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# Concrete - A study of the characteristic leaching behaviour of hardened concrete for use in the natural environment

Zement - Eine Untersuchung der bezeichnenden Auslaugungseigenschaften von ausgehärtetem Beton zur Verwendung in natürlichen Umgebungen

This Technical Report was approved by CEN on 20 December 2010. It has been drawn up by the Technical Committee CEN/TC 51.

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## Foreword

This document (CEN/TR 16142:2011) has been prepared by Technical Committee CEN/TC 51 “Cement and building limes”, the secretariat of which is held by NBN.

The work which the report refers to was developed by CEN/TC51-TC104 JWG12/TG6 in the period 1994-1999.

JWG12/TG6 has continued to work on this subject and has produced the CEN/TR 15678:2008 which is complementary to this TR.

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. CEN [and/or CENELEC] shall not be held responsible for identifying any or all such patent rights.

# Summary

At the initiative of CEN/TC 51 (Cement and building limes) and CEN/TC 104 (Concrete and related products), a task group (TG 6) of TC 51/WG 12 was convened in order to accompany or follow research work being carried out within the EC research programme which has the objective of establishing the effects, if any, of concrete on the natural environment and the potential effects of cementitious materials on the quality of drinking water.

This Technical Report deals only with developments, as officially reported, by a consortium of Dutch/German Institutes, to the European Commission in EUR 17869 EN [1], leading to a performance test method for characterising the leaching behaviour of hardened concrete for use in contact with the natural environment.

NOTE The standardisation of test methods for the use of cementitious materials (possibly including concrete) in contact with drinking water, although not fundamentally different in principle, is being developed within an adhoc group of CEN/TC 164/WG 3 and will be reported elsewhere.

The protection of the natural environment and the public's health and safety are matters of major importance. Also of significant importance, however, is the efficient and sustainable use of natural and secondary materials/resources. Many of these may be used as constituents of concrete. The need to appropriately balance these two issues within the concept of sustainable construction, provided the motivation for the investigations considered in this Technical Report.

The prenormative research, underpinning this Technical Report, included a literature survey and three progressively staged interlaboratory studies (ILS). These led to the refinement of a characterisation (sequential leaching) test, comprising a tank (diffusion) test and a separate availability (maximum leaching) test. A single-extraction compliance test was not developed. A range of inorganic components/species (anionic and cationic) was targeted; some with a potential environmental significance, others of a more mechanistic relevance. Overall, a statistical and mechanistic evaluation of the results within EUR 17869 EN [1] and an environmental analysis undertaken in this Technical Report, has lead to the following conclusions.

- The leaching of major components/species, which have no environmental significance (e.g. Ca, Na, K and $SO_4$) from monolithic hardened concrete is diffusion controlled.

- Diffusion control could not be demonstrated, even after 14 days of leaching, for most environmentally relevant elements (e.g. As, Cd, Co and Cu) even from a relatively weak and porous concrete, since concentrations were at or below the limits of detections (DTL) of the sensitive instrumental techniques employed.

- Leached levels of components from monoliths are not related, in any simple or consistent manner, to the total concentrations of components present in concrete, and are, typically, orders of magnitude smaller.

- Leached levels of components from monolithic specimens are not related, in any simple or consistent manner, to amounts apparently available for leaching as indicated from a leaching test on finely ground concrete and the appropriateness of using such a test in attempting to characterise the leaching behaviour of hardened concrete is subject to continuing discussion.

- The concentration levels found in almost all leachates from the different tests were very low and often near the limit of the chemical analysis, indicating the good environmental quality of the concrete mixes tested.

- Concrete, containing a bituminous coal fly ash constituent specifically selected for its relatively high content of trace/heavy metals, and designed to represent a worst case within EN 206-1 [2] in terms of permeability, did not show significant leaching of trace/heavy metals. Most components were at concentrations below the analytical limits of detection.

- The anomalous leaching behaviour shown by specimens where the mixing water was spiked with aqueous solutions of the very mobile oxyanions of As, Cr, Cd and V, indicates that they were not representative of real concretes, as acknowledged by the research investigators.

- The disproportionate effect observed in the investigations, between the relatively large amounts of trace/heavy metals added as spikes to fresh concrete and apparently available for leaching, versus the minimal amounts actually leached, suggests that substituting standardized recycled or more marginal, but standardized, novel materials for the traditional constituents of concrete, would not significantly affect concrete's environmental compatibility.

- Subjecting the solid constituents of concrete to test, in isolation, either on the basis of their total elemental composition, or their response to an availability test, or their individual performance in a compliance test, will give no indication of their potential performance (either relative or absolute) when chemically and physically bound in hardened concrete.

- The characterisation leaching method, reproduced in Part II of this Technical Report, demonstrates such poor reproducibility (R approximately 76 % at 14d for trace metals As/Cd/Cr/V) that without much further investigation and development, it should not proceed to CEN/TS status or become the precursor to a draft compliance test or be used for any regulatory purpose.

- Concretes within the envelope of compositions permitted in the EN 206-1 [2] will have an insignificant impact upon the natural environment under conditions of natural exposure.



# 1 Introduction

Traditionally, hardened concrete has not been perceived to be a material which has contributed emissions adversely affecting the quality of the natural environment. Indeed, concrete construction in contact with the natural environment constitutes the bedrock of infrastructure and the built environment. Additionally, hardened concrete has never been shown to be responsible for any incidence of environmental pollution. Accordingly, within the range of traditional compositions used in the EU Member States, concrete's environmental service record can be taken to be unblemished.

Concrete, unlike most other construction materials, is an active material, its chemical and physical microstructure develops in a continuous process as it ages. These changes give rise to a densification of the matrix, with attendant reductions in porosity/permeability and a more efficient/effective binding of chemical species within the hydrate structures. It would be expected that concrete's leaching behaviour would also be subject to age-related changes and that this would be dissimilar to many other materials. Much research indicates that this is the case and so calls into question whether protocols, derived as in this study, from those developed for testing inert materials, are at all appropriate for concrete.

Concrete is, however, in common with other construction materials, subject to continual product development. Its compositional complexity is increasing, as constituent materials, formerly considered to be marginal, are either now in use or being considered for use. In the absence of quantitative information, some of the more marginal materials (e.g. where a total analysis reveals an apparently high heavy metal content) can give rise to concerns about their potential emission levels.

In addition, environmental regulatory activity, although at different points in the cycle in different EU Member States, is more and more subject to centralised direction via instruments such as EU Directives and mandates, and is generally increasing in its pace and scope.

Within this operational framework, standardised leaching tests, whether national or international, have taken a range of forms:

- characterisation;
- compliance;
- verification;

each of which can be used to evaluate the environmental performance/compatibility of hardened concrete, under different specified conditions using different assessment criteria. Characterisation leaching tests consist of an availability (granular or pulverized specimen) procedure and a sequential/periodic tank (monolithic specimen) procedure which together provide the means for discriminating between the several transport processes such as:

- dissolution;
- wash-off;
- diffusion;

and for predicting the rate of leaching and long term behaviour of a material.

In addition, physical characteristics such as tortuosity, which is a measure of the prolonged path along which leached components have to travel, can be calculated.

Compliance leaching tests consist of single extractions of short duration, generally without agitation, and which permit a direct comparison with regulatory limits for individual analytical components. Such tests use the prior output from characterisation tests to establish and optimise their parameters.

Verification leaching tests are essentially second order compliance tests, modified for operation in the field and used to identify/assess changes in established performance of batches of a material.

A final, and desirable, element in any authoritative procedure designed to evaluate environmental performance would be the preparation and maintenance of a certified reference material (CRM), for example, a certified reference concrete, preferably used within the context of a proficiency testing scheme (PTS), in order to monitor the performance of a laboratory and validate the accuracy of its procedures. In the case of concrete, the preparation and robust certification of a CRM is unlikely to be either attempted or to be feasible given the continuous changes in microstructure to be expected, with the likelihood of associated changes in its leaching characteristics.

Accepting that a concrete CRM is unlikely to be developed, then the preparation of a standard leachate, again for use within a PTS, would be the minimum expected for validation of laboratory performance.

It should be understood that the complete analysis of a concrete (or any of its constituents) in order to give a total elemental composition, is generally acknowledged to be of little environmental value and would be rarely undertaken in testing given that the greater proportion of most analytical components, whether environmentally significant or not, is known to be insoluble under naturally occurring exposure conditions.

# Part I

## 2 Scope of the study

### 2.1 Summary of three interlaboratory studies (ILS)

As reported in EUR 17869 EN [1], the Dutch/German Project Team (see 6.2) carried out its investigations in three stages, each stage leading to an interlaboratory study (ILS); the final ILS involved European participation much broader than the Project Team's membership.

The starting point for each stage was that a method of short duration, for the basic characterisation of leaching of inorganic components, should be developed and finally, validated.

A literature survey had indicated that the main transport process from monolithic concrete should be diffusion controlled and that a diffusion (tank) test, together with a maximum leachability (availability) test would be required in order to derive effective coefficients of diffusion, in order to be able to predict long-term leaching behaviour of concrete in the field

#### 2.1.1 First interlaboratory study and its evaluation (ILS #1)

##### 2.1.1.1 Objective

The objective of the first ILS was to assess the effect(s) on the leaching of a range of inorganic components from concrete, made to a single mix design, of varying the major parameters within several different, nationally and internationally (ISO) standardised, availability and tank leaching methods; the work being carried out in up to five laboratories.

##### 2.1.1.2 Concrete used in the first ILS

**Table 1 a — Mix design**

| Constituent | Content (kg/m$^3$) |
|---|---|
| Portland cement (Class 42,5) | 302 |
| Coal fly ash | 60,5 |
| Water (demineralised) | 181 |
| W/(c + 0,4f) [a] | 0,55 |
| Gravel 16 mm – 8 mm | 573 |
| Gravel 8 mm – 2 mm | 743 |
| Sand < 2 mm | 483 |

**Table 1 b — Miscellaneous**

| | |
|---|---|
| Strength at 28 days (mean) | 48 MPa (N/mm$^2$) |
| Specimen type | 100 mm cube |
| Curing regime<br>Fog room:<br>Climate chamber: | Demould : 1 day<br>20 °C/100 % RH : 6 days<br>20 °C/65 % RH : 56 days |
| Age at start of test | 69 days |

[a] k-value concept for additions in EN 206-1 [2]; where c = Portland cement content, k = 0,4 for Class 42,5 Portland and f = coal fly ash content