RESEARCH ARTICLE



Leaching behavior of sustainable concrete made with coal ash wastes as replacement of cement and sand

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Received: 29 March 2022 / Accepted: 4 October 2024

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Abstract

The coal-fired power plant fly ash (FA) and bottom ash (BA) are gradually used as alternative materials in the concrete. However, knowledge of the leaching characteristics of using both incinerator ashes in concrete production is lacking. This work aimed to evaluate the leaching behavior of the FA and BA used in concrete production by employing batch and tank leaching tests. The outcomes of both leaching tests showed that there was no considerable leaching of any trace elements to the environment, and it remains much lower than standard limitations for utilization as construction materials. The results of cumulative mass discharge showed that the slope of flux time for all elements was less than 0.4 and because of that, primary surface wash-off was the main discharge process of all the heavy metals. Strength test results revealed that there was not much difference between coal ash concrete (CAC) and the control mix at the initial age of curing time. Despite that, at a long period of curing time (180 days), the compressive strength of CAC containing 20% FA as cement replacement and 100% BA as fine aggregate replacement increased by 76% due to the pozzolanic reaction of BA and FA in comparison to the normal concrete, while, due to the high porosity of BA, the workability of CAC decreased by 50%. The outcomes of the current work revealed that the combined use of FA and BA can be counted as a promising alternative in the production of sustainable concrete for structural applications toward sustainable development.

Graphical abstract



Responsible Editor: José Dinis Silvestre

Highlights

- Coal bottom ash and fly ash were added to concrete.
- The leachability and chemical properties of those concretes are discussed.
- There is no leaching of any trace elements to the environment.
- The release mechanism of all trace elements was the initial surface wash-off.
- Sustainable concrete can be produced using coal ashes.

Extended author information available on the last page of the article

Keywords Coal bottom ash \cdot Coal fly ash \cdot Leaching \cdot Sustainable concrete \cdot Sustainable developments \cdot Environmental science

Introduction

As the utilization of coal by coal factories escalates, so does the generation of industrial waste by-product materials known as bottom ash (BA) and fly ash (FA) (Mahedi et al. 2020). Based on the statistics, roughly 150-250 million tons of BA and around 650-800 million tons of FA have been generated worldwide annually (Wang et al. 2020). Nowadays, developing countries become more serious about the environmental problems of this waste material which is resulting in the regulation of new instructions for constructing new national projects such as coal-fired power plants (Rafieizonooz et al. 2016). On the other hand and according to the statistics, in 2024 fossil fuels plays an important role in the production of more than 83% of electric power in Malaysia which includes 58% coal and 42% gas. It indicates that the utilization of coal as a fossil fuel for the generation of electric power will be increased from 43 to 58% from 2014 through 2024 (Tenaga 2014). It can be said that by growing the consumption of coal, the amounts of coal ashes (BA and FA) will increase.

Besides, with the aim of growth sustainability in the construction industry, the recycling of different forms of waste materials is essential (Mohammadhosseini et al. 2017). It is also necessary to discover effective methods to decrease the utilization of non-renewable natural resources in the construction industry and reduction in the generation of high volumes of waste by industrial sectors. Therefore, toward sustainable development, the recycling of by-product materials such as coal-fired power plant wastes (BA and FA) as construction materials can be considered. An eco-friendly, green, and sustainable concrete can be manufactured by partial or total substitution of cement and aggregates with waste materials such as FA and BA, respectively (Rafieizonooz et al. 2016). It is reported that more than 80% of produced FA worldwide had been utilized in the production of cement or cement-based materials (Du et al. 2022; Meena et al. 2023). The utilization of BA and FA in the production of sustainable concrete showed a positive contribution in terms of higher quality and better strength (Hasim et al. 2022; Teixeira et al. 2022; Kaladharan and Rajabipour 2022). Also, there are other advantages of using BA as fine aggregate and FA as a supplementary cementitious material (SCM) in the production of concrete such as reducing the dead weight of the structure and increasing the final strength of concrete at a long period of curing time as well as mitigation of environmental pollution (Singh and Siddique 2014a,

b). The reduction in CO_2 emission and decreasing the use of natural resources are other advantages of using FA and BA as SCM and fine aggregate substitution (Rafieizonooz et al. 2017a; Lu et al. 2019; Çankaya and Pekey 2019; Liu et al. 2020).

Disposal of unused FA and especially BA is costly and places an extensive amount of burden waste on the power plant site. It can be said that the disposal of BA and FA in landfills can be a factor in the continuing problem of reduction in landfill areas which leads to environmental problems (Khankhaje et al. 2015, 2017; Nikravan et al. 2020). Additionally, in some countries, FA and BA are classified as hazardous waste due to the considerable amounts of heavy metals (Quina et al. 2018). On the other hand, it is also reported that heavy metals and trace elements in BA and FA in utilization have not been safely monitored and need more investigation (Jain et al. 2022; Wang et al. 2022a). In this respect, the potential risks to human health and the surrounding environment caused by the leaching of heavy metals and trace elements in coal BA and FA cannot be neglected (Yu et al. 2022; Wang et al. 2022b). Therefore, the presence of heavy metals in FA and BA must be evaluated before such waste can be used as construction replacement materials (Rehman et al. 2020).

The heavy metal concentrations and leaching performance of FA, BA, and conventional concrete (separately) were investigated in different countries such as Australia (Jankowski et al. 2006), the USA (Monroy Sarmiento et al. 2019; Mahedi and Cetin 2020), India (Sushil and Batra 2006; Prasad and Mondal 2008), South Africa (Musyoka et al. 2013), and Zimbabwe (Gwenzi and Mupatsi 2016). Some studies such as those of Sushil and Batra (2006) and Mahedi and Cetin (2020) mentioned that few amounts of heavy metals can be diffused out from coal ash, while other studies (Gwenzi and Mupatsi 2016; Rafieizonooz et al. 2017b; Nikravan et al. 2020) stated that the diffusing of the heavy metals from FA, BA, and normal concrete was not important for consideration. These contradictory findings could be due to various reasons such as using different leaching tests, varieties of boilers, the combustion temperature, diversity in the source and species of coal combusted, burning situations in the furnace, and finally ash managing performance.

FA compared to BA is a well-recognized material, and many studies addressed its advantages and disadvantages as a replacement construction material. On the other hand, the utilization of FA because of its negative effects on the strength performance of concrete is limited to a partial replacement, whereas BA can be utilized as a total replacement for fine aggregate. Furthermore, the environmental aspects of the utilization of coal ash concrete (CAC mixture) containing both BA and FA as replacement materials at the same time, including the leachability of heavy metals and trace elements, are not well documented. Thus, for the utilization of these kinds of industrial by-product materials in the production of sustainable concrete, there is a gap that needs to be investigated. Therefore, in this study, two independent leaching methods including toxicity characteristic leaching procedure (TCLP) and semi-dynamic tank leaching procedure (SDTLP) tests were utilized to investigate the probability of the leaching of heavy metals and trace elements from the CAC mixture, FA, and BA, separately. Besides, from the relationship between leaching time and leached concentrations of different elements, the main mechanism of leaching was derived and presented. Finally, the results of the mechanical performance of CAC mixtures including compressive, tensile, and flexural strength were reported to prove that these concrete mixtures can be utilized as a replacement for conventional and structural concrete in construction projects (Sam 2020; Tan et al. 2021). The structure of this paper is provided as follows. In "Materials and methods" section, materials and methods are discussed and the research methodology is presented. "Detailed discussion" section presents the results of this work and a detailed discussion. Lastly, in "Conclusions" section, the research findings are summarized, and future recommendations are provided.

Materials and methods

Materials

Following ASTM C150 (2007), ordinary Portland cement (OPC) was consumed in the current work. OPC consumed had final and initial setting times, specific gravity, blain surface area, and soundness of 210 min and 125 min, 3.15, 3990 cm²/g, and 1.0 mm, respectively. A single source of BA as substitution of fine aggregate and FA conforming to ASTM C618 and ASTM C618-15 (2012) as partially cement substitution were collected from a Malaysian coal-fired power plant. FA utilized in the current research had a soundness,

blain fineness, and specific gravity of 1.0 mm, $3450 \text{ cm}^2/\text{g}$, and 2.45, respectively.

The chemical structure of OPC, FA, and BA was provided in the previous study by the authors (Rafieizonooz et al. 2016) and is presented in Table 1. The chemical composition indicated that FA was mainly comprised of iron, alumina, and silica. According to the percentage sum of SiO₂, Al₂O₃, and Fe₂O₃ which was 78.82% and in agreement with ASTM C618 and ASTM C618-15 (2012), the FA was categorized as class F. BA was mainly composed of SiO₂, Al₂O₃, and Fe₂O₃ with minor amounts of MgO, SO₂, and CaO.

Fine aggregate was collected from the Sungai Sayong River quarry located in the north part of Johor, Malaysia. The fine aggregate utilized in this work was in agreement with the specification of ASTM C778 (2014). The particle size gradation of BA and river sand and their limitation were presented in the previous work of authors (Rafieizonooz et al. 2016). The upper and lower limits were referring to the river sand in agreement with ASTM C33 / C33M (2013).

The crushed coarse aggregates were obtained from a local quarry and 20 mm was the maximum size of crushed aggregates. The porosity and irregular shape of BA particles resulted in lower specific gravity and higher water absorption of BA in comparison to sand. From the results of the current work, water absorption of BA, fine, and coarse aggregate was 11.61, 7.4, and 0.61, respectively. It can be seen that the water absorption of BA is almost 36% higher than the water absorption of river sand. Moreover, the specific gravity of BA, fine, and coarse aggregate were 1.88, 2.62, and 2.69, respectively. In future studies, further experiments should be conducted to analyze the effect of adding BA on the density and water absorption of coal ash concrete.

Mix proportions

In the first stage, different mix ratios of concrete containing BA as river sand substitution and FA as a substitution for OPC were made. The mixtures' compressive strength and workability revealed encouraging results by employing 20% FA as cement substitution and a diverse portion of BA as river sand replacement. Therefore, CAC mixtures named CAC1, CAC2, CAC3, and CAC4 with a fixed percentage of FA (20%) and a diverse range of BA (25, 50, 75, and 100%, respectively) were chosen for additional experiments. The mix design with the results of the workability test is presented in Table 2. Saturated surface dry (SSD) fine aggregate

Table 1Chemical compositionof BA, FA, and OPC

| Component | CaO | Al ₂ O ₃ | Fe ₂ O ₃ | SiO ₂ | TiO ₂ | K ₂ O | MgO | P_2O_5 | SO ₃ | MnO |
|-----------|-------|--------------------------------|--------------------------------|------------------|------------------|------------------|------|----------|-----------------|------|
| BA | 8.70 | 18.10 | 19.84 | 45.30 | 3.27 | 2.48 | 0.97 | 0.35 | 0.35 | 0.29 |
| FA | 10.70 | 23.80 | 7.42 | 47.60 | 2.92 | 1.68 | 1.50 | 1.16 | 0.76 | 0.12 |
| OPC | 62.39 | 5.20 | 4.19 | 20.43 | - | 0.01 | 1.55 | - | 2.11 | - |

Table 2 Mix proportion

| Name | Binder (375 kg/m ³) | | Fine aggre- gate (780 kg/ m ³) | | Water to cement (%) | Coarse aggregate (kg/m ³) | Slump (mm) | |
|---------|---------------------------------|--------|--|--------|---------------------------|---|------------|--|
| | OPC (%) | FA (%) | Sand (%) | BA (%) | | | | |
| Control | 100 | 0 | 100 | 0 | 0.55 | 1035 | 73 | |
| CAC1 | 80 | 20 | 75 | 25 | 0.55 | 1035 | 92 | |
| CAC2 | 80 | 20 | 50 | 50 | 0.55 | 1035 | 76 | |
| CAC3 | 80 | 20 | 25 | 75 | 0.55 | 1035 | 53 | |
| CAC4 | 80 | 20 | 0 | 100 | 0.55 | 1035 | 37 | |

(sand) and BA were consumed, and a fixed value of water to cement ratio (w/c) equivalent to 0.55 was utilized in all the CAC mixes. The BA was utilized by mass in CAC mixtures as a substitution for river sand (fine aggregate). The volume of OPC was 375 kg/m³ and to calculate the percentage of the mixtures, the American standard for selecting proportions for concrete (American Concrete Institute (ACI) 211 2002) instruction was employed.

Testing procedure

Chemical properties

The chemical composition of BA and FA was carried out using X-ray fluorescence spectrometry (XRF). Samples of FA and BA with particle sizes lower than 90 μ m were submitted for chemical analysis. Moreover, for the elemental evaluation, the inductively coupled plasma-mass spectrometry (ICP-MS) method was used. Samples of BA and FA with less than 45 μ m particle sizes were submitted for chemical analysis. Also, all the extracted samples from both leaching methods were analyzed using ICP-MS.

Leaching test

SDTLP approach and TCLP method were employed in the current work for determining the leaching properties of BA, FA, and CAC mixture.

TCLP To evaluate the possibility of heavy metal mobilization, 50 g oven-dried specimens of crushed CAC mixture, FA, and BA were utilized. Then, all samples were placed in 2-L high-density polyethylene (HDPE) bottles, separately. In agreement with the TCLP procedure (US EPA 1311 2015), the maximum particle size of the BA and crushed CAC mixture was 9.5 mm. Subsequently, extraction liquid with a volume of 20 times the mass of the solid phase was inserted. The extraction liquid utilized was selected due to the alkalinity of the materials. The procedure for selecting the extraction liquid was provided based on the method in Rafieizonooz et al. (2022). After that, a rotary agitation machine was used to put the bottles inside it for 18 h with an average speed of 30 rotations per minute as indicated by TCLP. For more accurate results, triple samples were utilized and analyzed. To prevent any alteration in concentration, the leachates were preserved approvingly acidic by adding nitric acid (the pH should be lower than 2) and stockpiled at 4 °C for heavy metal analysis. Heavy metal concentrations in the leachate were measured using ICP-MS (Gwenzi and Mupatsi 2016).

SDTLP Three cubic CAC4 mixtures with the size of 100 mm for each side, involving 100% BA as river sand and 20% FA as a substitution of OPC, were completely immersed in separate 10-L containers according to the requirement of the SDTLP approach EPA 1315 (2017). The containers were filled with 5.4 L of distilled water as the leachate achieves a liquid-to-solid ratio of 9:1 (mL/cm²) of the monolith thickness. Leachate was collected and substituted with the fresh reagent water after 2 h and 1, 2, 7, 14, 28, 42, 49, and 63 days subsequently. The leachates were analyzed for pH and electric conductivity straightaway. Then, the leachates were acidified (pH < 2) with nitric acid and stored at 4 $^{\circ}$ C until final analysis with ICP-MS (da Silva et al. 2018). At the end of the arrangement of leaching intervals, the cumulative mass release, average interval flux, and interval mass (Garrabrants et al. 2012; US EPA 1315 2017) were calculated for every leaching interval employing Eqs. (1) to (3) in Rafieizonooz et al. (2022).

Strength properties and workability performance

For evaluation of the workability performance of all mixtures at the fresh state from the time they are mixed until they are set, the slump test was performed. At the hardened state, the compressive and splitting tensile strength of all mixtures were measured at 7, 28, 91, and 180 days of curing age following ASTM C39/39M (2021) and ASTM C496 (2017), respectively. Moreover and in agreement with ASTM C78/78M (2022), flexural strength was evaluated at 7, 28, 91, and 180 days of curing period utilizing beams of $100 \times 100 \times 500$ mm.

Detailed discussion

Leaching behavior

The performance of coal ash concrete, BA, and FA, in terms of leaching behavior, was analyzed. In this section, the results of the heavy metal analysis, TCLP, and SDTLP approaches were explained in detail based on the laboratory stipulation upon which the experiments were performed.

TCLP

The leaching performance of BA, FA, and pulverized pieces of CAC4 mix (incorporating 100% BA as a total substitution for fine aggregate (river sand) and 20% FA as a partial substitution for OPC) was investigated by using EPA method 1311 called the TCLP (US EPA 1311 2015). This method has been utilized by various previous studies to investigate the suitability of industrial by-product materials as replacements for construction materials (Kadir et al. 2016; Gooi et al. 2020; Prasetia et al. 2021; Petrović and Fiket 2022; Curpen et al. 2023). In this respect, the CAC4 mixture was selected because it contains the highest amount of BA (100%) as a substitution for river sand and consequently had the highest risk of heavy metal leachability compared with other CAC mixtures. For the TCLP method, extraction fluid 1 was chosen following the US EPA 1311 (2015) and came from the initial pH of the FA, BA, and CAC4 mixture. The results of the initial elemental analysis by ICP-MS are presented in Fig. 1.

As shown in Fig. 1, only mercury (Hg) in BA and crushed CAC4 particles exceeded the standard limitation (higher than 0.2 mg/L for Hg). According to the chemical composition of BA and CAC4 particles, they could be considered hazardous materials (before performing leaching tests). However, only the chemical composition of materials is not



Fig.1 BA, FA, and CAC4 mixture heavy metal experiment result (mg/L) $\,$

It must be considered that the results of the initial elemental analysis which was conducted without extraction fluid for BA, FA, and CAC mix (Fig. 1) should be compared to the leaching test results of the TCLP approach. In this respect, the outcome of the TCLP test employing extraction fluid for BA, FA, and CAC4 is presented in Fig. 2. It can be said that there is a big difference between the results of the initial elemental analysis and the leaching test. Based on these results, all the elements, even mercury (Hg), presented very low leaching in comparison with the standard regulation and initial elemental analysis. Furthermore, the leaching of the heavy metals in crushed particles of the CAC4 mixture decreased at a higher rate than those when compared to BA and FA. The main reason for this reduction could be attributed to the encapsulation development of the cement paste in the concrete matrix which results in the binding of heavy metals and trace elements (Yu et al. 2022; Wang et al. 2022b).

For all the materials (BA, FA, and CAC4 mixture) and in comparison with the standard limitation in the TCLP 1311 method, lower concentrations of all the heavy metals were found in the leachates. The greatest solubility values were detected for Pb, Cd, and Ag for FA and BA samples. However, CAC4 particles indicated lower solubility values when compared to the BA and FA and it should be related to the concrete matrix and its encapsulant process. Therefore, from the results of the TCLP method, BA, FA, and CAC mixture can be categorized as non-hazardous materials and cannot be regarded as a matter of concern. The elements of Hg and V in CAC4 particles showed lower concentrations than the limit value of detection by the ICP-MS. Derived from the results of the TCLP method, it can be reported that releases of the selected heavy metals were detected lower than the standard limitation values which are stated in EPA (US EPA 1311 2015).



Fig. 2 BA, FA, and CAC4 granule TCLP experiment outcome (mg/L)

Results revealed that for all the heavy metals the maximum extraction ratio is less than 20% of the total initial concentration and it remains less than the limitation value following the US EPA (2015). These results were achieved in the lab condition and the effects of exposure to the atmosphere such as carbonation and chloride penetration were not considered. Long-term leaching tests involving exposure to the atmosphere should be considered for further investigation in future studies. However, pH dependence in combination with monolith leaching must be conducted for identifying the exact leaching performance of concrete as a compacted and impervious material under field exposure conditions.

The outcome of this study is in agreement with previous research (Kosson et al. 2014; Mehra et al. 2016; Gwenzi and Mupatsi 2016). They discovered that the concrete matrix efficiently produces a chemical encapsulating and connecting for holding the heavy metals within the concrete. According to their findings, the leaching of the heavy metals from BA and CAC was unimportant. Due to the utilization of crushed particles of CAC4 mixtures in the TCLP method, it can be said the recycling process of CAC mixes as rubble can be accounted for without any concern about the leaching of heavy metals and trace elements into the surrounding environment.

On the other hand, it is reported that heavy metals and trace elements in BA and FA in utilization over a long period of time have not been safely monitored and need more investigation (Jain et al. 2022; Wang et al. 2022a). In this respect, the potential risks to human health and the surrounding environment caused by the leaching of heavy metals and trace

elements in coal BA and FA cannot be neglected (Yu et al. 2022; Wang et al. 2022b). Therefore, it can be concluded that by considering TCLP results and after conducting long-term leaching tests (SDTLP), recycling, loading, transporting, and application of FA and BA might be accounted as substitution construction substances.

SDTLP

The specimen was a CAC4 mix incorporating 100% BA as a replacement for small aggregate (sand) and 20% FA as a substitution for OPC. The initial results of the SDTLP test for total concentrations of all elements are shown in Table 3. The outcomes were obtained from the mean of three samples from the same mixes. In agreement with the result of the TCLP test, only a few amounts of Pb, V, Cd, and Ag were detected in all samples while the rest of the elements were lower enough to be considered. The negligible leaching of the heavy metals for the rest of the elements in all three samples has to be attributed to the slow release because of the indirect flow path among the solid and liquid phases as well as encapsulant development in the concrete matrix (Cao et al. 2017). In addition to that, other reasons for the less concentration of other heavy metals can be highly alkaline situations and pH-dependent stabilization of the experiment. In this respect, Zhang et al. (2022) reported that due to exposure to the atmosphere, the carbonation process can impact observed diffusivity, physical retention, and solubility of heavy metals and trace elements. Therefore, in future studies, the effects of carbonation on the diffusion of heavy metals from the CAC mix should be investigated. According

| results of | Element code | Tank leaching test (µg/L) | | | | | | | | | |
|------------|--------------|---------------------------|-------|-------------|------|------|------|------|------|------|--|
| | | T01 | T02 | Т0 3 | T04 | T05 | T06 | T07 | T08 | T09 | |
| | | 2 h | 23 h | 23 h | 5 d | 7 d | 14 d | 14 d | 7 d | 14 d | |
| | As | 1.2 | 1.7 | 0.38 | 0.24 | 0.84 | 0.58 | 0.75 | 0.48 | 0.54 | |
| | Ba | 0 | 0 | 0 | 7.1 | 8.8 | 14.5 | 12.7 | 0 | 0 | |
| | Be | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | Cd | 254 | 59 | 78 | 26 | 15 | 0.07 | 0.17 | 0 | 0 | |
| | Cr | 10.7 | 9.9 | 6.8 | 10.1 | 8.5 | 2.9 | 2.4 | 0.46 | 0.92 | |
| | Со | 0.03 | 0.001 | 0.003 | 0.06 | 0.05 | 0.04 | 0.03 | 0 | 0 | |
| | Cu | 0.6 | 0.01 | 0.67 | 1.6 | 0.5 | 0.4 | 0 | 0 | 0 | |
| | Pb | 33.5 | 1.9 | 4.5 | 4.6 | 10 | 2.9 | 3 | 2.2 | 2.3 | |
| | Ni | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | Se | 0.58 | 0.54 | 0.09 | 0.42 | 0.18 | 0.23 | 0.21 | 0.09 | 0.12 | |
| | Ag | 193 | 121 | 36.9 | 37.9 | 12.9 | 14.8 | 6.9 | 4.5 | 5.9 | |
| | Ti | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | V | 9.1 | 14.6 | 3.7 | 6.8 | 7.7 | 9.2 | 8.5 | 3.1 | 3.0 | |
| | Zn | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | |
| | Eluate pH | 10.3 | 11.1 | 10.6 | 11.5 | 11.4 | 11.6 | 11.3 | 11.1 | 11.2 | |
| | | | | | | | | | | | |

Table 3 Preliminary

SDTLP

to Rowe (2005), with an increase in the thickness of materials, the release process of heavy metals decreased. Therefore, utilizing thicker concrete than test samples (bigger than 100*100*100 mm) should result in an additional reduction in the risk of heavy metal leaching.

Considering the initial results of the SDTLP test, heavy metals including V, Pb, Ag, and Cd were selected for further analysis. The outcomes of the monolithic leaching experiment can be employed to calculate the heavy metal element release procedure from the CAC4 mixture. Figure 3 represents the result of concentration diffusion for the above-mentioned heavy metals versus leaching time (days). Cd, Ag, V, and Pb were found in concentrations less than 254, 193, 14.6, and 33.52 μ g/L in all extractions. Based on the results, the concentration release of all the heavy metals was less than their standard limitation following EPA 1315 (2017).

Figure 4 presents the cumulative concentration of the heavy metals in comparison with the standard limitation. It can be reported that all the heavy metals did not exceed their standard limitations following all the leaching interval periods. Thoroughly, the cumulative concentration discharge at the end of the last interval (after 63 days of immersion period) for Cd, Ag, V, and Pb were 0.434, 0.434, 0.065, and 0.062 mg/L as compared to the standard limitation of 1.0, 5.0, 24.0, and 5.0 mg/L, respectively.

Previous studies (Hayes et al. 2015; Torras et al. 2011) found that trace elements might have been emitted by a primary surface wash-off process and not with the diffusion of this analyte throughout the concrete matrix. After the primary surface wash-off, the existing trace elements were well encapsulated by the cement paste and were under the standard limitation specified by U.S. EPA 1315 (2013).

The insignificant leaching of the heavy metals observed in the specimen leaching experiment can be concluded that these elements are immobilized and encapsulated by the cement paste. These results can be verified by previous studies which have explained that concrete microstructure enriched with heavy metals was discovered to encapsulate these components (Hayes et al. 2015; Gwenzi and Mupatsi 2016).

According to Serclérat et al. (2000), the mortar bars of concrete were shown to leach low concentrations of Pb when immersed in distilled water. The addition of BA and FA had minimal effect on the diffusion of the heavy metals from the coal ash concrete. It explains that there is an insignificant risk attributed to the leaching of the heavy metals from the monolithic concrete involving BA and FA to the groundwater resources and surrounding area.

Figure 5 represents the accumulative mass discharge of the heavy metals versus the leaching time. For the CAC4 specimen, a positive non-linear connection was found between the accumulative mass release of all the heavy metals and accumulative time. According to Van der Sloot et al. (1987), Beard (2002), Goumans et al. (2012), Garrabrants et al. (2014), and Gwenzi and Mupatsi (2016), if the slope (s) of the curve for log–log plots were greater than 0.8, the mechanism of release was matrix dissolution. On the other hand, if it was between 0.8 and 0.4, the release mechanism was diffusion-controlled release and finally, if

Fig. 3 Concentration release of **a** Cd, **b** Pb, **c** Ag, and **d** V versus time (days)



10000



10000



it was lower than 0.4, the mechanism of release was initial surface wash-off.

Figure 5 shows the slope (s) of the curve for log–log plots for heavy metals. According to this figure, the slopes for Cd, Ag, V, and Pb were 0.159, 0.211, 0.070, and 0.043 (mg/m^2) , respectively. From these results, it can be concluded that the slope between the cumulative mass release and the cumulative leaching time for all the heavy metals was below 0.4. Therefore, based on these findings and

compared to the instruction mentioned earlier, primary surface wash-off was the main process of discharge for all the heavy metals. In other words, with increases in the experiment time, the mass release of the heavy metals decreased. The encapsulant process by concrete matrix should be the main reason for this phenomenon. This is in agreement with the result of the TCLP experiments which also found the same mechanism of encapsulant for heavy metals and also is supported by previous studies (Kosson et al. 2014; Hayes et al. 2015; Mehra et al. 2016; Gwenzi and Mupatsi 2016).

Moreover, these results are following the result of the mass interval flux discharge versus the experiment duration which is presented in Fig. 6. The mass interval fluxes of all the heavy metals across the subjected surface area of the CAC4 specimens showed reverse linear relationships to the experiment time. This indicated that with increasing contact time in subsequent leaching intervals, the mass interval fluxes of all the heavy metals decreased.

After each interval, the value of specific conductivity and pH were calculated in the eluates of the CAC4 specimens. Figure 7 presents the specific conductivity and pH which fluctuated from 10.3 to 11.6 after 63 days of leaching time. Differences between the pH values from the starting time of the test to the end of the experiment duration were lower than "1" which revealed that the pH was practically unchanged. Paulus et al. (2016) reported that sulfate ions were mainly involved with calcium in eluates, and those chloride ions were involved with sodium. It could be the main reason for high specific conductivity and a high range of pH in concrete. The release of sodium ions in our study indicates characteristics of the concrete matrix that may influence its conductivity. While tortuosity, which refers to the extended path length for diffusion due to the non-linear nature of pores, was not directly measured in this study, it is acknowledged as a factor that could potentially affect the conductivity of coal ash concrete (Garrabrants et al. 2014). Future studies



Fig. 7 pH and specific conductivity versus leaching time

could explore the detailed relationship between tortuosity and ion release in such matrices.

The electric conductivity of the CAC4 mixture ranged from 90 to 640 μ S/cm throughout the whole leaching time, with considerable growth. There are a limited number of studies about the electric conductivity performance of construction materials. However, the findings of the current work were in good agreement with those reported by Schiopu (2007), Mahedi et al. (2020), and Sun and Vollpracht (2020). Schiopu (2007) reported that the specific conductivity of conventional concrete ranged from 105 to 1340 μ S/cm.

Workability

Consumption of industrial by-products such as BA and FA as replacement materials in the production of eco-friendly





concrete not only affects the mechanical and leaching properties of concrete but also influences the fresh properties of concrete (Rafieizonooz et al. 2017a). The workability performance of CAC mixtures is affected by utilizing BA as a partial or total replacement of river sand and FA as a substitution for OPC. According to the results, both BA and FA affected the workability performance of all CAC mixtures. The result indicated that the slump value of CAC1, CAC2, CAC3, and CAC4 mixes were 92 mm, 76 mm, 53 mm, and 37 mm, respectively. The workability performance of all CAC mixtures decreased by growing the volume of BA as fine aggregate replacement. BA with 11.6% had higher water absorption than the fine aggregate (7.4%) which might be the reason for the lower workability of CAC mixes compared with normal concrete mix C0. Besides, due to the irregular shape and rough texture of BA particles, the texture of CAC mixes containing BA as a replacement for river sand was enhanced with many more porous, fined-shaped, and irregular particles which were normally very rough. Therefore, the inter-particle friction was increased and resulted in the reduction of the flow ability of fresh concrete. Thus, at the constant w/c, the workability of CAC mixes was reduced by increasing the content of BA as a substitution for fine aggregate. However, the substitution of FA as OPC had a contrasting effect on the workability of CAC mixtures and increased the workability. The outcomes of the current study can be supported by the findings of Singh and Siddique (2014a). They also revealed that by increasing the amount of BA as a substitution for river sand (fine aggregate), the workability of coal ash concrete decreased.

Compressive, tensile, and flexural strength

The results of the compressive, tensile, and flexural strength tests are indicated in Table 4. The results revealed that the progress pattern of compressive strength for CAC mixes after a certain curing time is just about similar to that of the control mix. During 7-day maturing period, concrete mixture CAC1 achieved 90.1%, CAC2 achieved 81.4%, CAC3 achieved 77.2%, and finally, CAC4 mix achieved 74.2% of the control mixtures' compressive strength. At the 91 and 180 days of curing time and due to the utilization of remaining portlandite by the pozzolanic activity of FA and BA, the

fine spread of calcium-silicate hydrate (C-S-H) paste, and production of secondary C-S-H paste, CAC mixes achieved greater compressive strength than normal concrete at the same curing time. In other words, the significant improvement in the compressive strength of the CAC mixes at long curing time can be attributed to the pozzolanic activity of FA and BA. Singh and Siddique (2015) reported that after 28 days of curing, the consumption of remaining portlandite in the concrete matrix by pozzolanic action of FA and BA was considerable. Therefore, at the long-term curing time, the compressive strength of CAC mixes increased due to the consumption of portlandite by the pozzolanic activity of FA and BA which resulted in the formation and well spread of secondary C-S-H. The idea of the encapsulation mechanism of trace elements inside the concrete structure by the formation of cement paste at a long time of curing age also can be supported by the production of secondary C-S-H paste.

At the 180 days of curing time, the compressive strength of concrete mixes CAC1, CAC2, CAC3, and CAC4 were 99.73%, 100.27%, 101.92%, and 100.49%, respectively, compared with control mix C0. The results of this study are well supported by previous research stated by Singh and Siddique (2014b) and Kim and Lee (2011). They similarly discovered that in the long term of curing time, there is no meaningful difference between the compressive strength of concrete involving BA as a substitution of fine aggregate and normal concrete. Besides, according to Kaladharan and Rajabipour (2022), the utilization of BA and FA in the production of sustainable concrete showed a positive contribution in terms of higher quality and better compressive strength.

At the 7 days of the curing period, the indirect split tensile strength of the CAC1, CAC2, and CAC4 mixtures was reduced by 3.55%, 8.87%, and 5.32%, respectively, compared with control mix C0. On the other hand, the tensile strength of the CAC3 mixture increased by 1.99% when compared to the control mix C0. The tensile strength of the CAC1 and CAC2 mixtures at 180 days of the curing period decreased by 1.29% and 2.51%, respectively. At the same time, the tensile strength of the CAC3 and CAC4 mixes increased by 24.07% and 12.68% when compared to the control mixture C0. At the early time of the curing period, except CAC3 mixture, all the CAC mixtures presented

Table 4Change in compressive,indirect split tensile, andflexural strength for control andCAC

| Code | Compressive (MPa) | | | | Tensile (MPa) | | | | Flexural (MPa) | | | |
|-----------------|-------------------|-------|-------|-------|---------------|------|------|------|----------------|------|------|------|
| Со | 20.45 | 31.03 | 34.48 | 35.95 | 1.86 | 2.54 | 2.81 | 2.95 | 3.18 | 3.98 | 4.46 | 4.63 |
| CAC1 | 18.41 | 26.49 | 33.44 | 35.85 | 1.79 | 2.29 | 2.73 | 2.91 | 3.16 | 3.67 | 4.18 | 4.39 |
| CAC2 | 16.64 | 26.33 | 34.55 | 36.05 | 1.70 | 2.16 | 2.70 | 2.87 | 3.18 | 3.77 | 4.36 | 4.58 |
| CAC3 | 15.79 | 25.01 | 34.20 | 36.64 | 1.90 | 2.66 | 3.48 | 3.66 | 3.12 | 4.10 | 4.68 | 4.88 |
| CAC4 | 15.16 | 24.59 | 33.85 | 36.12 | 1.76 | 2.48 | 3.13 | 3.32 | 3.01 | 3.84 | 4.57 | 4.77 |
| Curing time (d) | 7 | 28 | 91 | 180 | 7 | 28 | 91 | 180 | 7 | 28 | 91 | 180 |

lower tensile strength than the C0 mixture. Nonetheless, by increasing the curing time, all mixtures presented roughly similar split tensile strength when compared to control mixture C0. Similar to the compressive strength, the creation of secondary C-S–H gel because of the pozzolanic activity of FA and BA at the late curing time is the main reason for the increment in the tensile strength of CAC mixes at the higher rate than normal concrete. The outcome of the current study can be supported by Singh and Siddique (2014a) and Teixeira et al. (2022) who reported the same result for the tensile strength of concrete involving BA as a substitution of fine aggregate.

The results of the flexural strength test presented that all CAC mixes achieved just about the same strength when compared to that of the control mixture C0. With increasing in the volume of the BA as a substitution for river sand, the flexural strength of CAC mixes slightly changed. At 7 days of curing time, concrete mixture CAC1 achieved 99.43%, CAC2 achieved 99.81%, CAC 3 achieved 98.11%, and finally, CAC4 achieved 94.75% of the control concrete C0's flexural strength. In addition, at 180 days of the curing time, mixtures CAC1 achieved 94.83%, CAC2 achieved 99.07%, CAC3 achieved 105.51%, and CAC4 achieved 103.05% of the control concrete C0's flexural strength. The results of the flexural strength test were in agreement with the results of compressive and split tensile strength tests. At the early age of curing time, all the CAC mixes presented lower flexural strength than the C0 mixture. Nonetheless, by increasing the curing time, all mixes presented almost similar or higher flexural strength compared with control mix C0. It can be said that the improvement in the flexural strength of CAC mixes at a long period of curing might be attributed to the production and distribution of secondary C-S-H gel due to the consumption of portlandite by the pozzolanic activity of BA and FA. The outcome of the current work can be supported by Singh et al. (2015) who reported the same result for the flexural strength of concrete involving BA as a substitution of river sand (fine aggregate). In addition to that, the utilization of BA and FA in the manufacturing of sustainable concrete proved a convincing involvement in terms of higher quality and better flexural strength (Hasim et al. 2022).

Conclusions

In this study, two independent leaching methods including TCLP and SDTLP tests were utilized to investigate the probability of the leaching of heavy metals and trace elements from the CAC mixture, FA, and BA to the surrounding environment. Besides, from the relationship between leaching time and leached concentrations of different elements, the main mechanism of leaching was derived and presented. Furthermore, the workability and strength properties of such concrete were reported to complete the comparison between different mixtures and to support the idea of using coal ash concrete as a replacement for conventional concrete. Therefore, the following outcomes can be derived from current research:

- Initial investigation of the chemical composition of BA, FA, and crushed CAC4 particles showed that the concentration of all the heavy metals was lower than the standard limitation. On the other hand, the TCLP test revealed that none of the heavy metals, leached at a higher rate than the highest concentration of elements by standard regulation for toxicity properties.
- 2. SDTLP test results supported the outcomes of the TCLP test, which presented that all the heavy metals did not exceed their standard limitations after all the leaching interval times. The results of cumulative mass discharge showed that the slope of flux time for all elements was less than 0.4 and because of that, primary surface washoff was the main discharge process of all the heavy metals. Hence, with an increase in the leaching time, the mass release of the heavy metals decreased, and it could be due to encapsulant development by cement paste and the pozzolanic activity of FA and BA. From the results of pH and specific conductivity, a combination of chloride ions with sodium and sulfate ions with calcium in eluates are the main reason for high specific conductivity and high range of pH in the coal ash concrete, respectively.
- 3. Fresh properties of CAC mixtures revealed that with a growth in the volume of BA as a substitution of fine aggregate, the workability decreased. Some physical properties of BA such as irregularly shaped and porous particles might be the main reason for the reduction in the workability of CAC mixtures.
- 4. The coal ash concretes presented almost the same performance in terms of strength properties at the early age of curing time when compared to the normal concrete. However, by increasing the curing period, the compressive, tensile, and flexural strength of coal ash concretes increased at a slightly higher rate than the control mixture C0. The slow hydration process and delayed pozzolanic action of FA and BA at the initial days of curing time might be the feasible clarification for a reduction in the strength properties of CAC mixtures. In contrast, the production of secondary C-S-H paste due to the consumption of remaining cement by the pozzolanic activity of BA and FA might be the possible explanation for better performance of CAC mixtures than the control mix C0 in terms of compressive, tensile, and flexural strength.

From the results of this research, it can be concluded that recycling, loading, transporting, and utilization of BA and FA-collected from the same power plant-as construction material substitution should be considered to not only minimize their environmental problems but also reduce the unit cost production of environmentally friendly concrete. However, because of the nature of coal and different incineration technology in various power plants, BA and FA need to be investigated in terms of toxicity performance before they can be used as construction material replacements. In addition to that, the results of this study were achieved in the lab condition and the effects of exposure to the atmosphere such as carbonation and chloride penetration were not considered. Long-term leaching tests involving exposure to the atmosphere should be considered for further investigation in future studies. Besides, other kinds of leaching tests such as pH-dependant leaching procedure and column leaching test should be considered for further investigation.

Acknowledgements The authors gratefully acknowledge the support received from Yonsei University School of Civil and Environmental Engineering, Seoul, South Korea.

Author contribution Mahdi Rafieizonooz: conceptualization, data curation, writing—original draft, writing—review, resources methodology. Jang-Ho Jay Kim: supervision, editing, funding acquisition, project administration. Elnaz Khankhaje: writing—original draft, writing—review, resources methodology, validation, investigation. Shahabaldin Rezania: writing, review, editing, formal analysis, validation.

Funding This work is supported by the Regional Development Research Program funded by the Ministry of Land, Infrastructure, and Transport (Grant 22NANO-B158359-03). This work is supported by the Korea Agency for Infrastructure Technology Advancement (KAIA) grant funded by the Ministry of Land, Infrastructure, and Transport (Grant 21NANO-B158359-02).

Data availability Data will be made available on request.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare no competing interests.

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