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Recycling and utilization of high volume converter steel slag into CO₂ activated mortars – The role of slag particle size



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ABSTRACT

Converter steel slag is produced in large quantities as by-product of primary steel-making, which is mainly landfilled or used as an aggregate The high reactivity of converter steel slag with CO_2 makes it an ideal material for CO_2 capture by forming calcite even at ambient temperature and pressure. This reaction can also be used to for the production of green building materials. However, the carbonation of steel slag is strongly controlled by the particle size, which influences CO_2 transport and carbonation speed. In this study, low carbon footprint binders consisting of various fine converter steel slag powder (80%) and normal cement (20%) are applied to produce mortars using ambient carbonation. The results indicate that the variation of steel slag particle size influences gas transport and CO_2 uptake of carbonated steel slag blended mortars during the curing period, which effects microstructure, strength, and leaching. The application of optimal size range of steel slag (21.75–24.13 µm) in blended mortars leads to a higher compressive strength (31.21 MPa), CO_2 uptake (15.9%), faster carbonation rateas well as sustainability efficiency (0.486 MPa/(kg/m³)), which is obviously superior compared to the larger or smaller size of steel slags mortars. The leaching properties, especially V and Ca leaching of steel slag blended mortars are observed to be strongly influenced by the CO_2 uptake, rather than steel slag particle size. A green binder system is achieved by both the reduction of cement content and the high CO_2 uptake by mass of binder.

1. Introduction

Concrete is the most used building material worldwide, with almost 20 billion tons manufactured every year. Moreover, the demand for concrete keeps increasing in recent years. Cement is the binder normally used in concrete to meet the strength, durability and economic requirements. However, cement manufacture is an energy-intensive process (Feiz et al., 2015), and a large amount of limestone is calcined. As a consequence, a huge amount of CO_2 is emitted, both from the calcination as well as the essential fuel combustion to heat the kiln. This process accounts for 8% of global green gas emission (Andrew, 2018). In order to achieve CO_2 reduction via lowering the required cement content, various types of blended cement with supplementary cementitious materials and/or industrial by-products have been developed.

A potential material that can serve as such a substitute is converter steel slag which is produced in large quantities worldwide as byproduct of primary steel-making (~90 kg slag per ton of steel) (Fisher and Barron, 2019). In the EU alone, with a primary steel production of about 110 million tons, this leads to 10 million tons of slag annually (EUROFER 2018). In the Netherlands 0.7 Mtons of steel slags are produced, which is substantial in comparison to the demand for cement of 4M tons annually. (Das et al., 2007). Jiang et al. summarized previous studies of steel slag application in cement and concrete and recommended only 10–20% of steel slag as cement replacement due to the low hydration reactivity (Jiang et al., 2018). Converter slag is also used as aggregate, but volume stability problems due to the presence of free lime are common (Brand and Roesler, 2015). For this reason the majority of converter slag is used as backfill material or landfilled.

In contrast to the low hydration reactivity, converter steel slag was identified to show a high reactivity in a CO_2 rich environment (Huijgen and Comans, 2006a) because it contains α -C₂S, β -C₂S, and free MgO/CaO that can sequestrate CO₂ effectively according to Reddy's study (Reddy et al., 2019). EI-naas et al. also proposed a pre-hydration

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method for enhancing the carbonation degree of steel slag (El-Naas et al., 2015). A review by Humbert indicated that the CO₂ uptake ability of steel slag can be up to 200-400 g CO₂/kg depending on the carbonation conditions (Humbert and Castro-Gomes, 2019). The volume expansion during the carbonation of calcium silicate and the precipitation of calcite contributes to a significant strength development after pressurized CO2 curing (Moon and Choi, 2018). In Shao's study, a combination of poorly crystalline CaCO3 and C-S-H was confirmed after steel slag carbonation, which also has a potential to provide strength (Shao et al., 2013). Furthermore, the hydration products of cement, such as calcium silicate hydrates, calcium hydroxide, and ettringite were also identified to be reactive with CO₂ even at ambient conditions (Jang et al., 2016). Most of the previous investigations were carried out using a pressurized and pure CO₂ environment for fast strength development of steel slag concrete, conditions which add to the embodied energy of the products. On the other hand, the carbonation of steel slag was also confirmed to take place at ambient pressure and temperature (Ghacham et al., 2016). This provides a possible ecotreatment to activate steel slag blended concrete at ambient condition using CO₂.

At ambient temperature and pressure, the dissolution rate of CO₂ is lower and the CO₂ transportation mostly relies on the gas permeability and humidity (Fernández (Fernández Bertos et al., 2004)). This process can be influenced by the mix design of the concrete and binder, and their particle sizes. Baciocchi et al. found that the CO₂ uptake ability of steel slag was enhanced from 2 kg/t to 130 kg/t when the particle size decreased from 2 mm to 0.105 mm (Baciocchi et al., 2009). By selecting the optimal steel slag size in slurry carbonation, Polettini et al. reported a CO₂ uptake of 465 kg/t (Polettini et al., 2016). As a consequence, the produced carbonation products dominate the pore volume development, as opposed to the cement binder hydration. They also control the CO₂ diffusion (Kashef-Haghighi et al., 2015), and influence the strength performance and microstructure development. Therefore, a variation of steel slag particle size should affect the strength evolution of CO₂ activated steel slag concrete, the amount of reaction products and leaching properties. However, the particle size effect of steel slag carbonation was only investigated in thin-film and steel slag slurries (Polettini et al., 2016). There is no report related to the steel slag particle size selection in CO₂ activated production of green concrete building materials available yet.

Therefore, the objective of this study is to manufacture a potential green converter steel slag-cement based building material by ambient CO_2 curing (298K, 0.2 CO_2 atm, 1 bar, and 65% RH). To this end, different particle sizes of converter steel slag are investigated (298K, 0.2 CO_2 atm, 1 bar, and 65% RH). The investigated parameters include: particle size of steel slag, curing duration, compressive strength, mass change, microstructure, CO_2 uptake, and leaching properties. The observed results in this study contributes to the recleying and application of high volume steel slag in low carbon footprint building materials. The optimal selection of steel slag particle size and applying of ambient carbonation help to design concrete with high sustainablility coefficiency and low energy consumption.

2. Materials and methods

2.1. Materials

In this study, Ordinary Portland cement (CEM I 52.5 R) was used as a reference binder (ENCI, Netherlands, specific density is 3.1 kg/m^3), and inert quartz powder (provided by Quarzwerke Gruppe) was applied as cement replacement in reference mortars.

The investigated converter steel slag was provided by Tata Steel, IJmuiden, the Netherlands (specific density is 3.9 kg/m^3).

For the purpose of cement replacement the as-received converter steel slag was milled into powder using a disk milling (Retsch, RS 300 XL) with a power of 2200W. In order to create different particle size

 Table 1

 Chemical compositions of cement and converter steel slag.

Chemical composition (% mass)	Converter steel slag	CEM I 52.5 R
CaO	41.55	67.97
SiO ₂	11.47	16.19
Al_2O_3	2.24	3.79
Fe ₂ O ₃	31.35	3.59
MgO	3.78	1.71
MnO	4.78	0.09
TiO ₂	1.56	0.28
P_2O_5	1.30	0.42
SO ₃	0.03	4.05
V ₂ O ₅	1.14	/
Cr ₂ O ₃	0.35	0.01
Cl	0.01	0.04
LOI	0.72	0.51

distributions the milling durations were varied from 7, 9, 11–13 min (labeled as D1, D2, D3, D4).

The chemical properties of cement and steel slag were determined with X-ray fluorescence (XRF, PANalytical Epsilon 3) spectroscopy and their crystalline phases were detected using X-ray diffraction (XRD, Bruker D2 PHASER), the results are shown in Table 1 and Fig. 1.

The main oxides in steel slag are CaO, SiO_{2} , and Fe_2O_3 , and the detected mineral phases are larnite, brownmillerite, wuestite and magnetite, the latter three being Fe-related minerals, consistent with the high content of Fe_2O_3 (31.35 % in Table 1) in steel slag.

The particle sizes of cement, quartz powder and milled slags were measured using laser granulometry (Master sizer 2000) and the result are shown in Fig. 1(b). The median particle size (d_{50}) of cement, quartz powder, D1, D2, D3, and D4 are 8.08 µm, 10 µm, 28.98 µm, 24.13 µm, 21.75 µm and 16.17 µm, respectively. It should be noted that even after milling, the particle size of slag is coarser than that of cement and quartz powder.

2.2. Application of convert steel slag in sustainable mortars

The milled slags were used as cement replacement (80% wt.) in mortars and then their influence on mortar properties under different curing conditions was evaluated. The mixture design and labels of the mortars are shown in

Table 2. The water to binder ratio of the mortars was kept at 0.4, and a standard sand (DIN EN 196-1) with a particle size of 0-2 mm was used as aggregate (sand to binder ratio was 3). In the reference mortar (Ref.), quartz powder was used to replace 80% wt. cement.

2.2.1. Mortar preparation and curing conditions

The milled slag (or quartz powder) was mixed with cement in a Hobart mixer for 30 s, and then sand was added followed by 30 s mixing. Water was then added to the dry mixture and everything mixed for 30 s at medium speed. The fresh mixture was cast into moulds (40 mm \times 40 mm \times 160 mm) and then covered with plastic film for 24 hours before demoulding.

There were two types of curing conditions applied on the mortar samples: normal humidity curing and ambient $\rm CO_2$ curing.

One batch of mortar samples after demoulding was cured sealed by covering it with plastic film and storing it at room temperature before further tests.

The second batch of mortar samples was cured in a climate chamber (Fig. 2) at a temperature of 25°C. The relative humidity (RH) in the chamber was kept at 65% with a circular flow-through and aa concentration of 20% CO₂ was applied in the chamber continuously for the tests.

2.2.2. Mechanical property of mortars

After the corresponding curing, the compressive strength of mortar



Fig. 1. (a) XRD patterns of cement and converter steel slag; (b) particle size distributions of cement, quartz powder, and milled slags.

Table 2 Mixture design of mortars with milled slags (kg/m^3).

Samples	d ₅₀ of steel slag or quartz powder	Cement	Quartz powder	Steel slag	Sand	Water
Ref.	10.00 µm	100.78	403.12	0	1511.70	201.56
D1	28.98 µm	106.34	0	425.37	1595.12	212.68
D2	24.13 µm	106.34	0	425.37	1595.12	212.68
D3	21.75 µm	106.34	0	425.37	1595.12	212.68
D4	16.17 µm	106.34	0	425.37	1595.12	212.68

samples was tested according to EN 196-1 (BS EN 2005). The compressive strength of samples after demoulding was tested as 1 day strength. Then humidity cured samples were tested after 14 days, while carbonated samples were tested after 3 days, 7 days, and 14 days.

2.2.3. Characteristics of hydration/carbonation products

To further understand the influence of particle size of converter steel slag on the properties of mortar under different curing methods, the hydration and carbonation products in mortar samples with different slags were evaluated.

Firstly, the total mass of steel slag blended mortar prisms was recorded during the CO_2 curing, which reflects the mass change of mortar due to the water evaporation and hydration/carbonation reactions.

After curing, the mortar samples were dried at 105 °C and crushed

into powder for further testing. The X-ray diffraction (XRD) test was conducted with a Bruker D2 PHASER to identify the relevant hydration and carbonation products. Thermo-gravimetric (TG) analysis was performed (STA 449 F1) on the milled powders with a temperature increase of 10 $^{\circ}$ C/min up to 1000 $^{\circ}$ C starting from 40 $^{\circ}$ C with nitrogen as the carrier gas to quantify the reaction products.

The amount of CO₂ uptake was calculated according to the mass loss from 550°C to 800°C, which is related to CO₂ release from the decomposition of calcium carbonate (Mo et al., 2016). The binder to sand ratio was 1:3 and sand is inert to CO₂, so the amount of CO₂ uptake ability per gram of converter steel slag blended binders was calculated with the following equation:

$$CO_2 \ uptake = \frac{M_{550} - M_{800}}{M_{800}/4} \times \ 100\%$$

$$M_{550} \text{- mass at } 550^{\circ}\text{C}$$

$$M_{800} \text{- mass at } 800^{\circ}\text{C}$$
(1)

2.2.4. Evaluation of total CO_2 emission and sustainability efficiency

The CO₂ emission of energy consumption during converter steel slag milling process was calculated by the power of disk mill and milling duration according CO₂ emissions from public electricity production (295.8 g/kWh) in EU. The energy consumption of the CO₂ inlet was selected as 80.7kWh/ton (Monkman and Shao, 2010). The CO₂ emission from cement, sand, and water production are set as 930 kg/ton, 4



Fig. 2. Schematic of carbonation chamber.

kg/ton and 0.196 kg/ton, respectively (Li et al., 2019). The calculation of sustainability efficiency was done according to a previous study (Damineli et al., 2010).

2.2.5. Microstructure

The profile of the pore size of the cured mortars was evaluated by Mercury Intrusion Porosimeter (MIP) using Micromeritics AutoPore IV. The intrusion pressure was 0–227 MPa.

The microstructure of mortars after humid and CO_2 curing was observed using Scanning Electron Microscopy (Phenom Pro).

2.2.6. Environmental impact evaluation

One batch leaching (NEN 6966) was conducted for mortars. The cured mortar was crushed to below 4 mm and the leaching test was conducted using a dynamic shaker, at ambient temperature, and the leaching conditions were L/S 10, 250 rpm and 24 hours. The leachates were then filtered through the 0.017-0.030 nm membrane filters. After acidifying the leachates with HNO₃, the concentration of elements in the solutions were analyzed with Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-OES Spectroblue).

3. Results and discussion

3.1. Mechanical properties of mortars

The compressive strengths of all mortars are shown in Table 3. For all the mortar samples cured sealed in plastic, the strength increased with curing time. The mortars containing slags showed much lower strength after demolding. Mortars with D1, D2, and D3 showed around 30% lower compressive strength after 14 days hydration compared to the Ref. mortar with inert quartz powder, while D4 was only 3% lower than the reference mortar. This result demonstrates that steel slag has very low hydraulic reactivity, which is consistent with previous findings (Qiang et al., 2016a). It is also noticed that the strength of mortars containing steel slag increased with the decrease of slag particle size in hydration mortar samples. This may be due to the promotion of cement hydration due to the nucleation function of smaller particles (Lawrence et al., 2003).

Regarding the mortars cured under CO_2 conditions, it can be seen that all mortars showed increased compressive strength with increased curing time, and the mortars with steel slags achieved higher compressive strength after 7 days CO_2 curing than the 14 days normal cured samples. In addition, the compressive strength of mortars after 7 days CO_2 curing with steel slags is higher than the one with quartz powder, and their 14 days carbonated compressive strength is more than 2 times that of reference mortar. This observation indicates the specific contribution of steel slag during CO_2 curing for strength development, implying that steel slag blended mortars can be effectively activated under CO_2 curing conditions.

Fig. 3 also shows that the effect of the steel slag particle size on strength performance at various curing ages. As previously stated, in the hydrated mortar samples, the compressive strength of mortar decreased with the increase of slag particle size (Qiang et al., 2016b) that finer steel slag particles exhibited higher reactivity, while in carbonated mortar samples, the mortar strength after 7 days firstly increased and

Compressive strengths of mortars

then decreased with the increase of particle size. An optimal steel slag particle size value can be identified from the correlation between steel slag particle size and compressive strength. In comparison with D4 (23.41MPa), D3 exhibits an increase of 33.32% in compressive strength after 14d carbonation, while it is 29.43% and 8.87% for D2 and D1, respectively. To further explain the contribution of steel slag and influence of its particle size on mortar properties, hydration and carbonation products and microstructures of mortars are studied in the followed sections.

3.2. Determination of hydration and carbonation products

Fig. 4 shows the XRD patterns of samples before carbonation (after 24h hydration) and after 14 days of CO_2 curing. It can be seen in Fig. 4 (a), that there is no significant difference in XRD patterns between steel slag blended mixtures after 24 hours hydration. The presence of CH is mainly due to the cement hydration (Shi, 2002). The larnite, brown-millerite, and wustite are related to the minerals in the unhydrated steel slag. Fig. 4 (b) shows the XRD patterns of samples after 14 days carbonation curing. Calcite is observed in all carbonated samples, while peaks of calcium hydroxide only present in D4, indicating an incomplete carbonation, which is confirmed by the phenolphthalein test as shown in Fig. 3 (b). The detected calcite in carbonated samples demonstrates the activation of steel slag and its carbonation products under CO_2 curing conditions.

3.3. Mass change of samples during carbonation

The mass development of different mortars was measured and is shown in Fig. 5. It can be seen that the mass development of the reference sample (quartz powder blended) consists of two stages. The first stage is related to a high decrease of total mass, then the second stage corresponds to a stable period. However, the mass variation of the steel slag blended mortars consists of three stages. At first, there is a rapid decrease in mass (stage I), after that, a mass increase period can be observed (stage II), that gradually reaches a stable period (stage III).

Once mortars were moved to the carbonation chamber, pore water kept evaporating due to the difference of humidity between the inside and the outside of the mortar resulting in the mass decrease during the initial several days (stage I), Once the CO_2 uptake exceeds the water evaporation, the mass change of the mortars shows the profile of stage II. In stage II, mineral phases in the blended mortars, such as dicalcium silicate, calcium silicate, tricalcium silicate, and C–S–H react with CO_2 to form calcium carbonate, amorphous silica gel, or CSH, and new free water (Humbert and Castro-Gomes, 2019), which resulted in an increase of mass of the sample.

Generally, these two processes are simultaneous. During the initial days of carbonation, the water evaporation process dominates the mass decrease. After that the sample is less saturated and connected pores become available for CO_2 diffusion. Consequently, more carbonation products are formed. The mass increase by carbonation gradually exceeds the mass loss by water evaporation. As a result, a turning point is observed, and the total mass starts to increase. In other words, the turning point reflects the speed of mortar carbonation. As can be seen from Fig. 5, the turning points for steel slag blended mortars containing

Sample	Compressive strength (MPa After demoulding	a) 3 d CO ₂	7 d CO ₂	14 d CO ₂	14 d hydration		
Ref	5.90 ± 0.98	8.91 ± 0.44	10.46 ± 0.49	11.25 ± 0.40	16.68 ± 0.51		
D1 (28.98 µm)	0.20 ± 0.02	3.73 ± 0.22	10.44 ± 1.27	25.44 ± 0.89	10.12 ± 0.32		
D2 (24.13 µm)	0.51 ± 0.02	5.59 ± 1.4	14.99 ± 1.48	30.30 ± 1.25	11.22 ± 1.05		
D3 (21.75 μm)	0.89 ± 0.11	5.46 ± 0.25	13.57 ± 0.97	31.21 ± 1.71	11.35 ± 0.46		
D4 (16.17 μm)	1.26 ± 0.02	6.04 ± 0.35	12.62 ± 1.24	23.41 ± 0.89	16.19 ± 0.66		



Fig. 3. (a) Influence of steel slag particle size on mortar strength and (b) phenolphthalein test of mortars after 14 days CO₂ curing.



Fig. 4. XRD patterns of samples (a) before carbonation, and (b) after 14 days carbonation (1-larnite, 2-quartz, 3-brownmillerite, 4-portlandite, 5-C₃S, 6-wustite, 7-LDH like structure, 8-calcite).



Fig. 5. Mass change of mortars during the carbonation curing.

slags of larger particle size occurs earlier than those with finer slags. Additionally, they reached a stable stage earlier (stage III). This demonstrates that a larger particle size of steel slag benefits the carbonation speed during CO_2 curing of mortar samples.

The contribution of the different particle sizes of steel slag to the carbonation speed can be explained by the initial porosity of mortar samples, and a subsequent decrease in the evaporation speed of pore water during the CO_2 curing. Moreover, the small pores were filled by the newly formed carbonation products. Then the route for CO_2 transportation was blocked. Thus, CO_2 took a longer time to reach the inside of the mortar samples containing fine size steel slag.

3.4. CO₂ uptake ability

The thermal gravimetric results of the different carbonated samples are illustrated in Fig. 6. The sharp decline of mass in the temperature interval of 550–800 °C in the TG curve is corresponding to the decomposition of calcium carbonates and is used to quantify the carbonation degree of the steel slag samples (Mo et al., 2016). It is noticed that the converter steel slag blended samples exhibit a higher mass loss in this temperature range than Ref. Furthermore, there is a minor peak between 400-500°C for D4, which is related to the decomposition of calcium hydroxide. This indicates that residual calcium hydroxide is present. This observation agrees with the XRD analysis (almost no calcium hydroxide in D1, D2, and D3) and the phenolphthalein test (Fig. 3b).

To evaluate the carbonation rate at different ages, the CO_2 uptake was calculated with Eq. (1) as shown in Fig. 7.



Fig. 6. TG results of samples after 14 days CO₂ curing.

The results showed that all samples exhibit an increasing CO_2 uptake with the increase of CO_2 curing duration. In addition, the CO_2 absorption occurs at a constant rate.

The constant carbonation rate is consistent with the mass loss and subsequent gain shown in Fig. 7 (b), and confirms that water evaporation dominates over the mass gain from carbonation at first. This also agrees with the previous discussion of total mass change where D2 reaches the turning point faster. It is clear to see that D4 presents a slower CO₂ uptake during the whole CO₂ curing period. A smaller slag particle size shows no positive effect on the CO₂ uptake below 24.13 μ m for a carbonation duration of 14 days. This result may be due to the slow CO₂ diffusion in fine steel slag blended samples.

Generally, the carbonation rates of normal concrete exhibit a linear relationship between carbonation depth and the square root of time. However, in the present study, the carbonation shows a constant rate for all blended mortars even when it has reached the sample core. This indicates that CO_2 diffusion alone does not control the carbonation rate of these steel slag blended mortars.

The CO_2 permeability in the concrete sample is dominated by several factors, such as humidity, temperature, and pore structure. During early age, free water evaporation provides more and more unsaturated pores, which effectively promote CO_2 transportation as a gas. As a consequence, a constant carbonation rate was observed. In addition, the reaction of steel slag carbonation needs appropriate internal humidity and a necessary water film on the slag surface (Baciocchi et al., 2011). Even though the fast water evaporation could provide more pathways Table 4

CO ₂ emisssion of carbonated steel slag-cement concrete	(kg/	m	"]
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CO ₂ emission	OPC ^a	D1	D2	D3	D4
Cement Steel slag (milling) Sand Water CO ₂ uptake CO ₂ for curing Total emisssion Sustainbility efficiency (MPa/(kg/	495.59 0 6.39 0.04 / 502.02 0.112	98.89 32.24 6.38 0.04 -82.95 0.025 54.63 0.466	98.89 41.52 6.38 0.04 -84.54 0.025 62.31 0.486	98.89 50.74 6.38 0.04 -79.75 0.024 76.32 0.409	98.89 59.97 6.38 0.04 -44.45 0.013 120.84 0.194
m ³))	0.112	0.100	0.100	0.105	0.191

a- normal cement mortar from previous study (Liu et al., 2019)

for CO₂ diffusion, a lack of water on the slag surface would slow the carbonation rate. On the other hand, slow water evaporation could inhibit CO₂ penetration, as well as the carbonation. These observations are consistent with the results in Section 3.3. It is confirmed again that the optimal steel slag particle size is a critical factor for the CO₂ uptake ability of steel slag blended mortars.

The CO₂ footprints of normal cement mortar and converter steel slag blended mortars are evaluated and shown in Table 4. The energy of steel slag milling and CO₂ gas preparation are included. It can be seen that the increase of milling intensity contributes to a gradually increased CO₂ emission due to higher energy consumption. This increases the total CO₂ emission from 54.63 kg/m³ to 120.84 kg/m³. However, the CO₂ uptake ability of steel slag and the high volume cement replacement results in a lower l footprints of blended mortars than for normal OPC. As a the building material, sustainability efficiency reflects the correlation between strength and CO₂ emission (Damineli et al., 2010). As can be seen, the sustainability efficiency of carbonatated blended mortars is obviously higher than for normal OPC concrete. D2 shows the highest sutainability efficiency of all samples. This indicates that the optimal selection of converter steel slag particle size can efficiently help to enhance the sustainability of carbonated blends by enhancing strength and CO₂ uptake of samples.

3.5. Microstructural property

To further explain the observed mechanical properties of the CO_2 cured mortar samples, their microstructure were investigated with SEM and MIP, and the results are shown in Fig. 8.

As it can be seen from the MIP results, sample Ref. shows the highest total pore volume (0.0786 ml/g) of all samples, which is due to the fact that quartz powder cannot be carbonated, while steel slag does carbonate, filling in the pores (Zhan et al., 2013). Comparing the total pore



Fig. 7. CO2 uptake of different cement-steel slag binders.



Fig. 8. Pore size distribution and SEM of mortars after 14 days carbonation curing (a) cumulative pore volume (b) pore size distribution (c) hydrated area (d) carbonated area.

Table 5One batch leaching results of converter steel slag blended mortars.

	Leaching ions (mg/L)					
	Al	Ca	Mg	Cr	V	pН
Before CO ₂	0.363	156.723	L.D.	0.007	0.006	12.4
3 days CO ₂	0.449	157.704	0.002	0.018	0.004	12.47
7 days CO ₂	0.671	166.777	0.006	0.069	0.047	12.28
14 days CO_2	0.104	68.558	0.466	0.06	5.522	10.85
Before CO ₂	0.334	156.507	L.D.	0.005	0.005	12.4
3 days CO ₂	0.442	159.496	0.001	0.02	0.012	12.43
7 days CO ₂	0.624	124.72	0.038	0.081	0.753	11.9
14 days CO_2	0.100	64.916	0.547	0.052	6.278	10.7
Before CO ₂	0.312	156.547	L.D.	0.006	0.005	12.46
3 days CO ₂	0.427	160.068	0.003	0.022	0.013	12.45
7 days CO ₂	0.462	163.288	0.004	0.049	0.024	12.39
14 days CO_2	0.099	68.172	0.476	0.05	5.251	10.83
Before CO ₂	0.221	156.403	L.D.	0.013	0.001	12.55
3 days CO ₂	0.311	159.013	0.005	0.01	0.005	12.5
7 days CO_2	0.264	156.541	0.003	0.034	0.003	12.56
14 days CO_2	0.634	122.65	0.056	0.087	0.447	11.91
	$\begin{array}{c} Before \ CO_2\\ 3 \ days \ CO_2\\ 7 \ days \ CO_2\\ 14 \ days \ CO_2\\ Before \ CO_2\\ 3 \ days \ CO_2\\ 14 \ days \ CO_2\\ 14 \ days \ CO_2\\ 3 \ days \ CO_2\\ 7 \ days \ CO_2\\ 14 \ days \ CO_2\\ Before \ CO_2\\ 3 \ days \ CO_2\\ 14 \ days \ CO_2\ CO_2\\ 14 \ days \ CO_2\ CO$	Leachin Al Before CO2 0.363 3 days CO2 0.449 7 days CO2 0.671 14 days CO2 0.104 Before CO2 0.334 3 days CO2 0.442 7 days CO2 0.442 7 days CO2 0.442 7 days CO2 0.621 14 days CO2 0.100 Before CO2 0.312 3 days CO2 0.427 7 days CO2 0.462 14 days CO2 0.462 14 days CO2 0.312 3 days CO2 0.427 7 days CO2 0.262 14 days CO2 0.209 Before CO2 0.221 3 days CO2 0.311 7 days CO2 0.264 14 days CO2 0.634	Leaching ions (mg/Al Ca Before CO2 0.363 156.723 3 days CO2 0.449 157.704 7 days CO2 0.671 166.777 14 days CO2 0.104 68.558 Before CO2 0.334 156.507 3 days CO2 0.442 159.496 7 days CO2 0.624 124.72 14 days CO2 0.100 64.916 Before CO2 0.312 156.547 3 days CO2 0.427 160.068 7 days CO2 0.462 163.288 14 days CO2 0.099 68.172 Before CO2 0.221 156.403 3 days CO2 0.311 159.013 7 days CO2 0.264 156.541 14 days CO2 0.264 156.541 14 days CO2 0.634 122.65	Leaching ions (mg/L) Al Ca Mg Before CO2 0.363 156.723 L.D. 3 days CO2 0.449 157.704 0.002 7 days CO2 0.671 166.777 0.006 14 days CO2 0.104 68.558 0.466 Before CO2 0.334 156.507 L.D. 3 days CO2 0.442 159.496 0.001 7 days CO2 0.624 124.72 0.038 14 days CO2 0.100 64.916 0.547 Before CO2 0.312 156.547 L.D. 3 days CO2 0.427 160.068 0.003 7 days CO2 0.099 68.172 0.476 Before CO2 0.221 156.403 L.D. 3 days CO2 0.312 156.403 L.D. 3 days CO2 0.211 155.013 0.005 7 days CO2 0.221 156.403 L.D. 3 days CO2 0.311 159.013 0.005 7 days	Leaching ions (mg/L) Mg Cr Al Ca Mg Cr Before CO2 0.363 156.723 L.D. 0.007 3 days CO2 0.449 157.704 0.002 0.018 7 days CO2 0.671 166.777 0.006 0.069 14 days CO2 0.104 68.558 0.466 0.001 Before CO2 0.334 156.507 L.D. 0.005 3 days CO2 0.442 159.496 0.001 0.02 7 days CO2 0.624 124.72 0.038 0.081 14 days CO2 0.100 64.916 0.547 0.052 Before CO2 0.312 156.547 L.D. 0.006 3 days CO2 0.427 160.068 0.003 0.022 7 days CO2 0.422 163.288 0.004 0.049 14 days CO2 0.099 68.172 0.476 0.55 Before CO2 0.221 156.403 L.D. 0.013 3	Leaching ions (mg/L) Ng Cr V Al Ca Mg Cr V Before CO2 0.363 156.723 L.D. 0.007 0.006 3 days CO2 0.449 157.704 0.002 0.018 0.004 7 days CO2 0.671 166.777 0.006 0.069 0.047 14 days CO2 0.104 68.558 0.466 0.06 5.522 Before CO2 0.334 156.507 L.D. 0.005 0.005 3 days CO2 0.442 159.496 0.001 0.02 0.012 7 days CO2 0.624 124.72 0.038 0.081 0.753 14 days CO2 0.100 64.916 0.547 0.052 6.278 Before CO2 0.312 156.547 L.D. 0.006 0.005 3 days CO2 0.427 160.068 0.003 0.024 0.449 0.244 14 days CO2 0.099 68.172 0.476 0.055 5.251

L.D. - below the detection limit.

volume of mortar samples with steel slag, it can be seen that the sample with D4 had the highest total pore volume and the other three showed a slight decrease with the increase of slag particle size.

The pore size distribution of different carbonated mortars is shown

in Fig. 8 (b). It is noticed that there is an additional peak at around 100 nm in D4 (16.17 μ m) that is not present in the other steel slag samples. This peak is usually observed in cement samples, and is characteristic for capillary pores of C-S-H in cement based concrete (Liu et al., 2019). The presence in D4 indicates that there likely is a coexistence of C-S-Hhydration and carbonation products, as already identified in Section 3.2. In addition, the pore volume in this range (around 100 nm) is dramatically reduced in samples with a higher carbonation degree, such as D1 and D2. It appears that capillary pores were partially refined by the carbonation products in D1-D3. However, when the steel slag particle size increased from 21.75 μm (D3) to 28.98 μm (D1), the location of the main peak gradually shifts to the larger value. In addition, a small peak was present at 28.98 μm (D1) steel slag blended mortar. This gradual increase of pore may be caused by the poor particle packing of the mixtures. The increase of pore size also correlates with the mechanical performance of D1, D2, and D3. The mechanical performance is not only controlled by the amount of reaction products but also influenced by the packing of mixtures (Brouwers and Radix, 2005).

3.6. Evaluation of leaching properties

To study the leaching properties of steel slag blended mortars after carbonation a leaching test was conducted after 3 days, 7 days and 14 days. The results are shown in Table 5 and Fig. 9.



Fig. 9. The ions leaching and CO₂ uptake versus pH of different converter slag blended mortars.

It can be seen that for all steel slag samples, the leaching of Mg keeps increasing with progressive carbonation. At the same time, Ca concentration decreases gradually concurrent with the appearance of V in the leachate and a drop in pH. The Al and Cr concentration of D1, D2 and D3 also increase until 7 days carbonation. For D4, the Al concentration keeps increasing during the whole test duration.

The correlation between pH and leaching of steel slag blended mortars is shown in Fig. 9., which indicates that the leaching behavior of samples relies on the pH variation and phase assemblage rather than the steel slag particle size difference. In addition, the carbonation degree controls the leaching properties by changing the pH. Since portlandite and free-lime exist in the binder system before carbonation, a high pH can be kept during early carbonation. During the carbonation process, the portlandite from cement and steel slag hydration can serve as a pH buffer, and keep Ca leaching at a high level for the first 7 days. On the other hand, the leaching of Mg is limited under these condition (Huijgen and Comans, 2006b). Regarding further carbonation, once the residual portlandite is carbonated and cannot contribute to Ca leaching anymore, the pH drops significantly. At the same time, the Mg leaching increases (Baciocchi et al., 2015a). So, the decrease of Ca leaching corresponds with the increase of Mg leaching. The leaching of both Al and Cr increase at first and then decrease with the lowering of the pH. The increase of Al leaching can be attributed to the carbonation of AFt and AFm from cement hydration, while the Cr leaching stems from the carbonation of C₂S in steel slag (van Hoek et al., 2016). However, the leaching of Cr and Al decreases after the pH drops to below 12, which is related to the identical amphoteric behavior. A similar result also was observed in previous research ((Baciocchi et al., 2015b)b). The leaching of V also exhibits a strong correlation with pH, being mainly driven by the dissolution behavior of C₂S in steel slag. During the early carbonation, the portlandite saturation induces a high Ca concentration and high pH in the pore solution. At these conditions of portlandite saturated pore solution, the dissolution of C₂S is inhibited and the leaching of the V hosted by C₂S limited (Santos et al., 2012). The increasing CO₂ uptake correlates with a decrease of pH as shown in Fig. 9. With more advanced carbonation, pH decreases and C_2S dissolution starts to increase, corresponding with a significant rise in V leaching. This observation agrees with previously published results (Costa et al., 2016). These are the overall expected mineralogy changes. Attributing discrete element behavior to specific phases will require modeling of the complex aqueous-mineral equilibria using FREEQC or similar programms.

This is considered beyond the scope of this study at this stage but related investigation will be conducted in future research. Furthermore, the V leaching of crushed mortars after 14 days carbonation in the present study is higher than the limit set in the Dutch Soil Quality Decree (1.8 mg/kg). However, the leaching properties are influenced by many factors, such as mix design, curing strategy and porosity. For example, D4 still exhibts a low V leaching (due to its still high pH), which meets the requirement of SQD. So, in future research the influence of mix design and porosity on V leaching will be investigated to solve this problem. it is possible to improve the V leaching using optimal concrete mix design, as well as the CO_2 curing process to satisfy the regulation. This also will be investigated in future research.

4. Conclusions

The presented results in this study confirms that it is possible to produce green building materials by carbonating converter steel slag blends at ambient conditions and that optimizing the converter steel slag particle size is a crucial parameter. The variation of steel slag particle size influences the efficiency of CO_2 uptake in blended mortars, which directly results in a difference of mechanical performance. The leaching properties also depend on the carbonation degree and subsequent pH value. Based on the above results, the following conclusions can be drawn:

- (1) Ambient CO₂ curing is more effective than using normal, humid curing of converter steel slag blended mortars.
- (2) There is an optimal mean particle size of $21.75 \ \mu m$ (D3) for steel slag in the present study based on strength performance after 14 days carbonation. The compressive strength and sustainability coefficiency are found to decrease for both larger and smaller steel slag particle sizes.
- (3) The mass development of steel slag blended mortars during CO_2 curing showed three stages. A turning point at the transition from stage I to stage II could be defined, which reflects the carbonation speed. The finer the converter steel slag was, the longer the blended mortars needed to reach the turning point.
- (4) Between 8 to 15.9% of CO₂ can be taken up by the cement steel slag binder due to carbonation, with the slag size of 24.13 μ m (D2) showing the highest CO₂ uptake after 14 days CO₂ curing. Less carbonation is observed when the slag particle size is finer or larger

than D2.

(5) The leaching properties of steel slag blended mortars are controlled by the CO_2 uptake, and only indirectly by steel slag particle size. The Cr and V leaching increased with progressing carbonation once the pH and Ca concentration of the leachate dropped. The highest V leaching was related to the steel slag D2 (24.13 µm), which correlated with the highest CO_2 uptake and lowest leachate pH after 14 days CO_2 curing.

Declaration of Competing Interest

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

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