Toxicity Characteristics and Heavy Metals Leachability of Self Compacted Concrete Containing Fly Ash and Bottom Ash as Partial Cement and Sand Replacement

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Abstract This research aimed to examine the toxicity characteristics of self-compacted concrete (SCC) containing fly ash (FA) and bottom ash (BA) as a partial replacement of ordinary Portland cement and fine aggregate. In particular, the other objective was to identify the heavy metals leaching nature of FA and BA in SCC as well as to determine their possible use as construction materials. FA and BA derived from the phase of combustion in coal-fired power plants. It contains heavy metals within their compositions. SCC mixtures were prepared to have various percentages of FA (substitution of cement) and BA (substitution of sand) of 0, 10, 20, and 30% respectively for subsequent experiments. Several investigations were performed out, such as the characterization of the main composition and heavy metals of the materials through X-Ray Fluorescence (main composition and heavy metal characterization of the raw materials), the compressive strength test, the Toxicity Characteristics Leaching Procedure (TCLP) and the Synthetic Precipitation Leaching Procedure (SPLP). Results showed that SCC containing FA and BA replacement had obtained compressive strengths of a similar range or higher than the control SCC (without any replacement of FA and BA). Sample FA10BA10 or 10% substitution of FA and BA recorded the highest compressive strength value at 58.07 ± 0.50 MPa. From the results of TCLP and SPLP, it founded that the inclusion of ashes up to 30% was safe as the concentration of heavy metal leaching did not surpass the concentration of

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pollutants for toxicity characteristics. In conclusion, this research suggests that the disposal and use of FA and BA as a promising replacement of construction materials may be used to minimize their environmental issues, improve efficiency and reduce the cost of production of SCC in the future.

Keywords Toxicity characteristic · Leaching · Heavy metals · Construction materials · Fly ash · Bottom ash · Self-compacted concrete

1 Introduction

Fly ash (FA) and bottom ash (BA) are usually the excesses of waste created by the combustion process in coal-fired power plants. The total quantity of FA and BA produced represents 80% of the total residues produced from incineration [1]. Residual weights range from 5 to 30% before combustion, depending on their composition [2]. FA is more like a type of fine particulate matter with many heavy metals, organic compounds and chlorides [3]. Besides, BA can explain as particles consisting of different elements such as metals, minerals, ceramics and unburned materials [4, 5]. Both FA and BA contain several hazardous elements that must be recognized as a threat to the environment and its life-threatening effects.

Researchers Chang and Wey [6] and Aubert et al. [7] have previously identified the decreasing impact of the FA and BA disposal applications in Taiwan and France, respectively [6, 7]. Consequently, incineration of solid waste has contributed to a reduction in the amount of disposal, which, in turn, has resulted in the formation of ash as combusted by-products. BA had already been reused primarily as road ingredients and as aggregate or sand material in concrete. Meanwhile, due to the high content of heavy metals in the FA, ashes had very little use and were often disposed of in landfills. However, in recent years, FA is powerfully applicable as a substitute for cement in concrete industries due to its cement properties.

The purpose of this research was, therefore, to integrate scheduled wastes such as FA and BA into the use of SCC as building materials. Previous researchers have been committed to the exploration of the direct effects of the introduction of waste into their subject matter of study based on improvements in mechanical and chemical properties. However, environmental impacts were either ignored or less elaborated. Therefore, both the compressive strength and leachability effects of the integration were calculated, analyzed and presented in this paper.

2 Materials and Method

2.1 Study Location

The FA and BA were collected from a thermal processing plant in Peninsula, Malaysia. The material composition of FA, BA, and OPC was analyzed using the X-ray fluorescence (XRF) test performed using the Bruker AXS S4 Pioneer. The FA and BA samples used in the test were prepared in the form of pellets, with a sample at a wax ratio of 8:2 using the Pressed Pellet Technique. Between all the elements and compounds in the ash detected by XRF, only the metal elements measurable by the Atomic Absorption Spectrometry (AAS) as shown in Table 1.

Besides, aggregates and sand used in the manufacture of SCC usually complied with the specifications of BS EN 206-1 [8]. For aggregates, sizes of 14 to 20 mm prepared using a sieving process. OPC was used in compliance with BS EN 197-1 [9], although mixtures were tested to conform with EN 943-2 [10].

A series of mixtures were prepared accordingly for the control sample with 0% of FA and BA and mixture with different percentages FA and BA (10, 20, 30% of FA and BA). The overall binder for all samples ranged from 530 to 550 kg/m^3, respectively. Standard moulds of 150 mm^3 in compliance with BS EN 12390-1 [11] were used to contain fresh SCC items. The configuration of the mix for this research, as shown in Table 2.

A compression test assessed the compressive strength at 28 days in BS EN 12390-3 [12]. Next, the crushed cubes are smashed using a steel hammer to produce smaller pieces of the samples. The fragments were then further crushed using the Aggregate Impact Value (AIV) equipment to reduce the size of the solid particles to less than 9.5 mm. The crushed fragments were sieved and retrieved as samples for the Toxicity Characteristic Leaching Procedure according to Method 1311 [13].

TCLP was used as a set of guidelines for preparing collected samples of concrete specimens for the leachate analysis to be completed. Since the SCC cubes were

Table 1 Heavy metals of FA, BA and Ordinary Portland Cement (OPC)

Heavy metals	Formula	Concentration (mg/L)		
		FA	BA	OPC
Chromium	Cr	228	176	54
Manganese oxide	MnO	900	800	800
Iron (III) oxide	Fe_2O_3	41,600	52,600	30,200
Nickel	Ni	107	88	19
Copper	Cu	101	38	26
Zinc	Zn	52	31	164
Arsenic	As	38	12	37
Lead	Pb	62	18	60

Table 2 Proportions of SCC mix incorporated with FA and BA

Mix design	Total binder (kg/m^3)	Cement (kg/m^3)	FA (kg/m^3)	CA (kg/m^3)	Sand (kg/m^3)	BA (kg/m^3)	Water (kg/m^3)	SP (kg/m^3)
FA0BA0 (control)	550	550	0	593	914	0	228	5
FA10BA0	550	495	55	593	914	0	220	4
FA20BA0	540	432	108	593	914	0	216	4
FA30BA0	530	371	159	593	914	0	212	4
FA0BA10	550	550	0	593	822	67	228	5
FA0BA20	550	550	0	593	731	133	228	5
FA0BA30	550	550	0	593	640	200	228	5
FA10BA10	550	495	55	593	822	67	220	4
FA20BA20	540	432	108	593	731	133	216	4
FA30BA30	530	371	159	593	640	200	212	4

concrete specimens, the extraction method had to be performed under the Sample group containing more than 0.5% of the dry solids. The extraction fluid was prepared by diluting 5.7 mL of glacial acetic acid (CH_3COOH) with pure water to a volume of 1 L. A total of 50 g of the sample was prepared and placed in a 2 L extraction bottle, and the extraction fluid was then poured in the rotary agitation apparatus, and it was left to spin for 18 h from end to end. The solution in the extraction bottle was then diluted to dispel the solid particles. The fluid portion of the sample was held at a pH of less than 2.0 and stored in the refrigerator at 5 °C for a leachate determination analysis using AAS, which was performed using Perkin Elmer Analyst 800.

Also, the Synthetic Precipitation Leaching Technique (SPLP) according to Method 1312 [14] was used to provide information on the mobility (leachability) of organic and inorganic components from liquids, soils, and waste. Extraction fluids with pH 4.2 solution have been used for this study. The scale of the samples used was less than 9.5 mm.

3 Result and Discussion

Table 3 indicates that compressive intensity increased from day 7 to day 28. All SCCs incorporated with FA and BA attained compressive strengths with a similar range but higher than standard concrete. By comparison to BS EN 206 [15], the sample strength classes ranged from class C45 to class C70 at 28 days and met the criteria to be graded as normal-weight and heavy-weight concrete.

At various replacements of OPC with 0, 10, 20, and 30% of FA, strengths were observed to be in the range of 30–42 MPa at 7 days, 41–59 MPa at 14 days and 49–69 MPa respectively. All the samples incorporated with FA were higher than

Table 3 Compressive strength of SCC samples incorporated with FA and BA

Samples	Compressive strength (MPa)		
	7 (days)	14 (days)	28 (days)
FA0BA0	33.60 ± 0.68	41.88 ± 0.39	49.30 ± 0.61
FA10BA0	36.21 ± 0.67	58.04 ± 0.33	68.79 ± 0.24
FA20BA0	30.04 ± 0.32	49.46 ± 0.14	67.48 ± 0.56
FA30BA0	41.77 ± 0.72	45.42 ± 0.31	59.91 ± 0.46
FA0BA10	49.5 ± 0.64	56.40 ± 0.47	58.01 ± 0.17
FA0BA20	49.24 ± 0.91	50.90 ± 0.71	56.76 ± 0.91
FA0BA30	47.02 ± 0.14	50.64 ± 0.97	57.94 ± 0.50
FA10BA10	43.04 ± 0.26	51.71 ± 0.56	58.07 ± 0.50
FA20BA20	45.49 ± 0.51	52.04 ± 0.58	57.33 ± 0.11
FA30BA30	33.58 ± 0.67	38.1 ± 0.54	46.81 ± 0.33

the control sample, which is FA0BA0. It shows that the replacement of FA has improved the compressive strength of the SCC. The highest strength was recorded from FA10BA0 sample with 68.79 MPa at 28 days. The increase in strength of fly ash concrete may be attributed to continuous hydration and the filling of pores with Calcium Silicate Hydrate gel formed due to pozzolanic action of coal fly ash [16].

In the meantime, for compressive strength with a different sand replacement of 10, 20, and 30% of BA shown that strengths were observed range of 33–50 MPa, 41–57 MPa, and 49–59 MPa at 14 and 28 days respectively. SCC incorporated with BA gained higher intensity at the early age of the SCC, varying from 47 to 49 MPa, but steadily increased at 14 and 28 days with the highest reported value from the FA0BA10 sample at 58.01 MPa.

3.1 Effect on pH for Toxicity Characteristic Leaching Procedure (TCLP) and Synthetic Precipitation Leaching Procedure (SPLP) Tests

One of the crucial factors that affect the leaching of heavy metals concentrations is the pH value. Yu et al. [17] suggested that the leachability of heavy metals is much dependent on the pH of leaching. Therefore, all the samples were subjected to TCLP leachate (pH 2.88 ± 0.1) and SPLP leachate (pH 4.2 ± 0.1). The pH results were recorded after 18 h of agitation using a rotary agitation apparatus. The results were shown in Fig. 1.

The results showed that the pH value for TCLP leachant for all the samples was much lower than the pH value for SPLP. In TCLP leachants, the lowest value was recorded with pH 5.2 from the FA30BA30 sample. Meanwhile, the highest value for pH in TCLP leachants was recorded from FA0BA0 sample with pH 11.4. Samples with replacement in FA only were demonstrating the decreasing value of pH with

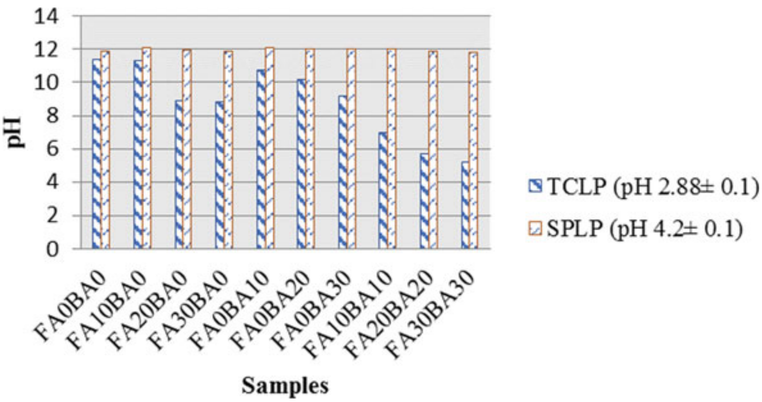

Fig. 1 Comparison of pH for TCLP and SPLP samples

the increment of the percentages of FA replacement. Similar trends were observed for samples with the combination of FA and BA.

The pH values of SPLP leachant in the samples were recorded in the range of 11.78 to pH 12.09. It is probably due to the alkaline nature of the materials that could significantly change the initial pH of the leachant [18]. It resulted in lower heavy metals were leached in SPLP compared to TCLP samples. Besides, Kim et al. [19] also suggested that most of the elements in FA and BA only slightly soluble. Heavy metals are most soluble in acidic leachant while those elements that form oxyanions are more soluble at high pH.

3.2 Comparison of Leachability of Heavy Metals Using TCLP and SPLP Method

The findings of the leachate analyzed were compared with the concentration limits for heavy metals set by the USEPA [20] and the value recommendations for chemicals of health significance in drinking water [21]. Also, both methods used identical particle sizes, which are smaller than 9.5 mm. However, there was a gap in the leaching fluid used in the SPLP and TCLP experiments. It was confirmed that most of the TCLP heavy metal concentrations were higher than the SPLP results.

Figures 2, 3, 4 and 5 shows a comparison of SCC heavy metal concentrations using SPLP and TCLP for all samples. Both test methods reported significant leachate concentrations of target metals for FA and BA content. However, due to the disparity in the leaching fluid used in the TCLP and SPLP experiments, the leaching concentrations with marginal variations can be observed. Concentration, volatility and solubility are several variables that assess the capacity for leaching [22]. Most of the TCLP heavy metal concentrations were slightly higher than the metal concentrations found in the SPLP test, particularly for As. It is because metal solubility usually decreases

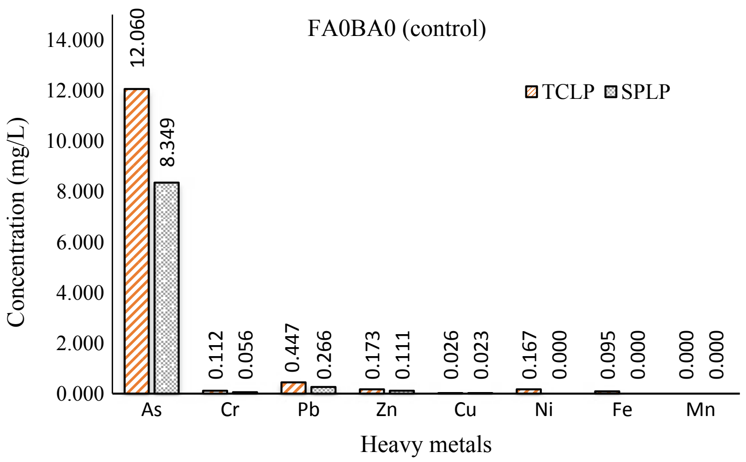

Fig. 2 Comparison of heavy metals concentrations for SCC using TCLP and SPLP for FA0BA0 (control) sample

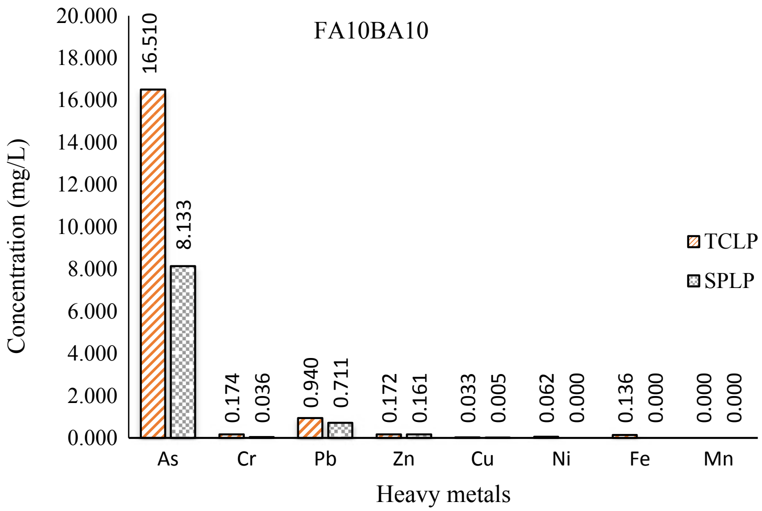

Fig. 3 Comparison of heavy metals concentrations for SCC using TCLP and SPLP for FA10BA10 sample

with a rise in pH. TCLP involved leaching under slightly acidic buffered conditions with pH 2.88 and pH was 4.2 in the SPLP test. Other heavy metals consisting of Pb, Zn, Ni, Fe and Mn have been leached at concentrations that are exceptionally low and do not reach the limit of the permissible concentration in leachate. That is because FA consists of aluminium oxide and iron hydroxide, which are common sorbents for the removal of Arsenic from water.

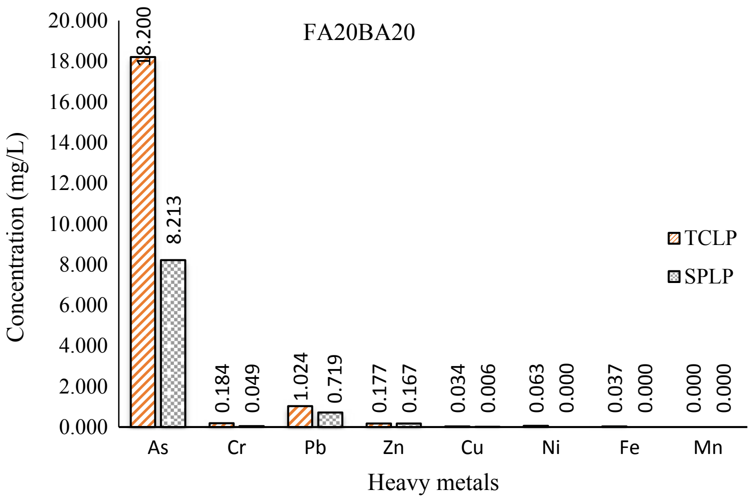

Fig. 4 Comparison of heavy metals concentrations for SCC using TCLP and SPLP for FA20BA20 sample

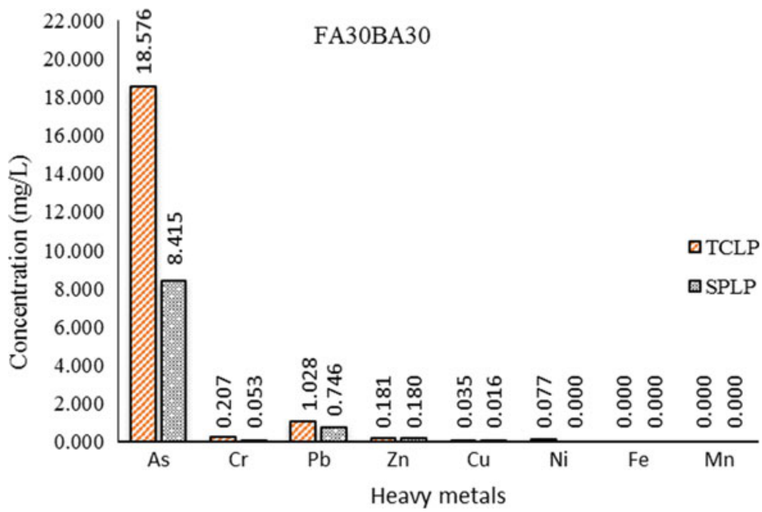

Fig. 5 Comparison of heavy metals concentrations for SCC using TCLP and SPLP for FA30BA30 sample

The element concentrations in all TCLP samples were below the acceptable limit set by the USEPA, except for Arsenic. In comparison, samples with the substitution of FA alone and samples with a combination of FA and BA resulted in higher leaching of As compared to samples with BA alone. The highest leaching of As was reported in the FA30BA30 sample at 18.576 mg/L. Arsenic has gained significant popularity as it is mobile over a broad pH range. Arsenic acid solution releases increase with pH, although this pattern is reversed in alkaline solutions [23]. The difference in leaching is caused by the pH dependency of most heavy metal elements [22].

A similar pattern observed in SPLP test which all elements were below the USEPA limit except for As. SPLP using a leachant with a pH similar to that of groundwater or surface water where the coals are used, stored, or disposed of, rather than the acetic acid of TCLP could be a more representative test for whether the waste materials might be hazardous based on its toxicity. Therefore, the concentrations of the elements in SPLP leachates, the results are compared to the World Health Organization limit for drinking water [21]. The results demonstrated that elements such as As and Pb exceed the WHO guidelines for drinking water quality. The highest concentration for As and Pb elements were recorded higher from sample FA30BA30 with 8.415 and 0.746 mg/L, respectively. Other than that, the FA0BA0 sample was the only sample that exceeds the WHO guidelines for Cr with 0.006 mg/L higher than the guidelines that consider extremely low concentrations. Elements such as Cu, Ni, and Mn were below the guidelines or not detected in SPLP. Meanwhile, Zn and Fe are not of health concern at concentration normally observed in drinking water as has been stated in the WHO guidelines.

4 Conclusion

Based on the characteristics of FA and BA, it was found that the concentrations of elements in FA were usually higher than in BA. FA thus displays higher concentrations of heavy metals compared to BA and OPC. As for compressive strength, it indicates that compressive strength increased from day 7 to day 28. By comparison to BS EN 206:2013, the sample strength classes ranged from class C45 to class C70 at 28 days and met the criteria to be graded as normal-weight and heavy-weight concrete. The other goal was to assess the leachability of SCC heavy metals incorporated with FA and BA. From TCLP, Arsenic leaching was the only heavy metal that leached out of the samples and exceeded the limit set by USEPA. The highest value for As was reported in FA30BA30 sample with 18.576 mg/L and indicated the highest value of all samples. Meanwhile, for SPLP, the findings again indicate that the highest concentration of heavy metals leached from the samples was As. The concentrations of Arsenic in the control sample are 8.349 mg/L. For FA and BA samples, which are FA10BA10 (8.133 mg/L), FA20BA20 (8.213 mg/L) and FA30BA30 (8.415 mg/L) of As concentration. The pH importance of the leaching agent is an important factor influencing the leaching of heavy metals. TCLP findings indicate higher value relative to SPLP results because the leaching agent is acidic compared to SPLP. In conclusion, a sustainable approach to the reuse of FA and BA in this research is useful for environmental and construction purposes.

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