RECYCLING OF AUTOCLAVED AERATED CONCRETE IN SCREED AND STABILIZED SAND

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ABSTRACT

Autoclaved aerated concrete (AAC) is a lightweight cellular concrete that has been used for more than 80 years. Currently, however, no good recycling options for AAC from construction and demolition waste exist. The amount of AAC waste that can be recycled in the production of new AAC is limited because of quality issues. Furthermore, recycling AAC into traditional concrete or as unbound aggregate causes both technical and environmental problems because of the low compressive strength (2-8 MPa) of AAC and its high amount of leachable sulfate: typically > 10,000 mg/kg dm (L/S = 10).

In this paper, recycled AAC waste was evaluated as a replacement of sand in a traditional screed (subfloor) and in cement stabilized sand products. A range of cements (CEM I, CEM II and CEM III), were used in combination with the crushed AAC waste aggregate (0-8 mm). During hydration a reaction of the AAC leachable sulfate and the aluminate contained in the cement resulted in the formation of (insoluble) ettringite. The main conditions influencing the formation of ettringite, and hence the leaching of sulfate, were examined in cement stabilized sand products. A sufficiently high pH was found to be crucial to meet sulfate leaching standards. The presence of additional sulfate as gypsum impurities in the AAC waste proved detrimental towards sulfate leaching.

Finally, the replacement of sand by recycled AAC improved the thermal properties of the screed, rendering the product suitable for niche products that target improved thermal insulation.

Keywords: Autoclaved aerated concrete; sulfate leaching; recycling; immobilization

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1. INTRODUCTION

1.1. Autoclaved aerated concrete: production and properties

Autoclaved aerated concrete (AAC) is a lightweight microporous building material with a density of 400-800 kg/m^3 and thermally insulating capacities (0.1-0.2 W/mK) [1,2]. Two types of AAC are commonly manufactured, white and grey AAC. Both types use combinations of CaO and Portland cement mixed with either ground quartz sand (white AAC) or Class F fly ash (grey AAC) [1]. These raw materials are mixed with water and aluminium powder and left to react at elevated temperature and pressure (450 K, 10-12 bars). The reaction of the hydroxyls released in the cement hydration process with the added aluminium results in the formation of H_2. This reaction involves a large volume expansion and causes a large increase in porosity which makes the material to rise like a cake [2,3]. Calcium sulfate (2-5 wt.%), in the form of gypsum or anhydrite, is added to the raw materials to facilitate the crystallisation of the calcium silicate hydrate phases into tobermorite. This results in higher strength and an end product less susceptible to shrinkage and carbonation [4,5,6].

Because of its high porosity and low energy and material consumption [7], AAC can be seen as a sustainable building product. However, the recycling of AAC still remains a challenge. AAC has a lower compressive strength (1-9 MPa) [1] than other stony materials in construction and demolition waste (C&DW) and is therefore not suitable for traditional recycling applications for the stony fraction of C&DW (e.g. the use in foundations). Moreover, the chemical composition of AAC can cause technical and environmental problems. AAC contains on average 12,600 mg/kg dm of leachable sulfate on an average total sulfate level of 2.1 wt.% [8,9]. The presence of excessive levels of sulfate can lead to technical problems (e.g. efflorescence and internal sulfate attack) in building materials [10,11,12] and ecotoxicological effects (salinity, sulfide formation, eutrophication) caused by leaching into the environment [13].

1.2. Current AAC recycling possibilities

Since AAC cannot be recycled in the applications that are used for the stony fraction of C&DW (mostly unbound applications) separate collection during demolition is required. Currently, it is possible to use AAC waste as a replacement for the sand fraction (maximal replacement of 20%) in the production of new AAC [3]. However, this application is limited to production and construction waste because of its more constant composition. AAC demolition waste can contain impurities (asphalt, plastics), resulting in visual contaminations and problems in the production process of new AAC. Other recycling options for AAC waste include the use of crushed AAC waste as oil absorbent or (low-grade) filler for cat litter boxes [3,14,15]. To date, a significant portion of AAC waste is, however, still landfilled [3].

1.3. New recycling opportunities by sulfate immobilization

A possibility for the creation of new recycling routes is by immobilization of the leachable sulfates in AAC, for instance by chemical binding in cement hydration products. Ambroise and Péra (2004, 2008), for example, used demolition waste containing calcium sulfate as a supplementary cementitious material in calcium sulfo-aluminate (CSA) cements. They showed that the calcium sulfate was entirely consumed and bound into insoluble hydration products in case the ratio of CSA to calcium sulfate was 4:1 or higher [16,17]. Brouwer et al. (2000) described a method for chemically immobilizing sulfate from screening sands, which contain up to 6 wt.% of sulfate, with either Portland cement, blast furnace slag cement or calcium aluminate cement [18]. The sulfate in this process is bound in ettringite [$Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 24H_2O$], a calcium trisulfoaluminate hydrate commonly found as an early-formed hydration product in hydrated cements. In ordinary Portland cements ettringite is formed in the hydration reaction of the aluminate (C_3A –cement chemistry notation) clinker phase in the presence of calcium sulfate proceeds according to reaction equation (1).

$$Ca_3Al_2O_6 + 3CaSO_4 + 32H_2O \leftrightarrow Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 24H_2O \text{ (ettringite)} \qquad (1)$$
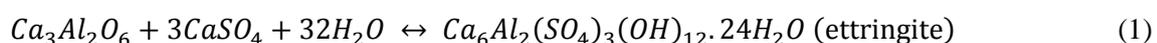
Ettringite formation can be associated with a loss of mechanical strength of concrete elements. However, this occurs in case ettringite is formed after cement hardening (delayed ettringite formation) [19]. Ettringite formation after hardening is accompanied with a solid volume expansion, which can cause crack formation and propagation in the already hardened cement matrix. In contrast, when ettringite is formed early on during hydration it contributes positively to the development of compressive strength.

In this paper we will produce recycling products with crushed AAC waste. To mitigate leaching problems, sulfate immobilization through ettringite formation will be investigated.

2. EXPERIMENTAL

2.1. Cement stabilized sand products with AAC waste

Cement stabilised sand products with recycled AAC were developed. In these cement stabilised sand products, the entire sand fraction was replaced by secondary materials: 60 wt.% mixed recycled aggregates (MRA) of C&DW and 40 wt.% recycled AAC.

First, AAC waste was crushed with a disc mill to aggregates 0-8 mm. Second, mixtures for a cement stabilised sand product were produced using recycled AAC (533 kg/m³ of product) and C&DW mixed recycled aggregates (0-8 mm, 800 kg/m³, produced by an industrial C&DW crushing installation), using different types of cement (150 kg/m³): CEM I 52.5N, CEM II B-M, CEM III/A 42.5N. Tap water (280 l/m³, used as mixing water, partially absorbed by the recycled AAC) was added and the materials were mixed in a concrete mixer. The mixture was casted into cylindrical molds and cured (100% relative humidity, 20 °C) (Figure 1).

Figure 1. Surface of the cement stabilized sand with crushed cellular concrete and mixed recycled aggregates (0-8 mm).

2.2. Sulfate leaching

The leaching of sulfate from the AAC waste, the mixed recycled aggregates and the recycling products was analysed using batch leaching tests. The batch leaching tests were run in accordance with the Flemish standard CMA/2/II/A.9.4 for determination of the leaching of inorganic compounds from granular materials and sludge. The only deviation from the standard was a size reduction of the materials to <4 mm. This particle size is also used in other leaching tests (e.g. the Dutch NEN 7383 standard) and enlarges the specific surface of the material, enhancing the leachability of sulfate.

The materials (<4 mm) were brought into contact with the leaching liquid (L/S = 10) with the aid of an overhead mixer (5 turns/min.). This method is based on the assumption that a state of equilibrium (or near-equilibrium) is reached between the liquid and the solid phase during the test period. Because ultrapure water is used as leaching liquid, the pH and ionic strength of the leachate are determined by the tested materials. After 24 h, the solid residue was separated from the liquid by filtration (pore size = 0.45 μm). The pH of the leaching liquid was measured immediately after filtration. The sulfate concentration of the leaching liquid was measured by liquid chromatography (ISO 10304-1).

2.3. Total sulfur concentration

The total sulfur content of the recycling products was analysed using inductively coupled plasma atomic emission spectroscopy (ICP-AES) (ISO 11885) after crushing (<100 μm) and closed microwave digestion by $HCl/HNO_3/HF/H_3BO_3$ (CMA/2/II/A.3).

2.4. Compressive strength measurements

The compressive strength of the recycling products was measured in accordance with EN 772-2 and more specific EN 771-3+A1, 5.5 standards, on cylindrical samples (h = 12 cm, d= 10 cm) after 28 days of curing.

2.5. Thermal conductivity measurements

Cement stabilized sand mixtures are usually not used in indoor applications. To examine possible thermal insulation effects that result from the use of recycled AAC in indoor applications, the thermal conductivity of a developed indoor floor screed with recycled AAC was measured.

In the floor screed product, part of the sand fraction was replaced by recycled AAC (0-8 mm). The used mixture contained 1100 kg/m³ river sand (0-4 mm), 210 kg/m³ recycled AAC, 220 kg/m³ CEM I and 150 l/m³ tap water. The materials were mixed in a concrete mixer. The mixture was poured in a square mold (90 cm²) and cured for 28 days (100% relative humidity, 20 °C). To exclude other factors than the AAC replacement, there was no replacement of river sand by mixed recycled aggregates.

The thermal conductivity of the screed product was measured with a heat flow meter (Figure 2) for samples of 90 cm² (ISO 8302) (h = 50 mm). The sample is positioned between a cold and hot plate. Between the plates a temperature difference of 10 K was imposed by water circuits. The heat flux through the sample was measured using heat flow meters and thermocouples positioned between the plates and the sample.
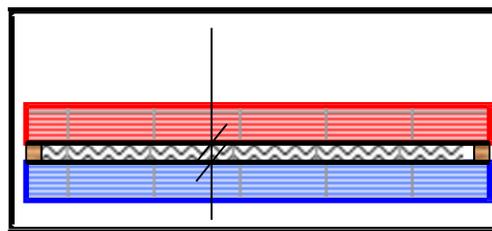
Figure 2. Structure of the heat flow meter apparatus. The sample is positioned between a hot (red) and cold plate (blue). The straight black lines between the plates represent neoprene layers with heat flow meters. The black box surrounding the structure represents an isolated structure.

Once the heat flux and temperatures are stabilized, these variables are measured every 10 minutes during 3 h. The thermal conductivity is calculated using equation (2).

$$\lambda = \frac{h(C_1 E_1 + C_2 E_2)}{2\Delta T} \qquad (2)$$

Where C_1 and C_2 are the calibration constants of the heat flow meters (in W/m²V), E_1 and E_2 are the electrical potential differences over the heat flow meters (in V), ΔT is the temperature difference over the top and the bottom of the sample (in K) and h is the thickness of the sample (in m).

The thermal conductivity is measured for different temperatures. There is a linear correlation between the thermal conductivity of a dry material and the average temperature in the material. Using this correlation and 5 measured values, the thermal conductivity at 296 K is calculated.

3. RESULTS

3.1. Sulfate leaching

3.1.1. *Leaching of the secondary resources*

The sulfate leaching of the used recycled AAC was 15,500 mg/kg dm. This value is similar to the average sulfate leaching (12,600 mg/kg dm) of AAC in 24 German production units, described by Lang-Beddoe & Schrober [8]. The used mixed recycled aggregates showed a lower, but still significant sulfate leaching (2,400 mg/kg dm).

3.1.2. *Leaching of the products*

Figure 33 shows the sulfate leaching of the cement stabilised sand products with recycled AAC. The leaching of sulfate is strongly dependent of the leachate pH. The leachate of the products with CEM I has an average pH of 12.4 and an average sulfate leaching of 840 mg/kg dm. If part of the Portland cement is replaced by supplementary cementitious materials (SCMs) such as coal fly ash or blast furnace slag (CEM II/B-M or CEM III/A) the pH of the leachate is observed to be lower. This is due to 1) the Portland cement dilution reducing overall alkali levels and 2) the pozzolanic reaction of the SCMs that consumes portlandite. This is accompanied by higher sulfate concentrations in the leachate.
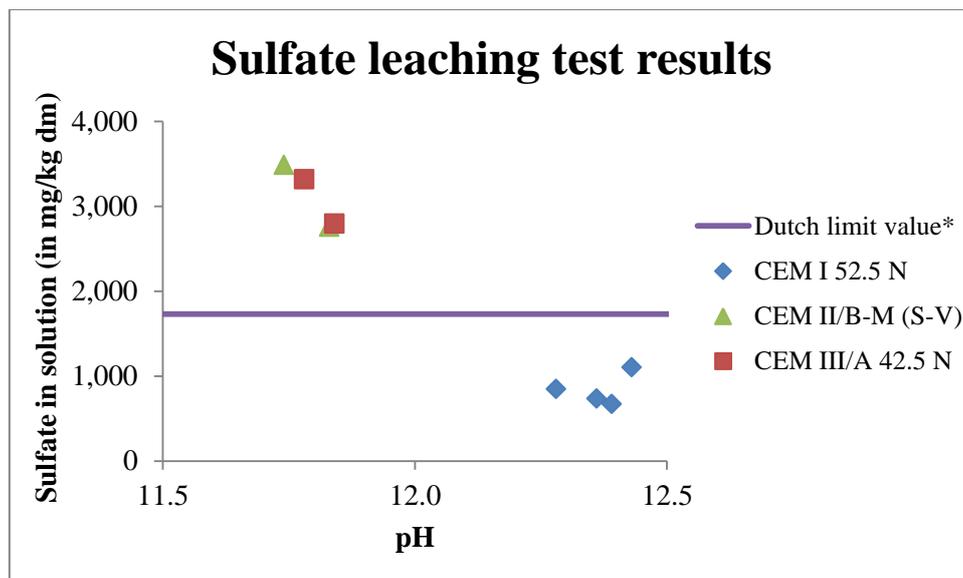
Figure 3. Sulfate leaching of the cement stabilised sand products with recycled AAC, determined by a batch leaching test.
*: Sulfate leaching limit value for column leaching test (NEN 7383, CMA/2/II/A.9.1) in the Dutch legislation for resources "Regeling Bodemkwaliteit".

The obtained sulfate leaching values were compared to the sulfate leaching limit value proposed by the Public Waste Agency of Flanders (OVAM) for the new Flemish environmental legislation (2,200 mg/kg dm). A sufficiently high pH of the cement stabilized sand products, best accomplished by using CEM I, is critical to comply with this limit value.

3.1.3. Relation with the total sulfur content

The total sulfur content is similar for the different samples produced with recycled AAC. Thus, the demonstrated lower leaching values for the samples with CEM I are not caused by a lower sulfur content.

Because of similar visual properties, mixing of AAC waste and gypsum (plaster board) waste can occur. To demonstrate the influence of gypsum contamination on the recycling products, a cement stabilized sand sample was produced with gypsum contaminated AAC. This gypsum contamination resulted in a large increase in total sulfur content and sulfate leaching of the products (Table 1).

Table 1. Average sulfate leaching (batch leaching test) for the produced samples in function of the total sulfur content in the cement stabilised sand products with recycled AAC.

| Cement stabilized sand samples	Variables		
	pH	Total sulfur content (in mg/kg dm)	Sulfate leaching (in mg/kg dm)
CEM I 52.5 N	12.3	5,800	760
CEM II/B-M (S-V)	11.8	5,700	3,100
CEM III/A 42.5 N	11.8	6,000	3,100
CEM I 52.5 N + gypsum contamination	12.1	12,000	9,800

The presence of gypsum particles in the cement matrix causes locally a very high concentration of leachable sulfate. This will result in a deficient amount of reactive aluminium to convert the available calcium sulfate into ettringite. This resulted in a strong increase in sulfate leaching.

3.2. Compressive strength

The cement stabilized sand products made using CEM I 52.5 N obtained an average compressive strength of 9.6 MPa. Depending on the intended application, this compressive strength can be increased by using more cement. It should be kept in mind that the used cement does not only provide strength, but also facilitates the immobilization of sulfate. Conversely, the use of less cement for applications that require a lower compressive strength could result in an increase of sulfate leaching.

3.3. Thermal conductivity

The floor screed sample with recycled AAC (ρ = 1,400 kg/m³) showed an average calculated thermal conductivity of 0.50 ± 0.01 W/(mK) at 23 °C (Figure 4). This corresponds to a heat resistance of 2.0 mK/W, which is higher than the heat resistance of traditional floor screeds (typically 1.0 mK/W).
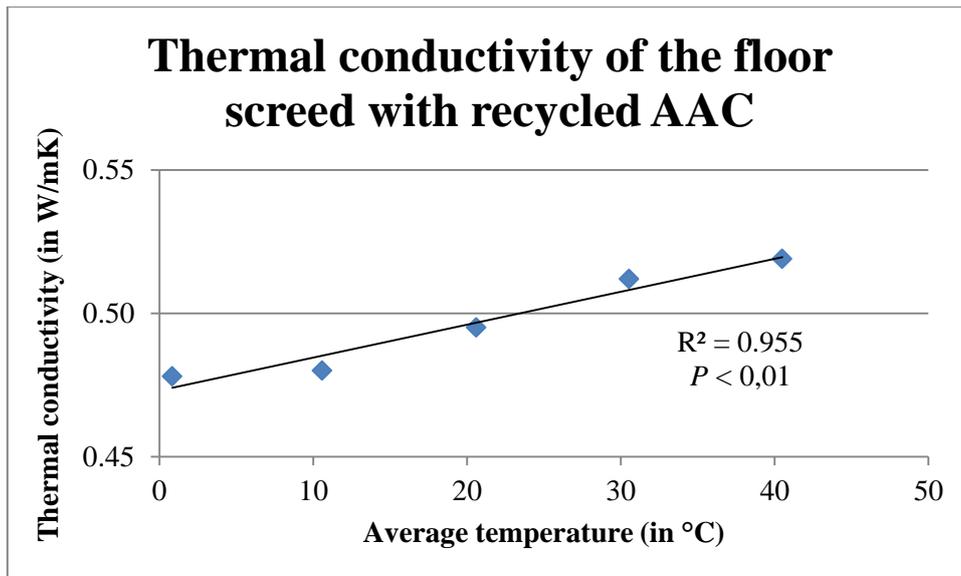
Figure 4. Thermal conductivity of the floor screed sample (ρ = 1400 kg/m³) with recycled AAC.

The use of this floor screed product is demonstrated in a case study in the Port of Antwerp (Figure).

Figure 4. Installed floor screed with recycled AAC in a case study in the Port of Antwerp.

4.   CONCLUSIONS

A new recycling route for AAC waste was developed. The main problem for the recycling of AAC is the presence of leachable sulfates. These sulfates were immobilized by the formation of ettringite by combination with Portland cement. Critical requirements for this immobilization are sufficiently alkaline conditions (>12.3) and the presence of sufficient Portland clinker aluminate (C_3A) to react with the sulfates contained in the AAC.

To reach a sufficiently high alkalinity in the developed cement stabilized sand products, the use of CEM I is crucial. The use of blended cements results in a lower leachate pH (<12) and a rise in sulfate leaching.

The developed products contained enough reactive aluminate to immobilize the available sulfate. However, when AAC waste was contaminated with gypsum particles, local hotspots of leachable sulfates created a depletion in reactive aluminium. This resulted in a strong increase in sulfate leaching.

The developed products meet the compressive strength requirements of the intended application. Furthermore, the use of recycled AAC can have a positive effect on the heat insulating capacities of constructions.

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