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The durability and environmental properties of self-compacting concrete incorporating cold bonded lightweight aggregates produced from combined industrial solid wastes

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HIGHLIGHTS

- Cold bonded lightweight aggregates (CBLAs) are produced from combined industrial wastes.
- Fine incineration bottom ash has been largely recycled in CBLAs.
- Durability of self-compaction concrete with CBLAs are evaluated.
- The SCC with three types of CBLAs complies with environmental legislation.
- A closed recycling loop of industrial solid wastes can be achieved.

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ABSTRACT

Several industrial solid wastes are integrally recycled to produce cold bonded artificial aggregates (CBLA) using the pelletizing technique, and incineration bottom ash fines (BAF, 0–2 mm) are innovatively used to strengthen the pellet strength. Three types of CBLAs are produced, in which BAF, nano-silica produced by olivine dissolution and polypropylene fibre are applied to improve the aggregates' properties (strength, etc.), respectively. The influence of these different types of CBLAs on the designed self-compacting concretes (SCCs) are experimental study and compared. The fresh and hardened properties of the concrete with and without CBLAs are investigated, including slump flow diameter, t_{500} time, V-funnel time, bulk density, flexural and compressive strength, etc. Moreover, the durability of the concretes is studied through water penetration and freeze-thaw tests. Additionally, the leaching behaviour of heavy metals and salts from the concretes are evaluated through different leaching tests according to environmental legislation. The results show that the roundish particle shape of CBLAs benefit the flow of concrete in fresh stage, and the strength of concrete with CBLAs has linear relation with its bulk density, and the cumulative mass loss profiles during freeze-thaw tests were influenced by the types of CBLAs. The leaching tests show that the concretes containing CBLAs are environmental non-hazardous.

1. Introduction

Construction and industrial solid wastes are nowadays attracting worldwide attention regarding to their dispose, or recycle and reuse as secondary resource [1,2]. The application of recycled concrete aggregates as alternative aggregates in construction field has been reported widely and promoted successfully [3,4]. The industrial solid wastes also have been used as building materials in recent years. For instance, coal fly ash (FA) from coal-fired power plants is generally used as filler or pozzolanic materials [5,6], and combustion bottom ash can be used as aggregates in concrete [7]. Municipal solid waste incineration (MSWI) bottom ash has been used as aggregates in concrete [8,9]. Paper sludge ash (PSA) from paper industry is reported to be used as pozzolanic additives or to produce hydrophobic concrete [10,11]. Washing aggregate sludge (WAS) from aggregate production has been used to produce lightweight aggregates under high temperature [12]. The MSWI bottom ash is a dominant by-product in waste-to-energy plant for the management of municipal solid waste and further treatments are needed to remove the salts and heavy metals from bottom ash to make its leaching properties comply with the legislation [13]. However, it is reported that these treatments have less

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efficiency on the fine fractions of MSWI bottom ash which contains higher amount of contaminants than the coarser fraction (>4 mm), hence, this fine MSWI bottom ash remains an issue that needs research.

It is worth to mention that those industrial solid wastes have their own intrinsic properties which may cause negative influence in application while might bring benefit in other situations. For instance, the presence of metallic aluminium in MSWI fly ash leads to the generation of hydrogen when used in concrete, which decreases the strength of concrete products [14], while when used in autoclaved aerated concrete, the metallic aluminium can be used as aerating agent [15]. Therefore, it is of interest to figure out the integral recycling possibility of these industrial wastes, in a way to transfer their drawbacks into benefits.

Pelletization techniques have been applied as a way to recycle the powdered wastes for producing artificial lightweight aggregates under high temperature with foaming agent or at room temperature with binders [16,17]. The production of artificial lightweight aggregates by a cold bonding pelletization technique based on solid wastes has been studied and reported in literature [18–22]. By using this technique, the industrial solid wastes can be recycled and reused to decrease the demand of space for landfilling; meanwhile, an artificial lightweight aggregate can be produced which could be used as aggregates in concrete, subsequently, the consumption of limited natural resource for aggregates can be reduced; in addition, most of the industrial solids contain contaminants (salts or heavy metals) which may pollute the environment and need to be stabilized before disposal, pelletization of these wastes with proper binders to produce artificial aggregates is a way to combine stabilization treatment and recycle of solid wastes. Hence, it is a promising method for the reuse and recycle of increasing industrial solid wastes. However, in most of the cases, unitary raw waste material (mainly powders) is used for pelletization except the binder [17] and the industrial solid wastes are produced daily and proper way to dispose them is in demand. On the other hand, these wastes can be very beneficial if being used in proper way. Hence, an integral recycle/reuse of those industrial wastes is of importance to be considered applying the pelletization technique.

The properties of the artificial aggregates have significant effects on their application in concrete as gravel replacement [23], and the crushing strength of the artificial aggregates is partially related to the strength of the concrete products. Therefore, methods for increasing the aggregate strength were studied [17,21]. It is addressed that the increasing of binder amount or curing under streaming condition can result in higher aggregates strengths [24]. However, there are rather few other methods for improving CBLA properties.

Hence, the main goal of this research work includes the recycling of combined industrial solid waste innovatively to produce cold bonded lightweight aggregates (CBLAs) and improving the properties of the produced aggregates. The methods of improving the pellets are: (1) use of MSWI bottom ash fines (BAF, 0–2 mm) to improve the skeleton strength of pellets; (2) adding polypropylene fibre (PPF) as reinforcement of the pellets; (3) applying nano-silica (nS) as binder replacement.

Several types of industrial solid wastes in the Netherlands are recycled integrally to produce CBLA by applying the pelletizing technique. Three types of CBLAs are produced with consideration of improving their aggregates' strength, including the use of BAF, nano-silica from olivine dissolution and polypropylene fibres. The properties of these CBLAs are evaluated and compared with the ones produced by others in literature. Subsequently, these CBLAs are used in self-compacting concrete to replace natural gravels and to study their influences on the concrete properties related to the properties of the CBLAs. The fresh and hardened properties of the concretes are tested and the durability properties of the concretes are studied through the water penetration and freeze-thaw tests. Finally, the environmental impact of the concretes with artificial aggregates are evaluated using two types of leaching tests and results are compared with the Dutch legislation.

2. Materials and experimental methods

2.1. Raw materials and their characteristics

The cement used in this study is CEM I 42.5 N (OPC) provided by ENCI (the Netherlands) for the production of aggregates and concrete mixtures. The powder coal fly ash (FA) used is obtained from a Dutch power plant and paper sludge ash (PSA) from a Dutch paper recycle company. The washing aggregate sludge (WAS) is provided by a gravel production company (Smals, the Netherlands). The municipal solid waste incineration (MSWI) bottom ash fine fractions (BAF, 0–2 mm) is provided by a waste-to-energy plant in Moerdijk (Attero, the Netherlands) by directly sieving from the bottom ash heap in the plant. The polypropylene fibre (PPF) with a length of 3 mm, density of 0.91 kg/m^3 is provided by FBG (the Netherlands) and Bonar (England). The nano-silica used in this study is the same as applied in [25] and [26] produced by olivine dissolution. The sand (0–4 mm) and gravels (2–8 mm and 8–16 mm) used for concrete are provided by Smals (The Netherlands).

The chemical compositions of the materials are measured by X-ray fluorescence (XRF) and the crystalline phases present in the materials are detected by the X-ray diffraction (XRD, Cu tube, 40 kV, 30 mA, 3–75°, 0.02°/step, 0.2°/min). Table 1 shows that the main chemical compositions of all the materials used belong to the SiO_2-CaO-Al_2O_3-Fe_2O_3 system. WAS contains mainly SiO_2, and a considerable amount of Al_2O_3 and Fe_2O_3, and PSA consists high amounts of CaO, and SiO_2 and Al_2O_3. The main chemical compositions in FA and BAF are SiO_2, Al_2O_3 and Fe_2O_3. Fig. 1 shows the crystalline phases in PSA, FA, WAS and BAF. It is detected that the main crystalline phases in WAS are quartz, magnetite, clay minerals of the chlorite family, and feldspar; in PSA there are calcite, portlandite, gehlenite, and calcium silicate found. FA consists of quartz and mullite, and BAF contains quartz, calcite, hematite, feldspar and anhydrite.

The particle size distributions (PSDs) of the powder materials are measured using laser diffraction (Mastersizer 2000 Malvern) and the PSD of the particle aggregates is measured following EN 933-2 [27]. A helium pycnometer (AccuPyc II 1340) is applied to measure the specific densities. The PSDs of the materials for producing aggregate are shown in Fig. 2 and their densities are listed in Table 1.

2.2. Artificial aggregates production and their properties

The artificial aggregates are produced using a disc pelletizer, which has a pan diameter of 100 cm and collar height of 15 cm as shown in Fig. 3(a). The vertical angle of the pan is 45° and the running speed is 15 rpm during the process. The raw materials used for the aggregates are firstly mixed homogeneously and then around 10 kg of the mixed materials are put on the running disc (Zone 1 in Fig. 3(a)). After around 5 min of running, about 2 kg of water is sprayed continuously on to the materials in the pan and then they are running again for 10 min. The next running cycle starts with the addition of another 10 kg of mixed materials to the running pan (Zone 1), and the rounded artificial aggregates from the last round will drop off automatically from the left lower corner of the pan (Zone 2 Fig. 3(a)). The freshly produced aggregates (Fig. 3(b)) are sealed in plastic bags for further use and tests.

Table 1
The chemical compositions and specific densities of the raw materials.

[% wt]	OPC	WAS	PSA	FA	BAF
CaO	67.85	1.46	54.94	6.18	18.58
SiO_2	14.85	73.31	13.64	45.20	39.13
Al_2O_3	3.62	11.29	8.64	27.49	7.60
Fe_2O_3	3.33	6.00	0.99	6.63	12.93
Na_2O	*	0.61	*	0.96	1.02
K_2O	0.82	1.85	0.46	2.17	1.09
MgO	1.57	0.89	2.09	1.36	1.93
P_2O_5	0.39	0.07	0.30	0.78	0.86
TiO_2	0.30	*	0.74	*	*
MnO	0.08	0.12	0.03	0.07	0.17
CuO	0.02	0.00	0.07	0.02	0.38
ZnO	0.07	0.01	0.08	0.05	0.66
Cl	0.11	0.00	2.24	*	0.29
SO_3	4.46	0.16	0.99	1.70	4.33
Others	0.36	0.59	0.19	2.08	1.89
LOI	2.16	3.64	14.59	5.31	9.17
Density [g/cm^3]	3.1	2.68	2.71	2.32	2.69

* Not detected.

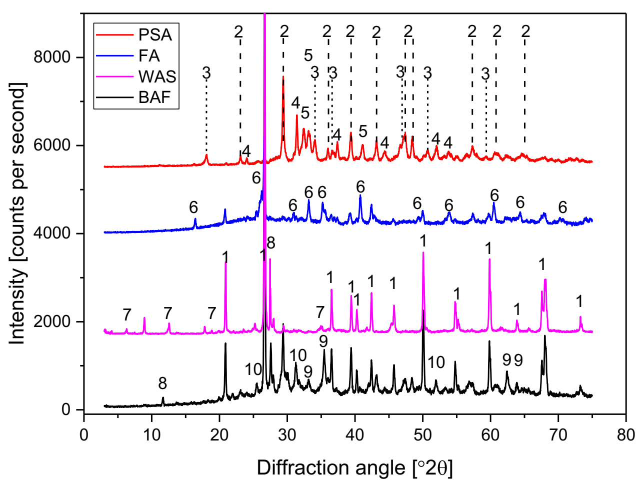


Fig. 1. The XRD pattern of the waste raw materials (1: Quartz; 2: Calcite; 3: Portlandite; 4: Gehlenite; 5: Calcium silicate; 6: Mullite; 7: Chlorite; 8: Feldspar; 9: Hematite; 10: Anhydrite).

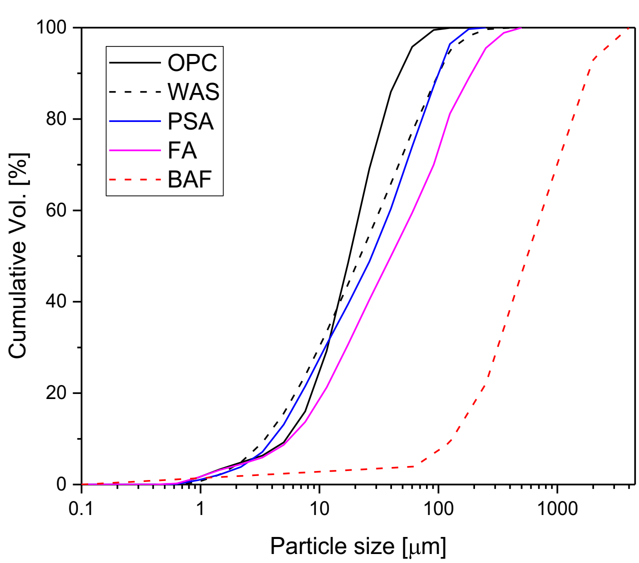


Fig. 2. The particle size distribution of the raw materials used.

There are three types of aggregates produced in this study with the purpose of modifying the strength of the pellets; the raw materials used for Sample 1 (S1) are OPC (10%), BAF (a value between 40 and 75%), WAS (5%), PSA and FA (around 10–45% in total) based on total dry mass. Sample 2 (S2), around 2.5% vol. polypropylene fibre (PPF) is added to the mixture of the raw materials, the other proportions are the same as for S1. Sample 3 (S3), around 0.8% of the cement is replaced by nano-silica, and the amount of the other materials are kept the same as S1.

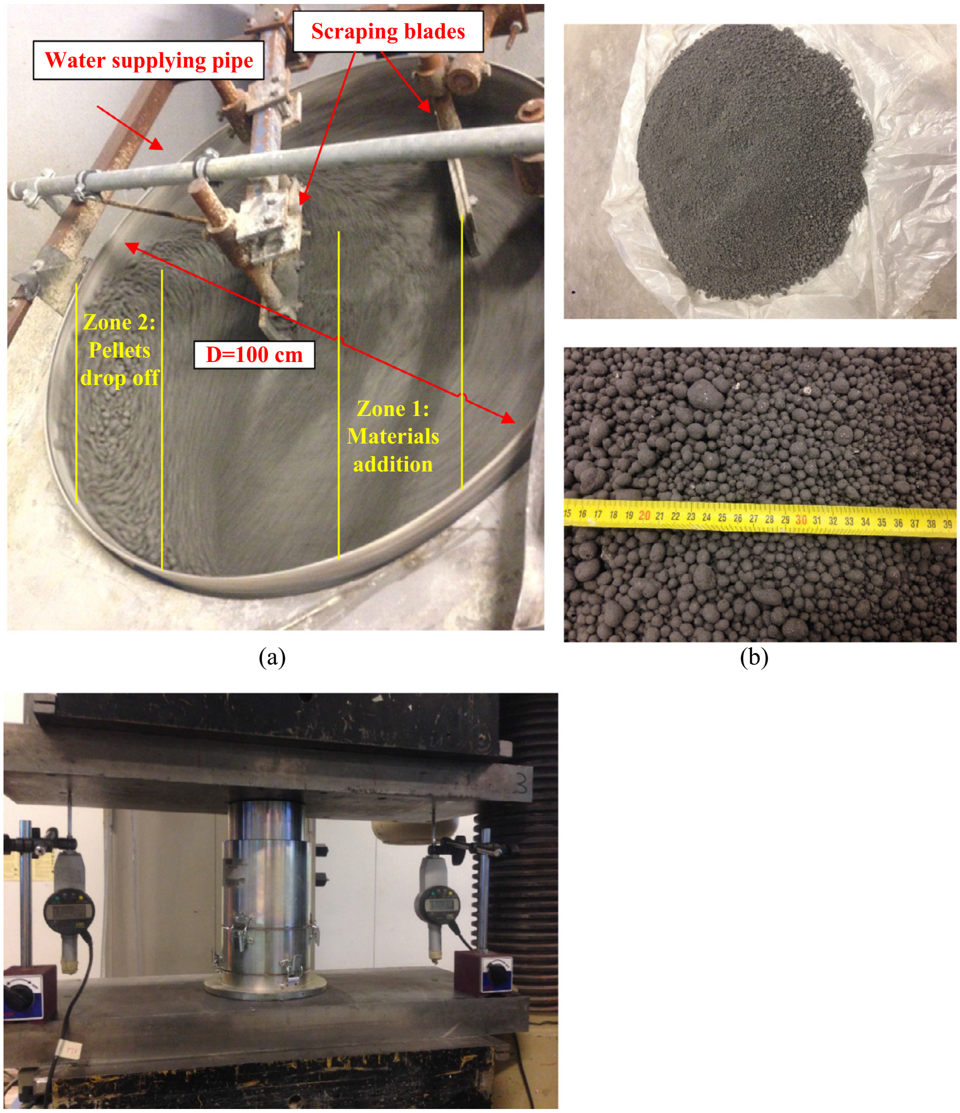
The particle size distribution (PSD) of the produced aggregates is determined using the sieving methods according to standard EN 933-2 [27] and their bulk densities are evaluated according to EN 1097-3 [28]. The water absorption (WA) of the artificial aggregates are tested following the procedure in EN 1097-6 [29] and the crushing resistance (CR) of the artificial aggregates are measured according to standards EN 13055-1 (Annex A, procedure 1) [30], the device used is shown in Fig. 3(b). The PSDs of the produced aggregates and gravels are shown in Fig. 4 and the fineness modulus (FM) of S1, S2, S3 tested according to EN 12620 [31] is 5.94, 5.93 and 5.92, respectively.

2.3. Concrete mixture design and test

A self-compacting concrete (SCC) is designed; the materials used are OPC, coal fly ash, sand with a particle size under 4 mm (FM is 2.90), crushed gravel with a particle size between 2 and 8 mm (FM is 5.69), and gravel with a particle size between 8 and 16 mm (FM is 5.92). The proportions of materials for each concrete mixture are listed in Table 2. A superplasticizer (SP, MasterGlenium 51con 35%, BASF) is used to adjust the flowability of concrete and its amount is fixed at 5.89% of the OPC for all the concrete mixtures.

The gravels (2–8 mm and 8–16 mm) in the concrete are replaced by the produced artificial aggregates (S1, S2, S3) with 30% and 60% by volume. The amount of each aggregates in concrete mixtures is calculated based on the idea that the particle size distribution of all the aggregates for the concrete should be similar to that of the reference concrete. Before mixing, the artificial aggregates are immersed in water for 24 h and then dried in open air for 2 h to avoid their absorption of water during concrete mixing.

The slump-flow of the freshly mixed concrete is tested following the procedure described in EN 12350-8 [32], the largest diameters of the flow spread on the two vertical dimensions are recorded and their average value is present as the slump-flow



(a) (b)

(c)

Fig. 3. (a) The disc pelletizer used for artificial aggregate production, (b) the produced aggregate and (c) devices for lightweight aggregate crushing resistance test.

(S-F) of the concrete. During this test, the time used for the concrete to first reach the 500 mm circle on the testing plate after the cone is lifted up is recorded and presented as t_{500}. The V-funnel test is performed on the fresh concrete according to the procedure defined in EN 12350-9 [33], and the time used for the concrete finishing flow out of the V-funnel is recorded as t_v. The bulk density of the fresh concrete is tested according to EN 12350-6 [34]. The fresh concrete is filled in cubic moulds ($150 \times 150 \times 150$ mm^3) and prism moulds ($100 \times 100 \times 500$ mm^3) and demoulded after 1 day, then cured in plastic buckets until testing date. The compressive strength tests are performed on the concrete cubes, and two-point loading flexural strength tests (Fig. 5(a)) are conducted on the concrete prisms after 28 and 56 days curing according to EN 12390-4 and EN 12390-5 [35,36], respectively.

The water penetration tests on the hardened concrete cubes under pressure are performed following the procedure described in EN 12390-8 [37] and the device used is shown in Fig. 5(b). The freeze-thaw resistance of the hardened concretes are evaluated by performing the freeze-thaw attack with de-icing salt on the concrete slabs sawn from concrete cubes (Fig. 5(c)) according to NPR-CEN/TS 12390-9 (part 5) [38], the mass loss of the tested samples after each testing cycling (7, 14, 28, 42 and 56 cycles) is recorded.

2.4. Leaching properties of BAF, artificial aggregates and concrete

Due to the fact that the MSWI BAF is a by-product from combustion of waste mixtures, it is reported to contain a certain amount of environmental contaminants which have the potential to leach out when in contact with water. From our previous studies [39,40], it is noticed that the leaching of antimony (Sb), molybdenum (Mo), copper (Cu), chloride and sulphate is the main leaching problem in the investigated MSWI bottom ash. Hence, the leaching test evaluation is mainly focused on these elements and salts in the current work as well. According to the SQD [13], for a non-shaped materials (materials which can be used as they are without further modification) the upper limit leaching value of Sb, Mo, Cu, chloride and sulphate is 0.32 mg/kg d.m., 1.0 mg/kg d.m., 0.9 mg/kg d.m., 616 mg/kg d.m. and 2430 mg/kg d.m., respectively. If the leaching value of sample exceeds the limit value as a non-shaped material, the sample should not be directly used as a building material. The leaching properties of the CBALs and the concrete which contained

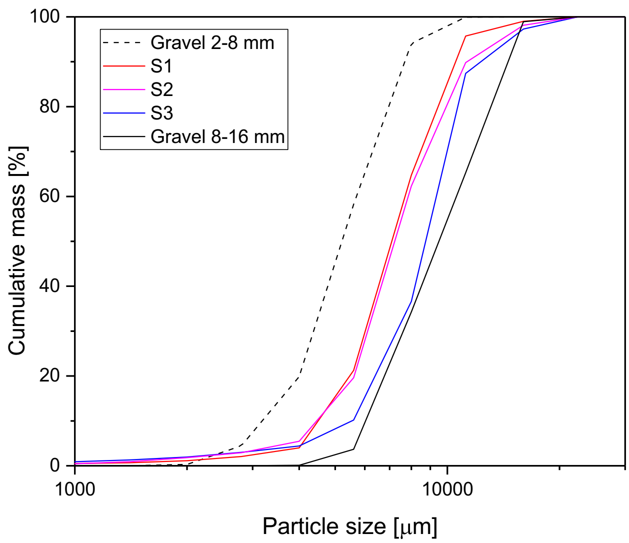


Fig. 4. The particle size distribution of the gravel used for concrete and the produced artificial aggregates.

the CBLAs are evaluated according to the applicable Dutch legislation – the soil quality decree (SQD) [13].

The samples before column leaching test according to NEN 7383 [41] is crushed under 4 mm and dried under 105 °C. The column used for this test has a diameter of 5 cm and length of 25 cm, the test temperature is 20 ± 2 °C. Around 800 g of representative sample is filled into the column and then water is forced to flow through the sample from bottom to the top for around 22 days, then the concentration of heavy metals in the leachate is determined by ICP according to NEN 6966 [42] and the salts are tested through high performance liquid chromatography (HPLC) following NEN-EN-IOS 10301-2 [43].

For determining the total amount of investigated elements, the testing samples are firstly crushed and then milled into powder, and then the mixture of nitric acid and hydrochloric acid (aqua regia) is used to digest the solid sample (NEN 6961 [44]). Then the digested solution is extracted and subjected to inductively coupled plasma mass spectrometry (ICP) to determine the total concentration of the observed heavy metals. The content of chloride and sulphate is determined according to EN 1744-1 [45].

The leaching behaviour of the investigated elements from the solid samples under column leaching tests were also described using their leachabilities, which demonstrates the leached-out degree of element from solid samples. The leachability of investigated samples is expressed and calculated according to the following equation:

$$\text{Leachability}(\%) = \frac{\text{Leached out concentration}}{\text{Total concentration}} \times 100$$

where the leached-out concentration means the detected concentration from liquid after the column leaching test on the solid samples, and the total concentration means the extracted concentration from the acid digestion of the corresponding solid samples.

The hardened concrete cubes were sent to commercial lab for tank leaching test according to NEN 7375 [46]. The dried concrete samples were merged in demineralised water for specified time, and then the leachate was renewed by new demineralised water. The volume ratio of liquid to tested sample was 2. The pH and concentration (ε) of components in the changed leachate were determined. The time to renew the water is 6 h after the samples were merged in water, and then 1, 2.25, 4, 9, 16, 36 and 64 days. The cumulative leaching concentration of a component after 64 days (ε_{64}) was a sum of its concentration in each renewed leachate. For each concrete mixture, duplicate samples were tested.

3. Results and discussions

3.1. Properties of three types of artificial aggregates

Plenty of research has been done on the recycling of powder coal fly ash or powder industrial wastes to produce cold bonded lightweight aggregates (CBLAs) [21,22,47]. Their results shown that the produced CBLAs have bulk densities (BD) of around 800–1600 kg/m^3, and water absorption (WA) between 14 and 27%, and a crushing resistance (CR) of approximately 1.9–6.4 MPa. The BD, WA and CR have a linear correlation, there is negative correlation between BD and WA, while positive correlation between BD and CR. The increase of the BD indicates a lower porosity of the CBLAs, subsequently a lower WA, and a higher CR can be achieved. Hence, increasing of binder content, and prolonging the pelletizing duration to obtain a denser microstructure, could guarantee the increase of CR. In this study, the bulk density (BD) of S1, S2 and S3 is approximately 923 kg/m^3, 870 kg/m^3, and 844 kg/m^3, respectively. The WA and CR of S1, S2 and S3 are 19% and 9.6 MPa, 22.4% and 9.9 MPa, and 23% and 7.56 MPa, respectively. Fig. 4 depicts that the particle sizes of the produced aggregates are mainly between 4 and 12 mm, and the sample S3 has coarser particles than S1 and S2, while samples S1 and S2 have very similar particle size distribution.

The BD and WA of S1 are quite similar to the ones in literature, while its CR is much higher. The primary difference between S1 and the CBLAs in literature is the use of municipal solid waste incineration (MSWI) bottom ash fines (BAF, 0–2 mm) instead of powered solid (fly ash, etc.). The beneficial use of BAF to produce CBLAs can be summarized as following: (1) the BAF can act as ‘aggregates’ in the pellets to strengthen the bonding and stability of the cementitious matrix due to the irregular particle shape of BAF; (2) the BAF is quite porous which will not increase the BD of CBLAs significantly, it even can reduce the BD if the proper amount of BAF is applied; (3) the BAF is innovatively recycled and reused in a profitable way without any future treatments,

Table 2
The design for concrete mixtures.

Materials	Ref.	Mix 1		Mix 2		Mix 3	
		30% Vol.	60% Vol.	30% Vol.	60% Vol.	30% Vol.	60% Vol.
	[kg/m^3]						
CEM I 42.5 N	310						
Coal fly ash	179						
Sand 0–4 mm	572.9						
Gravel 2–8 mm	697.8	566.52	435.24	577.07	456.33	667.05	558.38
Artificial aggregates	0	241.43 (S1)	483.00 (S1)	234.29 (S2)	468.71 (S2)	236.14 (S3)	472.14 (S3)
Gravel 8–16 mm	376	249.57	123.13	239.02	102.04	149.04	0
Water	139.5	139.5	139.5	139.5	139.5	139.5	139.5
SP	5.89						

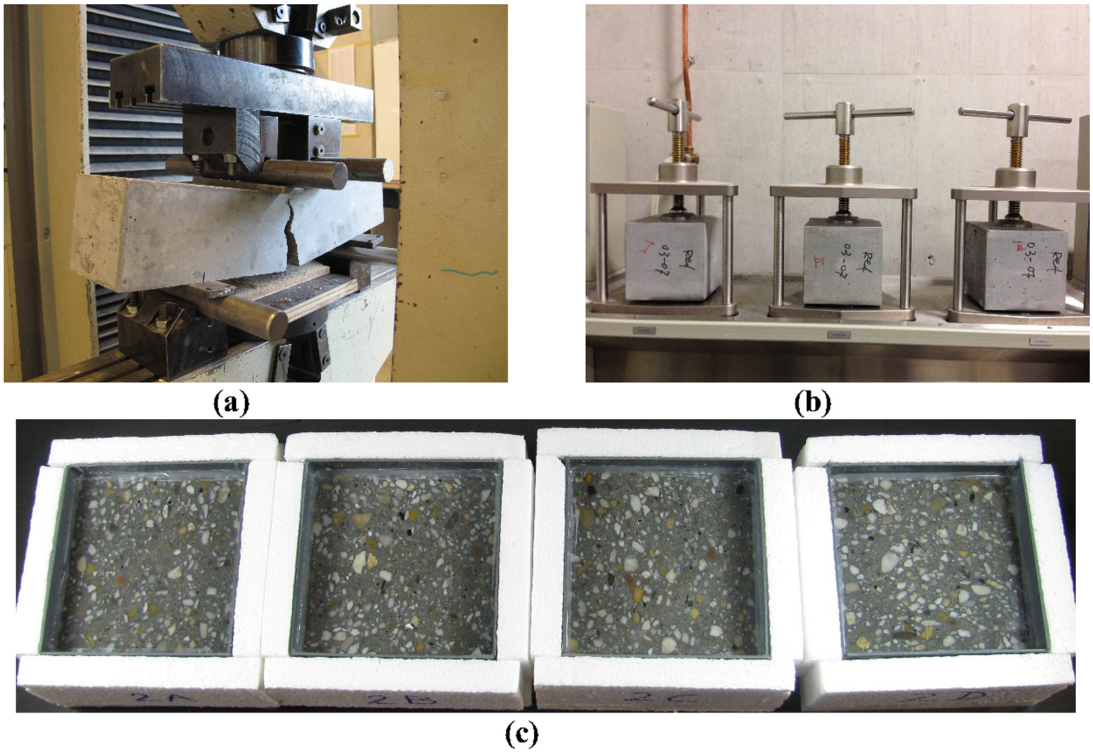


Fig. 5. (a) The device for flexural strength test, (b) water penetration test and (c) prepared samples for freeze-thaw test.

compared to its plain application in concrete whereby treatments are needed and/or deterioration effects are more pronounced. Therefore, the use of porous BAF to replace partially the powders used generally for producing CBLAs, with the purpose of increasing the CR of CBLAs, is proved to be an effective method, which is rarely investigated before.

Compared with S1, the compaction of the pellets during the pelletization of S2 is more difficult due to the addition of PPF, which results in a lower BD and then higher WA of CBLAs. However, the PPF in the cementitious matrix of S2 can serve as reinforcement to increase the strength of pellets from a mechanical point of view [48]. Subsequently, the CR of S2 is increased, even though its BD is decreased compared with S1. In this study, the PPF is for the first time used in a pelletization technique together with mixed industrial solid wastes, and it is proved that it is possible to use PPF as reinforcement to produce CBLAs with lower BD but higher CR. Moreover, considering the sustainable development of building materials, the use of waste recycled fibres in future study can be investigated during pelletizing.

Previous research studies have stated that the use of nano-silica (nS) in a cementitious matrix can improve its strength, durability, etc. [26]. Hence, it was expected that the use of nS can also bring profit for the strength of CBLAs. Compared with S1, sample S3 has lower CR, which is opposite to the nS function as reported. This result can be attributed to the following reasons: (1) the high specific surface area (SSA) of nS leads to very fast agglomeration of powders, subsequently a fast-growing speed of granulates; this phenomenon was observed during the pelletizing process. However, a thick moisture layer exists on their surface due to its high SSA, the water layer generates a considerable tension force which limits the compaction of the pellets during the pelletizing process. Furthermore, more capillary pores will be left after the evaporation of this water layer, leading to a porous microstructure of the pellets and lowering the CR. (2) it can be seen in Fig. 4 that S3 has a slightly coarser particle size than S1 and S2, which can be attributed to the high SSA. According to the research performed by Colangelo et al. [21], the CR of the smaller fraction is higher than that of the coarser fraction. Hence, the CR of S3 can be influenced partially by its coarse particle size. However, the quantification of this influence needs to be studied in the future. In addition, it was observed that the addition of nano-silica in gel form can cause heterogenization of granulates. In a future study, silica fume can be considered for a better distribution.

It is shown that the mixed industrial solid wastes are compatible to be integrally recycled to produce cold bonded lightweight aggregates with good crushing resistance. With the addition of BAF, PPF and nano-silica, the CR or the pelletizing process of the CBLAs can be further improved.

3.2. Properties of fresh concrete

3.2.1. Slump flow diameter

Fig. 6 shows the slump flow (S-F) diameter of the fresh mixed concrete with and without CBLAs. The reference concrete (Ref.) has a SF diameter around 753 mm, which belong to class SF2 according to EN 206-9 [49].

For concrete mixture 1, its S-F diameter has a slight increase (0.7% increase) compared to the reference when 30% vol. of artificial aggregates (S1) is added to replace natural aggregates, while a decrease of S-F diameter is observed when 60% vol. of aggregate is replaced by S1 (around 3% reduction). The S-F diameter of concrete mixture 1 with artificial aggregates (S1) with 30% and 60% vol. replacement still belongs to class SF2 according to EN 206-9 [49]. Concrete mixtures with CBLAs-S2 (Mix 2) shows a decrease of S-F diameter, and this diameter decreases with the increase of the S2 replacement level. The S-F diameter decreases by 15% compared to the reference when 30% vol. of S2 is used, and it belongs to SF1. Around 35% reduction of the S-F diameter of the concrete with 60% vol. of S2 is observed. For concrete with CBLAs-S3 (Mix 3), its S-F diameter has a slight decrease (about 3%) compared to the

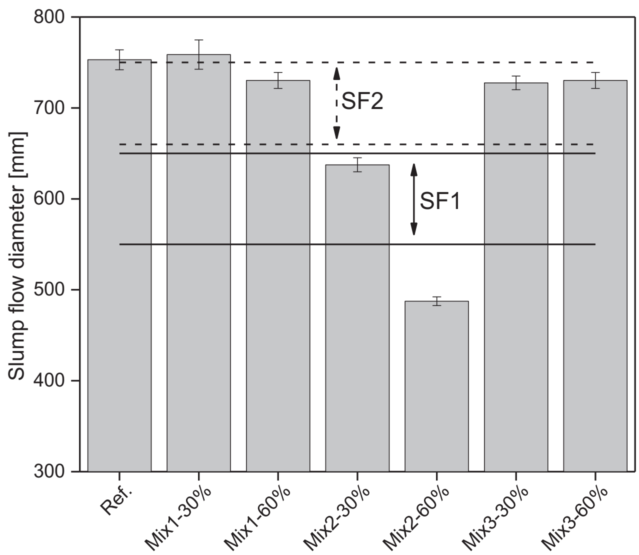


Fig. 6. The slump flow diameter of the fresh concrete mixtures.

reference for both mixtures with 30% and 60% vol. of S3, which belongs to SF2 according to EN 206-9 [49]. The increased use of S3 to replace natural aggregates does not show a significant influence on the SF diameter.

The slump flow diameter indicates the flowability and filling ability of the fresh self-compacting concrete (SCC). The above shown results demonstrate that the addition of artificial aggregates S1 and S3 up to 60% vol. will not influence the flowability of SCC dramatically. As reported by other researchers [50], the round shape of CBLAs promotes the flowability due to less friction between particles and matrix, hence, the use of chemical admixtures for increased flowability can be reduced. In this study, the dosage of SP for all the mixtures is kept the same, which contributes to less viscosity of the concrete when CBLAs are added. Furthermore, the CBLAs are lighter than the natural gravel. Due to the above-mentioned reasons, the S-F diameter of concrete with CBLAs is lower than that of the reference.

The addition of S2 to SCC reduces notably its SF diameter, which is contrary to the phenomena reported in some of the research related to application of artificial aggregates [51,52]. For CBLAs-S2, PPF is used to increase the pellets strength. During the pelletization, part of the fibres is embedded inside the pellets completely, to support the pellet strength as a role of reinforcement. However, there are also some fibres which are just partially embedded inside the pellets as shown in Fig. 7. During the concrete mixing, these fibres will increase the cohesive force between the artificial aggregates and the matrix, similar to the influence of PPF in concrete [53]. Hence, the flowability of the fresh concrete (Mix 2) is decreased dramatically compared to Mix 1 and Mix 3.

3.2.2. $t_{500.}$ time

During the slump-flow test, the time used by the concrete to flow to a diameter of 500 mm is recorded and the results are displayed in Fig. 8. The designed SCC reference has a t_{500} of around 8 s, which is in the range of viscosity class VS3 according to EN 206-9 [49].For the concrete mixture with CBLAs-S1 (Mix 1), the t_{500} of Mix 1 with 30% vol. of S1 is around 2% less than the reference, while Mix1 with 60% vol. of S1 has a t_{500} about 1% longer than the reference. The t_{500} of Mix 2 with 30% vol. of S2 is around 8% longer than the reference, while it is almost 13% higher than the reference when 60% vol. of S2 is used. The t_{500} of Mix 1 and Mix 2 with 30% and 60% vol. of CBLAs indicates that their viscosity class belongs to VS3 according to EN 206-9 [49], the same as the reference. The t_{500} of the concrete mixture with CBLAs-S3 (Mix 3) decreases significantly compared with the reference; the reduction for Mix 3 with 30% and 60% vol. of S3 is around 48% and 54%, respectively. The viscosity class of Mix 3 belongs to VS2 according to EN 206-9 [49].

The t_{500} of concretes with S1are not dramatically affected by the CBLAs' replacement level, while the t_{500} has a notable increase when more S2 aggregates are applied to replace natural aggregates, which is due to the blocking effect of fibres on the surface of the pellets as explained above. The reduction of t_{500} for concrete Mix 3 may be due to the following reasons: the sample S3 has coarser particles than S1 and S2, and it can be seen from Table 2 that all the coarse gravels are replaced by S3 in order to keep a similar total particle size distribution of aggregates; in this way, there is much less friction between the mortar matrix and the coarse aggregates, which indirectly decreases the viscosity of the matrix; thus, less time is needed to reach the 500 mm spread.

3.2.3. V-funnel test

Fig. 9 shows the time used for the V-funnel test on the fresh mixed concrete samples. It takes around 23 s for the reference concrete to flow out of the V-funnel, and this demonstrates that the viscosity class of reference concrete is VF2 according to EN 206-9 [49].

The t_v of Mix1 with 30% vol. of S1 increases with around 7% compared with the reference concrete, while it takes more than 2 times longer for Mix 1 with 60% vol. of S1 in the V-funnel test. Mix 2 with 30% vol. of S2 has a longer t_v than the reference, approximately 10%, and when 60% vol. of S2 is applied its t_v is almost 3 times longer than reference. The use of 30% vol. of S3 in Mix 3 results in a 40% reduction of t_v, while 60% vol. of S3 application in concrete causes a t_v around 1.7 times longer. The t_v for all the concrete mixtures with CBLAs increase with the increasing amount of aggregate replacing level. During the tests, it was observed that the artificial aggregates were pushed up and accumulated on top of the tested samples due to their roundish particle shape and lower viscosity. The lower viscosity may result in segregation of concrete. Thus, the roundish particles did not fell out of the V-funnel bottom until the mortar matrix finishing flow out, which is believed to result in the much longer t_v during test when 60% vol. of CBLAs are used. Therefore, with the artificial aggregates replacement, the V-funnel test cannot demonstrate the viscosity of the concrete mixture accurately when the concrete segregation tends to happen.

3.2.4. Bulk density of fresh concrete mixtures

Fig. 10 indicates the influence of artificial aggregates replacement on the fresh density of concrete mixtures. It can be seen that the use of S1, S2 and S3 leads to the reduction of the fresh density of the concrete, and the higher their replacement level, the lower the fresh density of concrete. The fresh density of concrete with S3 is the lowest compared with concretes with the same content of S1 or S2. The bulk density of sample S1 is higher than the other two, and S3 has the lowest bulk density. The fresh density of the concrete is related closely to the bulk density (as shown in Table 2) of the materials used for mixing; the results obtained in this study are consistent with literature [52].

It can be shortly summarized that the use of cold bonded artificial aggregates (S1and S2) does not influence the slump flow of the concrete, while the use of S2 decreases the slump flow due to the blocking effect of PPF. The use of CBLAs (S1 and S3) can reduce the viscosity of concrete due to their roundish particle shape, hence, the dosage to modify the water requirement can be reduced. The use of CBLAs in concrete reduces the bulk density of fresh concrete.

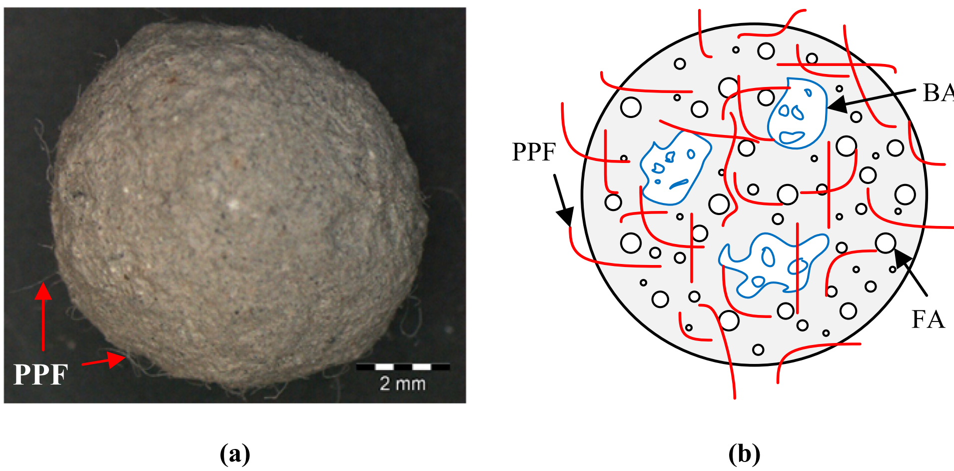


Fig. 7. (a) The artificial aggregate pellet with PPF addition under optical microscope and (b) the schematic graph of the pellet cross section.

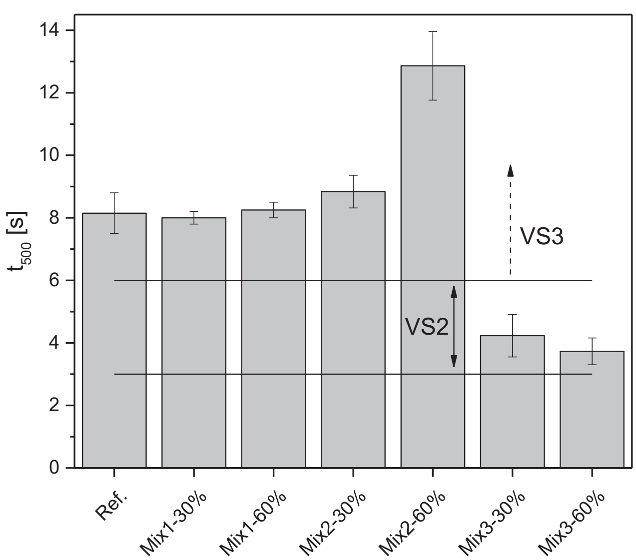


Fig. 8. The t_{500} time of the fresh concrete mixtures.

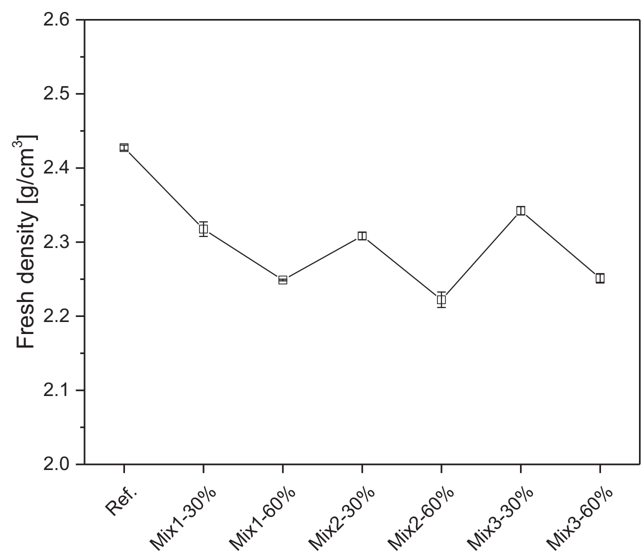


Fig. 10. The fresh density of the fresh concrete mixtures.

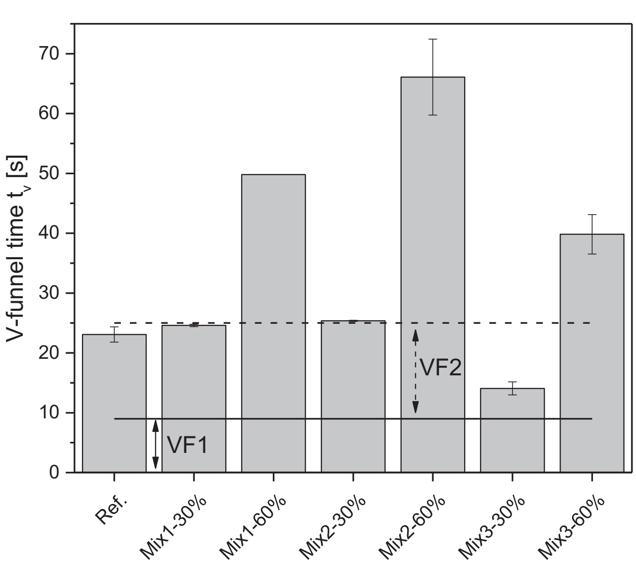


Fig. 9. The V-funnel time of the fresh concrete mixtures.

3.3. Properties of hardened concrete

Fig. 11 displays the flexural strength of all the hardened concretes with and without aggregates replacement. It can be observed that the use of CBLAs decreases the flexural strength of concrete, and the increasing replacement level of aggregates contributes to a higher reduction of flexural strength. For all the concrete mixtures after 56 days' curing, the reduction of flexural strength is around 16–27% of the reference value when 30% aggregates are replaced by CBLAs, while this reduction is around 36–40% when 60% vol. of artificial aggregates are used. Fig. 12 shows the compressive strength of hardened concretes after 28 and 56 days' curing. It indicates that the addition of CBLAs decreases the compressive strength of concrete, and this reduction increases with the increasing replacement level. This is similar as reported by other researchers [54]. The compressive strength after 56 days' curing has a 23–45% decrease compared to the reference, and higher replacement levels result in higher reductions. The higher crushing resistance of artificial aggregates does not give a high strength of the concrete as expected. Fig. 13 shows that the compressive strength and the fresh density of the concrete has linear relationship. It seems that when 30% Vol. of CBLAs is applied, the flexural and compressive strength have a linear correlation with

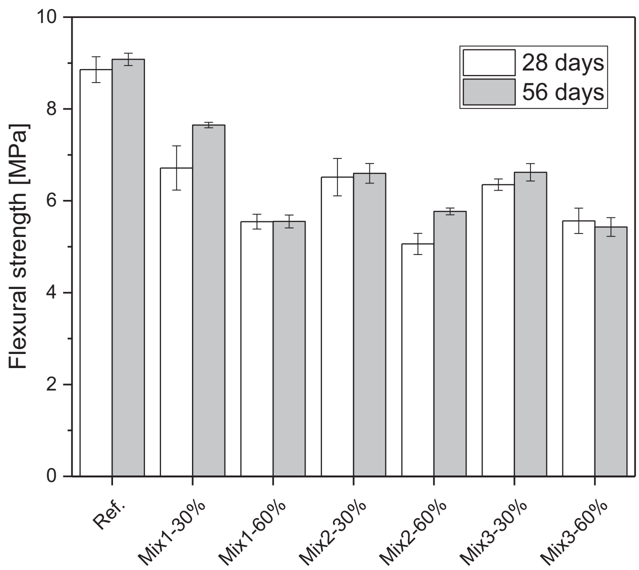


Fig. 11. The flexural strength of concrete after 28 and 56 days.

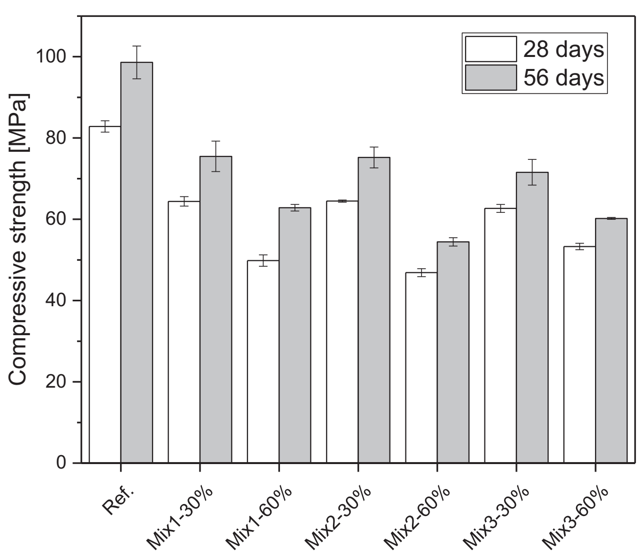


Fig. 12. The compressive strength of concrete cubes after 28 and 56 days curing.

the crushing resistance of the CBLAs; while when 60% vol. of CBLAs is used, there is no clear trend anymore between the concrete strength and the CR of CBLAs. The above present results and phenomena can be due to the following reasons: as observed during the test and the results from the fresh properties of the concrete, it is known that the use of CBLAs reduces the viscosity of concrete, which subsequently affects the strength of concrete. The slump flow diameter and the V-funnel time of the concrete is more influenced when 60% vol. of CBLAs is used, as shown in Section 3.2. Therefore, the influencing factors of concrete strength include the CR of CBLAs and the other parameters of the concrete (such as porosity, etc.).

3.4. Durability properties of hardened concrete

3.4.1. Water penetration

Fig. 14 shows the water depth measured after the water penetration test on the hardened concrete cubes. The measured water depths of all the concrete mixture with artificial aggregates are higher than of the reference, and a higher level of aggregates used causes a higher water penetrated depth. It is also noticed that the water depths of concrete with aggregates S3 are much higher than of Mix 1 and Mix 2. It is reported that the use of artificial aggregate causes an increase of gas permeability [52]. Fig. 15 shows the cross section of the concrete cube with 30% and 60% Vol. of S2. There are some pellets close to the edge of the concrete sample, and the distribution of the CBLA pellets is not evenly in the concrete ideally, due to the different particle shape of CBLA and natural aggregates. Moreover, it can be seen that the artificial aggregates are very porous, and some particles have voids inside. The water penetration depth in concrete with CBLAs consists of the penetration in the cementitious matrix in concrete, and the penetration in the CBLAs, while in normal concrete, it only includes the former. Therefore, the water penetration depth with CBLAs is higher than that of the reference.The water penetration depth reflects the porosity of concrete, and the results obtained demonstrate that concrete Mix 3 is more porous than the other two with CBLAs. This is due to the following reasons: (1) CBLA S3 is more porous than S1 and S2, which can be seen from its higher water absorption and lower

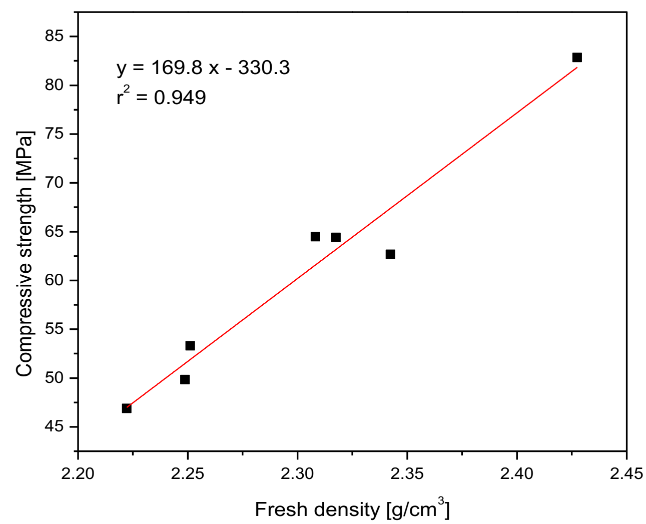


Fig. 13. The relation between compressive strength and fresh density of concrete mixtures.

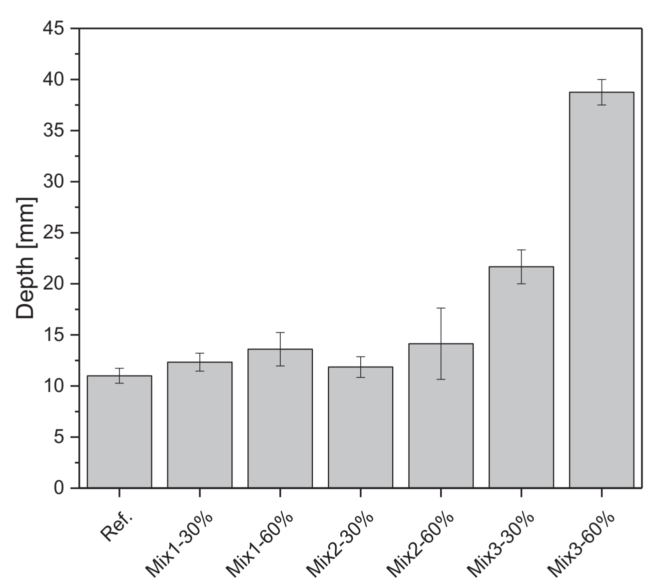


Fig. 14. The water penetration depth in hardened concretes after 56 days' curing.

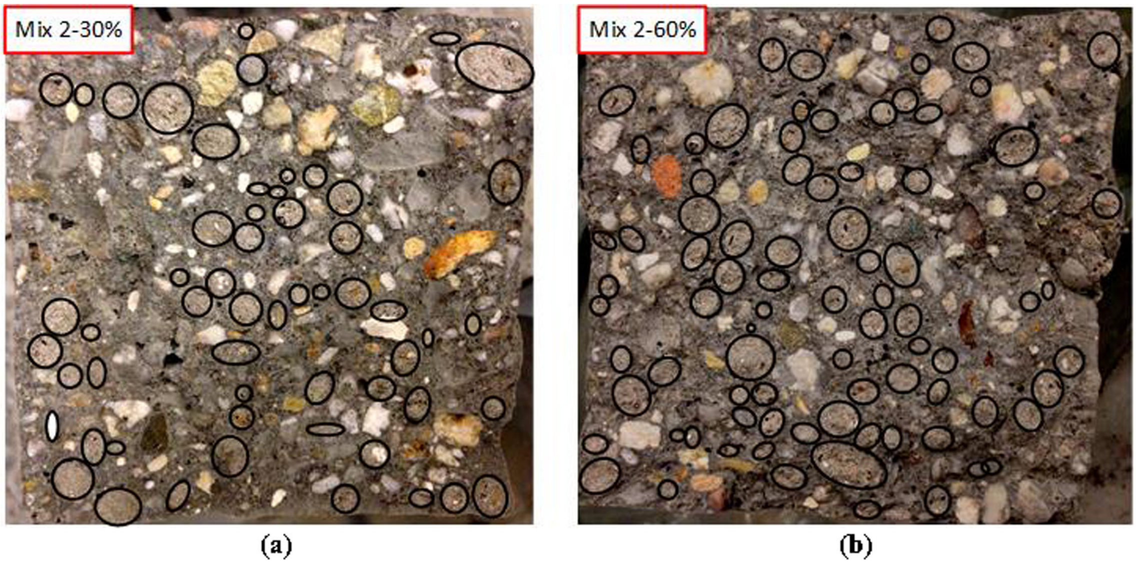


Fig. 15. The cross section of the concrete cube with (a) 30% and (b) 60% vol. (S2) aggregate replacement.

crushing resistance in Section 3.1; (2) the use of S3 reduces the viscosity of concrete significantly, as shown by the t_{500} and t_v, which subsequently influences the concrete microstructure.

3.4.2. Freeze-thaw test

Fig. 16 demonstrates the cumulative mass loss (CML) of concrete slab samples under freeze-thaw cycles in the presence of a de-icing agent.

The CML of the reference concrete increases slowly in the first 28 cycles, after which there is a sharp increase. The CML of Mix 1 with 30% vol. of S1 is increasing linearly, and it is 27% higher than that of the reference after 56 cycles. When 60% vol. of S1 is used, the CML of concrete increases during the first 28 cycles and is higher than that of the reference; after this, it is almost steady and it is nearly the same as the reference after 56 cycles. The CML of Mix 2 shows a linearly increasing trend during the test cycles, which is higher than that of the reference. Moreover, the increasing amount of artificial aggregates S2 does not lead to a significant difference in the CML profile. The CML of Mix 3 with 30% vol. of S3 is almost stable after 14 cycles, and it is 35% lower than that of the reference after 56 cycles. When 60% S3 is used, its CML is the highest among all the tested samples in this study, and it is stable after 28 cycles.

For normal concrete, the CML under freeze-thaw condition is due to: (1) the loss of cementitious matrix; (2) the loss of natural aggregates due to less binding by the matrix [55]. For the concrete with artificial aggregates, which have a higher porosity compared with natural aggregates, the CML of concrete also includes the mass loss of artificial aggregates due to expansion of water inside the saturated aggregates. This is one of the main reasons for the higher CML of Mix 1, Mix 2 and Mix 3 at early freeze-thaw cycles [55]. The higher CML of concretes with higher artificial aggregates in our study confirms this finding. The addition of PPF in aggregates S2 results in a slightly lower CML of concretes, which was also reported in [48] during soil treatment. This may be attributed to the gradual mass loss of concrete during the freeze-thaw test. For concrete with aggregates S3, the fast stability of CML could be resulting from the addition of nano-silica. Researchers have figured out that the use of a certain amount of silica in concrete could increase the freeze-thaw resistance of concrete [56]. Hence, the addition of silica in aggregates production results in the higher freezing-thaw resistance of concrete with S3. Another possible reason is the higher porosity of Mix 3 with 30% vol. of S3, which is reflected in the water penetration depth. The porosity of concrete leaves space for the expansion of water, which results in less damage during freeze-thaw tests. However, the Mix 3 with 60% vol. of S3 achieves the highest mass loss, which may result from the lower bonding strength of high a porous cementitious matrix; hence, the detaching of the mortar matrix is much easier during the freeze-thaw test.

The above results indicate the different influences of artificial aggregates on the freeze-thaw resistance of concretes. It seems that in this study the cumulative mass loss of concretes with artificial aggregates has two types of profiles: linearly increasing and a two-stage type (firstly increase, and then reaching a stable level). The correlation between the artificial aggregates properties (porosity, strength, etc.) and the freeze-thaw resistance profile needs to be investigated further.

3.5. Leaching behaviour of demolished hardened concrete

3.5.1. Column leaching test on crushed concretes containing CBLAs

According to our previous study [39,40], it was figured out the leaching of some heavy metals (Sb, Cu and Mo) and salts (chloride and sulphate) always exceed the limit value according to Dutch legislation [13] for the non-shaped materials category (which means the estimation for using the investigated materials as building materials directly as they are in granular form).

The leaching of Sb for the CBLAs is around 0.1–0.15 mg/kg d.m., which is under the limit value. The leaching concentration of Cu and Mo of the produced CBLAs is 1–1.4 and 1.5–2.3 mg/kg d.m., and exceed the limit values, respectively. The leaching of chloride is about 2500–4600 mg/kg d.m. and exceeds the limit value, while the leaching of sulphate (120–499 mg/kg d.m.) is well under the limit value. Hence, the CBLAs could not be directly used as non-shaped materials according to SQD, extra isolation protection is needed to prevent the potential pollution to environment during the application of these CBLAs. Similar statement was addressed by Colangelo, et al. [18].

Fig. 17 shows the total amount of heavy metals (Sb, Cu and Mo) and salts (chloride and sulphate) in the crushed concrete samples with and without artificial aggregates by mass. The amount of these elements in concrete increases when the artificial aggregates are used and higher replacement level leads to a higher concentra-

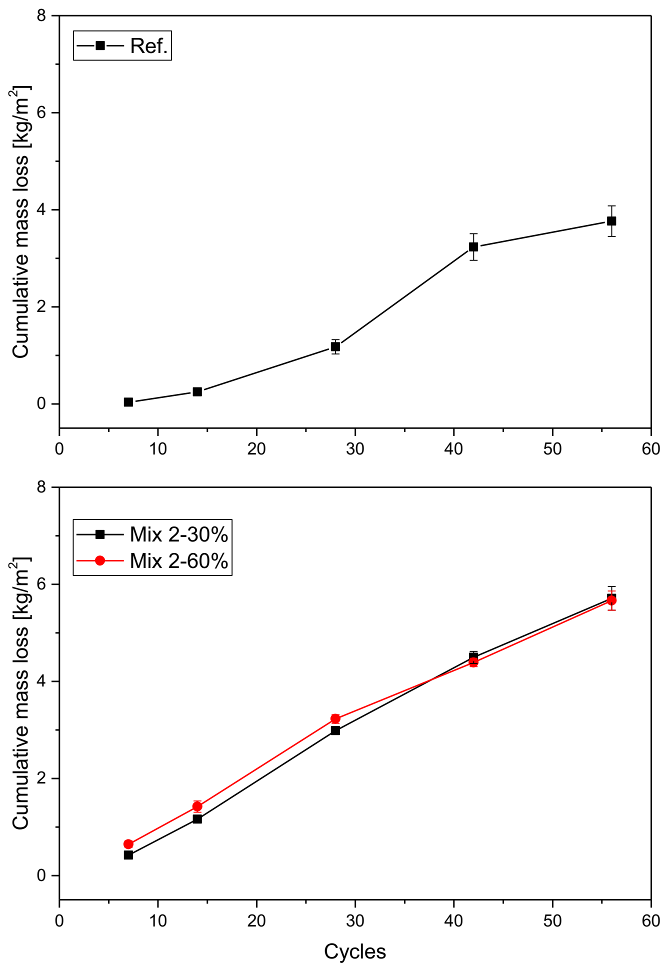


Fig. 16. The cumulative mass loss of hardened concrete samples after freezing thaw cycles.

tion of these elements compared to the reference. Fig. 18 shows the leaching concentrations of the investigated elements from the crushed concretes. It demonstrates that the leaching concentrations of these elements are all under the limit value according to the Dutch legislation and their leaching concentration increase with the increasing amount of artificial aggregates [13]. The leaching of Sb is lower than 0.004 mg/kg d.m. and below the detectable level, therefore they are not shown in the graph. This leaching

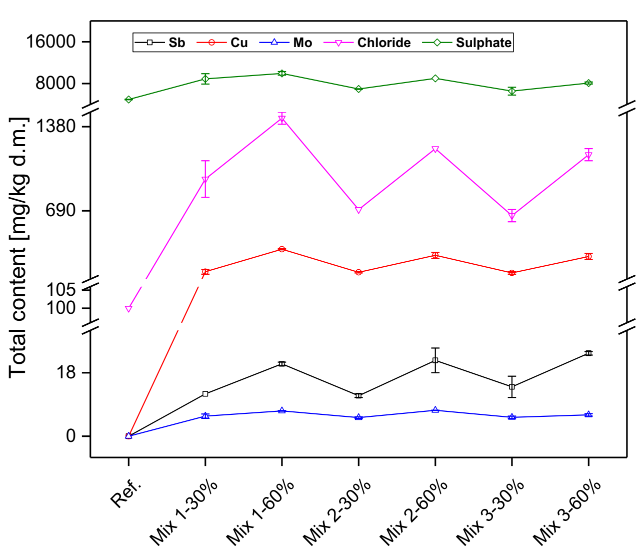


Fig. 17. The total concentration of Sb, Cu, Mo, chloride and sulphate in the concrete mixtures.

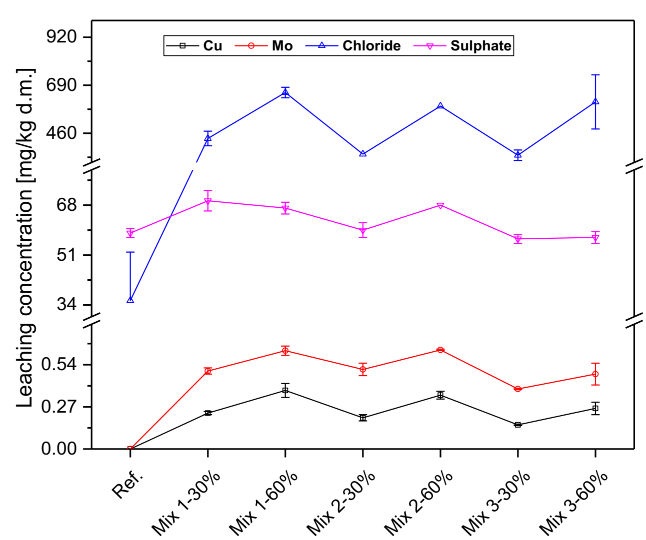


Fig. 18. The leaching concentration of Cu, Mo, chloride and sulphate ions from crushed concrete samples with and without aggregate replacement.

result means that these concretes can be recycled and reused as secondary building materials without environmental risks, in case recycling of these concretes is required when they finish their service life.

Due to the fact that the total amount of the investigated elements is influenced by the amount of artificial aggregates applied, the leachability of the concretes is calculated based on their total amount and leached out amount of elements as described in Section 2.4.

The leachability of the CBLAs is shown in Fig. 19(a). It shows that the sample S3 has a lower leachability of Sb, Cu, Mo and sulphate, while it has higher chloride leachability compared with sample S1. This demonstrates that the addition of nano-silica could promote the immobilization of the above-mentioned elements. For chloride, it seems that the use of PPF and nS has a negative influence on its immobilization, which may be explained by the higher porosity of S2 and S3, giving the highly soluble chloride more chance to be washed out.

Fig. 19(b) shows the leachability of Cu, Mo, chloride and sulphate of the crushed concretes based on the data of Figs. 17 and 18. The Cu leachability of concrete decreases with the increasing amount of artificial aggregates for all the concrete mixtures and concrete Mix 3 has the lowest Cu leachability compared with Mix 1 and Mix 2. The leachability of chloride has an increasing trend from reference to Mix 3, and a higher aggregates replacement level results in a lower leachability of chloride for each concrete mixture. The sulphate leachability of concrete with artificial aggregates is lower than that of the reference, and higher levels of aggregates being replaced contribute to a lower leachability of sulphates.

It seems that the concretes containing aggregates S3 have a better immobilization of heavy metals compared with the ones with S1 and S2. This may be due to the addition of nano-silica in the aggregates production which is reported to decrease the pH of the cementitious matrix in concrete [57]. As shown in Fig. 20, the pH of the tested leachate from the crushed concrete shows that the addition of artificial aggregates (S1 and S2) results in a higher pH of the leachate, while the pH of leachate from Mix 3 (with S3) is lower than that of the other samples; this also proves that the addition of nano-silica to the cementitious matrix leads to a lower pH. Moreover, the leaching behaviour of heavy metals in MSWI residues is highly related to the pH [58]. The use of nano-silica to produced CBLAs did not increase the crushing resistance of the aggregates, but it improves the leaching of the aggregates and the concrete with these aggregates.

Comparing the leachability of investigated components in CBLAs and concretes with corresponding CBLAs, the leachability of Sb, Mo, chloride and sulphate in concretes is lower than that in CBLAs, which demonstrates the further immobilization of these components when applied in concrete.

The leaching of the crushed concrete with the produced CBLAs in this study complies with the Dutch legislation, which means the recycling of these concrete as secondary building materials is environmental friendly after they finish their service life. Considering the leaching of concrete with CBLAs, the amount of CBLAs can be increased when additives are applied in concrete, such as fly ash, silica fume, ground granulate blast furnace slag, etc. which are reported to reduce the pH of the concrete [57]. Moreover, these additives can also be applied to be used as raw materials for CBLAs production when the leaching of the aggregates is critical.

3.5.2. Tank leaching test on the hardened concrete cubes containing CBLAs

The diffusion tests on the hardened concrete containing 30% and 60% vol. CBLAs were performed, and the cumulative concentration (ε_{64}) of components leached out from the concrete cube were shown in Table 3 and compared with the limit value for shaped materials according to SQD [13]. It shows that the ε_{64} of

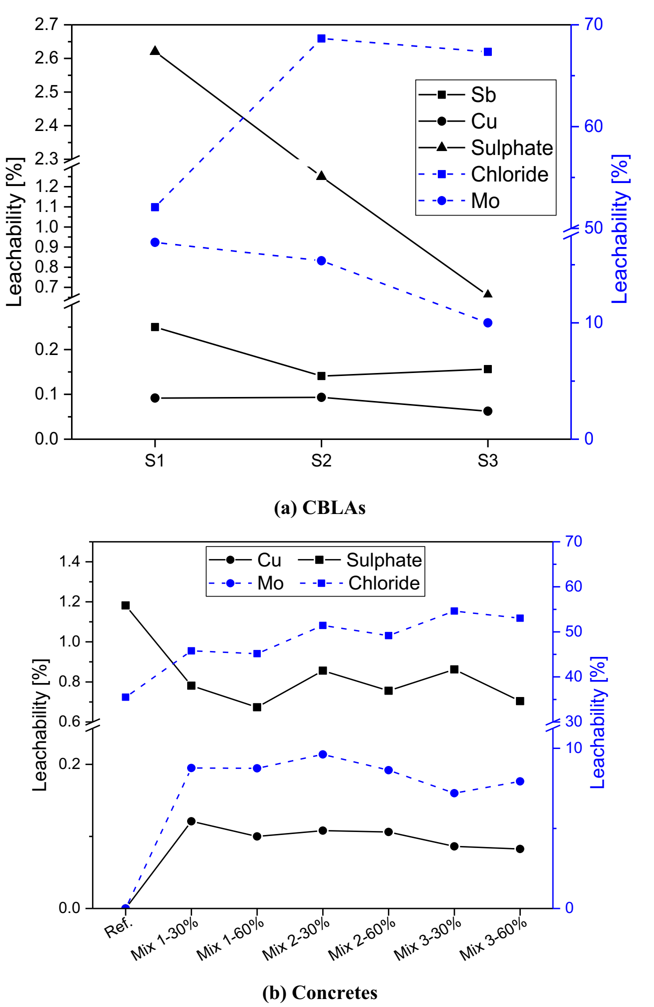


Fig. 19. (a) The leachability of Cu, Mo, chloride and sulphate from CBLAs and (b) the concretes containing CBLAs.

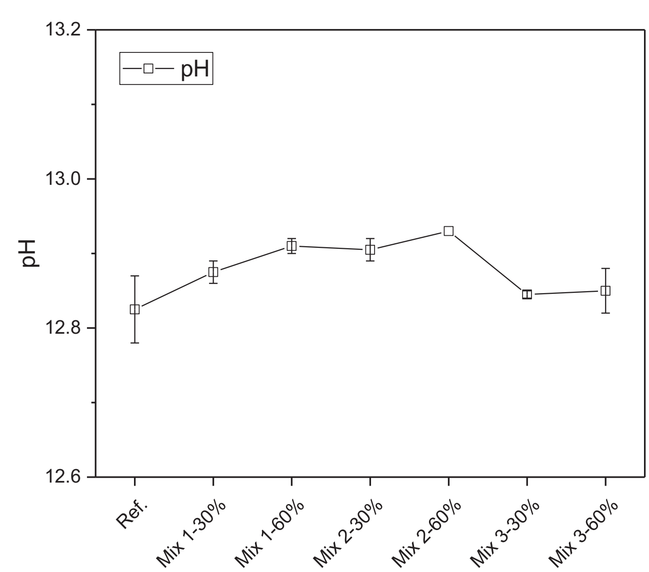


Fig. 20. The pH of the crushed concrete samples with and without CBLAs.

Table 3
The cumulative emission of components from hardened concrete containing CBLAs and the leaching limit for shaped materials according to SQD.

Components	SQD limit for shaped materials ε_{64} [mg/m^2]	Cumulative emission ε_{64} [mg/m^2]					
		Mix1-30%	Mix1-60%	Mix2-30%	Mix2-60%	Mix3-30%	Mix3-60%
Sb	8.7	0.19 ± 0.01	0.19 ± 0.01	0.17 ± 0.0	0.17 ± 0.0	0.25 ± 0.02	0.2 ± 0.01
Cu	98	2.0 ± 0.0	2.0 ± 0.0	2.0 ± 0.0	2.0 ± 0.0	2.0 ± 0.0	2.0 ± 0.0
Mo	144	0.62 ± 0.05	1.4 ± 1.1	0.53 ± 0.01	0.52 ± 0.02	1.3 ± 0.28	1.1 ± 0.15
Cl^-	110000	665 ± 78	3750 ± 2192	960 ± 57	1400 ± 0.0	1750 ± 71	2400 ± 283
SO_4^{2-}	165000	825 ± 21	1480 ± 877	880 ± 42	865 ± 21	1300 ± 141	1020 ± 113

all the components is far below the limit value in SQD, which demonstrate the non-hazardous of these concrete to environment, and the cumulative leaching of chloride increases with the amount of CBLAs. In concrete Mix 1, the cumulative leaching of Mo and sulphate also increase with the CBLA content. Hence, the amount of CBLAs applied in concrete needs to be aware regarding leaching of specific elements in CBLAs.

The column and tank leaching tests on the crushed and cubic concretes containing the three types of CBLAs produced from mixtures of industrial solid wastes show that these crushed and cubic concretes can be classified as non-hazardous materials, even though the CBLAs cannot comply with the leaching limit and then not allowed to be treated as building materials directly. These concretes containing CBLAs will be the final products which could be used in construction fields, in which around 13–27% industrial solid wastes due to the use of CBLAs are additionally recycled.

Therefore, the use of these concrete containing CBLAs produced from fine MSWI bottom ash and other industrial solid wastes are environmental non-hazardous according to legislation and they can be used directly. It can be concluded that, the pelletizing technique is successfully applied to produce artificial aggregates from integrated industrial solid wastes, and the concrete produced with these artificial aggregates is non-hazardous to the environment. In this way, the environmental burden related to disposal of industrial solid wastes and consumption of natural resource in construction field can be reduced. Moreover, the leaching of the crushed concretes complies the limits in legislation, which means after service life, they can be recycled and then reused again. Thus, a closed recycling circulation of several industrial solid wastes can be achieved.

4. Conclusions

In this study, the mixed industrial solid wastes are integrally recycled and innovative methods are used to improve the properties of the artificial cold bonded lightweight aggregates (CBLA) produced. Three types of aggregates are produced and used as aggregates in self-compacting concretes, and their influence on the concrete properties are studied and compared. The following conclusions can be drawn:

(1) Three types of CBLAs are produced from combined industrial solid wastes, including municipal solid waste incineration (MSWI) bottom ash fines (BAF, 0–2 mm), paper sludge ash (PSA), coal fly ash (FA), and washing aggregates sludge (WAS), with the ordinary Portland cement as binder. The BAF, polypropylene fibre (PPF) and nano-silica from olivine dissolution were used to improve the properties of the artificial aggregates, such as crushing resistance, bulk density, etc.

(2) The produced CBLAs were used as natural aggregate replacement in designed self-compacting concretes. It was found that the addition of CBLAs produced without (S1) and with (S3) nano-silica do not have a significant influence on the slump flow diameter, while the one with aggregates produced with the addition of PPF (S2) had a smaller slump flow diameter due to the blocking function of PPF. The t_{500} of concrete mixtures with S1 and S2 is slightly influenced, while the addition of S3 results in shorter t_{500} time. It is observed during the V-funnel test that the round shape of the artificial aggregates had a 'ball effect', which makes the artificial aggregate particles to be pushed on the top layer of the sample.

(3) The compressive and flexural strength of the concrete is decreased with the increasing amount of artificial aggregates. The concrete strength has a positive linear correlation with the crushing resistance of the CBLAs when 30% vol. aggregates is replaced, while this relationship is not found for sample with 60% vol. aggregates replacement, due to the significant influence of CBLAs on the fresh properties of concrete (such as viscosity). The compressive strength of the concrete containing CBLAs has linear relation with its fresh bulk density.

(4) The water penetration depth (WPD) of concretes shows that the use of CBLAs increases the water WPD, and the concrete with aggregates S3 has a much higher WPD which is attributed to the influence of concrete viscosity. Less water or chemical admixtures for modifying the water requirement is recommended when CBLAs are used.

(5) There are two cumulative mass loss (CML) profiles of concrete with CBLAs obtained in this study: one linearly increases with the test cycles, another one is a two-stage profile (increasing stage and stable stage). The use of aggregates S2 which contains PPF results in a linear increase of CML, and there is marginal difference when the amount of S2 is increased from 30% to 60% vol. The concrete with aggregates S3 has a two-stage CML profile, which is assumed to be influenced by the nano-silica in aggregates and the porosity of the concrete.

(6) The column and tank leaching tests show that the crushed and cubic concretes containing CBLAs comply with legislation, which demonstrate that they are non-hazardous and can be used as building materials. Furthermore, the leaching of crushed concrete complies the legislation demonstrates the reuse of them as building materials after service period, thus a sustainable recycling of industrial solid wastes can be realized.

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