

LEACHING OF HEAVY METALS FROM CONCRETE PAVERS CONTAINING WASTE-TO-ENERGY BOTTOM ASH AS AN AGGREGATE

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Introduction

In its waste-to-energy (WtE) plant, AEB Amsterdam annually combusts about 1.3 million metric tonnes (t) of residual household and comparable industrial waste, generating about 340 000 t of bottom ash (BA). This ash is submitted to several treatment steps, including washing and sieving, so that it complies with the emission and composition limits for use as building material set in the Dutch regulation on soil quality. This way, treated BA can be used as a replacement for conventional aggregates such as gravel in concrete. In that case, the finished concrete product has to comply with the emission limit values for shaped building materials, including limits for heavy metals, halides and sulphates. Previous research showed that carbonation, *i.e.* the reaction of CO₂ from the air with compounds such as CaO, decreases the pH of BA, increasing the solubility of certain heavy metal compounds.¹ So if BA is used as an aggregate in shaped concrete materials which are exposed to ambient air for many decades, this could increase diffusion of heavy metals from the BA in these materials. Furthermore, at the end of their life the concrete products are likely to be crushed and recycled as granulate. In this second life phase, further carbonation and thus increase of heavy metal emission can occur. Therefore, this study investigates the effect of carbonation on the leaching of regulated heavy metals from concrete pavers containing BA as aggregate, and from their granulates.

Materials and methods

The concrete pavers (0.3 m x 0.3 m x 0.045 m) used in the experiments consisted of a base layer of 39 mm covered by a 6 mm top layer. Treated BA provided by AEB Amsterdam was used to partially replace the aggregates in the base layer. Other aggregates consisted of fine and coarse sand, gravel and granite stone. CEM III/A and ground limestone were used as a binder. The used BA complied with the emission and composition limits for use as “IBC” (*Isoleren, Beheersen, Controleren* or Contain,

Control and Monitor) building material set in annex A of the Dutch regulation on soil quality and accounted for < 17% of the total aggregate mass.

Two concrete pavers were carbonated in an incubator (Heraeus Thermo BBD 6220) at 20 vol% CO₂, 30°C and 70% humidity. After 14 days, one of the pavers was submitted to a diffusion test according to CMA/2/II/A.9.2, in parallel with a non-carbonated paver. The second carbonated paver was crushed with a hammer until a 0-31.5 mm granulate fraction was obtained. Also, a non-carbonated paver was crushed and both the carbonated and non-carbonated granulates were, as such and after grinding them to a < 4 mm fraction, submitted to a shaking test according to CMA/2/II/A. The remaining granulates from the carbonated paver were placed back in the CO₂ incubator to simulate second life and samples were taken and submitted to a shaking test after 3, 7, 10, 14 and 17 days. The concentration of the heavy metals and sulphur in the eluates was determined with ICP-OES (Varian 720-ES). The granulates from the carbonated and non-carbonated paver and from the carbonated paver after 17 days in the CO₂ incubator were also submitted to a column test (NEN 7383 AP04).

Results and discussion

Diffusion of heavy metals from carbonated and non-carbonated paver

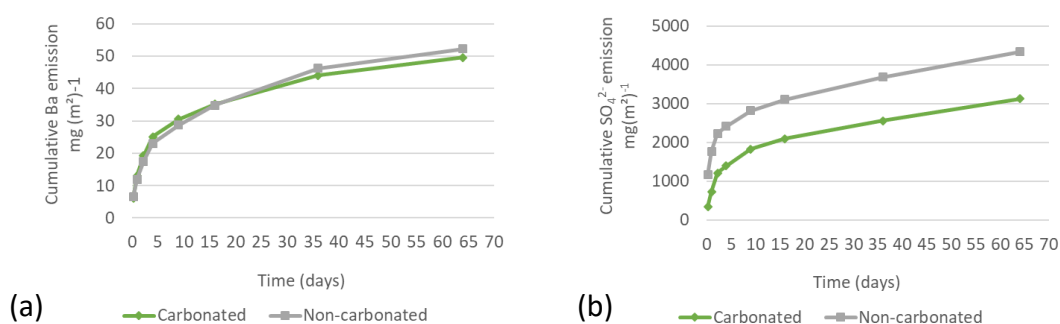


Figure 1: Measured cumulative Ba (a) and SO₄²⁻ (b) emission from the carbonated and non-carbonated paver (mg(m²)⁻¹)

The concentration of heavy metals was below the detection limit in most of the diffusion test eluates for both the carbonated and the non-carbonated paver. The detection limits ranged from 0.05 mg(m²)⁻¹ for Cd to 1.5 mg(m²)⁻¹ for Pb, but even considering the concentrations equal to the detection limit in all the eluates, the measured cumulative emission after 64 days was below the Dutch limit value for all the regulated metals. Ba was the only metal present in concentrations above the detection limit in all eluates and its emission rate quadratically decreased over time. Furthermore, the emission of Ba from the carbonated and non-carbonated paver showed little difference (Figure 1a). The emission of sulphate (SO₄²⁻) from the

carbonated paver was only about half of the emission from the non-carbonated tile (Figure 1b). The pH of the eluate at 64 days was 10.9 and 11.4 for the carbonated and non-carbonated paver, respectively.

Leaching of heavy metals from granulates

The pH of the shaking test eluate of the non-carbonated paver granulates was 12.1, whereas the pH of the shaking test eluate of the carbonated paver granulates was 11.5 and gradually decreased to 10.8 for granulates that remained 17 days in the CO₂ incubator. The sharpest pH decrease was noticed the first 7 days, after which the pH more or less stabilised, suggesting that a chemical equilibrium was already reached at an early stage. The conductivity showed a similar trend: it was about 3500 µS/cm for the eluate of the non-carbonated paver granulates, whereas it was about 1100 µS/cm for the eluate of the carbonated paver granulates and stabilised at around 900 µS/cm after 7 days of carbonation of these granulates. The carbonation process clearly decreased the dissolution of minerals from the granulates.

The concentration of As, Cd, Co, Pb, Se and Sn was below the detection limit in the shaking test eluates of most of the granulates, so it was not possible to draw conclusions on the effect of carbonation on their leaching behaviour. The leaching of Ba, Cr, Cu, Mo and V from the granulates did not show a trend as a function of the carbonation time but remained rather constant. The leaching of Zn clearly decreased with increasing carbonation time and could be linked to the decreasing eluate pH. The leaching of Sb from the granulates showed an increasing trend over time, but there was a significant difference between the leaching from the granulates (blue line — in Figure 2a) and from the granulates that were ground before the shaking test (orange line — in Figure 2a).

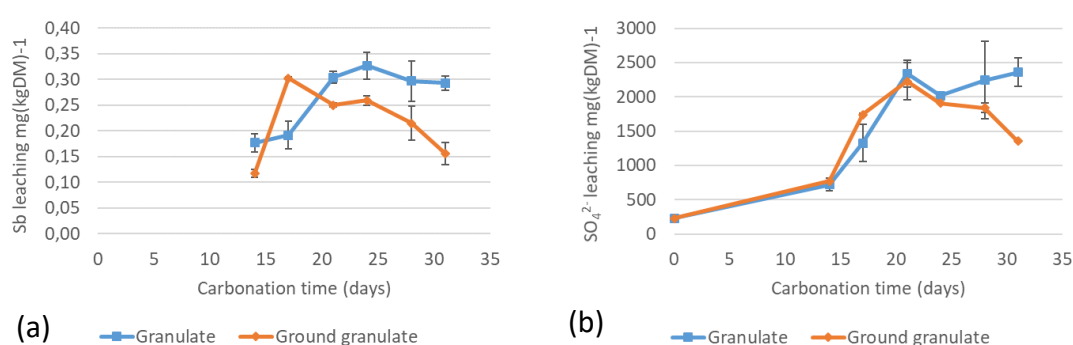


Figure 2: Sb (a) and SO₄²⁻ (b) leaching from granulates and granulates that were ground before submitting them to a shaking test as a function of carbonation time (mg(kg)⁻¹)

The Sb leaching from the granulates that were ground before the shaking test followed the typical Sb leaching trend as a function of pH that was established in previous work.² The increase of sulphate leaching with increasing carbonation time

(Figure 2b) can be explained by the breakdown of ettringite minerals through reaction with CO₂:³



The sulphate leaching trend was very comparable to the Sb leaching trend, suggesting that their immobilisation mechanisms are linked, possibly because Sb(OH)₆⁻ ions are incorporated in or adsorbed to sulphate containing ettringite minerals.²

Column tests showed that the leaching of regulated metals, halides and sulphates from granulates of the non-carbonated and carbonated paver and from the latter after 17 days of additional carbonation were below the Dutch limit values for use as uncontained building material.

Conclusions

Diffusion tests of concrete pavers in which part of the aggregates was replaced by WtE BA showed that these pavers complied with the Dutch legal emission limit values for shaped building materials, even after carbonation for 14 days in an incubator at 20 vol% of CO₂. Column tests confirmed that 0-31.5 mm granulates from these pavers comply with the Dutch legal emission limit values for uncontained building materials, even after 17 days of additional carbonation. So also in a second life as granulate these WtE BA containing pavers could be used safely.

Adsorption on or incorporation in ettringite minerals was shown a relevant binding mechanism for Sb in concrete applications and possibly also in BA at elevated pH. However, the differences in Sb leaching from granulates and from granulates that were ground before the shaking test showed that other, not yet fully understood binding mechanisms apply. Possibly also physical aspects such as particle size or composition and porosity of the contact surface influenced the water/solid phase interaction and by this also the Sb leaching.

References

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