

Leaching behaviour of solidified MSWI fly ash and APC residues: assessment of two different treatment methods.

Peter Nielsen, Veronika Dutré & Roland Dreesen

VITO, Flemish Institute for Technological Research, Boeretang 200, B-2400 Mol, Belgium

Fly ashes and air pollution control (APC) residues produced from the Incineration of Municipal Solid Wastes (MSWI) require to be treated before being landfilled in a waste disposal site. Two disposal sites, with different MSWI residue treatment methods have been investigated for their environmental impact. At disposal site 1 the MSWI residues were solidified with portland cement, at disposal site 2 with a combined extraction/solidification process, called Solatech[®]. Monolithic samples were collected from the top, middle and bottom of both disposal sites.

The environmental impact has been assessed using leaching tests complemented with microscopic techniques, such as optical microscopy (petrography), and mineral phase identification using X-ray diffraction, Energy and Wavelength Dispersive Spectroscopy. The microscopic characterization is also very helpful in assessing the potential long-term environmental impact of the material. The leaching behaviour has been assessed using the Dutch standard extraction test (NEN 7341) for assessment of maximum availability, the Dutch standard leaching test for monolithic materials (NEN 7345) and pH static tests. The eluates of the leaching experiments were analysed for As, Cd, Cr, Hg, Mo, Pb, Zn, Ca, K, Na, Cl, SO₄, and pH. The concentrations of As, Cd, Cr, and Hg were below the respective detection limits for most of the 8 eluates of the diffusion test (NEN 7345), and this for both treatment methods at the 2 disposal sites.

Here, we have focused on the leaching of Pb and Zn because the leaching concentrations are above the detection limits for all 8 fractions of the leaching test for at least one of the two disposal sites, and because they demonstrate remarkable differences between the two treatment methods. In table 1 and 2 the total concentration and leachate concentrations are represented for a selected number of elements. The leaching behaviour of Pb and Zn is characterized by diffusion followed, at the end of the experiment, by apparent depletion. The Pb concentrations in the eluates of the Solatech[®] samples are nearly all below the detection limits.

Table 1: Total concentration and leaching data from the MSWI -residues solidified with cement.

| Element | Total concentration (mg/kg) | Max. availability (NEN7341) (mg/kg) | Diffusion Leaching (NEN7345) (mg/m ²) |
|-----------------|-----------------------------|-------------------------------------|---|
| Pb | 425 - 2750 | 194 - 1032 | 18 - 112 |
| Zn | 1100 - 4640 | 1080 - 4505 | 7.3 - 13.0 |
| K | 612 - 9221 | 763 - 9767 | 5980 - 77933 |
| Na | 612 - 4635 | 563 - 4304 | 5559 - 49878 |
| Cl | not measured | 12968 - 54565 | 141812 - 749436 |
| SO ₄ | not measured | 25711 - 40177 | 362 - 906 |

Table 2: Total concentration and leaching data from the MSWI-residues solidified with the Solatech® process

| Element | Total concentration (mg/kg) | Max. availability (NEN7431) (mg/kg) | Diffusion leaching (NEN7345) (mg/m ²) |
|-----------------|-----------------------------|-------------------------------------|---|
| Pb | 1060 - 1690 | 51 - 257 | < 3 |
| Zn | 2380 - 5180 | 1738 - 3737 | 2.4 - 4.3 |
| K | 583 - 2640 | 762 - 2672 | 1862 - 8464 |
| Na | 1920 - 4140 | 1895 - 4161 | 8841 - 13314 |
| Cl | 47900 - 70700 | 2721 - 17962 | 33663 - 49058 |
| SO ₄ | not measured | 28843 - 55767 | 3582 - 3861 |

X-ray diffraction showed that in the cement solidified samples chloride, sodium and potassium are present as soluble salts of halite and sylvite. These salts are not present in the Solatech® samples. Here, most soluble salts were removed during the extraction process. The availability of sulphates in both disposal sites is comparable, yet significantly more sulphate is leached from the Solatech® samples. The pH static tests show that the leaching of sulphates is strongly pH dependant (Fig 1a). For some of the metals, such as Pb the pH static tests also show strong pH dependence (Fig. 1b). A similar but less pronounced pH dependence was observed for Zn.

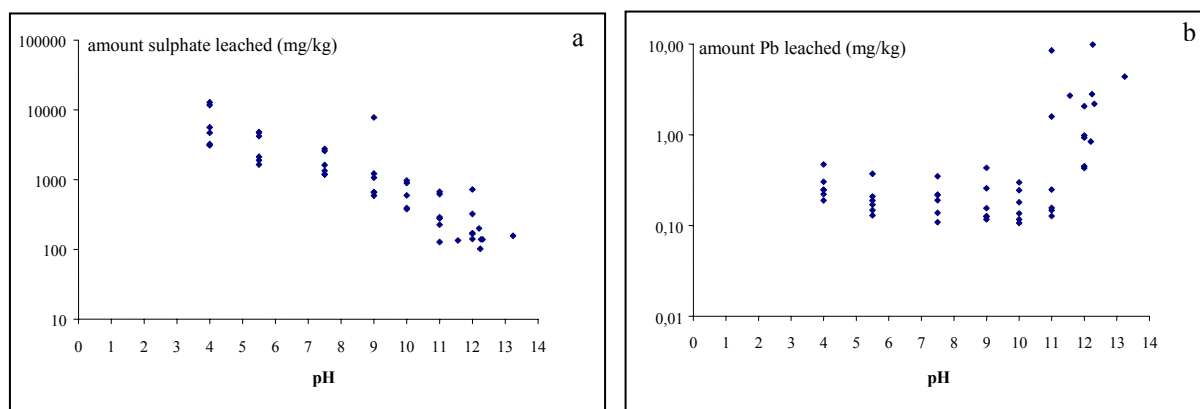


Figure 1: pH static tests on samples from the cement solidified landfill. Amount of Pb (left) and sulphate (right) leached (mg/kg) as a function of pH.

The petrographic characterization and X-ray diffraction data showed that the degree of carbonation differs significantly between the two landfills. Especially, the samples treated with the Solatech® process are carbonated. Here, calcite is present as microcrystalline patches, and as conspicuous fibro-radial carbonate crusts approximately 60 micron thick. X-ray diffraction data confirmed that calcite was one, if not the most prominent mineral in the Solatech® samples. Other minerals detected in large quantities are the hydrated salts ettringite and hydrocalumite, also known as Friedel's salt.

Within the cemented samples carbonation of the matrix is weak or even lacking as suggested by the abundance of portlandite. Carbonation is only pronounced in the samples from the top of the monolith. In the cemented samples portlandite, calcite, hydrocalumite and ettringite were the main minerals.

This difference in degree of carbonation is also reflected in the pH of the eluates of the NEN7345 diffusion test. The eluates of the Solatech® samples are characterized by a pH of 11.0 to 11.9, while leaching of the cement solidified samples resulted in a pH of 12.2 to 13.1. Carbonation is a process that results from the reaction of $\text{Ca}(\text{OH})_2$ in cement with atmospheric CO_2 . Carbonation increases with time. Here, however the cemented samples were exposed to atmospheric conditions for approximately 10 years, while the strongly carbonated Solatech® samples only have been exposed to atmospheric CO_2 for about 5 years, so the increased carbonation of the Solatech® samples can not be attributed to longer exposure times. Carbonation must have been enhanced by the Solatech® process.

The differences in leachate concentrations of Pb, Zn, and other elements can be largely explained by the more pronounced carbonation of the Solatech® samples. The heavy metals from the MSWI fly ash and APC residues are incorporated or adsorbed to similar phases in the two disposal sites. Apparently, the extraction that occurs during the Solatech® process mainly removes highly soluble salts of potassium and sodium chloride. The difference in leachate concentrations can thus not be attributed to speciation of the heavy metals or to the removal of highly soluble mineral phases. The pH static tests demonstrate however that a lowering of the pH due to carbonation may influence the leachability of the heavy metals from the solidified MSWI fly ashes and APC residues.

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