

Effects of carbonation and leaching on porosity in cement-bound waste

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Abstract

Porosity is possibly an important parameter with respect to leaching of constituents from cement monoliths. During its lifetime, the pore structure of cementitious matrices changes due to carbonation and leaching. This paper discusses the effects of both accelerated carbonation and continuous leaching on the porosity, and, conversely, how porosity affects leaching properties. Two sample types are investigated: a mortar with MSWI-bottom ash substituting the sand fraction and a cement paste with 30 wt% of the cement substituted by a flue gas cleaning residue. The samples have been intensively carbonated in a 20% CO₂ atmosphere for up to 60 days and were subsequently leached. The porosity was investigated by mercury intrusion porosimetry.

Accelerated carbonation decreases total porosity by 12% in the case of 60 days of treatment of bottom ash mortars, whereas continuous leaching during 225 days increases it by 16%. Both carbonation and leaching decrease the amount of smaller capillary pores.

Carbonation decreases both porosity and pH. Decreasing porosity diminishes leaching of sodium and potassium, while the decrease in pH increases leaching. However, the former process dominates the latter, resulting in a net decreasing effect of carbonation on the release of sodium and potassium from these cement matrices.

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1. Introduction

Leaching of heavy metals from wastes is a major concern for waste management. Depending on the heavy metal concentration and the physico-chemical characteristics of the waste, landfill disposal or reuse as construction material may be viable management options (IAWG, 1997; Sabbas et al., 2003). In both treatment options, the addition of cement and water in order to form a monolithic product is a well-known technique (Conner, 1990; IAWG, 1997). For landfill disposal the goal is to decrease heavy metal solubility and decrease permeability and geometric surface area to limit transport of contaminants to the environment. Increasing material strength is an additional requirement when recycling materials.

Total porosity and pore size distribution affect both the transport of metals to the environment and the strength of

the monolith and are in this respect important parameters to monitor (Bishop et al., 1992; Kumar and Bhattacharjee, 2003). The pore system in well compacted cement-based materials consists of two types of pores (Bishop et al., 1992; Garboczi and Bentz, 1996; Hewlett, 1998; Kumar and Bhattacharjee, 2003): (a) gel pores, which are micropores with pore radii smaller than 0.01 μm and are included in the volume occupied by calcium silicate hydrate (CSH) and (b) capillary pores, which are mesopores with average radii ranging from 0.01 to 10 μm. Capillary pores are the remnants of water-filled space during hydration and are responsible for the reduction in strength and elasticity of the cement monolith (Kumar and Bhattacharjee, 2003). The exact threshold between gel and capillary pores is rather arbitrary (Hewlett, 1998). In mortar or concrete a third type of pores, macropores, exists because aggregate particles are enclosed by cement particles with an interfacial transition zone occurring between cement and aggregate. The thickness of this interface is considered to be 30–60 μm and is thus included in the macropores (Hewlett, 1998). Macropores can also occur in cement paste due to

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inadequate compaction and may also affect strength (Kumar and Bhattacharjee, 2003).

Once the monolith has been produced, leaching and ageing, and more specifically carbonation, start changing the original characteristics of the material. In nature, carbonation and leaching occur intermittently and alternately during the lifecycle of a monolith.

Carbonation involves the chemical reaction of cement hydration products (e.g., portlandite, $\text{Ca}(\text{OH})_2$, and CSH) with carbon dioxide from the atmosphere in the presence of sufficient moisture leading to formation of calcite, CaCO_3 (Bin Shafique et al., 1998; Garrabrants et al., 2004; Van Gerven et al., 2004). This reaction lowers the pH in the pore water of the monolith from pH 13 to ultimately about pH 8. Heavy metals, originally present as hydroxides in the matrix, are progressively converted to carbonates, thus changing their solubility. In addition, matrix porosity can change. The molar volume of the calcite mineral is 11% larger than that of the portlandite mineral (Mollah et al., 1993; Gervais et al., 2004). An increase in mineral volume within the physical boundaries of the monolith decreases the pore volume, provided the strains accompanying this reaction do not cause microcracking of the monolith structure. Progressive carbonation eventually leads to depletion of portlandite and subsequently decalcification of CSH to provide new portlandite for the carbonation reaction (Gervais et al., 2004). This decomposition of CSH is reported to increase (Gervais et al., 2004) or decrease (Ngala and Page, 1997) porosity. Pore size distribution has also been shown to change by carbonation, possibly due to decalcification of CSH (Ngala and Page, 1997), with a decreasing volume of small pores ($<0.03 \mu\text{m}$) and consequently increasing proportion of capillary pores ($>0.03 \mu\text{m}$).

Leaching is a process, in which constituents in the solid phase are transferred to the liquid phase in contact with it, followed by transport of these constituents from the pore liquid out of the monolith into the surrounding leachant, mainly by a diffusion process. The rate of leaching of metals from porous matrices is dependent on, among others, its porosity (Bishop et al., 1992). The higher the porosity or the more pores are connected, the higher the effective diffusivity of the constituents (Houst and Wittmann, 1994; Garboczi and Bentz, 1996). Closed pores, i.e., pores not connected with the outer atmosphere, do not come into contact with the leachant (Bishop et al., 1992) and are thus not relevant with respect to leaching. It has also been reported that gel pores exhibit a conductance that is 400 times lower than that of capillary pores (Bentz and Garboczi, 1992), indicating they present a negligible contribution to leaching. Other factors affecting the rate and amount of leaching include the type and amount of the constituent investigated, the alkalinity of the matrix, and the chemical properties of the pore water and leachant (pH, E_h , ionic strength, etc.). Some of these parameters are further explored in a related paper (Van Gerven et al., 2006).

Although porosity influences the rate of leaching, leaching itself may also affect porosity. When leaching occurs, portlandite dissolves and new or larger pores develop (Garboczi and Bentz, 1996). Total pore volume, as well as pore size, increases, thus increasing the relative amount of capillary pores. Acid leaching for 90 days was reported (Bishop et al., 1992) to increase the percentage of large capillary pores ($>6 \mu\text{m}$) relative to total pore volume from below 5% to above 23%, while the percentage of small pores ($0.007\text{--}0.036 \mu\text{m}$) decreased by 25% or more. Leaching with deionized water affected porosity much less.

Previous papers have investigated the effect of carbonation or leaching on porosity. Both effects, however, have never been analyzed on the same specimen. This paper will, therefore, focus on the effect of both carbonation and leaching on the porosity of a monolith. The aim is to compare the importance of both effects. Moreover, the effect of carbonation on leaching is split into a porosity component and a pH component by bringing together results from this paper with data presented in a related paper (Van Gerven et al., 2006).

Two cement-based materials were produced using typical residues from the municipal solid waste incineration industry: (i) a bottom ash replacing the sand fraction in mortar and (ii) a flue gas cleaning residue substituting for a portion of the cement in cement paste. These waste residues were chosen because they represent two waste streams with distinctly different properties, but both containing heavy metals (Sabbas et al., 2003). Bottom ash is a coarse material consisting of mainly silica, alumina, iron and calcium oxides, as well as unburned organic matter. Bottom ash contains only a small amount of readily leachable salts, the amount of which is even lower in the investigated material than in untreated bottom ash due to the wet-sieving treatment. Flue gas cleaning residue is finer material including the reaction products and excess reagent of the air pollution control process, thus often containing a high proportion of calcium (hydr)oxide and chloride salts. Samples from the two sample sets were carbonated for 14, 30 or 60 days and were compared to samples that were not carbonated. Different types of samples were also leached during 225 days and compared to unleached samples. Porosity and pore size distribution is investigated by means of mercury intrusion porosimetry (MIP).

2. Materials and methods

2.1. Production of monoliths

Residues from a municipal solid waste incinerator (MSWI) were incorporated in cement matrices. One residue was a 0–2 mm fraction of a bottom ash that underwent a wet-sieving treatment as described in a previous paper (Van Gerven et al., 2005), while the other was a flue gas cleaning residue originating from a scrubber with semi-dry injection of lime (Geysen et al., 2004).

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