

Case study

Mechanical performance and microstructural properties of cement mortars containing MSWI BA as a minor additional constituent

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ABSTRACT

Along with an increase in the amount of municipal waste being incinerated, the amount of Municipal Solid Waste Incineration Bottom Ash (MSWI BA) is growing every year. Combined with limited landfill sites in the Netherlands, this necessitates new applications for the MSWI BA disposal. The larger MSWI BA fractions (> 3 cm) can be successfully used as a natural aggregate substitute in concrete. At the same time, the fine MSWI BA fraction (< 3 cm) has several drawbacks (high porosity, high contaminant content, metallic aluminum, etc.), which make it difficult to use this way. However, if this fine MSWI BA fraction is subjected to treatment that eliminates these shortcomings, it can become a product suitable as a Minor Additional Constituent (MAC) for cement (< 5 % w/w). The addition of MAC allows to use less cement without having a negative influence on its properties, therefore reducing the carbon footprint, and utilizing suitable waste materials (instead of landfilling them). In this paper, mortars and pastes containing the decontaminated (by sieving out the most contaminated fraction < 125 μm) and mechanically treated MSWI BA < 3 mm was used as a cement replacement in the range of 0–20 %. Three types of milled fines were studied, two of them had a particle-size distribution close to CEMI 42.5 and 52.5 to determine the optimum percentage of cement replacement, while the third was not optimized. Effects of MSWI BA fine replacements were studied in terms of mechanical properties, hydration (isothermal calorimetry), microstructure (optical microscopy), and the environmental impact (the standard leaching test by ICP–AES and ion chromatography). The results showed that the mildly milled fines, with a cement replacement of 1 %, are most suitable as a MAC as they have the lowest environmental impact, and no negative effects on the mechanical properties (compressive strength).

Abbreviations: APC, Air Pollution Control; BACF, Bottom Ash Coarse Fines, the bottom ash fraction 0.125–3 mm; FA, Fly Ash; IC, Ion chromatography; ICP–AES, Inductively Coupled Plasma Atomic Emission Spectrometry; LL, Legislation Limit; MAC, Minor Additional Constituent; MCF, Milled Coarse Fines, the bottom ash fraction 0.125–3 mm, milled; MSW, Municipal Solid Waste; MSWI, Municipal Solid Waste Incineration; MSWI BA, Municipal Solid Waste Incineration Bottom Ash; PTE, Potentially Toxic Element; SOF-42, Size Optimized Fines for CEM I 42.5 N cement, milled MCF; SOF-52, Size Optimized Fines for CEM I 52.5 R cement, milled MCF.

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1. Introduction

The Municipal Solid Waste (MSW) quantities generated worldwide increase every year. At this moment in the Netherlands, an average person produces about 0.5 ton of waste per year [1]. Most of that waste is incinerated in waste-to-energy plants, and the issue of Municipal Solid Waste Incineration (MSWI) residue recycling becomes particularly acute due to landfill space limitations, as well as the tightening of environmental legislation (such as the Dutch Green Deal 2012).

There are three types of MSWI residues: air pollution control (APC), fly ash (FA), and bottom ash (BA). MSWI APC and FA are highly contaminated fractions and require a special treatment before being landfilled [2]. MSWI BA, composing ca. 80 % w/w of all residues is a much cleaner product; although without treatment, it contains potentially toxic elements (PTE) in concentrations that significantly exceed the legislation limits (LL) [3]. However, if the most contaminated MSWI BA fraction ($< 125 \mu\text{m}$) is separated, the remaining material is clean enough to be used in various applications after washing out water-soluble salts [4].

The MSWI BA part $> 125 \mu\text{m}$ can be divided into two fractions: below 3 mm (the fine fraction; 50–60 % w/w of the total MSWI BA) and above 3 mm (the coarse fraction) [5]. Nowadays, the latter is being actively studied for applications as a substitute for natural aggregates in concrete [6,7]. The main challenges faced upon utilizing the fine MSWI BA fraction (0.125–3 mm) in a cementitious environment are its high porosity and, thus, a high water demand, and the presence of metallic aluminum particles producing hydrogen gas at high pHs, thus causing bubbles and cracks in hardened cement products [8]. Saikia et al. [9] found that using the untreated MSWI BA fraction (0.1–2 mm) as a fine aggregate replacement (25 % w/w) resulted in a significant deterioration of mechanical properties of mortars.

Another possible application for the fine MSWI BA fraction is a cement substituent. Multiple studies investigated a partial cement replacement with MSWI BA; however, fairly significant amounts (from 10 % to 50 % w/w of MSWI BA in MSWI BA–cement mixtures) are usually used, which severely deteriorate the resulting concrete/mortar mechanical characteristics [10,11]. There are also studies reporting acceptable mechanical properties, where a 10–30 % MSWI BA caused a strength reduction of 10–31 %, meaning the BA acted mostly as an inert filler. MSWI BA can also influence hydration rate negatively and a 20 % replacement of cement with five untreated MSWI BA particle size fractions; showed a delay in hydration up to 10 h for the finest fraction) properties [12,13].

However, it might still be possible to use MSWI BA as a minor additional constituent (MAC) because cement standards allow the use of an inorganic additive of up to 5 % w/w, and such low replacement levels should not have a negative effect on cement properties [14]. As large additions of MSWI BA to cement results in a noticeable deterioration in concrete/mortar properties, and most studies do not consider low replacement levels (below 10 %); there are currently no cement products containing MSWI BA within the MAC range on the market.

Using MSWI BA as a MAC requires milling because the standard specifies that it should be $< 125 \mu\text{m}$ in size. This has several advantages, because besides reducing the particle size, an optimized milling procedure might also help to remove pieces of metallic aluminum by flattening them and subsequently sieving them out and reduce the porosity and water demand of the MSWI BA itself [15].

However, previous research has mostly focused on milling MSWI BA without separating it into different fractions first, and the fine MSWI BA fraction (0.125–3 mm) has not been thoroughly studied. An unspecified ground ($< 0.150 \text{ mm}$) and activated with up to 4 % of CaCl_2 MSWI BA fraction was used to replace 20–40 % w/w of cement and showed results comparable with a 100 % cement reference for a 20 % replacement level, while the same MSWI BA without an activator demonstrated a 40 % loss in compressive strength (21 days curing, a 20 % replacement) [16]. In another study, a fraction of 0.04–0.25 mm was investigated as a low-density binder with wood–cement boards analyzing among others its properties as an insulation material [17]. Tang et al. focused on a combined milling and heating treatment at 550 and 750 °C (to increase reactivity and reduce the leaching) of a 0–2 mm MSWI BA fraction slowly milled to a size below 0.125 mm. Only a replacement level of 30 % was studied, and the reported mechanical properties were between 15 % and 50 % lower in comparison with a reference sample depending on the MSWI BA pretreatment prior to use [18]. The only paper dealing with different particle-size MSWI BA fractions (five fractions $< 6.3 \text{ mm}$) milled below $150 \mu\text{m}$ made a study on hydration of mixtures but did not report the mechanical properties [12].

To sum up, to the best of authors' knowledge, there is almost no published research on the applications of the fine MSWI BA fraction (0.125–3 mm) as a MAC [19], as it has the advantage of being significantly less contaminated than the finest fraction ($< 125 \mu\text{m}$) but cannot be recycled as a natural aggregate substitute. Therefore, it is topical to use milled MSWI BA fine fractions as a MAC. Thus, the aim of this study is to test small replacement levels of differently ground MSWI BA fines (the fraction 0.125–3 mm) in a cement mixture in the MAC range (0–5 % w/w) and to compare them with higher replacement levels (up to 20 %) to demonstrate negative effects on the properties of mortars and pastes, which might not be visible at replacements below the MAC range.

We targeted this study to determine the maximum MSWI BA content in cement mortars resulting in no detectable deterioration of properties (both mechanical and environmental). The second one is the maximum MSWI BA content that results in a satisfactory level of the deterioration of the final product (mortar) compared to the cement standard to discover whether the particle size distribution of MSWI BA fines tailored to a certain cement particle size influences a mortars mechanical performance.

To achieve these objectives, the mechanical properties of mortars containing MSWI BA fines (the MAC range: 1, 3, and 5; and the extended range: 10 % and 20 % replacement level), leaching properties of dry mixtures and hardened crushed pastes, and hydration properties of pastes containing MSWI BA fines were analyzed. A microstructural analysis of mortars containing MSWI BA fines is also provided. We provided the data on all the properties for the extended range of additives (up to 20 %) to make changes in the structure and quality of resulted pastes/mortars clearly visible because in the MAC range some changes are very difficult to observe even though there is a detrimental effect. Thereby, this data also provides the deeper understanding on the impact of added MSWI BA milled fines. It was also necessary to make sure of the limited or absent environmental impact of the mortars containing MSWI BA, therefore, the leaching properties of dry mixtures and hardened pastes with substitutes were examined. Because the topic of leaching in cement

products with similar substitutes is vastly discussed in the literature, an elaborate consideration of it has been avoided in this study [20, 21].

2. Materials and methods

2.1. Materials

Pre-treated (standard washed) MSWI BA fines (< 3 mm) were supplied by the municipal solid residues processing plant of Mineralz (the Netherlands). The material was sieved into two fractions: < 125 μm and 0.125 μm – 3 mm, MSWI BA coarse fines. They were subsequently milled generating three final fine products: 1) **MCF** (Milled Coarse Fines), the BACF fraction milled to reduce the particle size below 125 μm ; 2) **SOF-52** (Size Optimized Fines) milled to a PSD close to CEM I 520.5 R; 3) **SOF-42** (Size Optimized Fines) milled to a PSD close to CEM I 420.5 N. CEM I 52.5 R and CEM I 42.5 N supplied by ENCI (the Netherlands) were used as binders for references and mixtures with milled MSWI BA fines (**MCF**, **SOF-52**, **SOF-42**).

2.2. Reagents

Deionized water (a Milli-Q Academic system, Millipore) was used for the preparation of all the solutions and washing. Nitric acid (69 %, for analysis, ISO-grade, AppliChem Panreac) was used for all the operations with ICP–AES. All standard solutions were from High Purity Standards. Standard solutions of ICP-AM-6 (100 mg/L of Al, Sb, B, Ba, Be, Cd, Ca, Co, Cr, Cu, Fe, Li, Mg, Mn, Mo, Ni, K, Na, Si, Pb, Tl, Sr, V, Zn) and ICP-MS-68B (100 mg/L of Sb, Ge, Hf, Mo, Nb, Si, Ag, Ta, Te, Sn, Ti, W, Zr) were used for the calibration in the range 0.01–10 mg/L. An ICP-AM-15 standard solution (10,000 mg/L of Na, K, Ca, and Mg), P (10,000 mg/L), and S (10,000 mg/L) was used for the calibration in the range 1–100 mg/L. An internal standard solution of Sc (20 mg/L) was prepared from a standard solution (Inorganic Ventures, 1000 mg/L).

2.3. Methods and equipment

2.3.1. Inductively coupled plasma atomic emission spectrometry

Inductively coupled plasma atomic emission spectrometry (ICP–AES) provides rapid and highly sensitive multielemental quantification of leachates with high accuracy and precision [22,23]. An ICP–OES 5100 SVDV spectrometer (Agilent Technologies) was operated in the axial mode (the auxiliary gas flow, 1.2 L/min; coolant gas flow, 14 L/min; and nebulizer gas flow, 0.8 L/min). All emission lines were measured simultaneously.

2.3.2. Fused bead preparation

To prepare fused beads, the following procedure was used: 0.95 g of BA was mixed with 9.5 g of a flux (67.00 % $\text{Li}_2\text{B}_4\text{O}_7$ – 33.00 % LiBO_2 , w/w, Claisse), and 0.1 g of LiBr (Claisse) as a non-wetting agent. Mixtures were put individually into borate fusion ovens for 24 min at 1065 °C. The procedure was made for 3 replicates.

The borate-fusion technique provides rapid and easy sample preparation to conduct a subsequent multielement analysis using ICP–AES. Compared to microwave digestion, which frequently causes difficulties with a factually complete sample dissolution [23,24], a sample is completely dissolved. A blend of lithium metaborate and tetraborate was selected as a mixture of basic and acidic fluxes, which provides a complete and reproducible sample preparation of industrial waste BA including different incinerator types [25], various types of slag (blast furnace, steel) [26], and BA [27] as all these residues contain both types of oxides.

2.3.3. Sample preparation for ICP (fused beads)

Fused beads were ground for 2 min in a planetary ball mill, and 1-g weighed portions were dissolved in 50.00 mL of 5 % w/w HNO_3 (p.a. grade) in an ultrasonic bath for 1 h and subjected to ICP–AES. The estimated average concentrations of designated elements ($\mu\text{g}/\text{mL}$) were recalculated to mg per 1 kg of the raw material.

2.3.4. Optical microscopy

To obtain microscopic photographs of MSWI BA containing cement pastes a Zeiss Axio Imager Z1 was used with an Epiplan NEOFLUAR objective (5.0 \times /0.13) with a \sim 2 μm resolution and 1 μm pixel size.

2.3.5. Ion chromatography

Ion chromatography (IC) was selected as a method for chloride and sulfate determination in leachates [28]. A Thermo Scientific Dionex ion chromatograph 1100, 2 \times 250 mm AS9-HS ion-exchange columns, an isocratic flow (0.25 mL/min) was used. Ion detection was accomplished by measuring the suppressed conductivity making use of an electrolytically regenerated suppressor (Thermo Scientific Dionex AERS 500 2 mm). As eluent, a 9 mM solution of sodium carbonate was used.

2.3.6. Auxiliary equipment

A planetary ball mill (Fritsch PULVERISSETTE 5) fitted with zirconium oxide grinding bowls, and 20- and 10-mm mill balls were employed for the size reduction of the sample. An XP 504 analytical balance (Mettler Toledo), 15-D0438 riffle boxes (sample splitters, CONTROLS Group), an SM-30 shaking table (Edmund Bühler GmbH), a UF 260 drying oven (Memmert), and a Multiwave 3000 (Anton

Paar) microwave oven for digestion with a 60-bar rotor were used throughout. Automatic Eppendorf Research pipettes (Eppendorf International) were used for the preparation of calibration solutions. A-class polypropylene volumetric flasks (Vitalab) with volume (50.00 ± 0.12) mL and (100.00 ± 0.20) mL and polypropylene test tubes (Axygen) were used for the preparation of calibration and test solutions.

2.4. Procedures

2.4.1. Standard leaching test

The standard leaching test NEN-EN 12457-4 [29] (24 h shaking, L/S 10, 200 rpm) was performed on non-hydrated mixtures containing MCF, SOF-52, and SOF-42; as well as hardened cement pastes (crushed and sieved below 4 mm prior to testing) containing MCF (3 replicates of 30 g) to evaluate the level of PTEs, chloride, and sulfate, as well as to investigate the binding capacity of cement regarding the PTEs release at 28 days of curing. After the test, the leachates were filtered through 0.2- μ m filters to prepare solutions for IC and ICP-AES (for the latter, filtrates were stabilized with nitric acid).

2.4.2. Milling conditions

We aimed to reach the desired particle size with as little milling as possible to minimize the cost. Thus, the first milling procedure was carried out in four cycles (10–10–20–20 min), and after each cycle, the material was sieved through a 0.5-mm sieve to remove flattened Al particles and a 0.125-mm sieve to obtain the MCF fraction. This was done to remove softer particles that had already reached a desired size while harder ones were ground for longer to guarantee a uniform particle size. The MCF was then additionally milled again for 85 and 210 min (300 rpm) for SOF-42 and SOF-52, respectively, to obtain particles in the range of CEM I 42.5 N or CEM I 52.5 N. SOF-52 was then mixed with CEM I 52.5 R, and SOF-42 with CEM I 42.5 N. More information about the milling procedure can be found in [19].

2.4.3. Determination of hydration delay

The effect of MCF and SOF-52 as a partial replacement of CEM I 52.5 R in cement mixtures (0–20 % replacement) on the hydration process was studied by isothermal calorimetry (a TAM Air calorimeter). Mixtures were prepared with a water/powder ratio of 0.5. Calorimetry measurement outcomes were normalized per cement mass in the sample.

2.4.4. Mortar preparation

Mortars with cement–MSWI BA mixtures were prepared according to EN-197-1. One batch was made from 450 g of a binder (either pure cement or cement + MSWI BA milled fines), 225 g of water, and 1350 g of sand. All mixtures (containing 1 %, 3 %, 5 %, 10 %, and

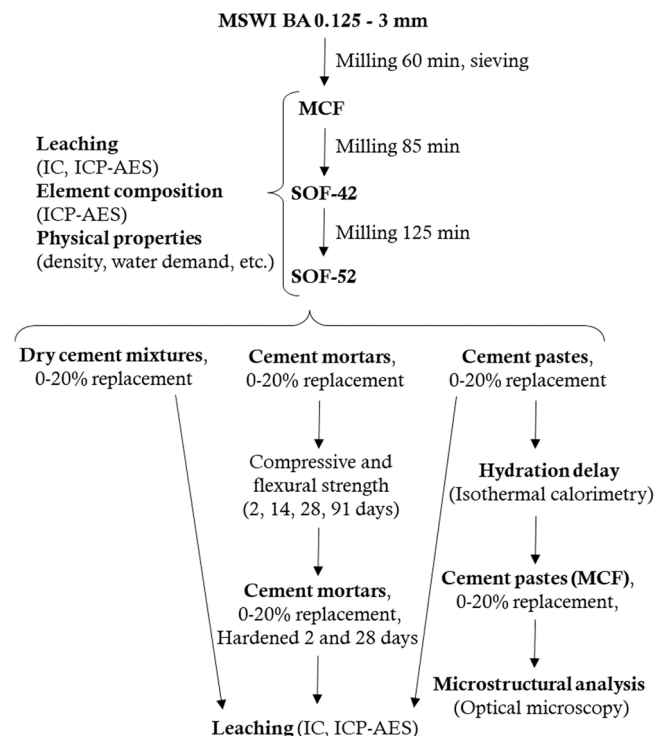


Fig. 1. Schematic representations of the methods and their purposes used in the study.

20 % w/w of MSWI BA milled fines) were premixed and homogenized with a mixer prior to adding water.

The mortars were cast in Styrofoam molds and cured in water. Mortars that were used for the leaching test were cured at a high humidity without a direct contact to water.

2.4.5. Flexural and compressive strength

All prepared mortars (4×4×16 cm) were submitted to testing of compressive and flexural strengths (a three-point flexural test, Zwick Z020). All samples were tested at 2, 14, 28, 91, and some also at 7 days of curing, additionally.

2.4.6. Sample preparation and microstructural analysis

For microstructural analysis, paste samples containing MCF were prepared by the procedure described below. In a glass jar, water and cement-MCF powders were mixed (prepared in advance with the replacement level by weight), a water/powder ratio 0.5. After 24 h, the jar was carefully removed, and the samples were placed in water for the remaining 6 days. After that, to stop the hydration, the samples were placed in isopropanol for 48 h and regularly mixed, and isopropanol was replaced every 12 h. After this procedure, the samples were polished using isopropanol to prevent hydration and provide a smooth surface without scratches.

Microphotographs of the samples were analyzed using ImageJ software, looking at representative areas, analyzing each area to differentiate three types of the matter in the paste: unhydrated cement, hydration products, and porosity. The results for each sample were averaged.

The outline of the study including material, methods, and procedures presented in Fig. 1.

3. Results and discussion

3.1. Summary of properties of MSWI BA fines

Tables 1, 2, and Fig. 2 provide a summary of the main chemical and physical properties of the three types of milled BA fines used in the study. Most physical properties appear to be similar to the cement, therefore, the BA milled fines can be considered a suitable cement replacement material [19].

3.2. Leaching of dry mixtures and hardened pastes, and mortars

To be able to use a material as a MAC, it should not influence the mechanical properties negatively and comply with local environmental LLs. Thus, it is important to examine the mortars, preferably at all stages of curing for their leaching properties.

To assess the environmental impact and potential risks of handling the cement containing BA, the leaching of the dry unhydrated cement mixtures was also determined even though the final assessment is made for a hydrated mortar or concrete. It is necessary to consider in what form and at what point, a cement product containing BA fines can be in contact with the soil (ground water) to prevent possible contamination.

3.2.1. Anion leaching of dry cement-BA mixtures

Fig. 3 demonstrates the chloride and sulfate leaching from unreacted cement-BA mixtures. Note that for all replacement levels of SOF-52, the chloride leaching was at the same level as the initial leaching from the corresponding cement (CEM I 52.5 R) and does not change. A very slight increase in concentrations is observed for SOF-42. For MCF (with CEM I 52.5 R), a slight decrease in chloride leaching for low replacement levels can be seen, and a slight increase in the large replacement level area (10–20 % w/w) can be observed. Chloride leaching for all the mixtures is below the LL [30].

Sulfate leaching gradually decreases in all the cases when the BA share increases in the mixture, and in mixtures containing SOF-52 and MCF sulfate leaching is noticeably reduced compared to the leaching of pure fines, therefore, the main source of the leaching is cement, not BA. For pure SOF-42, the reduction in leaching is negligible. For a 1 % w/w replacement, it is close to that of pure cement and then decreases with an increase in the share of SOF-42.

3.2.2. PTEs leaching of dry cement-BA mixtures

Table 3 shows the leaching test results for PTEs that were either in concentrations above the LL or were high in the pure BA fines.

Table 1

Leaching of sulfate and chloride, and cations present in the leachates for ground BA fines: MCF; PSD-optimized to CEM-I-42.5-N ground MCF (SOF-42); PSD-optimized to CEM-I-52.5-R, ground MCF (SOF-52); the units for concentrations given as mg per kg of dry solid. ($n = 3, P = 0.95$), [19].

Fines	Chloride mg/kg _{ds}	Sulfate	Sb
MCF	500 ± 50	5400 ± 500	0.70 ± 0.07
SOF-42	590 ± 60	5800 ± 500	0.80 ± 0.08
SOF-52	650 ± 70	6200 ± 600	0.90 ± 0.09
LL	616	2430	0.32

Table 2
Physical properties of BA fines and cement ($n = 3, P = 0.95$).

Property	Units	Cement		BA fines		
		I-52.5-R	I-42.5-N	MCF	SOF-42	SOF-52
Bulk density	(g/cm ³)	1.1 ± 0.1	1.7 ± 0.1	1.5 ± 0.1	1.6 ± 0.1	1.5 ± 0.1
Water demand	(% w/w)	22.0 ± 0.5	21.0 ± 0.5	20 ± 0.5	22.0 ± 0.5	24.0 ± 0.5
D (0.5)	µm	11	20	39	20	13

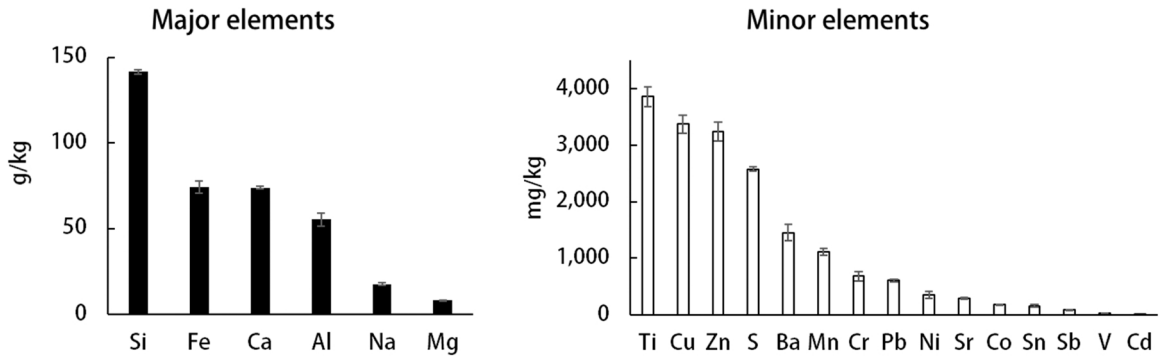


Fig. 2. Element composition of the MCF, SOF-52, and SOF-42 precursor, MSWI BA, fraction 0.125–3 mm, ($n = 3, P = 0.95$).

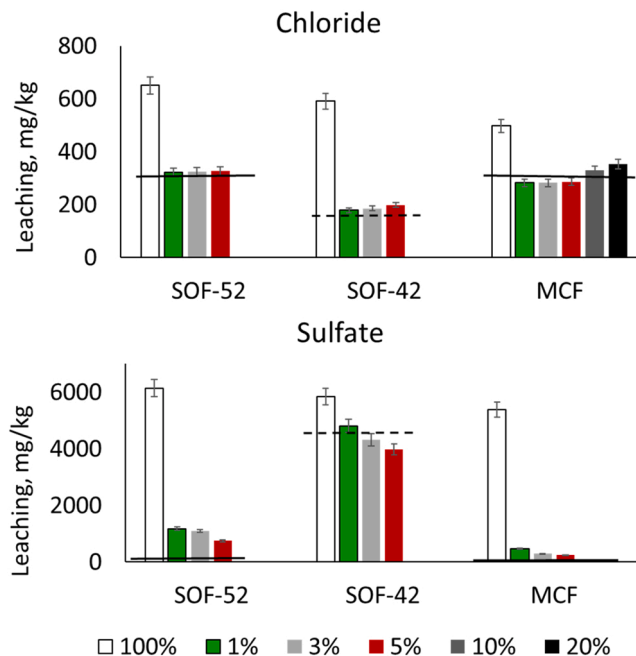


Fig. 3. Leaching of chloride and sulfate from mixtures containing 0–20 % w/w BA fines, only MCF were tested for replacements higher than 5 % w/w; 100 % corresponds to pure materials. The level of anion leaching for CEM I 52.5 R is expressed with a black solid line; for CEM I 42.5 N, with a black dashed line; the initial leaching values recalculated to mg/kg of the material.

CEM I 52.5 R has a relatively high level of barium leaching (30 mg/kg dm), which becomes only half (15 mg/kg dm) with the addition of 1 % w/w MCF, and even lower with the addition of 1% w/w SOF-52 (11 mg/kg dm). As the BA replacement level increases, BA leaching increases too; and for MCF replacement levels of 10–20 % w/w, they reach a level close to the leaching of pure CEM I 52.5 R.

This can partially result from an increase in sulfate concentration due to the addition of fines, the excess of which reacts with the available barium to form BaSO₄, which has a very low solubility [31]. A slight difference in sulfate concentration of mixtures with MCF and SOF-52 is concordance with it, as the sulfate leaching from MCF is slightly lower in comparison with SOF-52.

Table 3

PTEs leaching of mixtures containing 0–20 % w/w BA fines (recalculated to mg/kg of the material); all values for Sb are below the detection limit; “b.d.l.” denotes below the detection limit; ($n = 3, P = 0.95$) [30].

Sample	pH	Ba	Cr	Cu	Mo
CEM I 52.5 R	11 ± 0.1	30 ± 3	0.5 ± 0.05	b.d.l.	0.18 ± 0.02
MCF 1 %	11 ± 0.1	15 ± 2	1.8 ± 0.2	0.05 ± 0.005	0.19 ± 0.02
MCF 3 %	11 ± 0.1	18 ± 2	1.4 ± 0.1	0.07 ± 0.007	0.17 ± 0.02
MCF 5 %	11 ± 0.1	19 ± 2	1.4 ± 0.1	0.13 ± 0.01	0.16 ± 0.02
MCF 10 %	11 ± 0.1	31 ± 3	0.4 ± 0.04	0.16 ± 0.02	0.18 ± 0.02
MCF 20 %	11 ± 0.2	32 ± 3	0.3 ± 0.03	0.28 ± 0.03	0.19 ± 0.02
SOF-52 1 %	11 ± 0.1	11 ± 1	2.3 ± 0.2	b.d.l.	0.6 ± 0.06
SOF-52 3 %	11 ± 0.1	17 ± 2	2.9 ± 0.3	0.08 ± 0.008	0.87 ± 0.09
SOF-52 5 %	11 ± 0.1	19 ± 2	2.4 ± 0.2	0.11 ± 0.01	0.74 ± 0.07
CEM I 42.5 N	11.4 ± 0.1	6.0 ± 0.6	6.6 ± 0.7	b.d.l.	0.84 ± 0.08
SOF-42 1 %	11.4 ± 0.1	7.0 ± 0.7	8.0 ± 0.8	b.d.l.	1.0 ± 0.1
SOF-42 3 %	11.4 ± 0.1	7.7 ± 0.8	8 ± 0.8	b.d.l.	1.1 ± 0.1
SOF-42 5 %	11.4 ± 0.1	8.0 ± 0.8	7.7 ± 0.8	0.05 ± 0.005	1.1 ± 0.1

As for the subsequent increase in the Ba^{2+} concentration with an increased cement replacement level, it might be assumed that it happens because the main source of Ba^{2+} is cement, and its share is growing reaching a concentration peak at a 20% w/w replacement level. Additionally, in the case of SOF-42 and CEM I 42.5 N, barium concentrations are insignificant and are approximately at the same level for the entire range of additives.

Initially, chromium leaching was not detected in all the tested BA fines. Table 3 shows that the chromium leaching gradually increases in the row of MCF < SOF-42 < SOF-52 for the same cement substitution levels, although it is necessary to take into account that the CEM I 42.5 used for the SOF-42 samples leaches much more Cr (6.6 mg/kg) than the CEM I 52.5 (0.5 mg/kg). The emerging chromium leaching can be explained by a change in pH (from 8 ± 1 for pure BA fines to 11 ± 1 for cement–mixture leachates), and an increase in the solubility of chromium (III) hydrated oxide species in strong alkaline environments [32,33].

Copper was initially present in all the BA fines at the concentration level of 0.7 ± 0.1 mg/kg. Table 3 shows that the Cu^{2+} concentration in all the leachates is extremely low. Nevertheless, it is noticeable that in the MCF range of 1–20 % w/w, the Cu concentration in the leachate slowly increases.

Molybdenum leaching was within the limit for all the BA fines (0.4, 0.6, and 0.9 mg/kg for MCF, SOF-42, and SOF-52, respectively); in all the samples containing MCF, molybdenum leaching is at the level of Mo leaching from CEM I 52.5 R and does not change as the MCF share in the mixture increases (Table 3). For SOF-52, the concentrations for the same replacement level with the same cement are 4–5 times higher, although they are still below the legislation limit. For SOF-42, Mo concentrations are approximately the same for all the replacement levels, and slightly exceed the limit (1.0 mg/kg); however, it should be outlined that CEM I 42.5 N makes a significant contribution to the Mo leaching by itself.

Even though in all the samples (MCF, SOF-42, and SOF-52) antimony was present at a level exceeding the LL by 2–3 times, it was not found in any of the leachates of cement mixtures.

Table 4 shows how PTEs are leaching normalized by the substitution level of MCF in the cement mixture. As can be seen, all curves have a similar shape with an abrupt drop within the 3 % w/w replacement, then a gradual decrease until 10 % followed with a plateau or a further slight decrease. It should be noted that even though the level of PTEs is not that high, their main source is cement, and this explains a decrease in concentrations as the share of cement in the mixture becomes smaller. However, as can be seen from Table 4, the values decrease much more than the share of cement (which changes from 100 % to 80 %), while the leaching is reduced by 60–100 %.

3.2.3. Leaching of mortars containing MCF

No significant leaching of anions was detected for all pastes and mortars with a cement replacement level of up to 20 % w/w after 2 d of curing (chloride leaching, 40–100 mg/kg; sulfate leaching, 10–20 mg/kg), showing a high capacity of cement for chloride and

Table 4

Relative leaching of PTEs (normalized per 1 % w/w of added BA fines), mg/%. “b.d.l.” denotes below the detection limit.

	Ba	Cr	Cu	Mo
MCF 1 %	1.5	0.18	0.005	0.019
MCF 3 %	0.60	0.047	0.002	0.006
MCF 5 %	0.38	0.028	0.003	0.003
MCF 10 %	0.31	0.004	0.002	0.002
MCF 20 %	0.16	0.002	0.001	0.001
SOF-52 1 %	1.1	0.23	b.d.l.	0.06
SOF-52 3 %	0.57	0.097	0.003	0.029
SOF-52 5 %	0.38	0.048	0.002	0.015
SOF-42 1 %	0.70	0.80	b.d.l.	0.10
SOF-42 3 %	0.26	0.27	b.d.l.	0.037
SOF-42 5 %	0.16	0.15	0.005	0.022

sulfate binding. After 28 days of curing, the leaching of sulfate (10–40 mg/kg) and chloride (20–50 mg/kg) is even lower and in all cases well below the LLs (616 mg/kg for chloride and 2430 mg/kg for sulfate).

The leaching of PTEs in all mortars and pastes after 2 and 28 days of curing is also below the detection limits and, accordingly, below LLs. The only exception is Ba, the content of which is relatively high in the cement itself and is not caused by pollution from the BA.

3.3. Effect of MSWI BA fines on cement hydration (CEM I 52.5 R)

Figs. 4–6 depict the effect of MCF and SOF-52 on the hydration of cement pastes. Replacements of 1%, 3%, and 5% w/w were used to analyze the MAC range (Fig. 4), and 20% w/w to make detrimental effects more visible. MCF was also tested for a 10% w/w replacement. CEM I 52.5 R with 1%, 3%, 5%, 10%, and 20% w/w quartz powder replacements were used as additional references with the same water/powder ratio of 0.5.

The shapes of the existing peaks and no extra peaks indicate that there are no side reactions associated with a higher sulfate content in BA fines compared to cement (2.83% w/w for cement vs. 5.3% w/w for MCF and 6.1% w/w for SOF-52) [34].

Fig. 4 also shows that the cumulative heat (normalized per cement) is the highest for MCF for 1% and 5% w/w replacement for reasons unclear. A higher cumulative heat could have been expected for SOF-52 as it has the smallest particle size and could therefore be more reactive. However, it is possible that the milling and removal of the associated quenching layer releases not only more sulfate but also other components such as organic matter that might have a negative influence [35].

For a 3% w/w replacement, the cumulative heat is almost the same, and for 10% w/w (as MCF is only compared with quartz), MCF cumulative heat is slightly higher than a 0% replacement, and almost the same as for 10% w/w quartz. Altogether, in the range of the replacement for MAC, there is no significant difference.

Fig. 5 demonstrates more clearly the effect from BA fines on cement hydration. The addition of 20% w/w SOF-52 leads to a delay in hydration of ca. 4 h, the addition of 20% w/w MCF, ca. 2 h. The cumulative heat is almost the same for both types of fines and does not differ significantly from quartz (see Table 5).

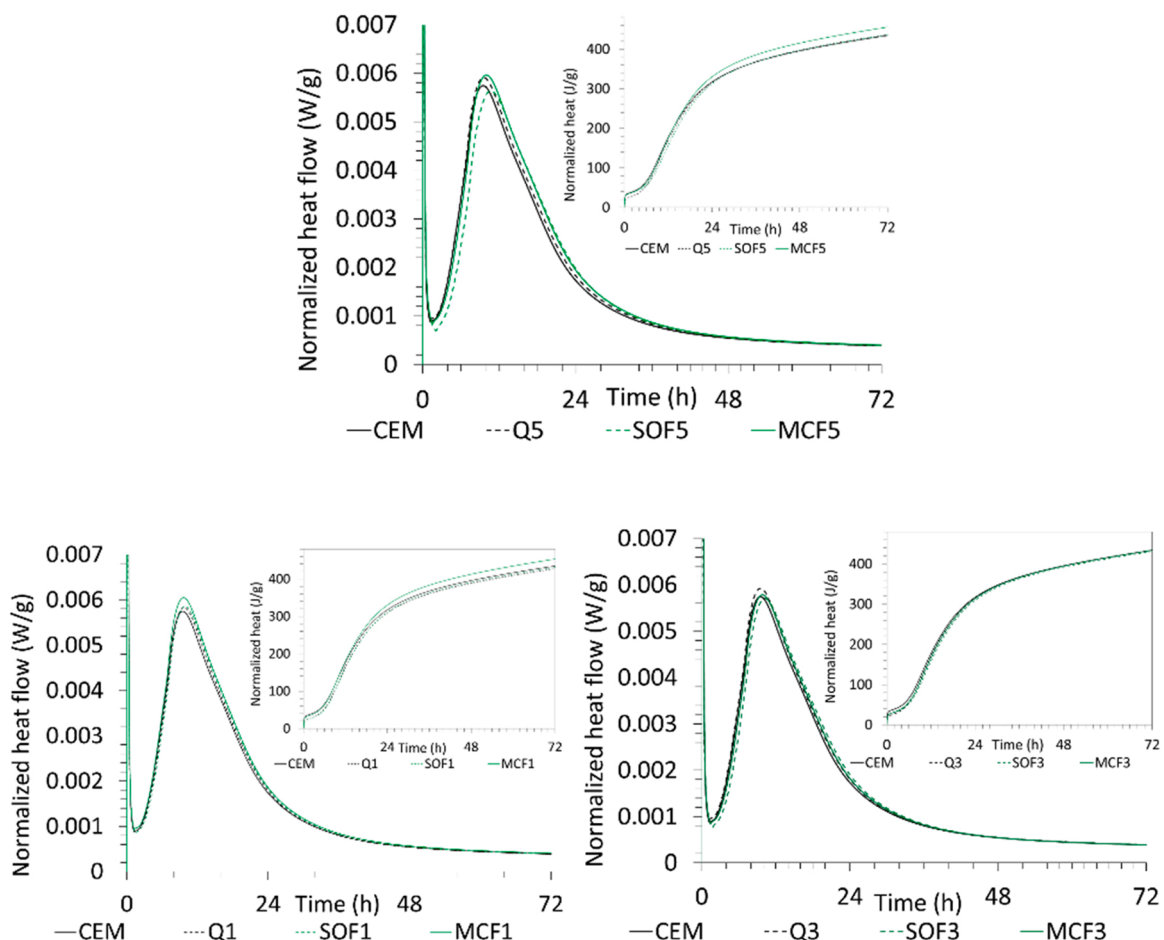


Fig. 4. Effect of added BA fines on cement hydration (0–10% w/w replacements) normalized by the cement weight.

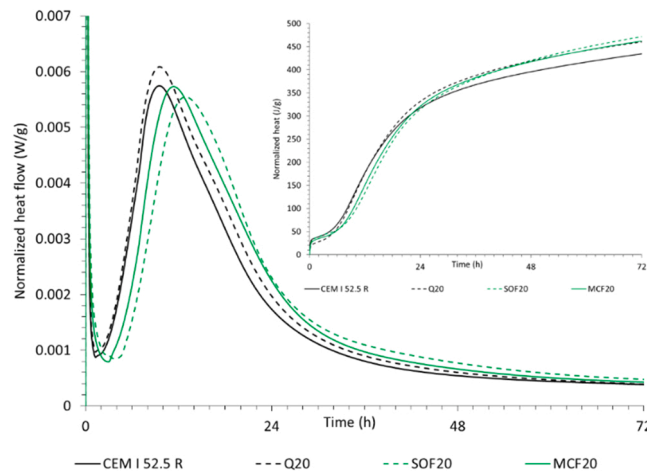


Fig. 5. Effect of added BA fines on cement hydration (a 20 % w/w replacement) normalized by the cement mass.

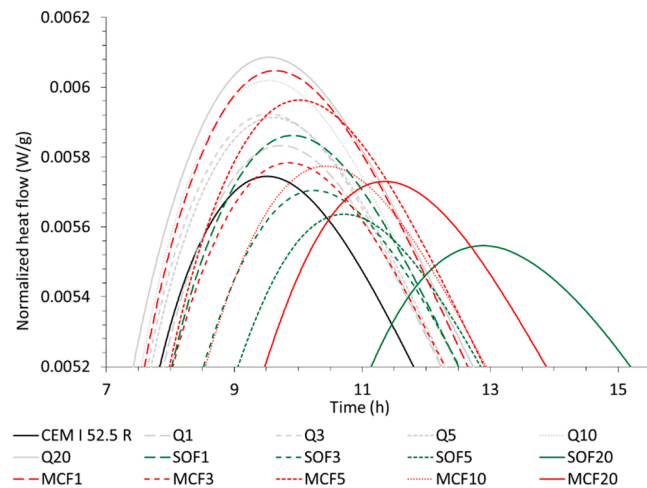


Fig. 6. Effect of added BA fines on cement hydration (0–20 % w/w replacements) normalized by the cement weight. The curves of those samples that show no significant differences compared to the reference (100 % CEM 52.5) are shown as dashed lines.

Table 5

Total heat at 72 h for pastes containing BA fines and quartz normalized by the cement weight (J/g).

% w/w repl.	Sample			
	CEM 52.5	Quartz	MCF	SOF-52
0	432	–	–	–
1	–	425	450	428
3	–	432	431	428
5	–	434	453	433
10	–	441	443	–
20	–	457	459	468

Figs. 5 and 6 show that the longest delay in the hydration is observed for a 20 % w/w replacement with SOF-52. This can be explained by additional sulfate (SOF-52 leaches about 13 % more than MCF), and by the fact that at this replacement level the impact of other components in BA that may contribute to the delay becomes more noticeable. The leaching of SOF-52 is higher, because very fine particles are generated when the quenching layer is removed during the longer milling time [36,37]. Further delays in cement reaction are approx. 2 h for MCF (20 % w/w), around 1 h for SOF-52 (5 % w/w), and MCF (10 % w/w), and around 30 min for SOF-52 (3 % w/w). It is noteworthy that there are no significant delays observed for MCF in the MAC range. Table 5 shows the cumulative heat

at 72 h for all the fines studied. Taking into consideration all above-mentioned data, it can be concluded that the most beneficial is the use of MCF with 1 % or 5 % w/w replacements.

3.4. Mechanical properties of MCF mortars (0–20 %)

The mechanical properties for mortars with a cement replacement level above the MAC range ($> 5\%$ w/w) were tested only for MCF and not for SOF-52 or SOF-42, as preliminary research showed that other properties of these fines would unlikely make them applicable in a large range of additives.

3.4.1. Strength development of MCF samples

In addition to the 28 d strength, the early strength (2 days) was tested to ensure that the reaction delays visible with calorimetry do not have a negative influence on the strength development. As can be seen from Fig. 7a, the 2-day compressive strength is acceptable up to a 10 % w/w MCF replacement level according to the standard for CEM I 52.5 R (minimum compressive strength 30 MPa). After 28 days, only the samples with a replacement level up to 5 % w/w show sufficient strength (the minimum compressive strength, 52.5 MPa).

For the flexural strength of 2-day hardened samples, a linear correlation with the replacement level is also observed (Fig. 7b); however, at 28 days, the same general trend is visible. As it is for the compressive strength results, the harmful effect is minimal or absent only at a 1 % w/w replacement level.

Additionally, it was noted that in most studies available on this topic, there is a place for some methodological changes related to testing certain replacement levels after a certain number of days of hardening. Almost none of the available papers revealed the data on mechanical properties of hardened cement products at the early stage (1–2 days), while for these terms most cements have the minimum requirements for the compressive strength [38].

It appears beneficial if the use of any additive at any quantities would begin with testing the early compressive strength to continue the experiments only in the “working” concentration range. Fig. 7a shows that the 2-day compressive strength is acceptable for this type of fines only within a 10 % w/w replacement; after 28 days, it shifts to less than a 10 % w/w replacement. It is possible that milling larger (coarser) fractions of ash would change the situation; however, this analysis suggests that with time the initial structural

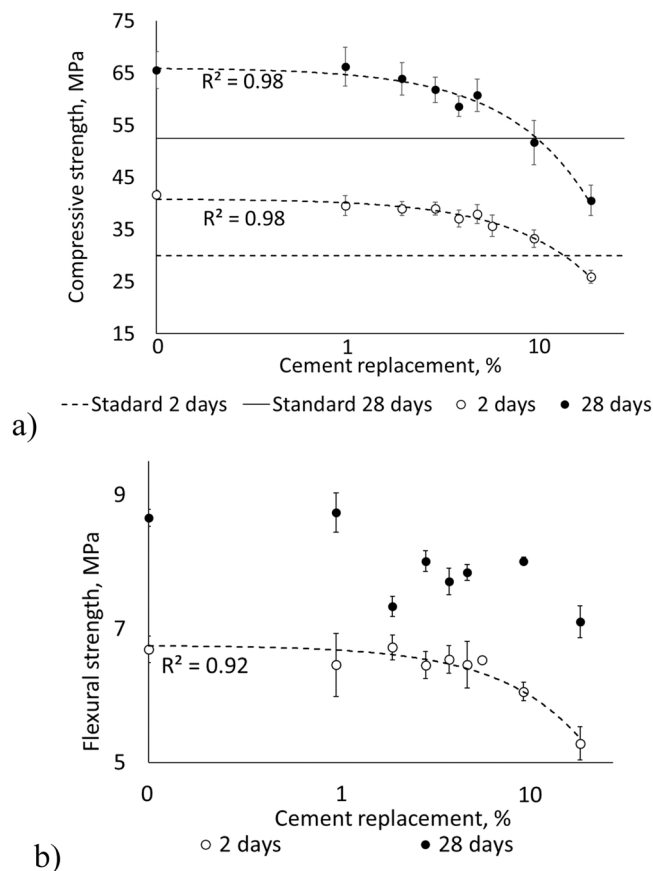


Fig. 7. Compressive (a) and flexural (b) strength of CEM I 52.5 R mortars containing MCF (from 0 % to 20 % w/w cement replacement). The X-axis is presented in a logarithmic scale. (n = 18, P = 0.95).

changes would lead to defect aggravation only. However, this issue requires further studies. Moreover, the compressive strength correlates with the replacement level, which may serve for predicting, and indicates the estimation of a potential effect of the additives on the mechanical performance.

Taking into consideration the fact that the strength analysis is made in the range of low replacement levels of fines in cement mixes, the difference between them is not expected to be significant. However, considering that one of the goals of this study is to determine the replacement level where no detectable difference with the reference sample can be observed, the insignificant difference of the absolute values will not be neglected in this case. Fig. 8 shows the trends of the compressive strength for all the replacements (MCF) from 2 to 91 days.

At the early stage (2 days), all the replacement levels show results slightly worse than the reference. The replacement levels of 1–3 % w/w show close results (a loss in compressive strength of 5–6 %). By day 7, the 1 % w/w sample is very close in strength to the reference; 2 % w/w is slightly inferior to it (a 5% loss compared to the reference); and 3–5 % w/w are still slightly lower (a 7–9 % loss). By day 14, the difference between all the replacement levels becomes slightly more obvious, with 1% w/w showing even better results than the reference (a 5 % gain), and 2 % and 4% w/w are at the same level with it. Levels of 3 % and 5 % w/w have already noticeably lower compressive strength (4 % and 10 % losses, respectively).

By day 28 (the standard for cement), the best performing mortars are 1 % w/w (a 1 % gain) and the reference; 2 % w/w is very close to them (a 3 % loss); 3 % and 5 % w/w are slightly worse (6 % and 7 % losses, respectively); and 4 % w/w shows the lowest strength (a 11 % loss). By day 91, the best results are for the reference and 2 % w/w (a 1 % loss); 1 % w/w is slightly worse (a 4 % loss); and 3–5 % w/w are approximately at the same level (a 7–8 % loss). The apparent absence of changes from 28 to 91 days for the level of 1 % w/w can be attributed to the experimental error (which can be within 10 % according to the testing standard [14]), which almost does not go beyond the range of differences for all the replacement levels in this case.

3.5. Mechanical properties of the size optimized fines SOF-52 and SOF-42

The performance of various replacement levels of three types of BA fines (MCF, SOF-52, and SOF-42) was assessed in the MAC range (0–5 % w/w). In the following subsections, two types (SOF-52, and SOF-42) are discussed separately, and MCF is compared with SOF-52 (as they were tested with the same cement type).

Because one of the objectives of this study was to find out whether the BA milling towards the particle size of the relevant cement beneficially affects the quality of fines as MAC, two types of fines with an increased milling time (SOF-52 and SOF-42) with two types of cement, which are similar in composition but differ in PSD were compared. The goal here was to determine whether the mortars containing these fines vary in trends for mechanical performance with the changing cement reactivity.

3.5.1. Compressive strength (SOF-52)

Fig. 8 shows the increasing trends of the compressive strength for all the replacement levels (SOF-52) from 2 to 91 days. Unlike for MCF, for SOF-52, at the early stage (2 days), all the replacement levels show a slightly higher compressive strength than the reference. Levels of 1 %, 3 %, and 4 % w/w show similar maximum gains (8–11 %). However, by day 14, levels 0–3 % w/w almost do not differ (no gain or loss), while 4–5 % w/w are noticeably worse (a 5 % loss).

By day 28 (the standard for cement), the best performing mortars were 1 % w/w (a 1 % loss), 3 % w/w (a 3 % loss), and the reference. Slightly lower, with close values are 2 (a 7 % loss) and 4 % w/w (an 8 % loss) and the level of 5% w/w showed the lowest compressive strength (a 11 % loss). By day 91, the highest compressive strength is observed for the reference, 2 %, and 4 % w/w (a 1 % gain and a 1 % loss, respectively). Levels of 1 % and 3 % w/w are slightly worse (6 % and 8 % losses). As in the previous case of MCF,

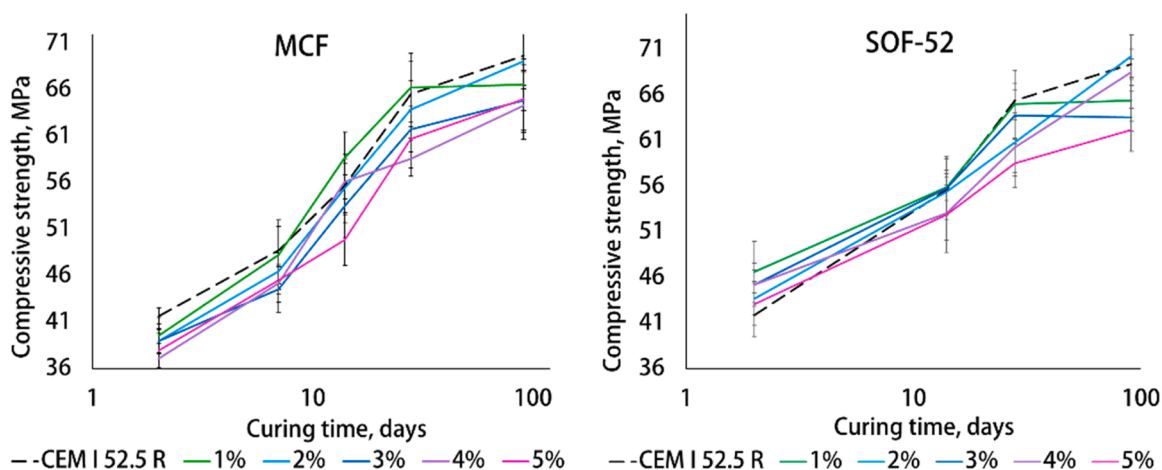


Fig. 8. Compressive strength of mortars (CEM I 52.5 R) containing MCF (right), and SOF-52 (left) (0–5 % w/w cement replacements); the data on the 5 % w/w replacement solely can be found in [19].

only for 2 % w/w the strength change between 28 and 91 days is not observed, while 5 % w/w has the worst performance (a 10 % loss). As it was discussed above, stagnation in the latter tested period of hydration can be attributed to the experimental error.

3.5.2. Compressive strength (SOF-42)

In the case of less reactive CEM I 42.5 N cement (Fig. 9), the outcome is slightly different from the previous cases, although the general trends are similar. Unlike MCF and SOF-52, after 2 days of curing, replacement levels of 1–2 % w/w show a compressive strength slightly higher than the reference (3 % and 9 % gains, respectively), while 3–5 % w/w show significantly lower strength (a 13–21 % loss in strength). However, after 14 d curing, all replacement levels show worse performance in comparison with the reference, while levels of 1–2 % w/w have a very similar strength (4 % and 6 % losses, respectively), and 3–5 % w/w are slightly lower (a 10–13 % loss).

By day 28 (the standard for cement), the strength for the level of 1 % w/w and the reference are almost the same (a 1 % gain). They are followed by 2 % w/w (a 2 % loss), then 4 % w/w (a 4 % loss), and 3 % w/w (a 6 % loss); the level of 5 % w/w shows the lowest compressive strength (a 7 % loss). By day 91, almost all the replacement levels show the same strength, with 3 % w/w being slightly above the reference (a 4 % gain), and all the rest are statistically indistinguishable (a 0–2 % loss). In this case, the stagnation is not observed for any replacement level for the period from 28 to 91 days. It is noteworthy that in the long-term view, the addition of this type of fines appears to be beneficial. This can be explained by the fact that a higher level of hydration is reached later for this cement than CEM I 52.5 R, it can be assumed that BA fines are incorporated into the cement matrix better, while with CEM I 52.5 R, with the ill-incorporated ash particles into the matrix, the negative effect of this phenomenon will become more noticeable over time.

3.5.3. Comparison of mechanical performance between MCF and SOF-52

Fig. 10 shows the final mechanical performance result comparison of the two types of fines, MCF and SOF-52. As can be seen at the early stage (2 days) for all the replacement levels, these fines show better results than MCF and the reference. By day 14, the same result is already observed only for 3 % and 5 % w/w, and for 2 % w/w the results are roughly the same for both types of fines. By day 28, SOF-52 is still better at 3 % and 4 % w/w replacements. For 91 days, this is also true for 2 % and 4 % w/w. The general conclusion is that only with a replacement level of 1 % w/w, a significant effect of fines on the final cement product properties is not observed. However, 2–3 % w/w replacement levels also do not seem to pose a threat. The replacement levels of 4–5 % w/w lead to a decrease in strength (28 days) by 8–10 %, which is also not critical because that is within the allowed measurement error range of 10 % but is no longer negligibly small. Taking into consideration that the preparation of SOF-52 requires additional time and energy costs, their use does not seem to be justified.

3.6. Microstructural analysis of pastes containing MSWI BA fines

To gain more insight into the behavior of MCF on the cement microstructure, optical microscopy was used on cement pastes with different MCF replacement levels. Fig. 11 shows the microstructure of a cement paste containing 0 % (top) and 20 % w/w MCF (bottom) after 7 days of hydration. Each original image was segmented into 3 parts (porosity, reaction product, and unreacted cement/MCF) based on grey values using ImageJ software. Further, the lightest part (unhydrated cement or BA particles) was shown in red, the light-gray part (hydration products) in white, and the darkest areas (porosity) in black. This allows the quantification (area, %) of porosity and the general reaction degree of the sample and may give some insight into a decrease in the mechanical performance of mortars containing BA fines. Table 6 sums up the results.

Fig. 11 and Table 6 reveal that an increase in the share of BA fines is accompanied with a significant increase in the porosity of the cement mixture. However, there is no linear dependence due to packing effects; and for the 5 % w/w replacement level, the porosity is even slightly reduced. It also should be considered that optical microscopy only detects porosity in the micrometer range. Nanoporosity that is a common C-S-H gel cannot be detected. But the heterogeneity of the hardened paste increases with the share of

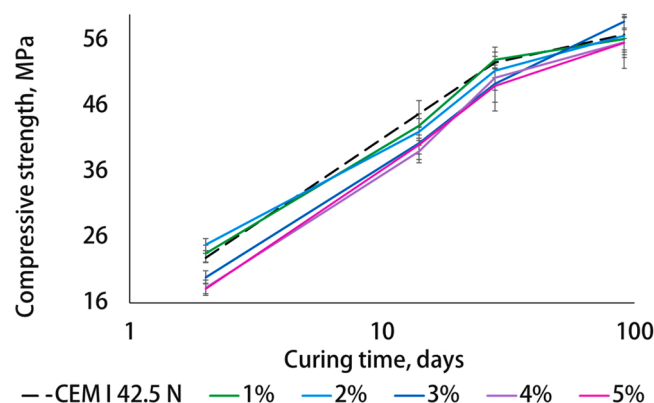


Fig. 9. Compressive strength of mortars (CEM I 42.5 N) containing SOF-42 from 0 % to 5 % w/w cement replacement.

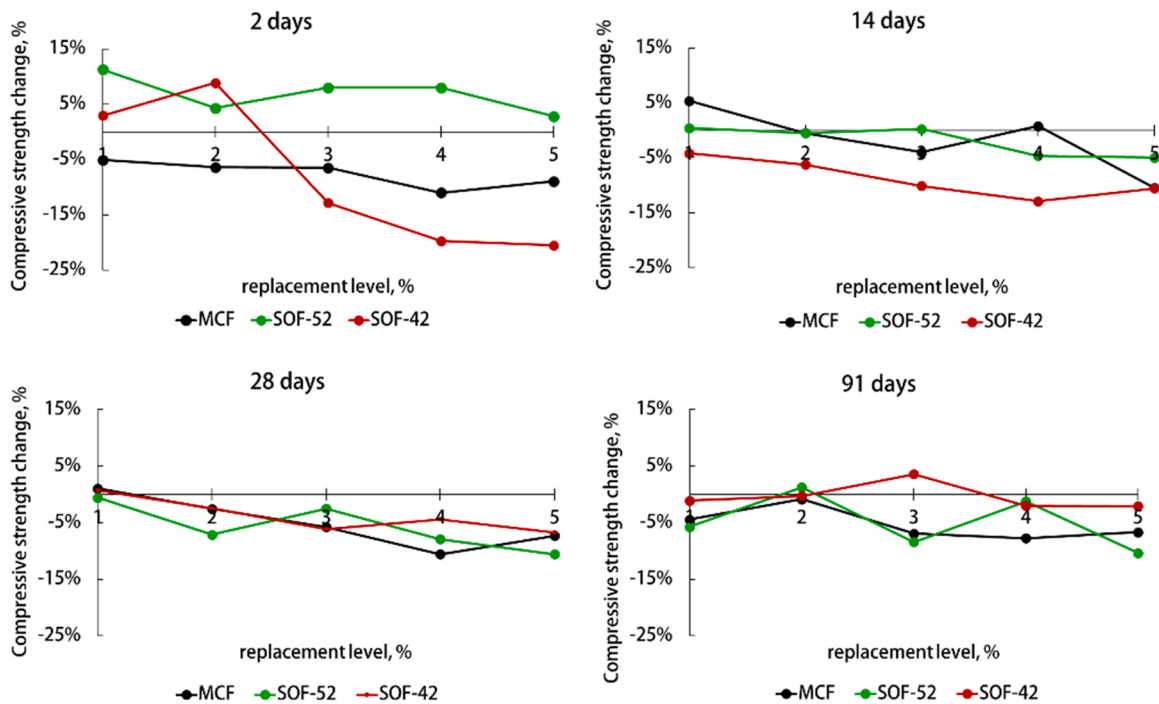


Fig. 10. The compressive strength change in CEM I 52.5 R mortars containing MCF and SOF-52 (from 0% to 5% w/w cement replacements) relative to the corresponding reference.

BA fines, and the material becomes extremely heterogeneous at a 20% w/w replacement (Fig. 11). Table 6 shows that at all the replacement levels, the share of unreacted cement/MCF particles remains the same.

Based on Table 6 data, the hardened paste structure of the reference and a sample containing 1% w/w MCF almost does not differ, neither in composition nor in homogeneity. The only difference is a slightly lower share of unhydrated cement, which can be explained by the fact that in the ash–cement mixture the water: cement ratio becomes slightly higher (as the water: powder ratio remains the same). This is confirmed by the mechanical performance (Section 3.4) and calorimetric data (Section 3.3). Even though for a 5% w/w replacement, the hydration product share increases, and the part of the porous zone diminishes, the heterogeneity of the mixture noticeably increases in comparison with the reference, which may cause the reduction in strength.

4. Conclusions

Thus, this study shows that milled MSWI BA fine fractions can be successfully used as a MAC in pastes and mortars. The tested milled BA fines are three derivatives from a fraction of coarse BA fines (0.125–3 mm): MCF (the coarse fraction milled to the size below 125 μm); SOF-42 (MCF milled to approximate the particle size of CEM I 42.5 N); and SOF-52 (MCF milled to approximate the particle size of CEM I 52.5 R). These three types of BA fines were compared on their performance to identify (1) the most suitable cement replacement level to be used as a MAC, the addition of which does not cause visible detrimental effects in the final products, mortars; (2) the maximum level of replacement by fines which would pass the cement requirements; (3) whether the approximation of the BA fine powder to the size of cement has a positive effect on the mechanical performance of mortars. Environmental properties of all types of BA fines as well as handling a potential risk related to the leaching of dry cement–BA mixtures are discussed.

As well, a methodological feature was stated. It is proposed to test the mechanical performance of MAC-added products on the early stage of hydration (2 days), which significantly shortens further tests as well as the use of the material, because the failure to satisfy these conditions will be critical for using any material as a MAC.

The findings as a whole suggest that the MCF fraction “as is” (mildly milled BA fines) appears to be most suitable for a MAC application. Further milling to produce SOF-52 does not seem to be beneficial from the standpoint of the compressive strength results. This is beneficial for a practical viewpoint as less energy and labor are required to produce this fraction compared to finer milled materials. The optimum cement replacement level where no changes in the material properties are observed is 1% w/w. However, a 2–5% w/w replacement also provides good results passing the cement requirements.

Thus, various properties tested in this paper suggest that all types of fines might be considered as potential MACs; however, milled coarse fines proposed in this study look most attractive from two aspects: (i) its effect on the cement hydration and, therefore, mechanical properties and (ii) their low environmental impact.

The main findings and limitations of the proposed approach can be summarized as follows:

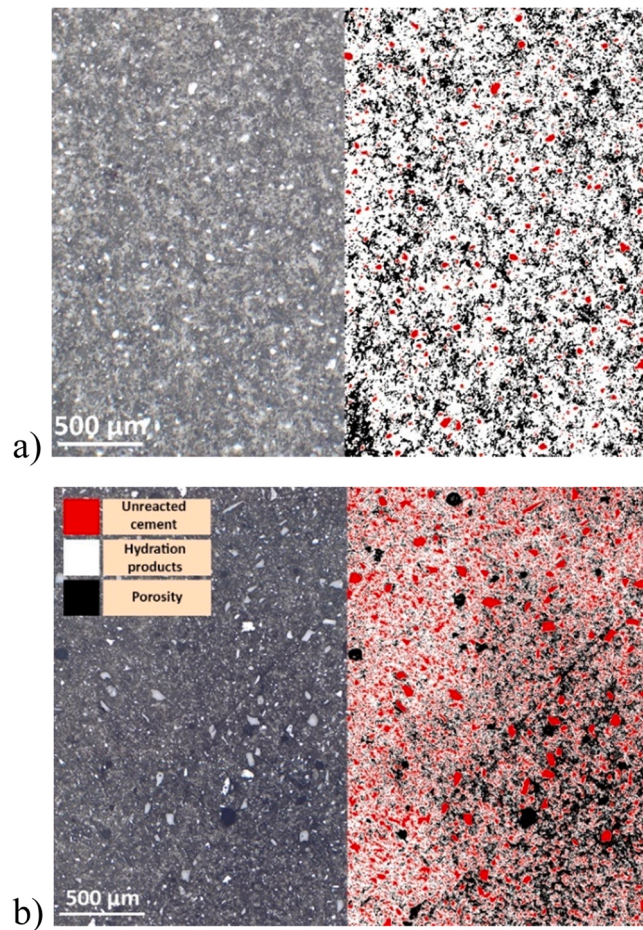


Fig. 11. Microscopic photographs of pastes (a) 100 % CEM I 52.5 R (b) 20 % of MCF + 80 % w/w of CEM I 52.5 R (7 days of hydration). The original photographs (left images) were segmented into 3 areas unhydrated cement or BA particles are marked red; white are hydration products; and black are pores.

Table 6

Distribution of differently hydrated products in cement pastes (7 days hydration), containing 0–20 % w/w cement replacement with MCF; On each photograph, 5 representative areas were selected and analyzed by ImageJ, the results were averaged ($n = 3, P = 0.95$).

% w/w MCF	Non-hydrated or BA %	Reaction product	Porosity
0	3.5 ± 0.8	78 ± 4	18 ± 4
1	2.5 ± 0.2	79 ± 2	19 ± 2
5	6 ± 2	82 ± 4	12 ± 3
20	3 ± 1	60 ± 20	40 ± 20

- The leachate analysis of hardened pastes (2 and 28 days) showed no cases when the legislation limit was exceeded; most elements were at a concentration level below the detection limit.
- Calorimetry showed that the detrimental effects are observed only with a large MCF share in the mixtures ($> 10\%$, a noticeable hydration delay); while for SOF-52 this was observed already at a 5 % w/w replacement. The smallest changes or beneficial effects were seen for MCF 1 % and 5 % w/w.
- Even though for some replacement levels, the compressive strength for SOF-52 appears to be better than for MCF (4 % w/w, 28 and 91 days of hydration, and 3 % w/w but only for 28 days), the reduction in this parameter is still greater for a 4 % w/w replacement (about 8 %) compared with 1 % w/w MCF (an 1 % increase in strength). The fact that the treatment of the material in the case of SOF-52 is more intense should be also considered.
- All investigated BA fines are satisfactory with respect to mechanical performance with a reduction in compression strength within 10 % (28 days). However, the best results were for MCF 1 % w/w with almost no change in the mechanical performance.

- Microstructural analysis of pastes containing MCF showed that on a significant increase in their share, the material porosity and heterogeneity increase significantly, which may deteriorate the mechanical properties of mortars containing BA fines.
- SOF-42, which was used only for the comparison of mechanical performance trends with a different type of cement (similar composition but less reactive), showed good results in the final stages of hardening, where 1–5 % w/w replacements almost do not differ from the reference; compressive strength 98–104 % of the reference at 91 days; 93–101 %, at 28 days; 87–96 %, at 14 days; and 80–109 %, at 2 days. This might be because CEM I 52.5 R is finer and, therefore, more reactive than CEM I 42.5 N; any additional reactions or interactions would result in a more noticeable (harmful for the strength properties) effect. Therefore, it is necessary to study this phenomenon further.

It would be beneficial for further research to compare various properties of different by-products including fly ash and steel slag to investigate the difference of the effect in more detail [39].

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.

Data Availability

Data will be made available on request.

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