PD CEN/TR 16142:2011

BSI Standards Publication

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

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National foreword

This Published Document is the UK implementation of CEN/TR 16142:2011.

The UK participation in its preparation was entrusted to Technical Committee B/516/12, Sampling and testing.

A list of organizations represented on this committee can be obtained on request to its secretary.

ISBN 978 0 580 71784 0

ICS 91.100.10; 91.100.30

This Published Document was published under the authority of the Standards Policy and Strategy Committee on 31 March 2011.

Amendments issued since publication

Date	Text affected

TECHNICAL REPORT

RAPPORT TECHNIQUE

TECHNISCHER BERICHT

CEN/TR 16142

March 2011

ICS 91.100.30; 91.100.10

English Version

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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Contents

Page

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.

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 Fog room: Climate chamber:	Demould : 1 day 20 °C/100 % RH : 6 days 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

2.1.1.3 Test procedures and data analysis

For the analytical (instrumental) techniques used, the leaching tests investigated and the statistical/mechanistic data analysis, see 6.2.2, 6.2.3 and 6.2.4, respectively, in EUR 17689 EN [1].

2.1.1.4 Results

2.1.1.4.1 General

The results of the first ILS did not contribute to an understanding of the mechanisms involved in leaching or in establishing the influence of varying the test conditions on the performance of the tests. The reason for this shortcoming is given in [1] by the investigators. They observe that the concentration levels found in almost all leachates from the different tests were quite low and often near the limit of detection of the chemical analysis, indicating the good environmental quality of the concrete tested.

The mean results of the first ILS, as obtained from the test procedures which were selected for all the subsequent work, are given in Table 2 a for total contents, available contents and amounts leached over 7 d and over 64 d (calculated). In Table 2 b comparisons are presented, where data permits, between amounts leached at 7 d and 64 d and available amounts and total contents, in order to place the degree of leaching, for individual components, in perspective.

2.1.1.4.2 Environmental analysis and discussion of the results

a) Introduction

Although these investigations did not set out to assess the environmental performance/compatibility of the concretes under test, much useful information can be obtained from an environmental analysis of the results obtained at each stage in the development of the research.

In this first ILS, points of particular significance are reported and discussed, below:

- A large amount of potential data is missing from Tables 2 a and 2 b much of it for components generally agreed to have an environmental significance. These omissions usually result from concentrations being at or below the limits of detection after 7 d of cumulative leaching during the tank test.

- Of the components for which 7 d leaching data is presented, Cl (chloride ion), Na (sodium ion) and S (sulfate anion) are naturally present in almost all soils and groundwaters and have no environmental significance; accordingly they can be disregarded in any environmental analysis of the results.

- For a small number of components, for example B (boron) and Cd (cadmium) in Table 2 a, the mean results recorded for amounts available for leaching, as deduced from a test on pulverized concrete, are greatly in excess of the total amounts determined to be present in the concrete. The most likely causes of such anomalies are the analytical difficulties of working at concentrations near to the limits of detection of the analytical techniques. Such results, however, can readily give rise to false conclusions.

In this ILS this leaves As, (arsenic), Cd (cadmium), Co (cobalt), Cr (chromium), Cu (copper), Pb (lead), Tl (thallium) and Zn (zinc) for further environmental consideration.

Simple inspection of the data reported in Tables 2 a and 2 b indicates that responses to the different leaching regimes, i.e. availability test or tank test, are component-specific; this is consistent with the body of research evidence in this area. An analysis of the data for each individual component could tend to obscure rather than illuminate any general trends. The discussion section, which follows, addresses characteristics of the data which have a broad significance for the assessment of the environmental compatibility of concrete with its environment.

b) Discussion

From Tables 2 a and 2 b some reasonably general points can be observed.

- Only a small proportion of the total amount of a trace/heavy metal has been leached during the sequential extractions (over either 7 d or as extrapolated to 64 d). The total amount, therefore, does not give the potential amount for leaching and gives no indication of what proportion of a component would be leached under either standard or field exposure conditions.

Table 2 a — Mean results from first ILS

Component	Total content		Available		Leached 7 d (measured)		Leached 64 d (calculated)	
	(measured) (mg/kg)	(calculated) (mg/m^2)	Lmax (mg/kg)	Emax (mg/m^2)	L7d (mg/kg)	E7d (mg/m^2)	L64d (mg/kg)	E64d (mg/m^2)
Al	9 755	-	71,4	-	-	n.d.	-	-
As	5,15	202	0,15	5,9	< 0,1	< 1,67	0,13	< 5,1
B	1,5	-	20,9	-	-	DTL	-	-
Ba	174,3	-	50,3	-	-	DTL	-	-
Br	n.d.	-	< 2,07	-	-	DTL	-	-
Ca	52 954	-	52 427	-	-	n.d.	-	-
Cd	0,16	6,3	0,44	17,3	< 0,01	< 0,1	< 0,01	< 0,3
Cl	103,5	4 068	103	4 048	9,8	383,3	25,1	987,6
Co	4,89	192	1,17	46	< 0,01	< 0,8	< 0,1	< 2,5
Cr	483,2	18 990	2,51	98,6	0,15	< 5,8	0,5	< 17,7
Cu	10,6	417	3,56	139,9	< 0,01	1,2	< 0,1	< 3,36
F	99,00	-	62,7	-	-	DTL	-	-
Fe	6 776	-	413,3	-	-	n.d.	-	-
Hg	n.d.	-	<0,022	-	-	DTL	-	-
K	3 046	-	853	-	-	n.d.	-	-
Li	n.d.	-	4,76	-	-	DTL	-	-

Table 2 b — Comparisons from first ILS: % leached vs. available/total

Component	% leached (measured or calculated)				
	7 d (measured) vs. available	7 d (measured) vs. total	64 d (calculated) vs. available	64 d (calculated) vs. total	Available vs. total
Al	-	-	-	-	0,7
As	28,3	1,9	< 86,0	2,5	2,9
B	-	-	-	-	?
Ba	-	-	-	-	28,9
Br	-	-	-	-	-
Ca	-	-	-	-	99
Cd	0,5	1,6	1,6	4,8	100
Cl	9,5	9,5	24,4	24,4	100
Co	1,8	< 0,2	< 5,5	2	23,9
Cr	5,9	< 0,1	< 18,0	0,1	0,5
Cu	0,9	< 0,1	< 2,4	<1	33,6
F	-	-	-	-	63,3
Fe	-	-	-	-	6,1
Hg	-	-	-	-	-
K	-	-	-	-	28
Li	-	-	-	-	-

Mg	1 064	-	777,9	-	-	n.d.	-	-	-	-	-	-	73,1
Mn	101,8	-	47,4	-	-	DTL	-	-	-	-	-	-	46,6
Mo	6,2	-	0,55	-	-	DTL	-	-	-	-	-	-	8,9
Na	397,9	15 638	156,8	6 162	10,4	408,3	22	862,9	6,6	2,6	14	5,5	39,4
Ni	121,3	-	11,5	-	-	DTL	-	-	-	-	-	-	9,5
P	201,5	-	1,16	-	-	DTL	-	-	-	-	-	-	0,6
Pb	13,6	535	0,65	25,5	0,25	9,85	0,81	31,7	38,6	1,8	100	6	4,8
S [a]	1 217	47 828	1 341	52 701	5,7	222,3	-	-	0,4	0,5	-	-	100
Sb	< 1,96	-	0,2	-	-	DTL	-	-	-	-	-	-	10,2
Se	0,36	-	0,094	-	-	DTL	-	-	-	-	-	-	26,1
Si	371 224	-	3 093	-	-	DTL	-	-	-	-	-	-	0,8
Sn	n.d.	-	3	-	-	DTL	-	-	-	-	-	-	-
Ti	704,3	-	< 0,21	-	-	DTL	-	-	-	-	-	-	< 0,1
Tl	< 1,22	48	0,085	3,3	< 0,1	< 1,67	0,13	< 5,1	49,9	< 8	100	10,7	7
V	18	-	0,16	-	-	DTL	-	-	-	-	-	-	0,9
Zn	80,9	3 179	55	2 162	0,21	< 8,3	0,64	25,3	0,4	0,3	1,2	0,8	68

[a] S is sulfate anion (SO_4)

NOTE 1 Lmax, in mg/kg is the maximum amount available for leaching.

NOTE 2 Emax, in mg/m^{2,} is Lmax. × 39,3; 39,3 is a geometrical factor for the 100 mm cube specimens, used to convert mass specific leaching into an equivalent value per unit area.

NOTE 3 E7d, is the measured cumulative release in the tank test, in mg/m^2, (L7d is calculated from E7d by dividing it by 39,3)

NOTE 4 E64d is a calculated extrapolation using the 7 d data.

NOTE 5 DTL means at or below the limit of detection.

NOTE 6 The amount available is determined on a sample of finely ground concrete (90 % < 125 µm) extracted consecutively in demineralised water at pH 7 then at pH 4.

- Much larger proportions of the available amount of the trace/heavy metals have been leached during the sequential extractions as extrapolated to 64 d. The actual proportion is component-specific but in the case of As, Pb and Tl it constitutes almost all of the available quantity.

On the evidence above, the available amount can give a good indication of the potential amount for leaching for some components (As, Pb, Tl) but is much less informative for other trace/heavy metals, being a particularly poor indicator of the potential amount for leaching for Cd, Cu and Zn.

- It can also be observed that there is no single, simple relationship between the amount of a trace/heavy metal, defined as available for leaching, and the total quantity in the concrete. The amount available as a proportion of the total ranges from < 0,1 % for Ti to 100 % for Cd.

- Most importantly, in terms of the environmental impact of this particular, but reasonably typical, concrete, the absolute amounts of the trace/heavy metals which gave measurable concentrations when leached over 7 d (or as extrapolated to 64 d), were in the sub-ppm (< 1 mg/kg) range; as recorded in Table 2 a.

2.1.2 Second interlaboratory study and its evaluation (ILS #2)

2.1.2.1 Objective

The objective of the second ILS was to validate the appropriateness of the test protocols (availability and tank) selected from those investigated in the first ILS for characterising leaching from concrete.

In the first ILS, the leaching behaviour of some environmentally insignificant components e.g. chloride ion, had suggested that transport could be diffusion controlled. The validation exercise was designed to try to establish whether or not diffusion control also applied to trace/heavy metals; this could not be established from the results of the first study because leached levels were too low.

Concrete of a different mix design (see Table 3 a) was prepared in order to give, what was considered would be, a worst case, within the scope of EN 206-1 [2], in terms of its potential leaching performance. The design included the use of a bituminous coal fly ash, specifically selected for its relatively high content of trace/heavy metals, in order to increase the intrinsic level in the concrete, together with a relatively high water/binder ratio in order to give a more porous/permeable, and hence potentially leachable, microstructure.

2.1.2.2 Concrete used in the second ILS

Table 3 a — Mix design

Constituent	Content (kg/m^3)
Portland cement (Class 42,5)	200
Coal fly ash	100
Water (demineralised)	180
W/(c + 0,4f) [a]	0,75
Gravel 16 mm - 8 mm	546
Gravel 8 mm - 2 mm	727
Sand < 2 mm	546

Table 3 b — Miscellaneous

Strength at 28 days (mean)	29,5 MPa (N/mm^2)
Specimen type	100 mm cube
Curing regime Fog room: Climate chamber:	Demould: 1 day 20 °C/100 % RH : 6 days 20 °C/65 % RH : 42 days
Age at start of test	52 day

[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.

NOTE The mix design was chosen to represent a worst case concrete; high permeability (high water/binder ratio) using a coal fly ash having a heavy metal content at the higher end of the compositional range.

2.1.2.3 Leaching tests

See Part II to this Technical Report for the test methodology finally adopted.

NOTE Some of the leaching periods were increased over those used in the first ILS, such that the longest period of immersion increased from 7 d to 14 d.

2.1.2.4 Analytical (instrumental) techniques

See 7.2.2 in [1].

2.1.2.5 Results

2.1.2.5.1 General

The results of the second ILS, like the first ILS, could not contribute to an understanding of the mechanisms involved in the leaching of trace/heavy metals from concrete. The very low concentrations of As, Cr and Cu, actually leached precluded my assessment of the leaching behaviour. Even the more major components (i.e. Ca, K, Na and S), although at high enough leached concentrations, could not contribute to the understanding, for a variety of mechanistic reasons. The investigators again made a formal note of the implications of the results on the environmental performance of the test concrete for the likely performance of concrete in the field indicating that it would present no dangerous emission to the environment with respect to leaching.

The mean results of the second ILS are given in Table 4 a for total contents, available contents and amounts leached over 14 d and over 64 d (calculated). In Table 4 b comparisons are presented, where data permits, between amounts leached at 14 d and 64 d and available amounts and total contents.

Table 4 a — Mean results from second ILS

Component	Total content		Available		Leached 14 d (measured)		Leached 64 d (calculated)	
	(measured) (mg/kg)	(calculated) (mg/m^2)	Lmax (mg/kg)	Emax (mg/m^2)	L14d (mg/kg)	E14d (mg/m^2)	L64d (mg/kg)	E64d (mg/m^2)
As	22,0	865	< 0,9	< 35,4	< 0,1	1,2	-	n.d.
Ca	38 491	1 512 696	30 918	1 215 077	52,5	2 065	-	variable
Cl	26,4	1 038	73	2 869	17,5	687	-	variable
Cr	46,0	1 808	1,2	47,2	< 0,1	0,8	< 0,1	1,3
Cu	11,0	432	3,6	141,5	< 0,1	0,8	< 0,1	1,2
K	2 263	88 936	1 047	41 147	256	10 079	552	21 690
Na	365	14 345	199	7 821	26	1 023	-	n.d.
S [a]	864	33 955	1 516	59 579	3,7	145	5,7	225

[a] S is sulfate anion (SO_4)

NOTE 1 The values for total contents will be inaccurate for some components since they exclude any contributions from the aggregates. They have been derived in [1] from analyses of the binder constituents, only.

NOTE 2 Results for amounts leached at 14 d for several of the components, as reported by six laboratories in [1], were variable.

NOTE 3 Results for Cl and S should be treated with considerable caution since the mean available contents are reported in [1] as between x2 and x3 greater than the total contents.

Table 4 b — Comparisons from second ILS : % leached vs. available/total

% leached (measured or calculated)				
14 d (measured) vs. available	14 d (measured) vs. total	64 d (calculated) vs. available	64 d (calculated) vs. total	Available vs. total
3,4	0,1	-	-	4
0,2	0,1	-	-	80
24	66	-	-	100
1,7	< 0,1	2,8	< 0,1	2,5
0,6	0,2	0,8	0,3	33
24	11	53	24	46
13	7	-	-	55
0,2	0,4	0,3	0,7	100

2.1.2.5.2 Environmental analysis and discussion of the results

a) Introduction

As in the first ILS the investigators did not set out to assess the environmental implications of the data sets, as summarised in Tables 4 a and 4 b, for the second concrete mix design under test. Nevertheless, some points of environmental significance can be noted.

- Of the components for which 14 d leaching data is presented, Ca, Cl, K, Na and S are naturally present in almost all soils and groundwater and have no environmental significance; as in the analysis for the first ILS they can be disregarded.

This leaves As, Cr and Cu from amongst the components selected for study in the second ILS, for any environmental considerations.

b) Discussion

A comparison, of the limited data presented in Table 4 b (second ILS) with that of Table 2 b (first ILS) for As, Cr, and Cu, indicates that the two data sets are broadly similar for each component and that:

1. the relationships between, amount available vs. total, or amount leached vs. amount available, seem to be characteristic of particular components in these hardened concretes;

2. the mobility of the components in this matrix is very similar to that found in the different concrete used in the first ILS;

3. the second ILS has provided a useful and accurate replication of the first ILS, which supports the general points made in the discussion section to 2.1.1.4.2 of this Technical Report.

The concrete mix was chosen to provide what was considered to be the most leachable concrete within the scope of EN 206-1 but the amounts of trace/heavy metals (As, Cr and Cu) leached over 14 d, or extrapolated to 64 d, were each less than 0,1 ppm (< 0,1 mg/kg) as shown in Table 4 a.

Overall, the results showed that the concrete used in the second ILS was not suitable for a full-scale precision experiment since the leached levels were too low. A third interlaboratory study was undertaken using concrete which had been artificially enhanced by spiking the mix with aqueous solutions of heavy metal oxyanions of As, Cd, Cr and V.

2.1.3 Third interlaboratory study and its evaluation (ILS #3)

2.1.3.1 Rationale for proceeding to a third ILS

2.1.3.1.1 Mechanistic significance of the third ILS

The results from the first and second ILS, for trace/heavy metals were too low to establish the mechanism(s) controlling their leaching. Although they had shown that higher total contents, as bound species, did not necessarily lead to greater amounts leached, it was considered that higher levels of initially unbound species could lead to measurable leaching.

2.1.3.1.2 Environmental significance of a third ILS

The first and second ILS had shown that for concrete within the scope of EN 206-1, no significant leaching of trace/heavy metals occurred. As other concrete compositions, currently outside the scope of EN 206-1, may be used in the future and recycled materials could be incorporated, a study of spiked concrete could give some indication of worst case performance.

For the third ILS:

a) participation was widened (18 laboratories/10 Member States) to include laboratories outside the Project Team's membership (see table in 6.3);

b) an additional leachant, low mineralised water (see 8.1 in EUR 17689 EN [1]) was introduced for comparison with demineralised. In the service condition such a water is much more likely to correspond to a realistic worst case condition than would demineralised (i.e. distilled/deionised) water;

c) a standard leachate (see table in 8.4 in EUR 17689 EN [1]) was included in the precision experiment in order to determine the precision of the analyses and the performance of the participants.

2.1.3.2 Concrete and solution spikes used in the third ILS

Table 5 a — Mix design

Constituent	Content (kg/m^3)
Portland cement (Class 42,5)	270
Coal fly ash	60
Water (demineralised)	162
W/(c + 0,4f) [a]	0,55
Gravel 16mm - 8mm	657
Gravel 8mm - 2mm	564
Sand < 2mm	509

Table 5 b — Miscellaneous

Strength at 28 days (mean)	38,5 MPa (N/mm^2)
Specimen type	100 mm cube
Curing regime	Demould: 1 day
Fog room:	20 °C/100 % RH : 6 days
Climate chamber:	20 °C/65 % RH : 56 days
Age at start of test	75 day

[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

Table 5 c — Amount of component added to concrete as an aqueous spike

Substance	Component	Amount of component added to mixing water (g/l)	Amount of component added to concrete (kg/m^3)	Amount added expressed in ppm of concrete (mg/kg) [a]
$Na_2HAsO_4{\cdot}7H_2O$	As	2,50	0,41	170
$Cd(NO_3)_2{\cdot}4H_2O$	Cd	1,00	0,16	65
$K_2Cr_2O_7$	Cr	1,50	0,24	100
NH_4VO_3	V	2,00	0,32	130

a In the case of As, Cd and V, the amounts of component added are between 5 × and 100 × greater than are likely to be present naturally in a concrete. See Table 2 a of this Technical Report for a comparison.

2.1.3.3 Leaching tests

See Part II to this Technical Report for the test methodology finally adopted.

2.1.3.4 Analytical (instrumental techniques)

See 8.3 in EUR 17689 EN [1].

2.1.3.5 Results

2.1.3.5.1 General

Overall, the results from the third ILS were taken by most of the investigators to indicate that the leaching of components from hardened concrete is generally diffusion controlled and that no further modifications to the test procedures were necessary. However, a minority view was expressed that the work had not, in fact, demonstrated this sufficiently in the case of the trace/heavy metals. In particular, it was observed that if diffusion was the controlling mechanism for leaching of trace/heavy metals, any increase in their total contents should be accompanied by an increase in amounts leached. This had not been observed.

The effects of spiking the concrete were varied. In the case of Cr and V, although the leachable amounts increased, an artefact was introduced into the system (S-shaped leaching curve) probably reflecting the different ionic mobilities of components bound in the solids versus the same species introduced artificially via the aqueous phase; the artefact obscured rather than illuminated the controlling mechanism for leaching.

In the case of Cd, although the total amount had been increased, the leachable amount changed little from the levels found in the earlier ILSs.

In the case of As, only the concentration of the first leachate fraction was enhanced while the next four fractions showed in many cases a leaching behaviour controlled by diffusion.

The mean results of the third ILS and the precision estimates obtained are given in Table 6 a for the availability test and in Table 6 b for the diffusion leaching test for which demineralised water was used i.e. for the tests as described in Part II of this Technical Report.

NOTE 1 The results obtained using the low hardness mineralised water were not significantly different to those obtained using demineralised and have, therefore, been excluded from this report for simplicity.

NOTE 2 The results obtained from the standard leachate have also been excluded for simplicity, but they revealed that the participants were capable of carrying out the necessary analytical procedures.

2.1.3.5.2 Environmental analysis and discussion of results

a) Introduction

As acknowledged by the investigators [1], the anomalous results (S-shaped leaching curves) obtained for the spiked concretes show that the specimens in the third ILS were not representative of real concretes. Their leaching behaviour, in the case of the added trace/heavy metals, did not show the same pattern found for the two coal fly ash-containing concretes used in the earlier ILSs. Consequently, it is difficult to use the results in any convincing way in a discussion of the environmental compatibility of real concretes. If, however, the results were to be discarded entirely on the basis of being non-representative, then some interesting observations, with, possibly, predictive implications would be overlooked and in addition the precision estimates, obtained from the third ILS, which may have validity for some concretes under some conditions, would also be discarded.

b) Discussion

In the third ILS, soluble salts of As, Cr, Cd and V were added to fresh concrete at levels equal to or higher, [much higher in the case of As, Cd and V, (see Table 5 c)] than levels found, typically in the solid constituents. The principal effect of this on the analytical results has been to increase the quantities which appear to be potentially available for leaching. However, where the limited data in Table 2 a (first ILS) and Table 4 a (second ILS), permit a comparison with that in Table 6 b (third ILS), it can be seen that the effect on actual leaching of As, Cd, Cr or V, at 7 d or 14 d, is small and not in proportion to the level of spiking. The leached levels are slightly higher but still in the sub-ppm range (As and Cd) or less than 3 ppm (Cr and V). Such a disproportionate effect, between amount of trace/heavy metal added to fresh concrete and apparently available for leaching in the hardened state, versus the minimal amount 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. This, environmentally, is a most important consideration.

3 The experimental precision and its implications

3.1 Introduction

Precision estimates were obtained from the third ILS. In Tables 6 a, availability test, and 6 b, diffusion leaching protocol, precision estimates are presented individually and also pooled in three distinct ways in order for comparisons to be made. The pooled estimates are identified as M_1, M_2 and M_3. with the definitions for statistical terms being in conformity with ISO 5725 [3].

Table 6 a — Mean results and precision estimates for the availability test

Component	Mean		Repeatability coefficient of variation CV	Reproducibility coefficient of variation CV	Repeatability limit r	Reproducibility limit R
	(measured) (mg/kg)	(calculated) (mg/m^2)	(%)	(%)	(%)	(%)
As	9,7	381,2	24,2	64,2	67,8	179,8
Cl	779,7	30 642	27,6	104,9	77,3	293,7
Cr	75,7	2 975	11,9	23,3	23,8	65,2
K	1 820,0	71 526	6,4	16,7	17,9	46,8
Na	307,3	12 077	7,2	28,4	20,2	79,5
S [a]	1 507,0	59 225	3,0	18,8	8,4	52,6
V	20,6	810	14,4	29,7	40,3	83,2
M_1) Overall means			13,5	40,9	37	114
M_2) Means (excluding As/Cl) [b] [See reference (1)]			8,6	23,4	24	66
M_3) Means for As/Cr/V (i.e. trace metals)			16,8	39,1	47	109

[a] S is sulfate anion (SO_4)

[b] The results given in [1] for means, exclude As and Cl. Data have been excluded where components gave average concentrations < 10 × DTL.

Table 6 b — Mean results and precision estimates for the diffusion test using demineralised water

Component	Mean		Repeatability coefficient of variation CV 14 d	Reproducibility coefficient of variation CV 14 d	Repeatability limit r 14 d	Reproducibility limit R 14 d
	14 d (calculated) (mg/kg)	14 d (measured) (mg/m^2)	(%)	(%)	(%)	(%)
As	0,21	8,47	9,7	26,8	27,2	75,0
Cd	< 0,01	0,05	15,8	41,0	44,2	114,8
Cl	5,4	210,6	11,3	56,0	31,6	156,8
Cr	2,9	112,7	15,3	20,7	42,8	58,0
K	34,2	1 3442	11,9	21,7	33,3	60,8
Na	37,9	1 489,4	9,1	24,8	25,5	69,4
S [a]	25,8	1 012,1	17,1	17,4	47,9	48,7
V	2,0	78,3	7,6	19,6	21,3	54,9
M$_1$) Overall means			12,2	28,5	34	80
M$_2$) Means (excluding As/Cd/Cl) [b] [See reference (1)]			12,0	22,0	34	62
M$_3$) Means for As/Cd/Cr/V (i.e. trace metals)			12,1	27,0	34	76

NOTE The data for leaching, extrapolated by calculation to 64d, in [1], is too limited to be of value and has been omitted from this table.

[a] S is sulfate anion (SO_4)

[b] The results given in [1] for means, exclude As, Cd and Cl. Data have been excluded where components gave average concentrations < 10 × DTL.

3.2 Discussion of the precision estimates

Under M$_1$, the overall means for four precision estimates, S_r, S_R, r and R, are presented with all the data pooled over all the components.

Under M$_2$, the means have been pooled as presented in [1], where data for the components As and Cl in Table 6 a and As, Cd and Cl in Table 6 b have been excluded.

Under M$_3$, the means have been pooled for those components which would be considered to have an environmental significance; in the third ILS these are the components, As, Cd, Cr and V, added as spikes.

A broad examination of the data for precision estimates, which include procedural and analytical variability, in Tables 6 a and 6 b leads to the following conclusions.

- Inter-laboratory variability, i.e. reproducibility, is very high even when, as in M$_2$, justifiable exclusion of certain components has been introduced. In the case of, M$_3$, where only environmentally significant components are included, reproducibility is extremely poor.

- The poor reproducibility observed, in both the availability test and the leaching procedures, has implications for the use of the test methodology. Without significant improvement, by way of isolating, identifying and correcting the major sources of variability, the method could not be standardised as a reliable characterisation method in its current form.

- Unless the major factors contributing to the poor reproducibility can be identified and corrected the leaching protocol could not be used as the precursor to a compliance method given the implications that high inter-laboratory variability has upon evaluation of conformity with any specified limit. Conversely, if the poor reproducibility cannot be improved upon, its implications would have to be taken into account during the process of setting any official compliance criteria for determining the environmental acceptability of concrete.

- The intra-laboratory variability, i.e. the repeatability, is also high when compared to that expected from test methods for concrete. However, analyses of materials for trace/heavy metals generally exhibit poorer repeatability than for major elements since the relationship between variability and concentration level is non-linear. Accordingly, it would be unrealistic to expect any significant reduction in intra-laboratory variability.

4 Standardisation of the characterisation leaching method

4.1 Introduction

There are a number of routes and qualifying criteria established by CEN whereby a test method can progress to a full European Standard (EN). Frequently, an EN will have had as its predecessor, a CEN/TS. The CEN/TS may have been based on a modified draft of a national (or international) standard. Where no appropriate national (or international) standard exists for such an adoption an entirely new method can be proposed, investigated, and validated via a robustness investigation and precision trial before submission to a CEN Technical Committee for consideration for full standardisation (EN), preliminary standardisation (CEN/TS) or for official reporting as a CEN Technical Report (CEN/TR).

As a response to such a submission having been made to a joint working group of CEN/TC 51 and CEN/TC 104, by the co-ordinator of the Project Team which undertook the research described in this Technical Report, and as reported to the European Commission in [1], CEN has approved the TCs' decision to publish the work as a CEN/TR without any commitment to proceed to either a CEN/TS or EN, on the following grounds.

- The research has demonstrated that the amounts of environmentally significant components leached from hardened concrete, produced within the scope of EN 206-1 [2], are at or below the limits of detection of the very sensitive available instrumental techniques.

- By the same fact the research has demonstrated that hardened concrete, within the scope of EN 206-1 is an environmentally compatible material.

- By extension, from the previous fact, there is no requirement for either a characterisation or a compliance leaching test in order to validate the performance of concrete, within the scope of EN 206-1, for use in contact with the natural environment.

- In addition, the poor precision, particularly for inter-laboratory reproducibility, demonstrated in the research, for species which could be quantified, indicates that the method is not, yet, sufficiently robust to proceed even to CEN/TS status for use in future in evaluating non-traditional concretes as they may become accepted into construction and into EN 206-1.

- Finally, the full testing protocol requires specialist skills to be employed at every stage, has an extended duration (14 d) involving two leaching procedures and needs expensive analytical instrumentation in order to be able to assess the extremely low concentrations of trace/heavy metals which might be leached from hardened concrete.

4.2 Potential applications for the method

The leaching method described in Part II of this Technical Report is a characterisation method. Using it, traditional concretes have effectively been characterised as leaching-resistant. Non-traditional concretes, for example concretes containing either recycled or novel constituents, have not been so directly characterised, with respect to trace/heavy metals.

In time, such materials may become standardised as construction products for use in concrete, by way of inclusion in EN 206-1. At the application stage for inclusion, an initial type test using the method documented in Part II of this Technical Report, on the potential leaching behaviour of concrete containing the product, could be made mandatory. The major shortcoming with such an apparently simple philosophy is the absence of pan-European regulatory criteria which represent acceptable/unacceptable leaching performance for concrete.

In addition, if at some stage, appropriate criteria were to be established it is unlikely that they would be set in isolation from a reference test method; traditionally, for related applications e.g. the drinking water environment, such tests have been single extraction compliance tests, not sequential extraction characterisation tests as described in Part II of this Technical Report.

4.3 Necessary developments before any method can be applied

From the foregoing discussions it should be apparent that use of the test method in Part II of this Technical Report for any purpose, in the absence of a number of regulatory and technical developments would be unjustified. The developments necessary to produce a coherent framework for its use can be tentatively advanced as follows.

a) The identification and correction of the major factors contributing to the poor reproducibility in the characterisation test, documented in Part II of this Technical Report, would need to be undertaken within CEN.

b) The conversion of the characterisation test into an appropriately precise European Standard compliance leaching test for hardened concrete, would need to be undertaken within CEN.

c) The setting of appropriate compliance criteria for leaching of potentially dangerous substances from hardened concrete could be undertaken either within CEN in consultation with the appropriate European authorities, or by the appropriate European authorities and subsequently documented in EN 206-1 as requirements in the initial type testing of concretes containing recycled/novel constituents.

5 Conclusions

On the basis of the scientific evidence presented in EUR 17869 EN [1] and its environmental interpretation undertaken within this Technical Report the following conclusions can be drawn.

5.1 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.

5.2 Diffusion control could not be demonstrated, even after 14 d 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.

5.3 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.

5.4 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.

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

5.6 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.

5.7 The anomalous leaching behaviour (S-shaped leaching curves) shown by specimens in ILS #3, 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.

5.8 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.

5.9 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.

5.10 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 yet proceed to CEN/TS status or become the precursor to a draft compliance test or be used for any regulatory purpose.

5.11 The results of ILS #1 and ILS #2, where concrete types were used that resemble construction concretes, indicate that concretes within the envelope of compositions permitted in EN 206-1 [2] will have an insignificant impact upon the natural environment under conditions of natural exposure.

6 Appendices

6.1 Members of the Project Team that undertook the investigations

A M H van der Veen (co-ordinator)	NMi Van Swinden Laboratorium B.V., The Netherlands
D A G Nater	NMi Van Swinden Laboratorium B.V., The Netherlands
W vom Berg	Technische Vereinigung de Großkraftwerksbetreiber e.V., Germany
W Wassing	Forschungsinstitut der Zementindustrie, Germany
G Bachman	Forschungsinstitut der Zementindustrie, Germany
H -M Sylla	Forschungsinstitut der Zementindustrie, Germany
R Bialucha	Forschungsgemeinschaft Eisenhüttenschlacken, Germany
G J de Groot	Energieonderzoek Centrum Nederland, The Netherlands
F J M Lamers	NV KEMA, The Netherlands
I Hohberg	Institut für Bauforschung der RWTH Aachen (ibac), Germany
R Rankers	Institut für Bauforschung der RWTH Aachen (ibac), Germany

6.2 Laboratories participating in the precision experiment in ILS #3

Participant	**Country**
ATILH	France
Castle Cement Ltd	United Kingdom
Cemkut-Technocem Kft	Hungary
Ciments d’Obourg S.A.	Belgium
ECN	The Netherlands
ENCI Nederland BV	The Netherlands
Euroc Research A.B.	Sweden
FEhS	Germany
Forschungsinstitüt der V.Ö.Z.	Austria
Hochschule für Architektur und Bauwesen	Germany
IBAC	Germany
Institute of Mineral Building Materials	Poland
KEMA (KET)	The Netherlands
Kernforschungszentrum Karlsruhe	Germany
Swedisch Geotechnical Institute	Sweden
UNINOVA	Portugal
University of Aberdeen	United Kingdom
VDZ	Germany

7 References

[1] EUR 17869 EN 1997 European Commission report, Development of a leaching method for the determination of the environmental quality of concrete, Directorate - General Science, Research and Development, DGX11/C/5 - Programme SM&T, ISBN 92-828-1327-4.

[2] EN 206-1, *Concrete — Part 1: Specification, performance, production and conformity*.

[3] ISO 5725 (all parts), *Accuracy (trueness and precision) of measurement methods and results*.

Part II

(informative)

TEST METHOD USED IN THE STUDY FOR CHARACTERISATION OF LEACHING

1 Scope

This Technical Report describes a method for characterising the time-dependent leaching behaviour of components from hardened concrete, for use in the natural environment.

This method specifies the procedures for determining the controlling mechanism(s) for leaching of components, their effective diffusion coefficients, in the case of diffusion-control and their cumulative release behaviour over any period of time.

This characterisation method consists of two leaching test procedures. A potential or availability (pulverised specimen) test and a diffusion (tank) [monolithic specimen] test.

The test procedures produce leachates, the analytical procedures for which are not included in this Technical Report.

This Technical Report does not comprise a compliance method.

NOTE 1 The information obtained from the method is quantitative. In the absence, however, of similar information for other construction materials or compliance criteria for acceptable/unacceptable performance of hardened concrete, the data obtained with neither permit a relative nor an absolute assessment of the environmental quality/compatibility of the concrete, unless, by default, in the case where leached concentrations of environmentally significant components are at, or below, their analytical limits of detection.

NOTE 2 In principle, this method could be used to characterise the leaching behaviour of hardened concrete, irrespective of the exposure conditions (e.g. natural environment, contact with drinking water etc.) which the concrete would experience in service. It should be noted, however, that a European standard test method for the extraction/migration of mineral micropollutants is also likely to be developed by CEN/TC 164 - Water supply.

NOTE 3 Analytical procedures for determining concentrations of components in leachates and which may be suitable for the purposes of this Technical Report are being developed by CEN/TC 292 - Characterization of waste and CEN/TC 230 - Water analysis.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

NEN 7341:1995[1)], *Leaching characteristics of solid earthy and stony building materials and waste materials — Leaching tests — Determination of the availability of inorganic components for leaching*

NEN 7345:1995[1)], *Leaching characteristics of soil and stony building and waste materials — Leaching tests — Determination of the leaching of inorganic components from buildings and monolithic waste materials with the diffusion test*

EN 12390-2, *Testing hardened concrete — Making and curing specimens for strength tests*

EN 12390-7, *Testing hardened concrete — Density of hardened concrete*

3 Terms, definitions, symbols and abbreviations

For the purposes of this document, the following definitions apply.

3.1
available amount
quantity of a component which can potentially leach from a finely ground material as indicated in a standard test using a specified leachant under specified conditions

3.2
characterisation test
standard leaching method, or group of related leaching methods, used to determine the controlling transport processes, the short and long term behaviour and the basic properties of a solid material, subject to specified sequential/periodic leaching

3.3
compliance test
standard leaching method used to determine if a material is either acceptable, or unacceptable, for a given application, by comparing its performance, under specified leaching conditions (usually single extraction, without agitation, under temperature control), against specified criteria for specified components

3.4
component
either an inorganic (e.g. metal ion, ionic radical) or organic species leached from a solid material, into solution in a leachate

3.5
effective coefficient of diffusion (De)
measure of the mobility of components in a saturated porous material; it is a function of total porosity, pore size distribution, tortuosity, concentration gradient and chemical interactions within the pores

3.6
environmentally significant
term applied to a component (inorganic or organic) of a solid material to indicate whether it is considered to be capable of damaging, or impairing the use of, an ecological system if the component is emitted into it

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1) Dutch standards, NEN 7341 and NEN 7345, are available in English, from the secretariat Nederlands Normalisatie-instituut (NNI) P.O. Box 5059, 2600 GB Delft, The Netherlands.

3.7
free coefficient of diffusion (D)
measure of the mobility of a component, free to move under a concentration gradient, in a liquid or gas; it is a function of temperature and the size and type of the component

3.8
headspace
volume of air between the upper surface of a liquid (leachate) in a container and the container's closure

3.9
inert component
inorganic component, e.g. sodium (Na), potassium (K) and chloride ion (Cl) which as a consequence of its chemical properties shows no reaction with a solid material's matrix

NOTE In the case of hardened concrete the inorganic components Na, K and Cl have proved not to be suitable for the purpose of characterising the leaching behaviour (see 9.1).

3.10
inorganic component
cationic (e.g. metal ion) and/or anionic (e.g. ionic radical) species leached from a solid material, into solution in a leachate

3.11
leachant
solvent used in a leaching test

3.12
leachate
solution (or eluate) obtained after leaching a solid material with a leachant

3.13
leaching
extraction, by a leachant (solvent) of inorganic and/or organic components of a solid material, into a leachate (solution) by one or more physico-chemical transport processes

3.14
L/S
abbreviation for liquid (volume) to solid (mass) ratio; L/S is expressed in (l/kg)

3.15
monolithic test piece
solid material in the form of a single piece for which specified criteria for dimensions and/or physico-mechanical properties, apply

3.16
tank (diffusion) test
standard method in which a solid material is in a saturated condition and surrounded on all sides by a leachant; the material and leachant are usually static

3.17
tortuosity (T)
measure of the prolonged path along which leached components have to travel through the pore system of a monolithic porous material; it can be determined from the ratio of free to effective coefficients of diffusion, usually using inert components Na or K

4 Materials and reagents

4.1 Materials

4.1.1 General

A specimen (test piece) of hardened concrete presented for determination of its characteristic leaching behaviour is likely to be one of the following three types:

a) a standard moulded specimen (of regular geometry), moulded from a sample of fresh (plastic) concrete, e.g. site or ready-mixed;

b) a standard moulded specimen (of regular geometry) moulded from a sample of fresh concrete the bulk of which has been used in the production of a precast product;

c) a precast product (of either regular or irregular geometry) or part thereof.

Irrespective of the specimen type, test pieces are required for two different purposes. Test pieces (P_D), shaped and monolithic are required for the diffusion (tank) test at 6.3 of Part II. Test pieces (P_A), which need neither be shaped nor monolithic, are required for the availability test in Annex A of Part II.

In the case of standard moulded specimens of types a) and b) the requirements for test pieces (P_D) and (P_A) are given at 4.1.2 of Part II.

In the case of specimens of type c), the requirements for test pieces (P_D) and (P_A) are given at 4.1.3 of Part II.

4.1.2 Requirements for standard specimens as test pieces (P_D) and test pieces (P_A)

4.1.2.1 General requirements

Test pieces (P_D) shall be prepared in duplicate and be monolithic, undamaged and representative in structure, composition and homogeneity of the fresh concrete from which they were prepared.

Test pieces (P_A) shall be representative in structure, composition and homogeneity of the fresh concrete from which they were prepared.

4.1.2.2 Dimensions of a test piece (P_D)

The minimum dimension of a test piece (P_D) shall be 40 mm.

NOTE 1 Standard cubes of 100 mm × 100 mm × 100 mm made in accordance with EN 12390-2 have proved to be suitable. See 4.1.2.5 of Part II, however, for additional requirements for moulds and for use of release materials.

NOTE 2 There are no specific requirements for the dimensions of test pieces (P_A).

4.1.2.3 Volume of a test piece (P_D)

The volume (V_p), in litres, of a test piece (P_D) shall be determined in accordance with EN 12390-7:2009, 5.5.4 (see Clause 2 of Part II).

NOTE The reference method (water displacement), given in EN 12390-7:2009, 5.5.2, should not be used, in order to minimize contact between water and a test specimen prior to leaching.

4.1.2.4 Oven-dried mass and oven-dried density of a test piece (P_D)

The oven-dried mass (m), in kg, shall be determined in accordance with EN 12390-7:2009, 5.4 (see Clause 2 of Part II).

The oven-dried density (Δ), in kg/m^3, shall be determined in accordance with EN 12390-7:2009, 5.4 (see Clause 2 of Part II).

4.1.2.5 Making and curing standard specimens

a) Standard specimens presented for test shall have been made in accordance with EN 12390-2 (see Clause 2 of Part II) and with the following additional requirements:

   1) irrespective of the type of material from which a mould is made it shall be thoroughly cleaned before each use by washing with soap and water, rinsing with copious amounts of tap water, given a rinse with demineralised water followed by a final rinse with isopropyl alcohol and then dried before use;

   2) irrespective of the type of material from which a mould is made neither mineral oil nor other non-reactive material shall be used to prevent the concrete from adhering to the mould.

b) Standard specimens presented for test shall have been cured and stored in accordance with the following requirements:

   1) standard specimens shall remain in covered moulds for 1 d ± 8 h. Covers shall be made of an impermeable material which does not react with cement;

   2) on removal from the mould the specimen shall be weighed to an accuracy of 0,1 % of the mass of the specimen and then placed in a polyethylene bag and sealed. The specimen in the bag shall be stored under environmental control at a temperature of (20 ± 2) °C and a relative humidity of 65 % RH for 6 d ± 8 h. The specimen shall be removed from the bag and stored under environmental control at a temperature of (20 ± 2) °C and a relative humidity of 65 % RH for at least 21 d ± 8 h.

4.1.2.6 Age and weight at test

a) Standard specimens shall not be tested at ages less than 90 d ± 24 h unless an earlier age is ordered by way of law, statute or regulation.

b) Where standard specimens are to be tested at later ages they shall continue to be stored under environmental control at a temperature of (20 ± 2) °C and a relative humidity of 65 % RH, until removed for testing.

c) The age at which a standard specimen is tested shall be reported.

d) Standard specimens shall not be used as test pieces (P_D) where the weight at test differs from that at the time of demoulding, by more than ± 1 % of the demoulded mass.

4.1.2.7 Packaging and transport of standard specimens

Where testing of standard specimens requires transfer from the place of making to a test laboratory, the following requirements apply:

a) a standard specimen shall be wrapped tightly in impermeable unplasticized plastics film, placed in a polythene bag and the bag sealed;

NOTE Where characterisation of a concrete includes determination of organic components known, or suspected, to be present within the plastics film material, alternative wrapping materials should be used.

b) a wrapped and sealed standard specimen shall be packaged securely in order to minimise damage in transit;

c) a mechanically damaged standard specimen shall not be used as a test piece (P_D) but may be used as a test piece (P_A).

4.1.3 Requirements for precast products (or parts thereof) as test pieces (P_D) and (P_A)

4.1.3.1 General requirements

In accordance with 4.1.2.1 of Part II.

4.1.3.2 Dimensions of a test piece (P_D)

In accordance with 4.1.2.2 of Part II.

4.1.3.3 Volume of a test piece (P_D)

In accordance with 4.1.2.3 of Part II.

4.1.3.4 Oven-dried mass and oven-dried density of a test piece (P_D)

In accordance with 4.1.2.4 of Part II.

4.1.3.5 Production and curing of precast products (or parts thereof)

Precast products (or parts thereof), presented for test, shall have been produced, cured and stored in accordance with the minimum requirements of the appropriate European standard product specification (or other appropriate specification) before transfer to a testing laboratory.

4.1.3.6 Age at test

a) Precast products (or parts thereof) shall not be tested at ages less than 90 d ± 24 h unless an earlier age is ordered by way of law, statute or regulation.

b) Where precast products (or parts thereof) are to be tested at later ages they shall be wrapped tightly, at 90 d ± 24 h, in impermeable unplasticized plastics film, placed in a polythene bag and the bag sealed and stored in an environment capable of minimising damage.

NOTE Where characterisation of a concrete includes determination of organic components known, or suspected, to be present within the plastics film material, alternative wrapping materials should be used.

c) The age at which a precast product (or part thereof) is tested shall be reported.

4.1.3.7 Packaging and transport of precast products (or parts thereof)

In accordance with a), b) and c) in 4.1.2.7 of Part II.

4.2 Reagents

4.2.1 General requirements

Use only reagents of analytical quality.

4.2.2 Leachant

Demineralised water with an electrical conductivity $\leq$ 1 µS/cm and a pH between 4 and 7.

4.2.3 Acids

4.2.3.1 Concentrated nitric acid (HNO_3) of density 1,40 to 1,42 (in g/cm^3 at 20 ° C).

4.2.3.2 Dilute nitric acid, at a concentration of (1,0 ± 0,1) mol/l.

4.2.3.3 Concentrated hydrochloric acid (HC1) of density 1,18 to 1, 19 (in g/cm^3 at 20 ° C).

4.2.4 Oxidising agent

4.2.4.1 Potassium dichromate (K_2CrO_7), solid.

5 Apparatus

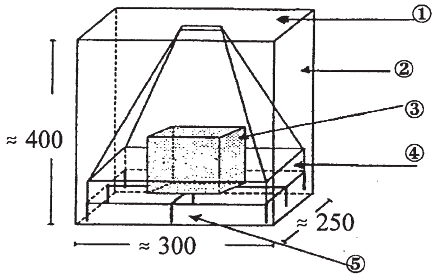
5.1 General

The apparatus shall neither emit nor absorb any of the components to be determined in the leachate.

NOTE A procedural blank test is not normally carried out to verify this requirement.

5.2 Sealable tank (or bucket)

Dimensions in millimetres



Key

1 cover
2 tank
3 test piece (P_D) (see 4.1.1 of Part II)
4 mesh support
5 drain/stopcock

Figure 1 — Typical tank and test piece arrangement during leaching

A sealable tank (see Figure 1 of Part II) of unplasticized plastics material shall have dimensions such that the test piece (P_D) (4.1.1 of Part II) shall be at least 20 mm beneath the surface of the leachant throughout the leaching procedure, in a volume of leachant, V, in litres, determined to an accuracy of ± 1 % where:

$$4(V_p) \geq V \leq 6(V_p) \quad (1)$$

and (Vp) is the volume (4.1.2.3 of Part II) of the test piece (P_D).

The tank shall contain either a plastics mesh supporting structure such that the test piece is in contact with leachant on all faces, or alternatively, be suspended on a plastics wire from the cover of the tank.

The tank shall be sealed with a suitable cover during the leaching procedure.

NOTE Covering the tank minimizes the uptake of atmospheric carbon dioxide.

The tank, cover and plastics supporting structure shall be rinsed, firstly with dilute nitric acid (4.2.3.2 of Part II) and then with the leachant (4.2.2 of Part II) before use.

5.3 Filtering equipment

Filtering equipment of either the low pressure or high pressure type has proved to be suitable.

5.4 Membrane filters

Membrane filters, suitable for operation with the filtering equipment (5.3 of Part II) shall have a pore size of 0,45 µm. Before use, they shall be rinsed successively with dilute nitric acid (4.2.3.2 of Part II) and the leachant (4.2.2 of Part II).

5.5 Plastics bottles

Sealable plastics bottles of appropriate size for storage of leachates, prior to analysis.

5.6 pH meter

A pH meter capable of measuring to an accuracy of ± 0,05 pH units.

5.7 Conductivity meter

A conductivity meter capable of measuring to an accuracy of ± 1 μS/cm.

6 Determining the leaching behaviour

6.1 General

The determination of the characteristic leaching behaviour of hardened concrete comprises three distinct laboratory procedures together with a number of calculation procedures.

A diffusion (tank) leaching test (6.3 of Part II) in order to provide information on time-dependent leaching characteristics from testing over a period of 14 d.

An availability leaching test (Annex A) in order to be able to define both a concentration gradient and an indication of the maximum potential for leaching of components from the material by diffusion.

A determination of the surface area (A) (Annex B of Part II) of a test piece (P_D) (4.1.1 of Part II), used in the diffusion (tank) test, in order to accurately determine the leaching of a component per unit area of the concrete and subsequently its effective diffusion coefficient, where leaching is diffusion-controlled.

A number of calculation procedures in Clause 9 of Part II which provide a means of quantitatively describing the leaching characteristics, in terms of the controlling mechanism and time-dependency of release of components from hardened concrete which is to be subject to exposure in the natural environment.

6.2 Principles

6.2.1 Diffusion (tank) test

The time-dependent leaching behaviour is determined by immersing a monolithic test piece (P_D) (4.1.1 of Part II), under static conditions, in a temperature controlled leachant (demineralised water), of known but uncontrolled pH.

NOTE The pH at which leaching takes place is controlled by the concrete test piece.

The initial leachant is replaced four times over periods of up to 14 d to produce five leachate fractions. The concentrations of components, pH and conductivity are determined in each leachate fraction.

6.2.2 Availability test

The available, or potential, amount of the components in the concrete available for leaching (Annex A of Part II), is determined on a finely ground sample (90 % passing a 125 μm sieve) by leaching twice with demineralised water set, initially, at a controlled pH 7 and secondly, at a controlled pH 4.

6.2.3 Surface area determination

The surface area (A) of a test piece (P_D) is determined (Annex B of Part II) by linear and/or radial measurement, as appropriate to test piece geometry.

6.2.4 Assessment of the characteristic leaching

From the results of the determinations (6.2.1 to 6.2.3 of Part II) the controlling mechanism for leaching of a component can be identified and assessed.

Where leaching is diffusion controlled an effective diffusion coefficient (D_e) is calculated for each component. Using the effective diffusion coefficient, the cumulative release (mg/m^2) of a component, after a period of leaching of 14 d, is calculated.

On the basis of the results, an assessment can be made of the time dependency of the leaching, the cumulative release of leaching, the cumulative release of components extrapolated to longer time periods and the effective diffusion coefficient.

NOTE Such information can be of use in any assessment of the environmental compatibility of the concrete for use in the natural environment.

6.3 Diffusion (tank) test

6.3.1 Test conditions

Carry out the diffusion (tank) test procedure (6.3.2 of Part II) at a temperature controlled to (20 ± 2) ° C.

6.3.2 Procedure

6.3.2.1 General

Carry out the procedure in duplicate.

6.3.2.2 First leaching period

Immerse a test piece (P_D) (4.1.1 of Part II) in the tank (5.2 of Part II) in a volume (V), in litres, of leachant (4.2.2 of Part II), determined to an accuracy of ± 1 %, in accordance with 5.2 of Part II.

Seal the tank, using the cover (5.2 of Part II). After a period (n = 1) of (6,0 ± 0,6) h, drain off the leachate. Record the drainage/replenishment time (t_1) for this first fraction as the time elapsed between first immersion and completion of draining, to an accuracy of ± 5 min.

Filter the first leachate fraction over the membrane filter (5.3 and 5.4 of Part II). Determine the pH, to an accuracy of ± 0,1, using the pH meter (5.6 of Part II) and the conductivity, to an accuracy of ± 1 µS/cm, using the conductivity meter (5.7 of Part II).

NOTE 1 The pH of the leachate fraction gives an indication of the level of the alkalinity of the test piece.

NOTE 2 The conductivity gives an indication of the dissolved solids content of the leachate fraction which may be of analytical significance.

If a leachate fraction is not to be analysed (see 6.3.2.5 of Part II) immediately, transfer a number of aliquots, sufficient for the analytical requirements, to sealable bottles (5.5 of Part II) and preserve and store them in accordance with 6.3.2.4 of Part II.

6.3.2.3 Subsequent leaching periods (second to fifth)

Immediately after draining the first leachate fraction in 6.3.2.2 of Part II and without rinsing or drying the test piece, refill the tank. Use the same volume (V) used in 6.3.2.2 of Part II and reseal the tank.

Continue the procedure in accordance with 6.3.2.2 of Part II but using a second period of immersion (approximately 18 h) which finishes, by drainage/replenishment, at (24,0 ± 2,4) h from the point at which the test piece was first immersed. Record this drainage/replenishment time (t_2) for the second fraction to an accuracy of ± 5 min.

Repeat the entire procedure of 6.3.2.2 of Part II a further three times, for periods of immersion which finish (by drainage/replenishment) at the times given for t_3, t_4 and t_5 in Table 1 for periods n = 3, n = 4 and n = 5.

Table 1 — Replenishment times of leachant and leaching periods

Time elapsed from first immersion, at which tank is to be drained/replenished with fresh leachant t_i [a] (hour)		Leaching period n_i	Leachate fractions (i = 1 to 5) in each leaching period
t_1	6,0 ± 0,6	1	1
t_2	24,0 ± 2,4	2	1+2
t_3	78,0 ± 7,8	3	1+2+3
t_4	168,0 ± 16,8	4	1+2+3+4
t_5	336,0 ± 33,6	5	1+2+3+4+5

[a] The time, t_i, at drainage/replenishment of leachant, for each period n_i, shall be measured to an accuracy of ± 5 min.

6.2.3.4 Storage of leachate fractions

Where analysis of leachate fractions is to be delayed, prevent precipitation, evaporation and deterioration of the components during storage by adopting one of the following procedures:

a) in the case of most cationic (metal) components (e.g. arsenic, cadmium, lead) minimize headspace in the storage bottles, before sealing, consistent with preserving the leachate by acidifying the aliquot with the minimum volume of nitric acid (4.2.3.1 of Part II) needed to reach pH 2.

b) in the case of mercury as a component, minimize headspace, consistent with preserving the leachate by acidifying the aliquot with the minimum volume of nitric acid (4.2.3.1 of Part II) and potassium dichromate (4.2.4.1 of Part II), needed to reach pH 2;

c) in the case of tin as a component, minimize headspace, consistent with preserving the leachate by acidifying the aliquot with the minimum volume of hydrochloric acid (4.2.3.3 of Part II) needed to reach pH 2;

d) in the case of anionic components (e.g. chloride, sulfate, fluoride) minimize headspace but use no acidification.

6.2.3.5 Analysis of leachate fractions

Analyse the leachate fractions, obtained in 6.3.2.2 and 6.3.2.3 of Part II, as soon as possible after each period from n = 1 to n = 5.

Carry out the analyses of the leachates for concentrations of appropriate components using appropriate analytical methods using, e.g. FAAS, GFAAS, ICP AES, ICP MS.

NOTE Methods are in preparation in CEN/TC 292 and CEN/TC 230 for the chemical analysis of leachates for a number of components.

7 Calculation of cumulative leaching and expression of results

7.1 Measured leaching of a component per leachate fraction

Calculate, for each component to be quantified, the measured leaching, E^*_i, for each leachate fraction, i, in mg/m^2, from the following equation:

$$E^*_i = \frac{c_i V}{fA} \qquad (2)$$

where

c_i is the concentration of a component in fraction i, in µg/l;

V is the volume of leachant, in l;

A is the surface area of a test piece (P_D), in m^2 (see Annex B of Part II);

f is a dimensionless factor: 1 000 (µg/mg).

In Equation (2), c_i, is the concentration of a component present in the leachate fraction before any preservative (see 6.3.2.4 of Part II) has been added. Where more than 4 ml of preservative per litre of leachant have been added, correct the concentration, as determined at 6.3.2.5 of Part II, for the volume of preservative.

Where the concentration, c_i, of a component in a leachate fraction is below the limit of detection (DTL) of the analytical method (see 6.3.2.5 of Part II) carry out two calculations to determine two limiting values for E^*_i .

Calculate the upper limit by equating, c_i in Equation (2) with the limit of detection for the component.

Calculate the lower limit by equating, c_i, in Equation (2) with zero thereby giving a zero value for E^*_i .

7.2 Measured and theoretical cumulative leaching of a component

7.2.1 General

The values for measured and theoretical cumulative leaching are used to assess whether the leaching behaviour can be described as diffusion controlled (see 9.2 of Part II). A comparison of the values can reveal the extent to which rinsing effects and dissolution and/or depletion of a component are involved in the leaching.

Where leaching of a component can be determined to be diffusion controlled, the development of ,$_n$ (the theoretical cumulative leaching) gives an indication (see 9.2 of Part II) of the part diffusion plays in the leaching behaviour.

7.2.2 Measured cumulative leaching of a component

Calculate, for each component, the measured cumulative leaching, ,$^*_n$, for each leaching period, from n = 1 to 5.

NOTE 1 The period, n = 1, begins at the point of the first complete immersion to the point at which the tank has just been drained i.e. it comprises leachate fraction i = 1. Period, n = 2, begins at the point of the first immersion to the point at which the tank has just been drained for the second time i.e. it comprises the sum of the leachate fractions i = 1 and i = 2; periods n = 3 to 5 are configured similarly (see Table 1).

Calculate ,$^*_n$ in mg/m^2, from the following Equation:

$$\varepsilon_n^* = \sum_{i=1}^{n} E_i^* \qquad (3)$$

for n = 1 to 5

where

E^*_i is the measured leaching of a component in fraction i, calculated using Equation (2), in mg/m^2;

n is the number of the appropriate leaching period.

NOTE 2 See Annex C of Part II for a diagrammatic representation of the diffusion (tank) leaching procedure.

7.2.3 Theoretical cumulative leaching of a component

Calculate, for each component to be quantified, the theoretical cumulative leaching, ,$_n$, in mg/m^2, for each of the leaching periods, n = 1 to 5, from the following equation:

$$\varepsilon_n = E_i^* \frac{\sqrt{t_i}}{\sqrt{t_i} - \sqrt{t_{i-1}}} \tag{4}$$

where

E^*_i is the measured leaching of a component in fraction i, calculated from Equation (2), in mg/m^2;

t_i is the draingage/replenishment time of fraction i, i.e. the time at the end of leaching of fraction i, in s;

t_{i-1} is the drainage/replenishment time of fraction i-1, i.e. the time at the start of leaching of fraction i, in s.

NOTE 1 See Annex C of Part II for a diagrammatic representation of the diffusion (tank) leaching procedure.

NOTE 2 The measured cumulative leaching ,*$_n$, always includes the measured leaching of previous periods. Accordingly, any particular deviation(s) in one period (e.g. wash-off effects during the first period) affect the following periods and add to the difficulties of assessment and interpretation of the general leaching behaviour.

NOTE 3 The theoretical cumulative leaching, ,$_n$, determines only the leaching up to and including period i on the basis of the measured leaching in period i.

8 Precision of cumulative leaching

8.1 General

Precision data were determined from a precision experiment (as part of an interlaboratory study (ILS)) conducted in 1996, involving 18 laboratories and one concrete mix. The concrete mix was artificially enhanced in its content of the environmentally significant components, arsenic (As), cadmium (Cd), chromium (Cr) and vanadium (V), by introducing solutions of standard additions (spikes) to the mixing water.

NOTE The approximate (i.e. rounded) quantities of the components added, in mg/kg of the concrete, were:

As at 170 mg/kg; Cd at 65 mg/kg; Cr at 100 mg/kg; V at 130 mg/kg.

Precision estimates for the standard deviation of repeatability, S_r, the standard deviation of reproducibility, S_R, the repeatability limit, r, the reproducibility limit, R, together with means pooled for each estimate over several ranges of components, are given in 8.2 and 8.3 of Part II, for the availability test and diffusion (tank) test, respectively.

8.2 Precision of the availability test

Table 2 — Mean results and precision estimates for the availability test

Component	Mean		Repeatability standard deviation S_r (%)	Reproducibility standard deviation S_R (%)	Repeatability limit r (%)	Reproducibility limit R (%)
	(measured) (mg/kg)	(calculated) (mg/m^2)				
As	9,7	381,2	24,2	64,2	67,8	179,8
Cl	779,7	30 642	27,6	104,9	77,3	293,7
Cr	75,7	2 975	11,9	23,3	23,8	65,2
K	1 820,0	71 526	6,4	16,7	17,9	46,8
Na	307,3	12 077	7,2	28,4	20,2	79,5
S	1 507,0	59 225	3,0	18,8	8,4	52,6
V	20,6	810	14,4	29,7	40,3	83,2
M_1) Overall means			13,5	40,9	37	114
M_2) Means (excluding As and Cl) [a]			8,6	23,4	24	66
M_3) Means for As/Cr/V (i.e. the environmentally significant metals)			16,8	39,1	47	109

[a] The results given for means M2, exclude As and Cl. Data have been excluded where components gave average concentrations < 10 x DTL (the limit of detection).

8.3 Precision of the diffusion (tank) test

Table 3 — Mean results and precision estimates for the diffusion (tank) test

Component	Mean		Repeatability standard deviation S_r 14 d (%)	Reproducibility standard deviation S_R 14 d (%)	Repeatability limit r 14 d (%)	Reproducibility limit R 14 d (%)
	14 d (calculated) (mg/kg)	14 d (measured) (mg/m^2)				
As	0,21	8,47	9,7	26,8	27,2	75,0
Cd	<0,01	0,05	15,8	41,0	44,2	114,8
Cl	5,4	210,6	11,3	56,0	31,6	156,8
Cr	2,9	112,7	15,3	20,7	42,8	58,0
K	34,2	13 442	11,9	21,7	33,3	60,8
Na	37,9	1 489,4	9,1	24,8	25,5	69,4
S	25,8	1 012,1	17,1	17,4	47,9	48,7
V	2,0	78,3	7,6	19,6	21,3	54,9
M_1) Overall means			12,2	28,5	34	80
M_2) Means (excluding As, Cd and Cl [a])			12,0	22,0	34	62
M_3) Means for As/Cd/Cr/V (i.e. the environmentally significant metals)			12,1	27,0	34	76

[a] The results given for means M_2, exclude As, Cd and Cl. Data have been excluded where components gave average concentrations < 10 x DTL (the limit of detection).

9 Characterising the leaching behaviour

9.1 General

Detailed procedures are given in NEN 7345 (see Clause 2 of Part II) for characterising the leaching behaviour of monolithic materials in general. In the case of hardened concrete, however, some modifications to the general procedures are required and these are indicated in the relevant clauses of this Technical Report. In order to establish the dominant mechanism for leaching from concrete (or any material) two or more ‘inert’ components need to be selected and their cumulative leaching profiles, assessed.

NOTE 1 An inert component is one, which due to its chemical properties, has no reaction with the concrete. Typically, these would be sodium (Na), potassium (K) and chloride ion (Cl). In the case of hardened concrete, however, these have proved to be unsuitable for characterisation purposes.

As established in EC report EUR 17869 EN (see Bibliography of Part II) components which have proved to be suitable for this purpose, from investigations on coal fly ash containing concretes, were silicon (Si), lithium (Li) magnesium (Mg) and barium (Ba); none of which are strictly inert, in the terms of the accepted definition.

NOTE 2 In order to be able to use a component for assessing the characteristic leaching behaviour, the concentrations measured in the leachate fractions used to determine the behaviour should be greater than three times the limit of detection (DTL) for the component, and should also be greater than three times the concentration of the component in the leachant.

Where diffusion-controlled leaching behaviour can be demonstrated for two or three inert components, it is generally assumed that the leaching of other components is also diffusion-controlled.

In the case of coal fly ash containing concretes the dominant leaching mechanism is said to have been determined (see Annex E of EUR 17869 EN) to be diffusion-controlled.

Supplementary calculation procedures, where particular characteristics are observed in the leaching behaviour, are given in Annex D of Part II.

9.2 Determining the controlling leaching mechanism

In order to determine the controlling mechanism for leaching of a component from the test piece (P_D) during the diffusion (tank) test carry out the assessment procedure in NEN 7345:1995, 9.3 (see Clause 2 of Part II).

At Stage 1 of the assessment procedure, note that for hardened concrete only five (n = 5) periods of leaching (not eight) apply.

At Stage 2 of the assessment procedure delete the four paths defined and substitute the following four paths:

a) “total” path : points 2 to 5, inclusive;

b) “start” path : points 1 to 3, inclusive;

c) “middle” path : points 2 to 4, inclusive;

d) “end” path : points 3 to 5, inclusive;

NOTE In the case of hardened concrete a smaller number of leaching periods are used when compared to the general case given in NEN 7345 (see Clause 2 of Part II). Accordingly, the definitions of the paths also change.

Carry out the procedure in NEN 7345:1995, 9.3 (see Clause 2) up to and including Stage 5.

9.3 Calculating the effective and mean effective diffusion coefficients of a component

9.3.1 Effective diffusion coefficient of a component

Calculate, for all (part) paths for which it appears that the leaching is diffusion-controlled, and for all leachate fractions within a (part) path, the effective diffusion coefficient, $D_{e,i}$, in m^2/s, for a component, from the following equation:

$$D_{e,i} = \frac{\pi (E_i^*)^2}{4(U_{bes}\rho)^2(\sqrt{t_i}-\sqrt{t_{i-1}})^2} \tag{5}$$

where

E^*_i is the measured leaching of the component (see **7.1**) in fraction, i, in mg/m^2;

U_{bes} is the quantity of the component, on the oven-dried basis, available for leaching according to NEN 7341 (see Annex A:1995, 9.3), in mg/kg;

NOTE U_{bes} is occasionally referred to as L_{max} (see Annex A and Annex E of EUR 17869 EN).

Δ is the oven-dried density of the test piece (P_D) in kg/m^3;

t_i is the drainage/replenishment time of fraction, i, (i.e. the time at the end of leaching of fraction i), in s;

t_{i-1} is the drainage/replenishment time of fraction, i-1, (i.e. the time at the start of leaching of fraction i), in s.

Express the effective diffusion coefficient of all fractions within the (part) path as a negative logarithm, from the following equation:

$$pD_{e,i} = -\log_{10} D_{e,i} \tag{6}$$

where

$D_{e,i}$ is the effective diffusion coefficient of a component, in m^2/s.

9.3.2 Mean effective diffusion coefficient of a component

Calculate, from the previously calculated values of $pD_{e,i}$ [for all fractions in the (part) path], the mean effective diffusion coefficient, pD_e, for the (part) path for a component, from the following formula:

$$pD_e = \frac{\sum_{i=1}^{n} pD_{e,i}}{n} \tag{7}$$

where

$pD_{e,i}$ is the negative logarithm of the effective diffusion coefficient of all fractions within the (part) path;

n is the number of data points in the path.

NOTE The mean effective diffusion coefficient is also expressed non-logarithmically in 9.3.3 of Part II.

9.3.3 Selection of the lowest value of the mean effective diffusion coefficient

Where it appears, for more than one (part) path that leaching is diffusion-controlled, select the lowest value of pD_e (indicating the highest mobility) as the most representative value.

Determine the standard deviation of the values for $pD_{e,i,}$ from the values determined for each leachate fraction.

Calculate the value of the lowest mean effective diffusion coefficient, non-logarithmically, as D_e, for the component, from the following equation:

$$D_e = 10^{-pDe} \tag{8}$$

where

pD_e is the negative logarithm of the lowest value of the mean effective diffusion coefficient for the component.

9.4 Calculating the cumulative leaching of a component per surface unit, per time interval

Calculate, the cumulative leaching of a component per surface unit, over any time interval, E_{xy}, in mg/m^2, from the following equation:

$$E_{xy} = 2\rho U_{bes}(\sqrt{t_x} - \sqrt{t_y})\sqrt{D_e/\pi} \tag{9}$$

where

Δ is the oven-dried density of the test piece (P_D), in kg/m^3;

U_{bes} is the quantity of the component, on the oven-dried basis, available for leaching according to NEN 7341 (see Annex A of Part II), in mg/kg;

t_x is the starting time of the interval concerned in relation to the start of the test, in s;

t_y is the end time of the interval concerned in relation to the start of the test, in s;

D_e is the effective diffusion coefficient of the component, in m^2/s.

9.5 Assessment of components for which no diffusion coefficient can be determined

Where it has been established at 9.2 of Part II that the test piece (P_D) shows diffusion-controlled leaching behaviour for two or more inert components, the leaching of the other components should also be diffusion-controlled, possibly in combination with initial wash-off and/or dissolution.

NOTE It is generally accepted that depletion of inorganic components other than the inert components occurs only if depletion of the inert components also occurs.

Where, however, no diffusion-control can be established, from testing, for specific components this could be a consequence of one or more of the following three phenomena:

a) chemical (equilibrium) changes in the solid or leachate;

b) low concentrations of the component in the leachate;

c) surface wash-off and/or dissolution of the component, which can occur temporarily at the interface of the test piece and the leachant.

In such cases, only the characteristics of release over 14 d (n = 5) can be determined using 7.1 and 7.2.2 of Part II. It is not possible to give a general method for calculating the characteristics of release for periods longer than 14 d.

9.6 Assessment of a diffusion coefficient

9.6.1 General

The value of the negative logarithm of the mean effective diffusion coefficient, pD_e, (see 9.3.3 of Part II) of a component, gives an indication of the components mobility and its propensity to be leached.

The minimum value of pD_e (see 9.3.3 of Part II) corresponds to the maximum rate of leaching, and vice versa, at constant availability, U_{bes}.

NOTE 1 In the case of sodium for example, the minimum value of pD_e is 8,88, which corresponds to the free mobility of sodium ion in demineralised water.

NOTE 2 Availability, U_{bes}, determines the concentration gradient under which diffusion can take place.

9.6.2 Assessment of the negative logarithm of the mean effective diffusion coefficient

Assess the value of the negative logarithm of the mean effective diffusion coefficient, pD_e, of a component, as follows:

a) low mobility : $pD_e > 12{,}5$;

b) average mobility : $11{,}0 < pD_e < 12{,}5$;

c) high mobility : $pD_e < 11{,}0$.

NOTE A pD_e value of less than 9,5 has no physical significance since it indicates that a material has no developed internal porosity (tortuosity). Where such a low value is found at 9.3.3 of Part II, the measured availability should be checked and redetermined (see Annex A of Part II), if appropriate.

9.7 Comparison of the mobility of a component with the free mobility of the same component in water

9.7.1 General

A comparison of the mobility of a component in a shaped or monolithic material, such as a concrete test piece, with the free mobility of the same component in demineralised water gives an indication of the tortuosity (physical retardation), 9.7.2 of Part II, and the retention factor (chemical retention), 9.7.3 of Part II, of the material’s matrix.

Tortuosity gives an indication of the path length which a component, under diffusion-controlled leaching, has to cover within a porous matrix. It is component-independent.

For the calculation of tortuosity an inert component has to be selected for which there is no chemical interaction with the matrix (see 9.7.2 of Part II).

NOTE In the case of hardened concrete it is unlikely that any component can completely fulfill the requirements of an inert component. In the general case, sodium ion (Na) is usually selected. Sodium has proved to be unsuitable for characterising concrete (see Annex E of EUR 17869 EN).

9.7.2 Calculating the tortuosity

Select the component which demonstrates the lowest value of pD_e (see 9.3.3 of Part II) in the concrete matrix, express it as a non-logarithmic diffusion coefficient and calculate the tortuosity, T, from the following equation:

$$T = \frac{D}{D_e} \tag{10}$$

where

D is the diffusion coefficient of the component in water, in m^2/s;

D_e is the mean effective diffusion coefficient of the component in the concrete, in m^2/s.

9.7.3 Calculating the retention factor

Calculate the retention factor, R, for a component, in the concrete matrix, from the following equation:

$$R = \frac{DT}{D_e} \tag{11}$$

where

D is the diffusion coefficient for the component in water, in m^2/s;

D_e is the mean effective diffusion coefficient of the component in the concrete, in m^2/s;

T is the tortuosity, calculated at 9.7.2 of Part II.

9.8 Calculating the quantity leached, per mass unit, in the diffusion (tank) test

Calculate the quantity, $U_{dif,\,t,}$ on the oven-dried basis, of a component leached, per mass unit up to time t, in mg/kg, from the following formula:

$$U_{dif,t} = 2A\rho U_{bes} \frac{\sqrt{D_e t}}{m\pi} \tag{12}$$

where

A is the surface area (see Annex B) of the test piece (P_D), in kg/m^3;

Δ is the oven-dried density of the test piece (P_D) in kg/m^3;

U_{bes} is the quantity of the component, on the oven-dried basis, available for leaching according to NEN 7341 (see Annex A of Part II), in mg/kg;

D_e is the mean effective diffusion coefficient of the component, in m^2/s;

t is the duration of the leaching, in s;

m is the oven-dried mass of the test piece (P_D), in kg.

9.9 Calculating the extent of depletion of a component

Calculate the approximate extent of depletion of a component, $UP_{\text{dif},t}$, leached in time t during the diffusion (tank) test, in percent, from the following equation:

$$UP_{dif,t} = \frac{U_{dif,t}}{U_{bes}} \times 100 \tag{13}$$

where

$U_{dif,t}$ is the quantity of the component, on the oven-dried basis, leached, per mass unit up to time t, in mg/kg (see 9.8 of Part II);

U_{bes} is the quantity of the component, on the oven-dried basis, available for leaching according to NEN 7341 (see Annex A of Part II), in mg/kg.

10 Test report

The test report shall include the following information:

a) reference to this Technical Report;

b) any deviation(s) from the test procedures described in this Technical Report;

c) a full identification and description of the test specimens, including;

  1) geometrical shape (e.g. cube, prism, section of a precast product);

  2) dimensions;

  3) details of the composition of the concrete from which the test specimens were prepared;

d) a description of the type of concrete presented for test e.g. site-mixed, ready-mixed, precast;

e) a confirmation that the concrete was designed to be of closed structure;

f) the date of production of the concrete and a description of the conditions of curing and storage where they deviate from the minima recommended in this Technical Report;

g) identification of the producer of the concrete, the place of production and, where different, the identification of the organization responsible for preparing the test specimens;

h) identification of the test laboratory;

i) the date of receipt of test specimens at the test laboratory;

j) the date of the start of testing;

k) the age of the test specimens at the start of testing;

l) the pH and conductivity of each leachate fraction;

m) concentrations of components determined in the leachate fractions and in the availability test, together with estimates of their precision;

n) estimates of the limits of detection (DTL) for each component, for the analytical method employed;

o) identification of the analytical methods employed;

p) the characteristics of the leaching behaviour e.g.

    1) the controlling transport mechanism for leaching;

    2) the mean negative logarithm of the effective diffusion coefficients for components;

    3) assessment of the mean negative logarithm of the effective diffusion coefficients;

    4) the cumulative leaching of components tested over 14 d;

    5) the cumulative leaching of a component per surface unit, per time interval.

Annex A
(normative)

Determination of the available (potential) amount of a component for leaching

A.1 Procedure

Determine the available or potential amount of a component for leaching in accordance with Dutch national standard NEN 7341 (see Clause 2 of Part II).

NOTE In the availability test finely ground concrete (90 % passing a 125 µm sieve) is consecutively leached with demineralised water initially set at a controlled pH 7 and secondly at a controlled pH 4, using a liquid (volume) to solids (mass) ratio (L/S) of 50 dm^3/kg.

A.2 Expression of results

Express the result as the maximum available amount for leaching, U_{bes} (sometimes identified as L_{max}), in mg/kg.

Annex B
(normative)

Determination of the surface area (A) of a test piece (PD) for use in the diffusion (tank) test

B.1 Procedure

Determine the surface area (A) of a test piece (P_D) for use in the diffusion (tank) leaching test (6.3 of Part II) by linear measurement of its characteristic dimensions such as length, width, height and by radial measurement of any curved surfaces.

Determine the characteristic dimensions to an accuracy of better than ± 1 mm.

B.2 Calculation and expression of results

From the characteristic dimensions, calculate, using classical geometrical principles the surface area (A) of a test piece (P_D).

Express the result in m^2 to the nearest 0,001 m^2.

Annex C
(informative)

Diagrammatic representation of the diffusion (tank) leaching procedure

The sequential (periodic) nature of the diffusion (tank) leaching procedure (6.3.2 of Part II) is represented diagrammatically in Figure C.1 of Part II.

It should be noted that the measured leaching of a component per fraction, E^*_i, is used to determine:

a) ,*$_n$, the measured cumulative leaching (7.2.2 of Part II) and;

b) ,$_n$, the theoretical cumulative leaching (7.2.3 of Part II).

In Figure C.1, E^*_i is the measured leaching of a component in leachate fraction i, in mg/m^2. It is calculated in accordance with Equation (2) in 7.1 of Part II. The fractions i = 1 to i = N indicate the successive leachates and the period n = N corresponds to the sum of the number of fractions from i = 1 to i = N.

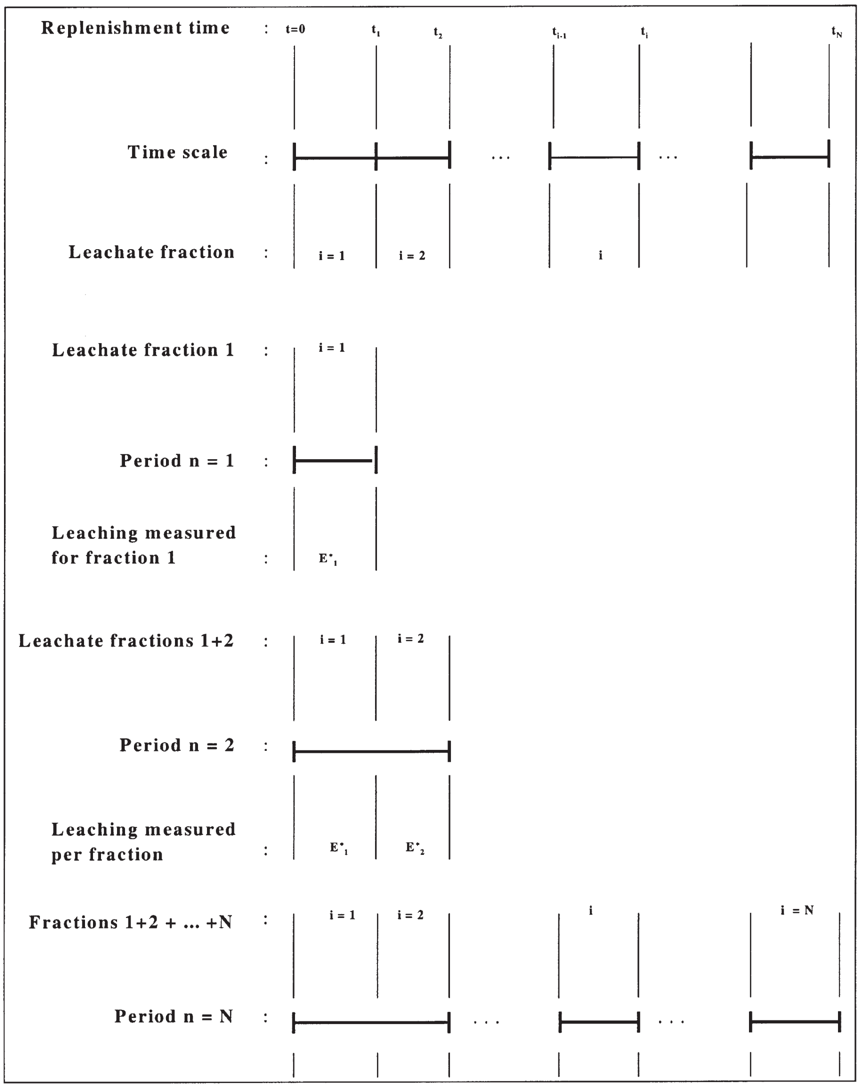


Figure C.1 — Diagrammatic representation of the diffusion (tank) leaching procedure

Annex D
(informative)

Supplementary procedures for calculating the indicative upper limit for leaching for particular characteristics of the leaching behaviour

D.1 General

In Clause 9 of Part II a description is given of the procedure to be used in establishing whether the leaching of a concrete test specimen is diffusion-controlled. This can be established from the leaching of several inert components or other components which are available in sufficient quantity to give measured concentrations which lie well above the limit of the detection (DTL) of the analytical method for the component.

Where it appears that the leaching of these components is diffusion-controlled, the concrete can be treated as a porous matrix. It can then be assumed that all components which are evenly distributed in the matrix initially have leaching which is diffusion-controlled.

In the case of a number of components it will often not be possible to determine diffusion coefficients. This occurs mainly for components which have a low availability and/or a high pD_e value (see 9.6.2 of Part II). For such components, rinsing, dissolution from only the outer layer of the test specimen, chemical speciation, complex leachate compositions etc., can mean that no diffusion coefficient can be determined. In such cases it is not possible to accurately determine the leaching levels of all the components either up to 14 d or beyond.

This annex describes calculation procedures which should be used to give indicative upper limits of leaching after a period of 14 d and after a period T days, for a range of characteristics in the leaching behaviour. Useful information can be obtained where the leaching of the matrix is generally diffusion-controlled and shows rinsing/wash-off (or direct depletion) for all components. In particular, the order of magnitude of the likely release of components by leaching, can be assessed.

NOTE In cases where a more accurate characterisation of the leaching is required, rather than the calculation of an indicative upper limit, modifications to the diffusion (tank) test procedure, such as, a longer test duration, longer periods between replenishments, a lower leachant to specimen volume ratio, would be required.

D.2 Diffusion-controlled leaching of components for which no diffusion coefficient can be established

Where the leaching behaviour of the concrete is diffusion or wash-off controlled and displays the characteristics described in summary in Table D.1, calculate the indicative upper limits of leaching at 14 d and at longer periods, T days, in accordance with the procedures and formulae given in the table.

NOTE For a detailed description of the terms used and the derivation of the formulae presented in Table D.1 of Part II, refer to NEN 7345 (see Clause 2 of Part II).

Table D.1 — Particular characteristics and calculation procedures for determining upper limits for leaching at 14 d and longer periods

Particular characteristics of the leaching behaviour	Upper limit for leaching after 14 d $_{,14}$ (mg/m^2)	Upper limit for leaching after T days [c] $_{,T}$ (mg/m^2)	Equation
Measured concentrations of components are 'low' in each (part) path. *Characterised by:* CF$_{1\text{-}5}$ [a] < 1,5DTL [b]	Let $_{,14}$ = $,^{*}_{5}$ [d] Calculate $,^{*}_{5}$ in accordance with 7.1 and 7.2.2 (In addition, set: c$_i$ = DTL , in the case where the measured concentration (c$_i$) is < DTL.)	Calculate $_{,T}$ from: $\varepsilon_T = \sqrt{(T/14)\varepsilon_{14}}$	(D.1)
Surface wash-off is observed and measured concentrations are low. *Characterised by early fractions dissolving or washing-off and for later fractions by:* *CF < 1,5 DTL.*		Calculate $_{,T}$ from: $\varepsilon_{\mathrm{T}} = \varepsilon_{\mathrm{afsp},0-2}{}^{\mathrm{e}} + \sqrt{(\mathrm{T}/14)\varepsilon_{\mathrm{oher},0-5}}{}^{\mathrm{f}}$	(D.2)
Apparent depletion is observed for inert and other components. *Characterised by diffusion-controlled leaching for an inert component in the second and/or third fraction and for another component, by:* *rc [g] < 0,35 in second and/or third fraction.*		Calculate $_{,T}$ from: $\varepsilon_T = \sqrt{(T/14)\varepsilon_{14}}$	(D.3)
Dissolution of a component. *Characterised for the component concerned, by:* *rc > 0,6*		Calculate $_{,T}$ from: $\varepsilon_T = 2\sqrt{(T/14)\varepsilon_{14}}$	(D.4)
Large variations are observed in the concentrations of measured components. *Characterised, for the total path and the (part) paths by:* *sd [h] > 0,5*		Calculate $_{,T}$ from: $\varepsilon_T = 5\sqrt{(T/14)\varepsilon_{14}}$	(D.5)

[a] CF is the measured concentration divided by the limit of detection (DTL).

[b] DTL is the limit of detection of a component using the selected analytical method.

[c] T is the period of leaching ,in days.

[d] ,$^{*}_{5}$ is the measured leaching of a component in leachate fraction five, in mg/m^2.

[e] ,$_{afsp,\ 0-2}$ is the quantity of a component rinsed from the surface of a test specimen, between time t = 0 and t_2 (see Table 1) calculated in accordance with NEN 7345 (see Clause 2 of Part II), in mg/m^2.

[f] ,$_{other,\ 0-5}$ is calculated as; $E^{*}_{1} + E^{*}_{2}$ +..+ $E^{*}_{5,}$ where E^{*}_{i} is the measured leaching of a component per leachate fraction, in accordance with 7.1.

[g] rc is the slope of the curve of the logarithm of the theoretical cumulative leaching ,$_n$ (7.2.3 of Part II) versus the logarithm of time t_i (table 1) for n = 1 to 5.

[h] sd is the standard deviation, calculated using linear regression, of log ,$_n$ versus log t_i relationship, described at footnote7).

Bibliography

[1] EUR 17869 EN 1997 European Commission report, Development of a leaching method for the determination of the environmental quality of concrete, Directorate - General Science, Research and Development, DGX11/C/5 - Programme SM&T, ISBN 92-828-1327-4

[2] EN 206-1, *Concrete — Part 1: Specification, performance, production and conformity*

[3] ISO 5725 (all parts), *Accuracy (trueness and precision) of measurement methods and results*

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