***Keywords*-Cigarette butts; Recycling waste; Fired clay bricks; Leachate; Heavy metals**

I. INTRODUCTION

Worldwide, cigarette butts (CBs) are the most common type of litter. The United States Department of Agriculture estimates that in 2004 over 5.5 trillion cigarettes were produced in the world [1]. This is equivalent to an estimated 1.2 million tonnes of cigarette butt waste per year. These figures are expected to increase by more than 50% by 2025, mainly due to an increase in world population [2]. In Australia alone, an estimated 25 to 30 billion filtered cigarettes [3] are smoked each year; of these, an estimated 7 billion are littered [4].

Most cigarette filters are made of cellulose acetate. Cellulose acetate filters in CBs are slow to biodegrade and can take up to 18 months or more to break down under normal litter conditions [5,6]. Filters have long term effects on the urban environment, especially in waterways and run-offs [7]. Toxic chemicals trapped in the CB filters can leach, thus causing serious damage to the environment [8,9,10]. There are up to 4000 chemical components in cigarette smoke, of which 3000 are in the gas phase and 1000 in the tar phase. Polycyclic aromatic hydrocarbons (PAHs), N-nitrosamines, aromatic amines, formaldehyde, acetaldehyde, benzene, and toxic metals such as cadmium and nickel combine to form more than 60 chemicals that are known to be carcinogenic [11, 8,9,10,11,12].

Landfilling and incineration of CB waste are not, universally, environmentally sustainable nor economically feasible disposal methods. Even when correctly binned and sent to landfill far from natural waterways, CBs remain an environmental hazard [13]. Also, landfilling of waste with high organic content and toxic substances is in general becoming increasingly costly and difficult [14,15,16]. Incineration of CBs is also a seemingly unsustainable solution as emissions from the burning waste contain various hazardous substances [17]. Recycling CBs is difficult because there are no easy mechanisms or procedures to assure efficient and economical separation and recycling of the entrapped chemicals. An alternative could be to incorporate CBs in a sustainable composite building material such as fired bricks.

Brick is one of the most accommodating masonry units as a building material due to its properties. Attempts have been made to incorporate waste in the production of bricks. For instance, the use of rubber [18], limestone dust and wood sawdust [19], processed waste tea [20], fly ash [21,22] polystyrene [23] and sludge [24]. Recycling of such wastes by incorporating them as inert components into building materials is a practical solution to a pollution problem. Nevertheless, leachability of waste is a matter of concern. This paper presents and discusses on the leachability of heavy metals from fired clay bricks incorporated with CBs.

II. MATERIALS AND METHOD

A. *Preparation of Clay Brick Samples Incorporated with CBs*

The CBs (of different brands and sizes) used in this study were provided by Buttout Australia Pty Ltd. The butts had been collected from dry receptacles. Upon delivery, the CBs were disinfected at 105^{o}C for 24 hours and then stored in sealed plastic bags. The soil used was brown silty clayey sand prepared for making fired clay and provided by Boral Bricks Pty Ltd, Australia. The classification tests including liquid limit, plastic limit, plasticity index and particle size distribution were carried out according to Australian Standard [25]. Chemical analyses were carried out to determine the main chemical components of the experimental soil. Chemical composition of the raw clay samples was determined using X-ray Fluorescence (XRF).

Proctor standard compaction tests were conducted, according to Australian Standard [26], to determine optimum moisture contents (OMC) and maximum dry densities for the experimental soil (control sample) and the mixed soil-CBs samples. Four different mixes were used for making fired brick samples. CBs (2.5, 5, and 10% by weight, about 10 – 40% by volume) were mixed with the experimental soil and fired to produce bricks. The mixes were made using a Hobart mechanical mixer with a 10 litre capacity for 5 minutes. The samples were compacted manually in appropriate moulds using predetermined masses corresponding to the maximum density (found from standard compaction tests).

B. *Leachate Analyses*

It is known that heavy metals such as arsenic, chromium, nickel and cadmium can be trapped in the filters of cigarette butts [27]. Hence, leaching tests were carried out to investigate the levels of possible leachates of heavy metals from the manufactured CB bricks. Two types of brick samples were investigated: crushed bricks and whole solid brick samples. Two different procedures were employed for the crushed samples: the Australian Bottle Leaching Procedure (ABLP) [28] and the Toxicity Characteristics Leaching Procedure (TCLP) [29]. ABLP is commonly used for regulatory analysis in Australia, while TCLP is commonly adopted in many studies [22,30,31].

For the whole solid brick samples, leaching testing was carried out to investigate the long-term leachate characteristics of the samples. A procedure modified from the Static Leach Test [32] and called the Static Leachate Test (SLT) was used in this study. The original method is generally used to investigate the mechanism of leaching from solidified waste [33,34].

Samples of leachates were prepared in triplicate and analysed for heavy metals using an Inductive Coupled Plasma Mass Spectrophotometer (ICP-MS). However, the laboratory leaching tests conducted in this study could only be used to define the intrinsic leaching characteristics of the waste. This is because the controlled conditions of the laboratory test simulate the leaching behaviour of the waste at its ideal or worst case that could occur in landfill, but this condition is not equivalent to actual landfill conditions [35].

1) *Australian Bottle Leaching Procedure (ABLP) and Toxicity Characteristic Leaching Procedure (TCLP)*

Both ABLP and TCLP use similar preparation procedures for regulatory analysis but differ in the limit of particle size used in the sample. In the ABLP test, the brick sample was crushed and a representative sample finer than 2.4 mm produced, while in the TCLP test, a crushed sample finer than 9.5 mm was prepared for the analysis. While the ABLP places no restriction on the minimum particle size [28], the TCLP requires particle sizes to comply with the standard method [29]. Both the ABLP and TCLP were adopted in the present study for comparative purposes.

Use of leaching fluid (leachant) is important for both tests. The category of landfill in which the final waste is intended for disposal dictates the type of leachant to be used. Two leachants (acidic leachant and deionised water) are essential in the ABLP, whereas only one, acidic leachant, is essential for the TLCP test. In order to compare the results, only one leachant was used in this study. As metals were the focus of the study, an acid was required to be added to the leachant to expedite the extraction of metals from the brick sample. For this purpose, acidic leachant was used.

Laboratory leaching fluids for both tests were prepared in screw-capped polyethylene bottles which were filled with crushed brick samples (20 g) and leachant (400 ml) at the ratio of 1:20. The bottles were agitated at 30 rpm for 18 hours in an end-over-end manner. An extraction fluid (5.7 mL of glacial acetic acid per litre) was initially added to the leachant. The leachate collected was then filtered using 0.7 μm glass fibre filters before being analysed using ICP-MS to determine the dissolved metals.

2) *Static Leachate Test (SLT)*

The static leachate test (SLT) used in this study was modified from the static leach test [32]. According to Dutre and Vandecasteele [33,34], the static leach test provides a better procedure than the semi-dynamic leach test as the latter may give imprecise information if no preparatory study is made of the waste-leachate equilibrium. In the static leach test, no leachant renewal was administered and this was intended to ensure leachate concentrations were produced to the maximum possible and equilibrium was near complete without much interruption which might otherwise be caused by leachate renewal. This arrangement is thought desirable in long-running leaching tests such as those required in the study. Furthermore, this procedure is considered to be a closer simulation of field conditions, although no equilibrium had to be reached completely during the static phase of the test.

Four whole solid brick samples, respectively containing 0% (control), 2.5%, 5% and 10% of CB content were prepared and fired. The brick samples were each suspended in a closed vessel under quiescent conditions for 134 days. The brick sample was positioned in the centre of the leaching fluid made up of deionised water in the proportion of 5 parts of leachant (deionised water) to 1 part of whole brick by mass. A lower ratio was employed in this investigation due to the low concentrations of the heavy metals. Note that the leachant was initially mixed with glacial acetic acid at the rate of 5.7 ml per litre. The leachant used in this test was consistent with the ABLP and TCLP tests for comparison purposes between the crushed and the whole solid brick results. Fig. 1 shows the experimental set-up for leachate collection from the SLT. The experimental preparation for the SLT of the brick samples is presented in Table I.

In the SLT method, leachant was not renewed by a fresh solution in order to produce the maximum leachate concentrations, and leachates were collected over long durations of 25, 41, 71 and 134 days. The progressively-spaced intervals between collections were necessary to allow leaching to take place smoothly as the leaching process was observed to be very slow. During leaching, samples were periodically taken out of the solution in the vessel and analysed on the specified days using triplicate samples. This was to determine the reproducibility of the results before the mean of three measurements was calculated. After sample collection following the 134-day test, the vessel contents were stirred vigorously, followed by sampling of the leachate solution that had presumably reached near equilibrium if not complete equilibrium. Results were compared between the leachate solution collected after the 134-day test period and after the vessel contents were agitated upon removal of the sample bricks on completion of the test. All the samples were first filtered using 0.7 μm glass fibre filters before the filtrates were analysed for dissolved heavy metals using ICP-MS.

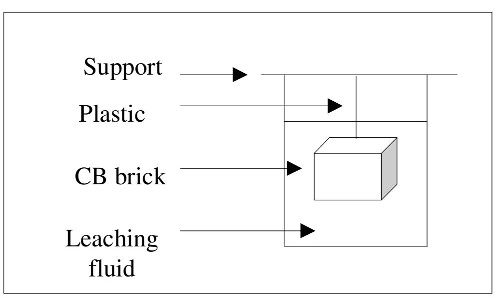

Figure 1. Experimental set up for static leachate test

TABLE I. COMPOSITION OF MIX OF LEACHING FLUID FOR WHOLE BRICK SAMPLES

Mixture Identification (%)	Weight (kg)	Distilled water (L)	Glacial acetic acid (mL)
CB (0.0)	3.145	15.7	89
CB (2.5)	3.060	15.3	87
CB (5.0)	2.295	11.5	66
CB (10.0)	2.165	10.8	62

III. RESULTS AND DISCUSSIONS

A. *Australian Bottle Leaching Procedure (ABLP) and Toxicity Characteristic Leaching Procedure (TCLP)*

Results of leachate analyses using ABLP and TCLP (Tables II and III) were compared against the concentration limits for heavy metals set by the TCLP Regulatory Levels according to the United States Environmental Protection Agency [35] and the Guidelines for Hazard Classification of Solid Prescribed Industrial Wastes, approved by the Environmental Protection Agency of Victoria [36]. Both methods yielded similar leachate concentrations of target metals for clay bricks with 0%, 2.5%, 5% and 10% of CB content. However, due to the difference in the particle size used in the ABLP and TCLP tests, marginal differences can be observed in the leachate concentrations. Most of the heavy metal concentrations from the ABLP tests (with smaller particle size) were slightly higher than the corresponding TCLP results. Different particle sizes control the surface of contact between the waste and the leaching fluids, thus influencing the contact time and contact area between the leachant and the metals that can leach and influence the leaching concentrations [31,37].

Table II summarises the ICP-MS results for heavy metals using the ABLP (particle size < 2.4 mm). The As, Zn and Cu concentrations in the 10% CB brick (0.123 mg/L, 1.285 mg/L and 1.090 mg/L respectively) were slightly higher than the control brick (0% CB) (0.007 mg/L, 0.965 mg/L and 0.190 mg/L respectively). The Cr and Pb concentrations were lower by 73% and 74% respectively with 10% of CB content compared to the control brick, while Ba and Ni concentrations were marginally low throughout the leaching period.

Similar trends were observed for most of the heavy metals in the case of the TCLP method. Table III shows that As and Zn concentrations increased at 10% CB content measured up to 0% of CB content with 0.010 and 0.890 mg/L difference respectively. The TCLP test also leached lower levels of Cu and Pb at 10% of CB content against control samples with 0.035 mg/L and 1.909 mg/L differences respectively. For Ba, Ni and Cr, the values obtained were low with only marginal differences during the leaching test. Se, Hg, Cd and Ag were not detected in the samples for both tests.

From observation of the results (Tables II and III), the increasing values of the metals could possibly originate from the metals trapped inside the CB waste incorporated in the manufactured brick. On the other hand, the decreasing concentration levels could possibly emanate from the clay soil [38] as the heavy metal values decreased with increased CB content. It should be emphasised that the concentrations of the metals in the leachate from both tests were in trace amounts which do not exceed the regulatory thresholds specified by the USEPA (1996) or the EPAV (2005) (Table IV).

TABLE II. CONCENTRATIONS OF HEAVY METALS USING ABLP

Heavy metals	Percentage of CBs by weight			
	0%	*2.5%*	*5%*	*10%*
	Concentration (mg/L)			
Arsenic (As)	0.007	0.019	0.093	0.123
Selenium (Se)	-	-	-	-
Mercury (Hg)	-	-	-	-
Barium (Ba)	0.590	0.440	0.605	0.510
Cadmium (Cd)	-	-	-	-
Chromium (Cr)	0.033	0.005	0.028	0.009
Lead (Pb)	0.130	0.019	0.058	0.034
Silver (Ag)	-	-	-	-
Zinc (Zn)	0.965	0.180	7.750	1.285
Copper (Cu)	0.190	0.320	0.680	1.090
Nickel (Ni)	0.007	0.006	0.016	0.009

- not detected

TABLE III. CONCENTRATIONS OF HEAVY METALS USING TCLP

Heavy metals	Percentage of CBs by weight			
	0%	*2.5%*	*5%*	*10%*
	Concentration (mg/L)			
Arsenic (As)	0.025	0.012	0.045	0.035
Selenium (Se)	-	-	-	-
Mercury (Hg)	-	-	-	-
Barium (Ba)	0.270	0.280	0.295	0.275
Cadmium (Cd)	-	-	-	-
Chromium (Cr)	0.007	0.003	0.006	0.008
Lead (Pb)	1.941	0.044	0.037	0.032
Silver (Ag)	-	-	-	-
Zinc (Zn)	0.255	0.115	0.670	1.145
Copper (Cu)	0.190	0.295	0.210	0.155
Nickel (Ni)	0.004	0.002	0.004	0.003

- not detected

TABLE IV. REGULATORY THRESHOLD BY USEPA (1996) OR THE EPAV (2005)

Heavy metals	Concentration Limit (mg/L)[a]	Concentration Limit (mg/L)[b]
Arsenic (As)	5	2.8
Selenium (Se)	1	4
Mercury (Hg)	0.2	0.4
Barium (Ba)	100	280
Cadmium (Cd)	1	0.8
Chromium (Cr)	5	20
Lead (Pb)	5	4
Silver (Ag)	5	40
Zinc (Zn)	500	1200
Copper (Cu)	100	800
Nickel (Ni)	1.34	8

a. United States Environmental Protection Agency (USEPA) (1996)

b. Environmental Protection Agency (EPA) Victoria (2005)

B. Static Leachate Test (SLT)

The SLT was used to assess leachability at the respective sampling times of 25, 44, 71, and 134 days. Table V summarise the results of the cumulative heavy metal concentrations at the specified sampling periods. The As, Ba, Zn and Cu concentrations, as observed, leached out gradually with time and as the CB content increased.

The As and Ba concentrations were highest (0.215 mg/L and 0.525 mg/L) at the 134-day leaching period for the 5% of CB content whilst the Zn and Cu concentrations were highest (respectively, 0.425mg/L and 0.090 mg/L) for the 10% of CB content. The Pb concentration on the contrary was highest at 0.008 mg/L for the control brick compared to other bricks. The Cr and Ni concentrations were negligible with marginal differences between them throughout the leaching periods up to 134 days for the control brick as well as the other bricks with CB content.

The four whole bricks were each removed from the leaching vessel upon completion of the tests, leaving the leachate solution in the vessel. The solution was stirred and left to settle (under gravity) before the supernatant was taken out of solution and filtered; then the filtrate (in triplicate) was analysed for heavy metals. The final leachate samples were each measured after the 134-day leaching period to confirm the cumulative amount of the target metals. This was done to determine the effect of any precipitates that could have formed in the leachate solution and also to check whether the leachate concentrations had reached equilibrium at the end of the leaching process. The metals concentrations were compared before sampling and after the vessel contents were agitated upon removal of the brick samples. Their difference was marginal (less than 0.005 mg/L). Se, Hg, Cd and Ag were however not detected in the samples. Overall, the metals that leached out from the brick samples were very low in concentration and below the regulatory limits set by the USEPA (1996) and the EPAV (2005).

TABLE V. CONCENTRATIONS OF HEAVY METALS (10% OF CB) USING SLT

Heavy metals	Leaching periods (days)			
	25	*25*	*25*	*25*
	Concentration (mg/L)			
Arsenic (As)	0.163	0.163	0.163	0.163
Selenium (Se)	-	-	-	-
Mercury (Hg)	-	-	-	-
Barium (Ba)	0.197	0.197	0.197	0.197
Cadmium (Cd)	-	-	-	-
Chromium (Cr)	0.009	0.009	0.009	0.009
Lead (Pb)	0.002	0.002	0.002	0.002
Silver (Ag)	-	-	-	-
Zinc (Zn)	0.383	0.383	0.383	0.383
Copper (Cu)	0.009	0.009	0.009	0.009
Nickel (Ni)	0.008	0.008	0.008	0.008

- not detected

All heavy metal leachate values determined in ABLP, TCLP and SLT tests (Tables II, III and V). were insignificant and comply with the concentration limits set by USEPA (1996) and EPAV (2005) (Tables IV). The ABLP and TCLP tests yielded similar leachate concentrations of target metals for clay bricks with 0 and 10% CBs. However, due to the difference in crushed particle size, the ABLP test (using smaller particle size) produced slightly higher values than the TCLP test for most concentrations.

IV. CONCLUSIONS

Results of three different leaching tests on fired clay bricks with 0%, 2.5%, 5% and 10% CB content are presented and discussed. The acidic leachant method (based on the ABLP and TLCP) using crushed brick yielded comparable low leachate concentrations of the target metals in the clay-CB bricks with different CB content (2.5%, 5% and 10%). SLT was intentionally designed for long term leachability at 134-day continuous leaching. The results show that for most of the heavy metals investigated, the concentrations of the heavy metals leached out increased gradually with leaching time but subsequently decreased slightly at the final leaching time of 134 days. The concentrations apparently fluctuated or remained nearly constant throughout the leaching time while some of the metals could not be detected at all.

On the whole, due to the differences in the types of samples used, the ABLP and TCLP tests (using crushed samples) produced slightly higher values than the SLT (solid samples) tests for most cases, even though the SLT measurements were made after 134 days. The SLT is clearly a less aggressive leaching test compared to ABLP and TCLP which are more destructive by virtue of the requirement for crushed particles to be used in the test. The particle size specification in the test plays a role in facilitating the homogeneity of mixing, which could possibly result in precipitation of the leachate components during the vigorous end-over-end mixing method, which in turn could result in higher concentrations of the leached metals, even for short leaching time frames. Equally, despite the differences in the test mechanisms, the purpose intended for every test conducted in this research was achieved, as evidenced by the results obtained. All concentrations of metals leached out were insignificant and far from exceeding the Guidelines for the Hazard Classification of Solid Prescribed Industrial Wastes [35] and the TCLP Regulatory Levels [36].

The low concentrations of the heavy metals from almost all the leaching tests conducted can also be attributed to the high firing temperature (1050ºC) that may have converted the metals in the bricks to metal oxides during the firing process [22,31]. These results indicate that the minimal leaching of the target metals from the incorporation of CBs in fired clay bricks could effectively minimise the potential of CB contamination problems. The results found so far show that cigarette butts can be regarded as a potential addition to raw materials used in the manufacturing of light fired bricks.

ACKNOWLEDGMENT

The results presented in this paper are part of ongoing research on Recycling Cigarette Butts. The authors would like to thank Buttout Australia and Boral Bricks Pty Ltd for supplying the cigarette butts and soil, respectively, for this study.

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