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Untreated municipal solid waste incineration ashes for cement replacement

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ABSTRACT

This paper characterised the two different ashes, namely Boiler Ash (BOA) and Residue Ash (RA), collected from Qatar's Municipal Solid Waste Incineration (MSWI) plant through a range of detailed analyses (chemical, physical and morphological). The potential utilisation of these raw MSWI ashes as supplementary cementitious material (SCM) in cement-based composites has been investigated by replacement with 0%, 10%, 20%, and 30% by weight of cement. The effect of the replacement levels on workability, setting time, and strength was investigated. Test results showed that setting time increased and workability decreased with the increase of BOA content. On the contrary, RA substitution decreased the setting time and increased the workability. The highest compressive strength was obtained in RA-incorporated mortars at a 10% replacement ratio. Beyond the 10% replacement ratio, RA incorporation significantly reduced the strength. Due to BOA's high unburned carbon content, BOA substitutions reduced the strength of the mortars. However, the impact of the replacement ratio was not as strong as those in RA mortars. Considering setting time, workability, and compressive strength, BOA and RA's optimum percentage of cement replacement was 20% and 10%, respectively. Heavy metal and salt leaching from MSWI ash-incorporated mortars were evaluated by the monolithic tank test. Results indicated that most toxic metals and salts, except Ba and Cl⁻, were stabilised in the cement matrix.

Introduction

Municipal solid waste (MSW) generation has been increasing worldwide [1] due to the increase in population and economy and rise in living standards hence requires proper management [2]. Incineration (energy-from-waste) is a commonly used technique for MSW treatment. Incineration substantially reduces the volume (up to 90%) and mass (70–80%) of municipal waste and recovers energy by generating heat [3,4]. Despite these advantages, the incineration process results in residual wastes, i.e. bottom ash, boiler ash, and air pollution control residue ashes. These residual wastes, hereafter would be called municipal solid waste incinerator ashes (MSWI ashes), contain a significant amount of toxic chemicals, such as heavy metals and toxic organic compounds, as well as alkaline salts [5–7]. Most MSWI ashes are considered hazardous wastes and threaten the environment [8–10]. Therefore, before landfilling, some pre-treatment techniques, including solidification/stabilisation (S/S), thermal treatment, and resource recovery (i.e., separation and leaching) must be applied [7,11,12]. Among these pre-treatment techniques, S/S has been applied worldwide as it needs a simple operation and low processing

costs [13,14]. Despite its advantages, S/S treatment significantly increases the mass and volume of residual waste disposed into landfills due to consuming cement and water for stabilisation [15]. S/S treatment may not be the best option where land availability is scarce (e.g. densely populated areas and small countries with limited land). Therefore, considerable efforts are being made to discourage landfilling and promote the valorisation of these residues by recycling, reusing and recovering.

MSWI ashes could be used in the construction industry as secondary raw materials depending on their physical and chemical properties. MSWI ashes could be utilised in cement production, concrete, ceramics, glass and glass-ceramics, road pavement and embankment [16,17,8,18,19,11,20–22]. Due to its high fineness, MSWI ashes are usually preferred for cement replacement (as supplementary cementitious material (SCM)), given that it satisfies most of the requirements in ASTM C618 [23]. Fresh and hardened properties of cement-based materials can be deteriorated by the presence of excessive amounts of heavy metals, organic compounds, chlorides and sulfuric anhydrides in MSWI ashes [24]. On the contrary, the high lime content of MSWI ashes may contribute to the cementitious properties [25].

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The characteristics of MSWI ashes vary depending on waste composition, the type of incineration and air pollution control technologies used, and at which location of the MSWI process it is collected [26]. In this paper, for the first time, two different MSWI ashes collected from different locations at the MSWI plant in Qatar were characterised through a range of detailed analyses (chemical, physical and morphological). This early stage of the research is aimed to examine the possibility of using untreated (without any grinding and washing) MSWI ashes as cement replacement to identify the limiting conditions of its use. The effect of the replacement levels on workability, setting time, and strength are investigated. Leaching of heavy metals and salts from the cement matrix is also presented to evaluate the environmental performance of mortars containing MSWI ashes.

Experimental program

Material collection

MSWI ashes were collected from Qatar's Domestic Solid Waste Management Centre (DSWMC), a refuse-derived incineration facility. Approximately 1500 tons of municipal waste is incinerated (at a minimum temperature of 850 °C) daily, of which 16% and 4% end up as bottom ash and fly ash, respectively [27].

MSWI ashes were collected separately from the heating boiler and the flue gas treatment system (Fig. 1). The fly ash collected from the heating boiler is called "Boiler Ash (BOA)", while the fly ash collected from the flue gas treatment system (i.e. semi-wet scrubber, powdered active carbon and bag house filters) is called "Residue Ash (RA)" hereafter. These MSWI ashes were analysed as received, i.e. without any processing (e.g. grinding, sieving and washing). The Portland cement (PC) used in this study was locally produced ordinary Portland cement (OPC) CEM I 42.5 R, corresponding to ASTM Type I cement. River sand with a maximum size of 4.75 mm was used as fine aggregate.

Preparation of specimen

The cement pastes and mortars were prepared with MSWI ashes as cement replacement at 0%, 10%, 20% and 30% by weight of cement. The mix proportions of the mortars are given in Table 1. A rotary mixer was used for mixing. The solid ingredients (i.e. PC, MSWI ash and sand) were first blended in the mixer to achieve a homogenous mix. Then, water was added and mixed for 2 min [27]. After completing the mixing procedure, the flow of mortar was measured using a flow table. Twelve 50 × 50 × 50 mm cube specimens were cast and hand-tamped from each

mortar mixture. After 24 h, the specimens were removed from the moulds and were cured in water until the testing date as required.

Leaching test

Different leaching tests (e.g. TCLP, EN-12457, and NEN 7345) can be applied to evaluate metal leaching from the mortars incorporating MSWI ashes [28]. Leaching of pollutants from the cement-based materials is mainly controlled by diffusion mechanism [29]; therefore, the Dutch tank test (NEN 7345) [30] was selected to determine the on-site leaching behaviour of the MSWI incorporated mortar specimens. After 28 days of water curing, hardened mortar specimens, i.e. cube samples (50 mm) from each batch, were immersed into a closed tank filled with 625 ml deionised water (i.e. the volume of deionised water is five times the volume of the mortar) without acidification and kept in static conditions. The leachate was collected after 8 h, 1, 2, 4, 9, 36, and 64 days. Note that the eluent (i.e. deionised water) was renewed after each leachate collection period. The collected leachate was filtered through a 0.45 µm membrane filter to remove solids and larger colloidal particles. Then, its pH was measured and stored in polyethylene test tubes and kept in the refrigerator until ion and heavy metal analysis.

Analysis conducted on materials and specimen

The methods and equipment employed for the analyses of PC and MSWI ashes along with the tests conducted on paste and mortar specimens are presented in Table 2.

Results and discussions

Characterisation of raw materials

The summary of the detailed chemical and physical characterisation of the PC and MSWI ashes are presented in Table 3. The main compounds present in PC are lime (CaO), silica (SiO₂), iron oxide (Fe₂O₃), and alumina (Al₂O₃), having values of 65.7, 18.5, 4.49 and 4.04, respectively. These four compounds constitute more than 90% of PC weight. Although both MSWI ashes are found to be enriched in calcium (i.e. lime contents are 47% and 42.5% for BOA and RA, respectively), they contain limited amounts of silicates, aluminates, and iron oxide. The sum of these three oxides is 18.2% for BOA and 4.54% for RA. In ASTM C618, the minimum value for the sum of these three oxides is specified as 70%. The main compositional difference between PC and both MSWI ashes is in terms of the latter having a much higher content

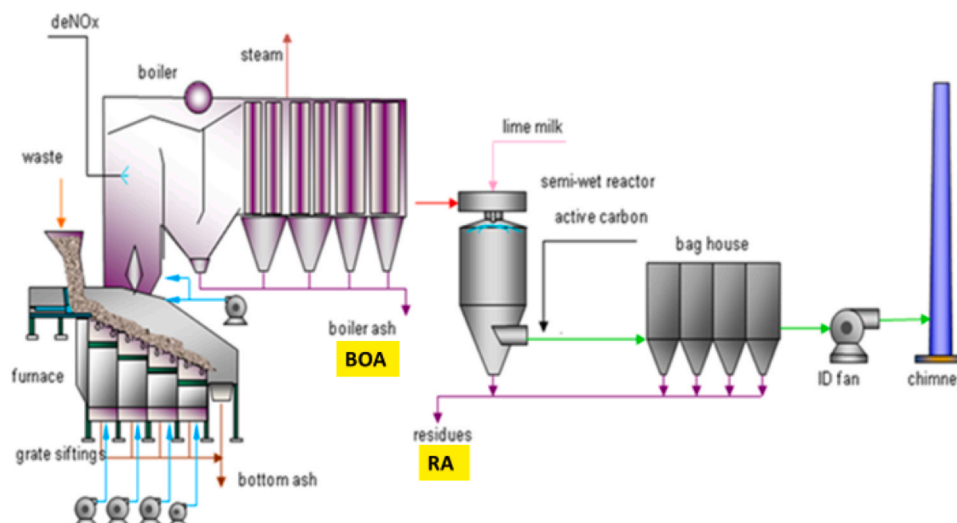


Fig. 1. Schematic illustration of incineration process in DSWMC and the generation of MSWI ashes (Boiler ash (BOA) and residue ash (RA)).

Table 1
Mortar mix design and workability of the mortars.

Mix	Mix proportion of the specimens (kg/m ³)					Workability (%)
	PC	BOA	RA	Water	Sand	
PC	400	0	0	181	1851	58
10BOA	360	40	0	181	1851	53
20BOA	320	80	0	181	1851	50
30BOA	280	120	0	181	1851	43
10RA	360	0	40	181	1851	72
20RA	320	0	80	181	1851	75
30RA	280	0	120	181	1851	83

Table 2
Test methods used.

Property	Test method/equipment or instrument
Raw Material	
Chemical Composition	Wavelength-dispersive X-ray fluorescence (XRF) spectrometer, ZSX Primus II
Loss on ignition (LOI)	ASTM C114
Particle size	Laser diffraction, LS 13 320 Particle Size Analyser
BET surface area	Chemisorption Analyzer, ChemiSorb 2750
Morphology	Scanning electron microscope (SEM), NeoScope benchtop SEM (JCM-6000)
Pastes	
Setting times	ASTM C191
Mortars	
Workability	ASTM C1437
Compressive strength	ASTM C109
Diffusion leach test	NEN 7345
Leachate	
Ion Analysis	Ion chromatography ICs 5000
Element Analysis	ICP-OES/ICP-MS/AAS

Table 3
Chemical compositions and physical properties of PC, BOA, and RA.

	PC	BOA	RA
	Weight %		
CaO	65.7	47	42.5
SiO ₂	18.5	10.4	2.74
Fe ₂ O ₃	4.49	1.7	0.602
Al ₂ O ₃	4.04	6.15	1.2
SO ₃	3.36	9.19	7.43
MgO	2.99	3.51	0.974
TiO ₂	0.27	2.14	0.492
K ₂ O	0.265	2.69	5.86
Na ₂ O	0.145	5.88	14.6
MnO	0.0954	0.0861	0.0178
P ₂ O ₅	0.0692	2.47	0.595
SrO	0.0577	0.116	0.0505
V ₂ O ₅	0.0316	-	-
Cr ₂ O ₃	0.0206	0.065	0.0224
Cl	0.0173	7.39	21.2
ZrO ₂	0.0091	0.0165	0.007
F	-	0.23	0.307
NiO	-	0.0104	-
CuO	-	0.0525	0.0756
ZnO	-	0.528	0.765
Br	-	0.0295	0.141
Y ₂ O ₃	-	0.0036	-
SnO ₂	-	0.0427	0.0652
Sb ₂ O ₃	-	0.0476	0.0559
BaO	-	0.112	0.0691
PbO	-	0.0849	0.238
LOI	1.0	9.8	2.0
Physical properties			
BET surface area (m ² /g)	1.46	8.51	5.52
Mean Diameter (μm)	28	166	33
Median Diameter (μm)	17	123	23

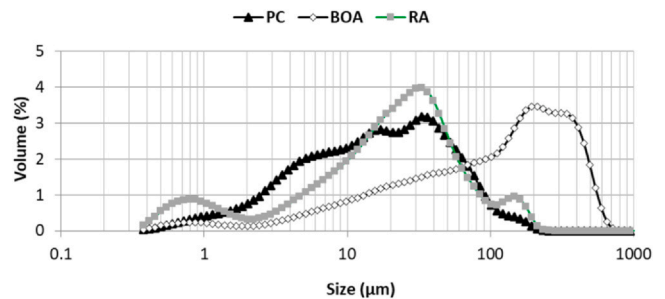


Fig. 2. Volume-based particle size distribution for PC, BOA, and RA.

of alkalis (Na₂O + K₂O), chlorides (Cl), and sulfur trioxide (SO₃) but a much lower content of silicates and iron oxides. The loss on ignition (LOI) values of PC and RA are similar (around 2%), while BOA has the highest LOI value (9.8%). The higher LOI value of BOA indicates the presence of unburned or partially burned organic particles [25,31]. Besides chemical and mineralogical composition, particle size distribution is one of the most important parameters that significantly influence the rate of hydration and strength development [32]. It has been known that the finer a by-product is, the more reactive it is. The particle size distribution of PC and RA is relatively similar (Fig. 2), with median diameters of 17 and 23 μm, respectively. However, BOA is found to have a high proportion of coarse particles (especially in the 100–600 μm size range) with a median diameter of 123 μm. These coarse particles could be made up of unburned or partially burned organic wastes and constitute unburned carbon [33]. The higher LOI value (9.8%) and large surface area (8.51 m²/g) of BOA also confirm this statement [31,25,34].

The Scanning Electron Microscope (SEM) images of PC and MSWI ashes at different magnitudes are shown in Fig. 3. It is observed that both ashes are composed of inhomogeneous, non-spherical, porous, and vesicular particles, which could provide a highly reactive surface for chemical and physical reactions [25]. It is worth noting that the colour of RA is off-white due to unreacted hydrated lime particles, which are injected for acid gas neutralisation in the air pollution control process (i.e. semi-wet scrubber).

Portland cement-MSWI ash pastes

According to ASTM C150 [35], the initial setting time of cement pastes should be longer than 45 min, and the final setting time should be shorter than 375 min. As seen in Fig. 4, the addition of BOA retards the setting times, i.e. increases both the initial and final setting times compared to the setting times of PC paste. The final setting time of the 30BOA mix is longer than 375 min. BOA's high unburned carbon content could be the reason for the observed longer setting times. In the literature, fly ashes with high carbon content are reported to retard setting times [36]. In contrast to BOA, the setting times of pastes with RA are shorter than PC paste. A relatively quick setting of mixes with RA could be attributed to its high chloride content of about 21% [25].

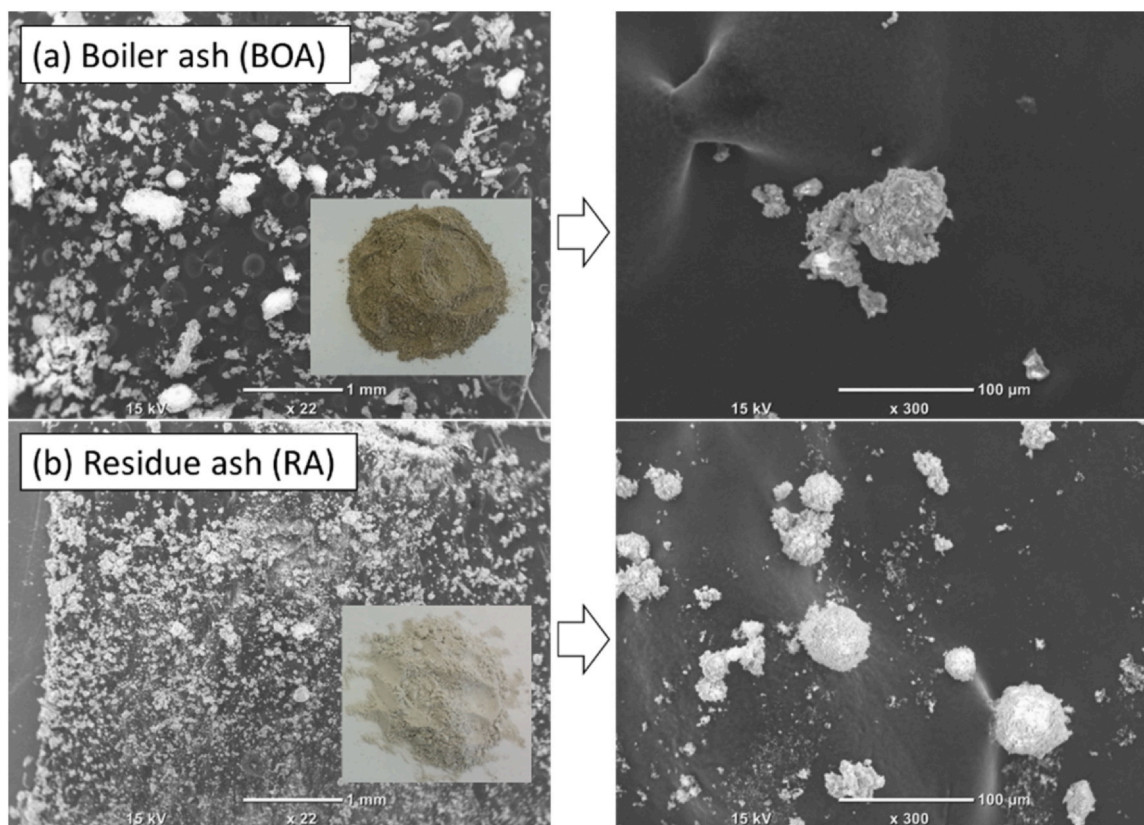


Fig. 3. MSW ashes used in this study: (a) BOA and (b) RA and their SEM images at two different magnitudes.

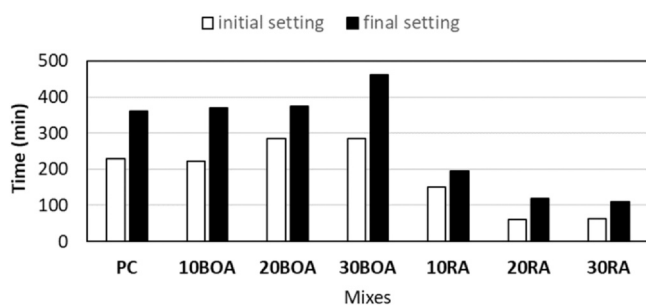


Fig. 4. Effect of BOA and RA on the setting time of pastes.

Portland cement-MSWI ash mortars

Workability

The 10BOA, 20BOA, and 30BOA mixes showed 53%, 50% and 43% flow values (Table 1), indicating that the workability of BOA-incorporated mortar mixes is lower than that of PC mortar mix (58%). Although RA is finer than BOA, the flow of mortars with RA is higher than PC mortar and mortars containing BOA. Contrary to BOA, adding RA increased the flow as RA content increased. Unburned carbon can adsorb water [31,34]. BOA has more unburned carbon content than RA. Therefore, BOA could adsorb more water and limit the free available water, reducing the workability of the mortars.

Compressive strength

After 7 and 28 days of water curing, the PC-MSWI ash mortars were tested for their compressive strength, and the results are given in Table 4. The analysis of these results indicates that all mortars gained strength with curing age. The 30RA showed significant deterioration in strength evolution (relative strength of 30RA to PC mortar at 7 and 28

Table 4

Compressive strength of mortar mixes at 7 and 28 days water curing.

Mix	Compressive Strength (MPa)	
	7 days	28 days
PC	12.7 ± 0.3	15.0 ± 0.5
10BOA	9.2 ± 0.3	12.7 ± 0.8
20BOA	8.5 ± 0.9	13.0 ± 0.5
30BOA	7.2 ± 0.6	11.7 ± 0.8
10RA	13.3 ± 0.8	16.5 ± 0.5
20RA	9.8 ± 0.3	12.7 ± 0.6
30RA	5.3 ± 0.3	7.7 ± 0.3

days are 42% and 51%, respectively), while the 20% replacement showed relatively better strength properties (relative strength of 20RA to PC mortar at 7 and 28 days are 77% and 85%, respectively). It is worth noting that the 10RA showed the highest compressive strength among all mixes at both ages. The relative strength of 10RA to PC is 105% at 7 days, and increased to 110% at 28 days. From the above results, it could be stated that RA reduces strength beyond 10% replacement ratios. Therefore, the optimum mixing ratio of RA should be 10% in terms of the compressive strength of the mortars.

The increase in BOA content is found to decrease the compressive strength. The impact of 10% and 20% replacement ratios is nearly the same for all ages. However, the impact is more pronounced with age for a 30% replacement ratio, i.e. relative strength of 30BOA to PC mortar is 57% and 78% at 7 and 28 days, respectively. The lower performance of BOA-incorporated mortars, at 7 days, could be due to their relatively coarser particle content which carries the unburned carbon. The unburned carbon particles do not provide cementitious reactivity [37] and could limit the free available water for hydration.

At the end of 28 days of curing, BOA and RA incorporated mortars are found to gain more strength compared to their 7 days strength. Even

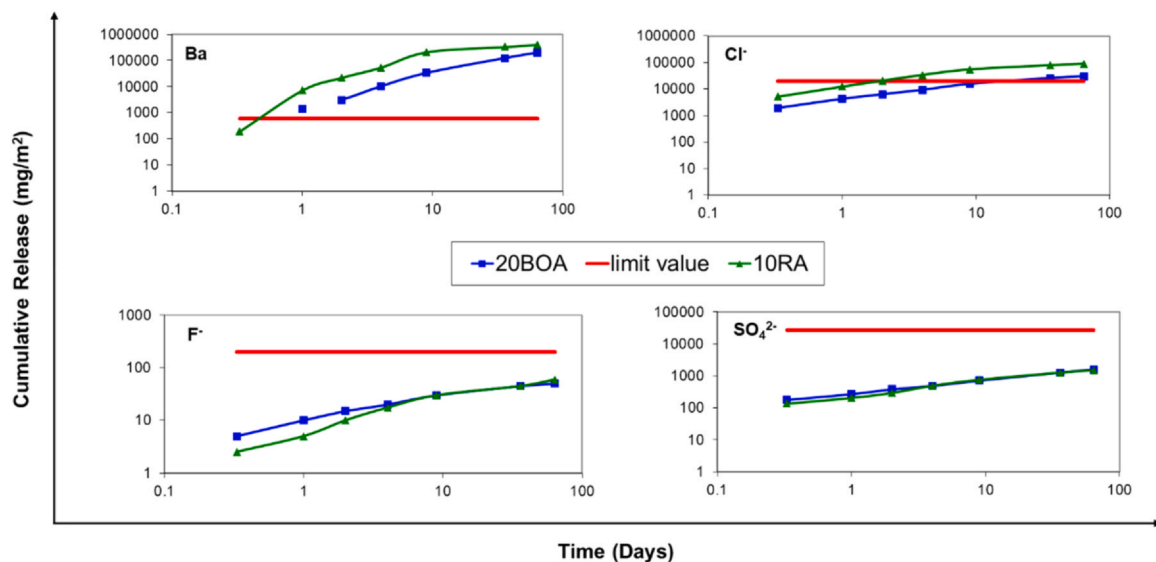


Fig. 5. Cumulative leaching of Ba, F⁻, Cl⁻ and SO₄²⁻ over 64 days and corresponding limit value specified in NEN 7345.

though both ashes had low silica content, which limits their pozzolanic properties, the high lime content of MSWI ashes would contribute to cementitious properties [27]. The impact of increasing BOA content is less intense than those observed in RA. This could be ascribed to the relatively higher silica content of BOA (i.e. silica content of BOA is about four times that of RA), which provides more hydration and pozzolanic reactions and results in less strength loss with continuous curing. Considering the compressive strength of the mortars, the optimum mixing ratio of BOA should be at most 20%.

Environmental performance of mortars

The leaching test was conducted for all mortar mixes; however, the chemical analyses of leachates were performed only for the mixes showing the best workability, setting time and strength properties. The best mix was selected as 20BOA for mortars with BOA, and 10RA was selected for mortars with RA. Leachate samples were analysed for Ba, Cr, Cu, Ni, Pb, Sb and Zn elements. All other elements except Ba were below the detection limit (0.005 mg/L), indicating that Cr, Cu, Ni, Pb, Sb, and Zn were well immobilised in the cement matrix. The MSWI ashes used in this study have relatively higher salt content; therefore, the leaching of major anions (i.e. F⁻, Cl⁻ and SO₄²⁻) from the mortars were also investigated. The results of NEN 7345 for 20BOA and 10RA are presented in Fig. 5. The released amount of Ba is more than 3 orders of magnitude higher than the allowable limit for both mortars. Regarding F⁻ and SO₄²⁻ content, leachates are below the limits. However, the released Cl⁻ content for both mortars exceeds the limits.

Conclusions

A detailed characterisation of two different MSWI ashes, i.e. BOA and RA, collected from different locations of the MSWI process of Qatar is performed in this study. After the characterisation of BOA and RA, an experimental study is carried out to investigate the effects of incorporating these MSWI ashes (0,10,20%, and 30% by weight of cement) as SCM in cement-based composites. Based on the experimental results, the following conclusions can be drawn:

- Due to high unburned carbon content, BOA substitution increased the setting time and decreased the workability. The highest delay in setting time (> 375 min) was observed in the 30BOA mixture (30% by weight of cement replaced with BOA).

- Due to the high chloride content, substituting RA decreased the setting time. The workability of RA-incorporated mortars increased workability as the RA content increased.
- Incorporation of RA in the mortar as cement replacement beyond 10% reduced the strength with the increase of RA content (> 10%). The most adverse effect was observed in the 30RA mixture.
- Incorporation of BOA in the mortar as cement replacement reduced the strength with the increase of BOA (10, 20%, and 30%) content.
- The impact of strength reduction with ash content was more pronounced in RA-incorporated mortars than in BOA mortars.
- Considering setting time, workability, and compressive strength, the optimum mixing ratio for BOA and RA was 20% and 10% by PC weight, respectively.
- The monolithic leaching test indicated that most metals and ions were stabilised in the cement matrix. However, leached Ba and Cl⁻ amounts were significantly above the allowable limits.
- Considering not only valorisation but also reducing the usage of cement and the volume and mass of ashes disposed into landfill, this study indicated that there is a possibility for utilisation of untreated BOA and RA ashes in cement-based composites. However, before any potential applications, pre-treatment is highly recommended due to Ba and Cl⁻ leaching.

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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