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Robustness validation of two harmonized European leaching tests for assessment of the leaching of construction products, including waste-based construction materials

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Abstract

Two leaching tests, a tank leaching test (TS-2) and a percolation test (TS-3), developed by CEN/TC 351 for use in assessing environmental properties in relation to CE marking of construction products, are undergoing robustness validation as part of the standardisation procedure. This paper provides an overview of the activities included in the robustness validation programme.

Keywords: Tank leaching test; Percolation test; Robustness validation; Construction products.

1 Introduction

The Construction Products Directive (CPD) addresses health and environment aspects of the service life (will be extended to include the entire lifecycle when the on-going replacement of the CPD by the Construction Products Regulation (CPR) is fully implemented), including the potential release of "dangerous" substances to soil and water, in relation to CE marking. The development of appropriate standards for assessment of the release to soil and water has been taken up by the European standardisation committee, CEN/TC 351: "Construction products: Assessment of the release of dangerous substances" where a horizontal approach to testing is promoted to avoid multiplication of test protocols per sector. The test standards will subsequently have to be adopted by the appropriate CEN product standardisation committees. Two technical specification, TS-2 and TS-3 have been developed by CEN/TC 351, largely based on adjustment of existing technical specifications produced for waste materials by CEN/TC 292: Characterisation of waste. TS-2: "Generic horizontal dynamic surface leaching test (DSLT) for determination of surface dependent release of substances from monolithic or plate-like or sheet-like construction products" is a tank leaching test, and TS-3: "Generic horizontal up-flow percolation test for determination of the release from granular construction products" is a column leaching test. In order to become a proper standard (EN), a technical specification must undergo validation. The first stage of a validation is the robustness testing where the sensitivity of the TS to changes in test conditions is determined. Based on the results, the TS may be modified prior to the next stage of the validation, the intercalibration, where the accuracy and precision of the method is determined.

Funded by the EU Commission and managed by CEN/TC 351 and NEN (the Dutch Standardisation Organisation), a consortium consisting of 7 European organisations has undertaken the robustness validation of TS-2 and TS-3. The project started in April 2011 and is scheduled to be completed in 2012. This extended abstract describes the tests and robustness validation programme. The project will not be entirely finalised before the WASCON 2012 conference, but some examples of results will be presented then.

2 Methods

2.1 The tank leaching test

The tank leach test CEN/TC351/TS-2: "Generic horizontal dynamic surface leaching test (DSLT) for determination of surface dependant release of substances from monolithic or plate-like or sheet-like construction products" provides different exposure configuration options, depending on the nature and shape of the product to be tested (monolith, sheet, plate). In its most common configuration, the specimen (minimum size in any dimension = 4 cm) is subjected to leaching with periodically renewed demineralised water at 22 °C in a closed tank with minimum requirements on the distances to the walls and bottom of the test vessel and to the surface of the water. The default liquid-to-surface area ratio (L/A) – the ratio of the volume of water added at each renewal and the surface area of the test specimen - is 8 cm^3/cm^2. The leaching solution is renewed after 0.08, 1, 2.25, 8, 14, 15, 28, 36 days. The pH, electrical conductivity (EC) and, optionally, Eh are measured in all eluate fractions which are subsequently filtered (0.45 µm) and subjected to chemical analysis. A special variation of the tank leaching test incorporated into CEN/TC351/TS-2 is the compacted granular tank test (based on the Dutch Standard NEN 7347) where the granular material to be tested is placed in a beaker in the tank with the surface exposed to the water. The test is primarily aimed at the determination of the release of inorganic substances, but may, with proper choice of test equipment and use of centrifugation instead of filtration, also be applied to determination of the release of non-volatile organic substances (e.g. PAHs).

Results are reported as a flux (e.g. $mg/m^2/day$) as a function of time or as leached amounts per unit surface area (e.g. mg/m^2) as a function of time. A slope of -0.5 or 0.5 in a log-log plot, respectively, indicates that the controlling release mechanism is likely to be diffusion.

2.2 The up-flow percolation test

The percolation or column leaching test CEN/TC351/TS-3: "Generic horizontal up-flow percolation test for determination of the release of substances from granular construction products is performed on granular or size reduced material (95% < 4 mm). It is based on the Technical Specification CEN/TS 14405 (2004), which was developed by CEN/TC 292 for testing of granular waste, and ISO 21268-3 developed for testing of soil. In this percolation test, the granular material is placed in a column of 5 or 10 cm diameter (the wide column is used if more than 5 % - but less than 20 % - of the material has a particle size exceeding 10 mm). The filling height of the material is 30 cm. The column is placed in an upright position, saturated with leachant (demineralised water) and left to pre-equilibrate for at least 48 hours. Demineralised water is then passed through the column in up-flow at 22 °C at a flow rate of 15 cm/24 hours (calculated for an empty column). 7 eluate fractions are collected within the range of L/S = 0.1-10 l/kg (L/S = 0-0.1, 0.1-0.2, 0.2-0.5, 0.5-1.0, 1.0-2.0, 2.0-5.0 and 5.0-10.0 l/kg). Each fraction is submitted to measurement of pH, electrical conductivity and, optionally, redox potential, and subsequently filtered (0.45 µm) and subjected to chemical analysis. The test is primarily aimed at the determination of the release of inorganic substances, but may, with proper choice of test equipment and use of centrifugation instead of filtration, also be applied to determination of the release of non-volatile organic substances (e.g. PAHs).

Results are reported as concentrations (mg/l) or accumulated leached amounts per unit mass of material tested (mg/kg) as a function of the liquid-to-solid (L/S) ration (l/kg). A constant concentration level or a slope of 1 for the accumulated release as a function of L/S is an indication of solubility controlled release.

3 Materials

The virgin and recycled construction products that have been tested in the robustness validation programme are listed in Table 1. Pictures of some of the materials are shown in Figure 1. Most of the materials were supplied by members of the various CEN Product Technical Committees. All the materials were delivered to BAM where they were pre-treated to the extent necessary (e.g. to get the required particle size distributions of granular materials). The total content of a wide range of inorganic substances were determined, and the physical properties such as e.g. particle size distribution of granular materials were also determined. The materials were then pre-tested for leaching properties for 24 hours (at L/A = 8 cm^3/cm^2 for monolithic materials and at L/S = 2 l/kg for crushed granular materials) before they were distributed to the different consortium members for robustness validation.

Table 1. Overview of the materials tested in the robustness validation programme

Monolithic, sheet- or plate-like materials used in tank leaching test (TS-2)	**Granular or granulated materials used in up-flow percolation test (TS-3)**
Clay masonry, CMA	Phosphorous slag, PSL
Steel slag (armour stone), MSS	Masonry, MAS
Autoclave aerated concrete, AAC	Steel slag, GSS
Roofing felt, ROF	Cement stabilised road-base material, CRB
External renders with organic binders, EXR	Natural aggregates, NAG
Tiles, TIL	Recycled concrete, RCC
Cement stabilised coal fly ash, CSC	Coal fly ash, CFA
Treated wood, TRW	Expanded clay, ECL
Timber structures, TIS	Reclaimed asphalt, RAG
Bricks made from construction & demolition waste, CDWC	Porous asphalt, PAS
Natural stone, NST	

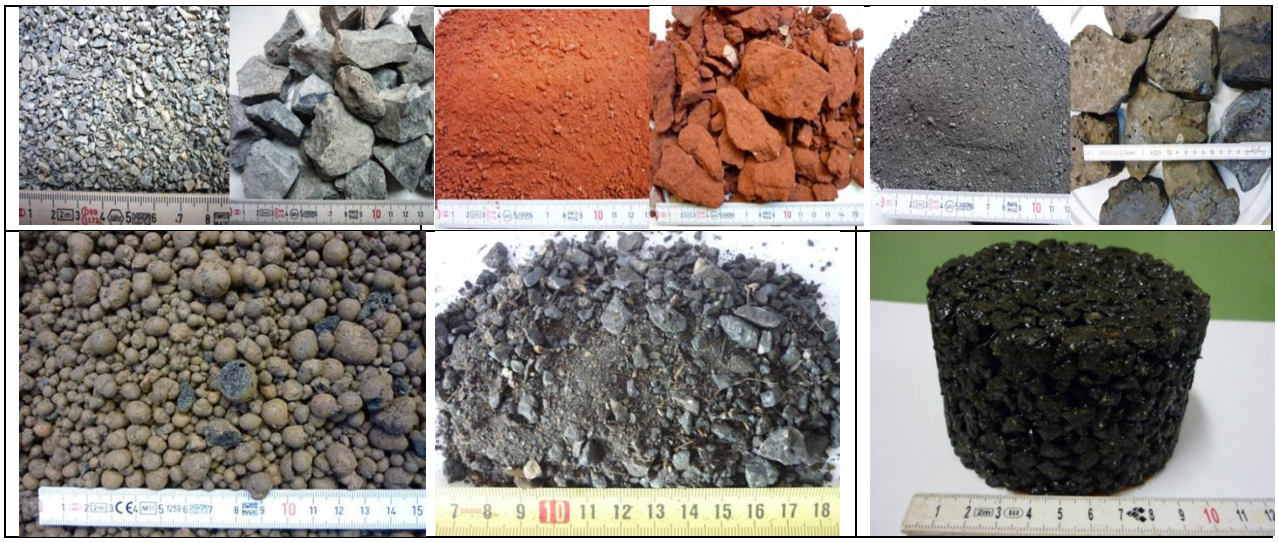

Figure 1. Some of the materials tested. Upper row from left to right: Phosphorous slag, masonry, steel slag. Lower row from left to right: Expanded clay, reclaimed asphalt, porous asphalt. Photo: BAM.

4 Conditions tested

The conditions tested for the tank leaching test, TS-2, were the liquid volume to surface area (L/A), the leachant or water renewal times (comparison of the default values for TS-2 with the renewal times of the widely use Dutch standard tank leaching test, NEN 7575:2002), and temperature. In addition, the repeatability of the test was assessed using two materials, CDWC and NST. The eluates from CMA; MSS, AAC, TIL, ROF, CSC, CDWC and NST have been/will be analysed for sulphate, chloride, fluoride, bromide, Ca, Mg, Si, Na, K, Al, P, Fe, Mn, As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Se, Sn, Sr, V, Zn, and in some cases also DOC. The eluates from EXR, TRW and TIS have been analysed for biocides, and the eluates from ROF will be analysed for PAHs. An overview of the conditions tested for TS-2 is shown in Table 2.

Table 2. Conditions tested for TS-2

Test condition	**Standard/default**	**Variation of conditions**	**Materials tested**
Liquid volume to surface area (L/A)	8 cm^3/cm^2	2 cm^3/cm^2, 5 cm^3/cm^2, 8 cm^3/cm^2, 9 cm^3/cm^2	CMA, MSS, AAC, ROF, EXR, TRW
Water renewal times	2hrs, 1d, 2.25d, 8d, 14d, 15d, 28d, 36d	Standard and 6hrs, 1d, 2.25d, 4d, 9d, 16d, 36d, 64d	CMA, MSS, TIL
Temperature	22 °C	10 °C, 17 °C, 22 °C, 27 °C	CMA, CSC, TIS

The conditions tested for the up-flow percolation test, TS-3, were the particle size distribution (which together with the flow rate may influence the degree of local equilibrium achieved in the test), the pre-equilibrium time, the temperature, and the flow rate. The eluates from PSL, MAS, GSS, CRB, NAG, RCC, CFA and EXL have been/will be analysed for sulphate, chloride, fluoride, bromide, Ca, Mg, Si, Na, K, Al, P, Fe, Mn, As, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Se, Sn, Sr, V, Zn, and in some cases also DOC. The eluates from RAC, RCC, and PAS have been/will be analysed for PAHs and in some cases also phenols. An overview of the conditions tested for TS-3 is shown in Table 3.

Table 3. Conditions tested for TS-3

Test condition	**Standard/default**	**Variation of conditions**	**Materials tested**
Grain size distribution	95 % below 4 mm	4 or 5 different grain size distributions ranging from 100 % < 4 mm to 100% 45-63 mm	PSL, MAS, GSS, CRB
Pre-equilibrium time	At least 48 hours	2 hours, 48 hours, 72 hours	GSS, NAG, RAG
Temperature	22 °C	10 °C, 17 °C, 22 °C, 27 °C	GSS, RCC, CFA, RAS
Flow rate	15 cm/day (empty column)	15 cm/day, 30 cm/day, 60 cm/day	GSS, CFA, ECL, RCC, PAS

Whereas the performance of the tests, TS-2 and TS-3, were distributed between six of the consortium partners, the subsequent chemical analysis of eluates for inorganic substances have been centralised and performed exclusively by the analytical chemical laboratory AZBA in Berlin. This was done in an attempt to minimise variability and errors originating from the chemical analysis of the eluates, since the robustness testing aims at assessing the sensitivity of the test results to deviations from the standard or default conditions and not at assessing the reproducibility of the methods. A major challenge has been the very low level of release of substances from several of the materials tested, since this increases the uncertainty and in some cases makes it difficult to determine the influence of varying conditions.

5 Final remarks

The results of the robustness validation will be used to confirm or suggest changes to the current default or standard conditions applied in TS-2 and TS-3, before the second stage of the validation process that will qualify these Technical Specifications to become full CEN standards – ENs – namely the round-robin test to determine reproducibility (between laboratory uncertainty) and repeatability (within laboratory uncertainty).