TEST METHODS TO ASSESS ENVIRONMENTAL PROPERTIES OF AGGREGATES IN DIFFERENT APPLICATIONS: THE ROLE OF EN 1744-3

H.A. van der Sloot*
E. Mulder**
*ECN
**TNO-MEP

Revisions

<table>
<thead>
<tr>
<th></th>
<th>A</th>
<th></th>
<th>B</th>
</tr>
</thead>
<tbody>
<tr>
<td>Made by:</td>
<td>H.A. van der Sloot</td>
<td>Approved:</td>
<td>R.N.J. Comans</td>
</tr>
<tr>
<td>Checked by:</td>
<td>J.J. Dijkstra</td>
<td>Issued:</td>
<td>C.A.M. van der Klein</td>
</tr>
</tbody>
</table>

ECN-Clean Fossil Fuels
Acknowledgement

This study was carried out with financial support from NOVEM under contract 376110/0001.
CONTENTS

LIST OF TABLES 4
LIST OF FIGURES 4
SUMMARY 5
1. INTRODUCTION 7
2. EXPERIMENTAL 9
3. RESULTS AND DISCUSSION 11
  3.1 Contact time in prEN1744-3 11
  3.2 Leaching as a function of pH 11
  3.3 Percolation test 12
  3.4 Particle size distributions of aggregates 12
  3.5 Diffusion rate from particles 14
  3.6 Relevant test methods for leaching 15
  3.7 Consistency of data for the same material class 15
4. RELEVANT ASPECTS CONCERNING THE DEVELOPMENT OF LEACHING TESTS FOR CONSTRUCTION MATERIALS 17
  4.1 List of Harmonisation projects and other relevant international projects 17
  4.2 CPD – Product families, technical specifications and leaching tests 17
  4.3 Life cycles of construction products 19
5. EN 1744-3 PERFORMANCE 21
6. CONCLUSIONS ON EN 1744-3 23
REFERENCES 24
APPENDIX A EN 1744-3 DATA WITH LONGER CONTACT TIME AND DIFFERENT PARTICLE SIZE DISTRIBUTIONS 27
APPENDIX B PH DEPENDENCE TEST DATA (PREN 14429) IN COMPARISON WITH EN 1744-3 DATA (ON COARSE AND FINE MATERIAL) AND EN 12457-2 DATA (CEN TC 292) 35
APPENDIX C PARTICLE SIZE EFFECTS 42
LIST OF TABLES

Table 1  Particle size distribution of test materials for EN 1744-3  9
Table 2  Differences in leaching conditions between EN 12457 and EN 1744-3  14
Table 3  Applications and test use  20

LIST OF FIGURES

Figure 1  Comparison of pH dependence leaching test data on cement mortars (size reduced to < 4 mm) with "concise" compliance test [27] data on material < 10 mm with undefined size distribution  16
Figure 2  Integration of test methods for environmental quality at different stages of the Building Cycle  19
SUMMARY

Aggregates are widely used in the construction sector in both bound and unbound form. Any bound application may lead in the long-term to an unbound application in the form of demolition debris. The assessment of environmental impact in such widely different applications requires different test methods, as the mechanisms of release and exposure conditions are not the same. In CEN/TC 154 Aggregates a new leaching standard has been worked out for judging the environmental properties of aggregates in connection with requirements on dangerous substances in the Construction Products Directive. The Standing Committee on the Construction Products Directive (SCC) has expressed the need for harmonisation of tests for dangerous substances in construction materials, among which leaching tests. This intention has consequences for the development of standards in the entire construction field. This position may also have consequences for the applicability of this new standard. In this report, the development of the CEN TC 154 standard is addressed in light of development of leaching tests in other areas. The key to environmental consequences of application of materials in or on soil is related to the manner in which materials are exposed to direct infiltration or other means of contact with water. This is generally addressed in a scenario type of approach. The following limitations of the CEN/TC 154 standard have been identified:

- The assumption of reaching equilibrium in 24 hours for particle sizes up to 32 mm is not fulfilled.
- The fraction of fines in the test portion is insufficiently addressed as only a high upper limit for particle size is given. The results are therefore more a reflection of the particle size distribution in the test portion than of the leaching of the aggregates from an environmental impact assessment perspective.
- Reproducibility of testing of the same material is jeopardized, when the particle size is so poorly defined. Depending on the strength of the material abrasion may lead to different particle size distributions during handling.
- As the test is not aimed at representing a relevant scenario of application, the test results can hardly be used in relation to the development of regulatory criteria. To develop criteria a link between test result and its meaning in practise is crucial. The intent to use the test for compliance purposes is therefore compromised.
- The release of constituents from aggregates using EN 1744-3 is artificially reduced by the short contact time and in addition by packing the material in a pile upon a screen. Both choices in the test lead to diffusion resistances, for which there would be ample time in any practical field application.

The prEN 1744-3 in fact is a mix between dissolution phenomena and diffusion processes. The manner, in which the EN 1744-3 test operates is to a large extent focussed on diffusion restrictions.

The specifications for aggregates to be used as unbound and hydraulically bound material for road base application require relatively fine-grained material as specified in prEN 13242 with at least 15 – 40 % of the material smaller than 4 mm. EN 1744-3 does not address these requirements. Besides, the test is not equipped for testing materials with a large fraction of fine particles, because of the fact that particles < 2 mm may fall through the screen and will be leached more intensively, caused by agitation and attrition.

In view of the position taken by the SCC in relation to dangerous substances in the CPD, it is recommended to evaluate the consequences of a horizontal approach before embarking on development of new leaching tests for individual types of construction products. In addition it is recommended to evaluate the needs for different materials and different applications of all construction materials from a more generic point of view. Already many tools are available from which methods can be selected. Then based on the identification of needs, appropriate characterization tools can be identified and associated compliance methods selected.
For dense coarse slag, the EN 1744-3 method can provide useful answers for a few specific applications. In case of dense slags with low porosity, wash-off effects may determine the net leaching. The current broad scope needs to be narrowed considerably.
1. INTRODUCTION

Aggregates are used widely in the construction sector in both bound and unbound form. The use of fine and coarse aggregates in concrete is an example of a bound application. Aggregates in structural fill or in road base applications are examples of unbound applications. Any bound application may in the long-term lead to an unbound application in the form of demolition debris. The assessment of environmental impact in such widely different applications requires different test methods, as the mechanisms of release and exposure conditions are not the same [1]. In the case of monolithic materials, such as concrete, diffusion and surface-related processes are important, whereas in unbound application percolation based release dominates.

In CEN/TC 154 Aggregates a new leaching standard [2] has been worked out for judging the environmental properties of aggregates in connection with requirements on dangerous substances in the Construction Products Directive [3]. In meetings of the Standing Committee on the Construction Products Directive (SCC) the need for harmonisation of tests for dangerous substances in construction materials, among which leaching tests, has been stressed [4]. This intention has consequences for the development of standards in the entire construction field. In relation to aggregates, this raises the question to which extent the test developed by CEN/TC 154 is suitable for the problem posed and which limitations the method has in relation to its possible use in a regulatory framework.

Guidance on the development of environmental standards in the field of construction is very limited or lacking, which may very well lead to a situation that standards do not fit the requirements of regulatory bodies. The wider range of environmental standards (composition, asbestos, and radioactivity) is not addressed here.

In this report, the development of the CEN TC 154 standard is addressed in light of development of leaching tests in other areas. In the framework of studies on harmonisation of leaching tests, it has been found that leaching behaviour of constituents is governed by basic rules of chemistry, which are not limited by boundaries set by material definitions. This implies that basic concepts of leaching are generally applicable to soil, waste, sediments, wastes and construction materials [1,5]. The main differences come from specific chemical environment created/dictated by such materials. Organic matter may dominate behaviour of elements in soil and sediments, whereas high pH conditions may be very relevant in many cement-based construction products. In spite of these differences generic behaviour of constituents can be identified.

The key to environmental consequences of application of materials in or on soil is then related to the manner in which materials are exposed to direct infiltration or other means of contact with water. This is generally addressed in a scenario type of approach [1,2,6,7,8]. Recent developments in CEN/TC 292 have led to a first guideline to address the environmental question to be answered and the tools needed to provide that answer [9].

Concerns on the development of the CEN/TC 154 standard have been raised at different occasions [10,11]. The main reasons for this concern were:

− The assumption of reaching equilibrium in 24 hours for particle sizes up to 32 mm is not fulfilled.
− The fraction of fines in the test portion is not addressed as only an upper limit for particle size is given. The results are therefore more a reflection of the proportion of fines in the test portion than of the leaching of the aggregates as a whole.
− Repeatability of testing of the same material.
− Interpretation of results in view of development of regulatory criteria.
− Inconsistency with a current regulatory approach, in which a wide range of construction.
− Materials are covered rather than developing tests for all different construction materials.
The experimental work that forms the basis for EN 1744-3 is rather limited (for instance no validation). As indicated below several aspects have been covered, but it was not possible to investigate all properties of EN 1744-3 that would have been relevant. Therefore tests have been executed, especially concentrating on the following questions:

a. **Verification of the assumption of reaching equilibrium in 24 hours for particle sizes up to 32 mm**
Test results for compliance need to be robust and are only relevant if equilibrium is approached, especially in a one-step test. Release from larger particles will be dominated by diffusion, rather than by percolation or dissolution mechanisms. Diffusion takes more time than the time a one-day test offers; so the test will likely lead to an underestimation of release. Crushing larger particles will increase surface area and so may speed up leaching.

b. **Contribution of different particle size fractions to the leaching result in this test**
What is the influence of fraction of fines in this test and how repeatable and reproducible is the test?
The fraction of fines in the test portion is not addressed clearly, as only an upper limit for particle size is given in the standard. How this condition is checked in practice is not specified. Size reduction of larger particles than 32 mm leads to non-reproducible samples due to the open formulation on the way of size reduction.
Another question depends the fines that may fall through the screen in the test facility. The mechanism of leaching of these fines may change. This change will also depend on the amount of fines that fall through the screen and the way they will behave under the screen.
For materials with a substantial fraction of particles < 2 mm, the results are therefore expected to be more a reflection of the fraction of fines in the test portion and the number of them falling through the screen than of the leaching of the aggregates as a whole.
Based on these considerations, the release of constituents from aggregates may suffer from poor repeatability and between laboratory variability. This aspect can only be verified through experimental work. Materials to be used as aggregates in concrete and asphalt should not be tested separately, but rather as part of a final product.

c. **Consistency of data for the same material class**
How comparable are results for the same material class in subsequent measurements, if too much flexibility is left in the sample preparation? Material from the same class or type can generally give very comparable leaching results. However, if a crucial factor determining the outcome of the test is insufficiently defined results are likely poor or not comparable. From a frequency of testing and quality control point of view this is a less desirable condition.

The information provided in this report addresses some of the critical aspects raised above.
2. EXPERIMENTAL

The experimental work carried out to verify the concerns by the Dutch experts consisted of testing 5 materials by means of the EN 1744-3 procedure with different contact times from 24 hours up to 1 week and different particle size distributions falling within the specification of the standard. Besides the EN-1744-3 test, also a pH dependence test (prEN14429 [12]), which is under development in WG-6 of CEN/TC292, was carried out to illustrate the differences in testing conditions relative to pH. In addition, a percolation test (prEN14405 [13], comparable to NEN 7343 [14]) and the batch shake test (EN 12457-2 [15]) test were carried out. The pH dependence test and the percolation test are so called characterisation tests, in which the release is measured as a function of respectively the pH and the L/S ratio. The latter can be related to a time scale based on infiltration rate, density of the material and the height of application. In both cases the release is determined at several (7 or 8) points. The other two tests (EN 1744-3 and EN 12457-2) are single batch tests, in which the release is measured at one test condition only.

The material selected for testing were:
- Phosphorous slag (coded P slag)
- Steel slag
- Municipal solid waste incinerator bottom ash (MSWI BA)
- Construction and demolition waste (C&DW)
- Artificial aggregate (Aggregate).

These materials represent a range of relatively porous and relatively low porosity materials.

The materials were prepared in a particle size meeting the requirements for the standard (< 32mm). The artificial aggregate material was partially size reduced using a jaw crusher to constitute a fraction of fines representing about 10 % of the total mass at particle size < 4 mm. MSWI bottom ash, C&D debris, P slag and steel slag were tested as received and after removal of the fraction < 4 mm. The slag samples had an original size distribution 4 – 16 mm. This, however, does not imply there are no fines in the test portion. The particle size specifications for EN 1744-3 are given in table 1.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Particle size distribution of test materials for EN 1744-3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Material</td>
<td>Notification</td>
</tr>
<tr>
<td>P slag</td>
<td>Coarse</td>
</tr>
<tr>
<td></td>
<td>Fine</td>
</tr>
<tr>
<td>Steel slag</td>
<td>Coarse</td>
</tr>
<tr>
<td></td>
<td>Fine</td>
</tr>
<tr>
<td>MSWI bottom ash</td>
<td>Coarse</td>
</tr>
<tr>
<td></td>
<td>Fine</td>
</tr>
<tr>
<td>C&amp;D waste</td>
<td>Coarse</td>
</tr>
<tr>
<td></td>
<td>Fine</td>
</tr>
<tr>
<td>Aggregate</td>
<td>Coarse</td>
</tr>
<tr>
<td></td>
<td>Fine</td>
</tr>
</tbody>
</table>

To illustrate the important role of different particle sizes on leaching data from an earlier study [16] on three of the five material types studied here are compared with the pH dependence test. This comparison shows that the materials studied then and now have the same leaching...
behaviour and thus the data on leaching of different particle sizes can be used to evaluate the role of different particle size distributions on leaching.
3. RESULTS AND DISCUSSION

3.1 Contact time in prEN1744-3

In Appendix A, test data for EN 1744-3 are presented for the 5 materials studied after 24 hours (as specified in the standard), 48 and a long contact time of 7 days. In the same figures data are presented for two different size distributions of the same material (coarse material and same material with an increased level of fine particles – see table 1 for particle sizes).

From the results, it follows that in general the concentrations of leached constituents increases significantly with longer exposure time (till often more than 50 % in 7 days) for materials with the same particle size distribution between test portions.

An even more significant difference exists between the “coarse” material and the samples with increased “fines”. This shows that it is rather important to be unambiguous and representative in the amount of fines and the size distribution used in the test. Apparently, a factor of 2 differences between two supposedly the same samples can easily occur.

These figures also indicate that on the short term (1 day) leaching of the fines will directly dominate the height of the leaching values, but the leaching of bigger particles by diffusion needs more time [16,17,18]. On the longer term (days till weeks) the larger particles may contribute significantly (by diffusion from larger particles) to the long term leaching level. In applications of unbound aggregates there is sufficient time for this slow process of release to make a contribution to the environmental impact.

3.2 Leaching as a function of pH

In Appendix B, pH dependence test data are given for all five materials, in which data are compared with EN 1744-3 data and further a comparison is made with prEN 12457-2 (shake batch test L/S=10, d <4 mm). This pH dependence test shows the leaching behaviour at L/S=10 at controlled pH ranging from pH=2 till pH = 12 using material size reduced to < 2 mm. The batch test at each pH level takes 48 hours. It gives an impression of the level of leaching that may be expected at different environmental pH conditions (the leaching level corresponds to the specified conditions). In the judgement of aggregate use, particularly coarse aggregate, exposure to the atmosphere and/or soil atmosphere is very likely. Due to the large voids between the particles, uptake of carbon dioxide from the atmosphere and/or from decaying organic matter in the soil is relatively fast; particularly since diffusion in air is 5 orders of magnitude faster than in water. This then leads to neutral conditions at the exposed surface of the materials. In many cases, the leachability at neutral pH is significantly different from that under high pH conditions often found in non-natural aggregates.

The release at a given L/S can be related to a time scale through the relationship between L/S, the infiltration rate, the density of the material and the height of the application under consideration. Generally, in an uncovered application of limited height L/S=10 may correspond to about 100 years of leaching in practice. Coarse granular alkaline materials used in for instance road application [19] will generally be carbonated fairly rapidly, which implies that the leaching conditions may be governed rather by a neutral than a high pH for most of the life time of the material. The pH dependence test provides the means to define an envelope of environmental conditions relevant for the material in a specific application.

It is shown that the leaching level of the prEN 12457-2 batch test is performing mostly on about the same level as the pH-dependence leaching test. The leaching of most components in the prEN1744-3 tests often appears to be much lower, often a factor 2-10. For some constituents
leaching performs on the same level. It may be expected that if the differences are high, leaching will increase if more time will be given to the leaching process in the prEN1744-3 test. Delayed release from coarse granular material by diffusion, which is not completed in 24 hours will lead for all constituents to higher release levels than reflected by the prEN 1744-3 test.

In case elements leach in manner that is largely solubility controlled, then results of different leaching tests may prove to be very similar as the concentration at which solubility control sets in may be reached fairly rapidly. Examples are of this are: V, Zn and Cu in C&D waste, Cr, Cd and Ba in MSWI bottom ash.

3.3 Percolation test

In the figures given in Appendix B the leaching with the column test NEN 7343 [14] is presented at L/S=10 (Cumulative leaching) only, to make leaching data comparable. The test results from this procedure are comparable with the percolation test under development in CEN/TC 292 (prEN14405 [13]). This test is more generally applicable as it is now also standardized for soil (ISO TC 190 [20]).

For the same pH condition, the leaching in the pH-dependence leaching test (prEN14429), the batch test (EN 12457-2) and the percolation test are quite comparable for many constituents. Sometimes the figures show that in the column test at own pH of the material performs at a very high pH-level, which even exceeds the levels investigated in the pH-dependence test and measured in the prEN 12457-2. This means that under percolation conditions a high pH-level of leaching may be expected in the field, at least in the beginning of the leaching process, resulting for some elements in an increase in release and for other elements in a decrease in release.

3.4 Particle size distributions of aggregates

Particle size issues have several aspects, which lead often to conflicting requirements. This implies that a choice needs to be made on preferred conditions of testing. Size reduction is advantageous for obtaining more repeatable test results. In addition, smaller vessels can be used for testing.

Size reduction of large particles may lead to exposure of fresh surfaces to leaching, which may lead to a deviation from practice in terms of carbonation and oxidation state. The latter aspect is particularly relevant for non-porous materials, such as industrial slag. Most materials used in road construction are relatively porous and in that case the effects are much less pronounced.

From the results presented in Appendix A it is clear that the difference between the materials as received and material with a different fraction of fines (< 4 mm) is significant for all materials and most elements.

Due to the choice of test conditions the field of application of EN 1744-3 is primarily focussed on non-porous materials. When a material contains a significant fraction of fines, then size reduction will generally not have a major effect on the leaching result. For larger particle sizes, diffusion from the interior of particles contributes significantly to the overall release. Since this process is slow, the testing time generally needs to be much longer than 24 hours. From the viewpoint of practical use of the information, one would rather not size-reduce. Size reduction due to transport and handling may occur and may affect the test results. However, the testing time needs to be rather long to compensate for the slow matrix diffusion. For practicality reasons it may therefore be better to reduce the size of the largest particles.

In the pH-dependence test the particle size is reduced to a size < 2 mm. In the percolation test and in the prEN 12457-2 the size is reduced to < 4 mm. (In the prEN 12457-1,2 and 3 the size reduction is < 4 mm, whereas for EN 12457-4 the particle size is < 10 mm). The advantage of size reduction to less than 4 mm is that a representative sample can be produced easier than without size reduction. In addition, the quantity of sample and the size of the leaching apparatus can be smaller. The procedure is still representative. When using these sizes the leaching will
not be dominated by diffusion, so specific information is obtained about leaching dominated by percolation and solubility control.

Many materials have particle sizes, which are normally below 4 mm or 10 mm, such as clay, sand, fly ash, etc. The specifications for aggregates to be used as unbound and hydraulically bound material for road base applications require relatively fine-grained material as specified in prEN 13242 [21]. This standard specifies that at least 15 – 40 % of the material used in that application must be smaller than 4 mm. In the present study only about 10 % fines was used. Larger amounts of fines would lead to results with a closer resemblance to EN 12457 type test results. Fines in the sample may have the effect that leaching in the 1744 – 3 test is more diffusion controlled than it should be.

Development of the prEN 1744-3 was among other reasons based on the thought that size reduction will increase the surface area, which will influence the leaching rate too much by exposing a relatively larger surface to the leachant. It is also considered that leaching may be influenced by carbonisation or other chemical reactions on the surface of particles. Size reduction may create new fresh surface area, which may lead to a different leaching behaviour. Size reduction (comparison between EN 12457 and 1744-3) can lead to important differences in eluate pH and redox conditions and consequently to differences in leaching behaviour. For example, P slag may show more than 1 pH unit difference between size reduced and non-size reduced material.

These aspects of chemical exposure conditions are true for some types of materials [22], especially for certain kinds of slag with a very low porosity. Many materials considered as aggregates for applications relevant to TC 154 have a high porosity, so there is no real difference between the longer term release of small (0 - 10 mm) and bigger particles (0 - 40 mm). The EN 1744-3 therefore gives a wrong impression of release in this case. The leaching of the bigger particles will only take longer. After a slightly longer period, the total leaching effect will be about the same as the effect of leaching of small particles. Only if the materials are much bigger (> 50 - 100 mm) leaching time will be such that it is important to understand the diffusion process better. For such big particles a tank leaching test should be carried out, since a batch test and a test like prEN1744-3 do not provide this information.

This means that the prEN 1744-3 may give a better performance for especially larger particles of low porosity materials, like as some kinds of industrial slag’s (e.g. P-slag, steel slag). However, if batches of these slag’s contain some % of fines as well, the difference will be much lower. In these cases it should also be considered that the prEN1744-3 only gives a 24 hours leaching performance. It doesn’t give insight in the leaching risks on the longer term, since it provides no information on the diffusion rate and so the data from this test cannot be extrapolated due the arbitrary nature of the method.

From previous studies on batch leaching test development [16] information on the influence of particle size can be obtained for the same materials as those studied here using the prEN 1744-3 test. In Appendix C the batch test data for P slag, cement stabilized MSWI fly ash and MSWI bottom ash are given for a range of elements and a wide variety of particle size ranging from 0 - 0.2 up to 40 mm. These data obtained a few years ago on similar materials are compared with data from the present study. The materials leaching behaviour is quite similar, therefore the results obtained in the previous work are relevant here. In Appendix C the pH dependence test data are used to illustrate that the corresponding materials studied in the previous work and in this work have the same leaching behaviour. Thus observations on particle size effects from the previous work are equally relevant for this work. For the stabilized MSWI fly ash (low waste loading) the comparison with cement mortar fails for some contaminants high in fly ash (Cd, Zn, Pb). However, for many other constituents the leachability is strikingly similar.
Following the pH dependence test data for each material, bar graphs illustrate the effect of particle size distributions on leaching in a batch test (L/S=10). In a batch test, the contact between particles and leachant is intensified by stirring or end-over-end tumbling. In case of larger particles, slow release by diffusion out of larger particles is a main release restriction. The observed effects of diffusion restrictions in a batch test with liquid solid agitation will most likely be enhanced in a test like EN 1744-3. In addition, the notion of increased leaching with smaller particle sizes is a misconception, as for some element-material combinations leachability decreases with decreasing particle size. This is caused by a significant pH difference after particle size reduction, which is associated with a decrease in leachability of specific constituents (examples V and Zn in MSWI bottom ash; Si, Al, SO₄ and V in cement-based products).

Besides the release restriction by diffusion out of larger particles in prEN 1744-3, the placement of the mass to be leached in a pile means that leached components need to diffuse to the bulk solution, which depending on the packing in the pile, implies an additional release restriction. The manner, in which the EN 1744-3 test operates, is therefore to a large extent, focussed on diffusion restrictions.

Summarizing, in table 2 the main differences between EN 12457-4 and EN 1744-3 are given.

Table 2  Differences in leaching conditions between EN 12457-4 and EN 1744-3

<table>
<thead>
<tr>
<th>Test</th>
<th>Agitation</th>
<th>Release process</th>
<th>Equilibrium</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch shake test EN 12457</td>
<td>Intense interaction</td>
<td>Contribution to release by diffusion from the larger particles (up to 10mm in Part 4) is limited due to the short testing time.</td>
<td>Equilibrium approached as evidenced by test results</td>
</tr>
<tr>
<td>series</td>
<td>between leachant and all</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>particles in suspension.</td>
<td></td>
<td></td>
</tr>
<tr>
<td>EN 1744-3</td>
<td>Stirring of surrounding</td>
<td>Contribution to release by diffusion from large particles is limited due to the short testing time and by the placement of particles in a compact heap.</td>
<td>Equilibrium between external leachant and inner core of particle heap not achieved in testing time</td>
</tr>
<tr>
<td></td>
<td>leachant only.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3.5 Diffusion rate from particles

Based on mobility data for relatively mobile constituents and less mobile species, the time needed for full exchange between (mobile) constituents in a particle of specific size with the surrounding solution can be estimated. If we assume an effective diffusion coefficient (Dₑ) for mobile species such as salts of 10⁻¹⁰ m²/s and a value of 3.10⁻¹² m²/s for retained species, then the time it takes to reach equilibrium between the inside of the particles and the surrounding solution can be estimated (x = √(2Dₑt)).

<table>
<thead>
<tr>
<th>Particle size (mm)</th>
<th>Mobile (10⁻¹⁰ m²/s)</th>
<th>Retained (3.10⁻¹² m²/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Time for equilibrium (days)</td>
<td>Time for equilibrium (days)</td>
</tr>
<tr>
<td>4</td>
<td>29</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>184</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>736</td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>2942</td>
<td></td>
</tr>
</tbody>
</table>
It follows that with particles sizes in excess of 4 mm it will take very long before equilibrium is established. In the field, however, this time is no limitation at all.

### 3.6 Relevant test methods for leaching

The characterization leaching tests developed in CEN/TC 292 are generic characterizations leaching tests that address leaching behaviour of granular materials. From the EU project Harmonization of Leaching/Extraction Tests [1,5] it follows that these test are more generally applicable than just to waste materials. The pH dependence test gives the relationship between the release at the own pH of the materials and at pH conditions that may be imposed on the material by the surroundings. The percolation test reflects the percolation behaviour of granular material, in which local equilibrium in the percolate from the column is likely to occur. These methods are equally relevant for aggregates as well as for soil or granular waste materials. For large monolithic materials, diffusion from the interior is the main release controlling process. This can be assessed in a tank leach test, which has been developed for construction materials and has been applied for the evaluation of release from concrete. The prEN 1744-3 in fact is a mix between phenomena assessed in a percolation test and those assessed in a tank leach test, and therefore it is neither representative for one nor the other situation. The contribution of both mechanisms of release (percolation and diffusion) in the end result is uncontrolled. Besides, the duration of the test is too short to see any changes in course of time.

### 3.7 Consistency of data for the same material class

Based on the results presented in Appendix C on the various materials, a significant data consistency can be noted for the same type materials being tested at different times. This type of data consistency is very helpful in deciding whether material tested deviates from normal behaviour or not. Allowing a wide scatter in particle sizes in a test creates significant differences in test results, which can not be linked to the material class or type anymore. In figure 1 this situation is illustrated for cement mortars, where the pH dependence data for widely different cement mortars are compared with compliance test results (batch shake test) on size reduced materials with poorly defined or undefined particle size. The pH dependence test data show a very consistent pattern, whereas the single test data are an order of magnitude lower and more scattered. If the test were run at the same particle size the results would have been internally consistent. In another manner allowance can be made for particle size effects on leaching occurring in practice. Results as obtained with EN 1744-3 would show even greater scatter as the diffusion from within the pile of particles to the surrounding solution is an additional delaying factor compared with a batch test. Better control over the particle size distribution in a leaching tests is a prerequisite for repeatable and reproducible testing.
Figure 1  Comparison of pH dependence leaching test data on cement mortars (size reduced to < 4 mm) with “concise” compliance test [27] data on material < 10 mm with undefined size distribution. Sample codes correspond with mortar samples from different origin.
4. RELEVANT ASPECTS CONCERNING THE DEVELOPMENT OF LEACHING TESTS FOR CONSTRUCTION MATERIALS

The development of leaching tests for aggregates can be placed in the broader perspective of leaching test development for other construction materials and related materials.

4.1 List of Harmonisation projects and other relevant international projects

In recent years, large international research projects have been carried out in the framework of EU DG Research aimed at better understanding of the leaching process in a variety of construction materials and related materials. Harmonisation of test methods across a wide range of materials has been studied (SMT4-CT96-2066 [5]) as well as validation of compliance test methods as specified in EN 12457 (CEN/TC 292 and CEN /TC 308 validation [23]). Some relevant new projects addressing these aspects will be started soon. For further selection and development of tests it is important to use the information from these projects as much as possible to avoid unnecessary duplication of work. Besides, it is important to check whether the results of newly developed tests are comparable with the results of other leaching tests, especially the more generic characterisation tests.

For an impression of these projects some of these projects are listed here:
- Technical work in support of the Network Harmonisation of Leaching/Extraction Test (SMT4-CT96-2066 [5])
- Utilising innovative kiln technology to recycle waste into synthetic aggregate BRST-CT98-5234 Report ECN – C-01-014 [24]
- ECRICEM I – Industry funded project on concrete prepared from alternative raw materials and alternative fuels in cement ECN-C-01-069 [25].
- ALTMAT (finalized in 2000)
- EU projects in preparation:
  - ECO-SERVE - FP 5 Concrete, blended concrete, asphalt (Kick-off in November 2002)
  - SAMARIS – FP 5 Paving materials (Contract preparation).

4.2 CPD – Product families, technical specifications and leaching tests

The Construction Products Directive was published in 1989 [3]. This Directive gives the framework for development of Technical Specifications for construction products. It was decided that for 40 ‘families’ of construction products Technical Specifications should be developed.

It was decided that the Technical Specifications should be developed according to the ‘New Approach’. This means that the technical specifications will be developed by organisations as CEN and EOTA. After 1989 first documents were made on 6 main criteria by the Commission. Then Mandates were given, mainly to CEN and EOTA
Some of these Technical Specifications are ready and published. Others will be published and will come into force in 2002.

The Technical Specifications contain test methods. Producers of products should use these test methods and should give the relevant information on the topics that are tested. If they do so they are allowed to bring their products on the market, with a CE-mark. If the product doesn’t
comply with the Technical Specification, the producer is not allowed to bring the product on the market.
In the Technical specifications, in general, no limit values are given. Member States, other authorities or users may set limit values. In some cases quality classes are given in the Technical specifications. In these cases, Member States, other authorities and users may choose from these classes.
The process of development of this directive and the Technical Specifications are guided by the Standing Committee on the Construction Products Directive (SCC.) The CPD is developed by DG-Enterprise.

It was decided in the SCC that for ‘dangerous substances’ a special procedure will be followed. It appeared to be too complicated to introduce this completely in the Technical Specifications within the available time frame. It was felt not adequate to develop test methods for these materials in each standard individually by each of the CEN-TC’s. Most of them didn’t feel equipped for this. It was discussed that a horizontal approach would be appropriate to solve this problem. Test methods should be developed in co-operation and then being used by all relevant TC’s in the relevant Technical Specifications. This was discussed in a workshop of 7 November 2000 in Vienna [4], by the commission, member states and industry.

It was decided that in the relevant Technical Specifications (as far as was felt necessary) could and would be indicated that on this point the actual legislation and limit setting and relevant test methods in the Member States will stay in force. In the SCC, it is investigated in co-operation with CEN how to proceed in organising this horizontal development of test methods further. The results of this horizontal development of test methods can be introduced in the next generation of the Technical Specification (e.g. in the revisions, which may be developed within about 5 years after publication of the Technical Specifications).

CEN/ TC 154 has completed the leaching test (EN 1744-3), which has been put up for formal vote early in 2002. In spite of the concerns, it was formally approved. If the EN-1744-3 is advised for dealing with dangerous substances in Aggregates and may even be suggested for other applications for which it is not validated, such a step now would complicate further horizontal development of test methods in the filed of construction in Europe. It would burden many authorities, producers and others with a new test they don’t know yet. They would have to change their procedures and limit setting now and retest all their materials. Further developments on a horizontal approach may lead subsequently to changes in selection of test methods by further development. This would lead again to introduction of other tests or changes in tests and would burden all the concerned organisations again.

It must be considered as well that in the field of EU environmental legislation test methods on leaching are necessary as well. In the existing EU-Landfill Directive [26], the Commission has the task to set standards by July 2002 and to select test methods. The Commission (DG-Environment) has asked CEN to continue development of standards for at least a number of Directives in the field of the Environment in a horizontal and more efficient way. (No repeated development of the same tests in different area). It would make sense to set a further step in horizontal development of tests on these environmental aspects in CEN. It would be of great help for the process to set a program for this by CEN and the Commission. It may be discussed then how to plan the development of these tests, how to discuss, select and validate test methods and how to act in the mean time in these cases where it is strictly needed to start with some (preliminary) sets of new tests.

As mentioned earlier in this document, leaching properties of many materials are primarily dependent on the matrix and not on the status of the product. In the fields of use and reuse of materials and in the field of waste management many environmental aspects can be adequately tested by the same tests. So it is proposed to include this in further discussions and planning of horizontal approaches. It should be discussed how CEN standards can get a broad scope without
being connected to one specific area or Technical Committee even when preparations are mandated to one TC or some co-operating TC’s. Several solutions may be possible for questions in this area. In relation to a first horizontal standardization activity in the field of sludge, soil and treated biowaste in support of EU Directives in these fields, discussions at CEN level have led to agreement at BT level how to proceed with horizontal standards in the largely vertically organized CEN structure.

4.3 Life cycles of construction products

Aggregates as addressed in CEN/TC 154 may be of natural origin, in the case that the material is being excavated and crushed to meet the particle size specifications for use, or be derived from alternative materials. This may be either from recycling of sorted demolition waste or from slag generated in industrial processes (steel slag, phosphorus slag) or from waste incineration (incinerator bottom ash). The environmental aspects of the materials are different and relate to the possibility of release of undesired constituents to the environment. This aspect is relevant for all materials, only the level of release can be different for non-natural materials. In an evaluation of environmental impact not only the immediate future is at stake, but also the long-term behaviour of the materials under consideration. If a material behaves properly in the short term, but performs badly in recycling and as such creates limitations in the long-term, it is appropriate to reconsider its use. In figure 2 an evaluation of the life cycle of construction products in relation to potential release of dangerous substances is given.

![Life cycle of construction products diagram](image)

**Figure 2** Integration of test methods for environmental quality at different stages of the Building Cycle

In evaluating aggregate use in fields other than road construction, aspects such as incorporation of aggregates in cement leads to different scenarios of use, reuse and recycling, which can impossibly be addressed by the method proposed by CEN TC 154.

It is crucial for the evaluation of construction products to use a scenario type of approach, in which a generic basis provides the relevant background to select appropriate conditions for testing materials in specific applications. A hierarchy in testing is very useful, as the characterization provides the relevant background information to make decisions on quick tests suitable for compliance purposes [1,5]. However, it is of major importance that the quick tests /
compliance tests are linked to the generic characterisation tests. For the test proposed by CEN TC 154 this has not been demonstrated.

In the framework of an EU project on artificial aggregates [24] different scenarios of use were evaluated and the methods associated with them identified. In table 3 the results are given.

Table 3  Applications and test use

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Stage</th>
<th>Characterisation</th>
<th>Compliance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Aggregate production</td>
<td>Production and quality</td>
<td>pH dependence test on size reduced material</td>
<td>Compliance batch test at own and imposed neutral pH (L/S=10) after size reduction</td>
</tr>
<tr>
<td>Aggregate in temporary storage</td>
<td>Service life</td>
<td>Percollation test</td>
<td>Compliance leaching test (L/S=2, own pH)</td>
</tr>
<tr>
<td>Aggregate in concrete in contact with surface water and constructions on land (pilars, quays, breakwaters, locks)</td>
<td>Service life</td>
<td>Tank leach test (imposed neutral pH) *</td>
<td>Monolith compliance test (imposed neutral pH) *</td>
</tr>
<tr>
<td>Aggregate in concrete in contact with groundwater (pilings, walls)</td>
<td>Service life</td>
<td>Tank leach test at own pH</td>
<td>Monolith compliance test at own pH</td>
</tr>
<tr>
<td>Aggregate in concrete in contact with sea water (oil rigs, quays, breakwaters)</td>
<td>Service life</td>
<td>Tank leach test (seawater)</td>
<td>Monolith compliance test (seawater)</td>
</tr>
<tr>
<td>Aggregate in concrete from demolition recycled as aggregate for concrete</td>
<td>Recycling</td>
<td>Tank leach test (imposed neutral pH) *</td>
<td>Monolith compliance test (imposed neutral pH) *</td>
</tr>
<tr>
<td>Aggregate in concrete from demolition reused unbound in roadbase or embankment</td>
<td>Reuse</td>
<td>pH dependence test on crushed material and percolation test</td>
<td>Batch test (L/S=10) at own pH and imposed neutral pH using crushed material</td>
</tr>
<tr>
<td>Aggregate in road base from demolition reused in road base of embankment</td>
<td>Recycling</td>
<td>pH dependence test on crushed material and percolation test</td>
<td>Batch test (L/S=10) at own pH and imposed neutral pH using crushed material</td>
</tr>
<tr>
<td>Aggregate in concrete or in road base from demolition going to landfill</td>
<td>Disposal End-of-life</td>
<td>pH dependence test on crushed material and percolation test</td>
<td>Batch test (L/S=10) at imposed neutral pH using crushed material</td>
</tr>
</tbody>
</table>

* Most application on land or in contact with surface waters are exposed to neutral pH conditions due to either neutralization from CO2 in the air or pH buffering in surface water.

The table covers fields relevant to TC 154 as well as TC 227 (aggregate in concrete). The methods specified are also consistent with recent work on cement mortars [25]. The Dutch Building Materials Decree [28] addresses release from construction products in a more generic manner consistent with a scenario type of approach.
5. EN 1744-3 PERFORMANCE

The arbitrary choice of the contact time in EN 1744-3, without taking the role of the particle size specified in the EN 1744-3 standard into consideration, leads to results which do not reflect equilibrium nor a well-defined stable end point.

In view of the upper limit of the particle size specified in the standard (d < 32 mm), the particle size distribution in the test portion is not sufficiently defined. A small percentage of fines in the test portion can significantly change the leaching results, which will likely lead to poor repeatability and reproducibility of the standard. The specification of < 32 mm particle size is thus not sufficiently precise.

Major difference in leachability will occur as a result of different size distributions all meeting the specification in the standard. Experimental work points at significant differences between the “coarse” material and the samples with increased “fines”, which still fall within the specification of the standard. A factor of 2 difference between two supposedly the same samples can easily be obtained. These concerns relate most strongly to relatively non-porous aggregates, which are most sensitive to the mentioned factors (particle size, contact time). Size reduction for these materials would be justified, as "equilibrium” will not be reached in 24 hours. After size reduction a test of 24 hours is suitable. Alternatively, the test should last longer, when such large particles up to 32mm are allowed.

The 2-mm openings in the bucket leads to fines falling into the stirring section of the apparatus proposed. This leads to disproportionate leaching of such fines relative to the bulk material within the bucket.

From a leaching point of view it is important to realize that when leaching is governed by solubility control the same leaching may be obtained in spite of significant differences in particle size distribution. This is inherent to the leaching process. To the contrary, for the more mobile contaminants the largest differences in release can be expected, as these are affected most by the slow diffusion out of coarser particles.

Another aspect related to the chemistry of leaching is connected to the leaching behaviour as a function of pH and redox (slag). Differences in pH and / or redox, as a result of differences in particle size distribution, may lead to significant differences in leachability.

Not in all cases will size reduction lead to higher release. Due to pH change as a result of size reduction, leachability may in fact decrease as particle size decreases. An example of this is Mg in P slag. Another example is sulphate leachability from C&D waste, where the coarse material has a lower pH and higher sulphate leachability. This is consistent with the leaching behaviour of concrete.

For dense (low internal porosity) slag type materials, the particle size is a crucial parameter as release from these materials is surface area related and size reduction would lead to changes in material condition. For instance size reduced steels slag and phosphorus slag would lead to a much higher pH and possibly reducing conditions, which lead to significantly different leaching results than relevant in the field with mostly oxidised conditions and a more neutral pH as a result of carbonation. Size reduction in that situation may not be desirable. However, in case of larger particles a longer contact time is needed to reach stable end conditions in the test.

Material used in road construction, which is one of the aimed fields of application of the standard, has to meet technical specifications, which stipulate the use of well graded material
containing a significant amount of fines (15 –40 % of materials < 4 mm, see prEN13242). This implies that aggregates, used in that type of specification, are not adequately tested by the EN 1744-3 method, because of the fact that the leaching process will predominantly be ruled by the fine fraction.

The field of application of the standard is too broad. As a minimum, it needs to be narrowed down and described more precisely. It is definitely not suitable for judging aggregates to be used as aggregate in concrete (work field of TC 227).

For the evaluation of environmental aspects of aggregates in the framework of TC 154 or in that of TC 227 a scenario approach is recommended, in which the relevant use and associated exposure conditions are addressed. A hierarchy in testing is advised where, based on the evaluation of the intended use of the material, and based on more elaborate testing, a limited selection of suitable methods is defined for compliance. Such methods will not be limited for use in the field of aggregates, but be more generally applicable in the field of construction applications.

A life cycle evaluation, which is currently not addressed in the CPD, is important from an environmental point of view, as long-term behaviour of materials in subsequent stages of its life cycle needs to be addressed. If the use of alternative materials is not critical in primary use, but is definitely critical in a size-reduced form as unbound aggregate, then one may want to reconsider such application.
6. CONCLUSIONS REGARDING EN 1744-3

Leaching characteristics
The EN 1744-3 method does not reflect leaching behaviour in a specified scenario, nor does it give insight in leaching behaviour of aggregates. The method underestimates release in practice. It is not possible to relate test data to exposure risks. Due to poor definition of particle size the repeatability is questionable. In terms of release, the method provides an undefined mix between dissolution and diffusion phenomena.

Quality check on the test by developers of the test
The background information provided as basis for justification of choices on test conditions is very limited. General leaching principles are not discussed or only in a very limited way. An (open) European validation, even with a limited number of laboratories has not been performed and/or reported. No comparison with other similar tests has been made. A discussion with leaching experts from Member states has not taken place.

Function of the test as ‘compliance test’
The method has been classified as compliance test. It is not clear, however, to which method the test complies. As a clearly defined scenario as basis for the method is lacking, it is not clear how the results can be used in environmental risk evaluation, which is the basis for regulation. Therefore, it is not clear how to relate test results to actual or new legislation.

Consequences
The scope of the method is formulated too widely. If this test is improperly recommended, producers may have to test several products again, which would lead to double cost. The relevance of test results for the wide scope is questionable from an environmental impact point of view. If the method is implemented as part of the CPD requirements on dangerous substances, EU Member states will have to use this test and come up with limit values without an adequate basis for risk assessment. The place of the test relative to tests for other construction materials such as concrete, bricks and the like is not clear. In addition, specific situations as redox changes and external pH influences are not properly dealt with. In view of the requirements, the application of the test is questionable for use of aggregates as unbound and hydraulically bound material for road base application. For those applications the specification of prEN 13242 requires that at least 15 – 40 % of the material must be smaller than 4 mm.

In view of the position taken by the SCC in relation to dangerous substances in the CPD, it is recommended to evaluate the consequences of a horizontal approach before embarking on development of new leaching tests for individual types of construction products. In addition, it is recommended to evaluate the needs for different materials and different applications of all construction materials from a more generic point of view. Already many tools are available from which methods can be selected. Then based on the identification of needs, appropriate characterization tools can be identified, and associated compliance methods selected.
REFERENCES


[14] NEN (1995): Leaching characteristics of solid earthy and stony building and waste materials; Leaching tests; Determination of the leaching of inorganic components from granular materials with the column test, Dutch standard NEN 7343, NNI.


APPENDIX A  EN 1744-3 DATA WITH LONGER CONTACT TIME AND DIFFERENT PARTICLE SIZE DISTRIBUTIONS

EN 1744-3 carried out on synthetic aggregate, MSWI bottom ash and demolition debris in two particle size distributions for each material at different contact times for elements Al, B, Ba and Ca.
EN 1744-3 carried out on synthetic aggregate, MSWI bottom ash and demolition debris in two particle size distributions for each material at different contact times for elements Cr, Cu, K and Li.
EN 1744-3 carried out on synthetic aggregate, MSWI bottom ash and demolition debris in two particle size distributions for each material at different contact times for elements Mg, Mo, Na and SO4.
EN 1744-3 carried out on synthetic aggregate, MSWI bottom ash and demolition debris in two particle size distributions for each material at different contact times for elements Si, Sr, EC and pH.
EN 1744-3 carried out on steel slag and P-slag in two particle size distributions for each material at different contact times for elements Al, B and Ba.
EN 1744-3 carried out on steel slag and P-slag in two particle size distributions for each material at different contact times for elements Pb, S and Si.
EN 1744-3 carried out on steel slag and P-slag in two particle size distributions for each material at different contact times for elements Ca, Cr and Fe.
EN 1744-3 carried out on steel slag and P-slag in two particle size distributions for each material at different contact times for elements V, EC and pH.
APPENDIX B PH DEPENDENCE TEST DATA (PREN 14429) IN COMPARISON WITH EN 1744-3 DATA (ON COARSE AND FINE MATERIAL) AND EN 12457-2 DATA (CEN TC 292)

Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for MSWI bottom ash on K, Ba, B, Cd, Cr and Cu.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for MSWI bottom ash on Mo, SO₄ as S, Pb, Sb, V and Zn.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for C&D waste on Cr, Cu, SO₄ as S, K, V and Zn.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for steel slag on Cr, B, Ba, SO$_4$ as S, K, and V.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for steel slag on Mg, Na, P, Ca, Si and Sr.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for phosphorus slag on Al, As, B, Ba, Ca and Mg.
Single step leaching tests EN 1744-3 (CEN TC 154) and EN 12457-2 (CEN TC 292) with the pH dependence test (prEN 14429) for phosphorus slag on SO4 as S, V, K, P, Si and Sr.
APPENDIX C   PARTICLE SIZE EFFECTS

Illustration of similarity of in leaching behaviour by pH dependence test data for MSWI bottom ash from previous work [16] and this work.

- **B**
- **K**
- **Ni**
- **Sb**
- **Mg**
- **Ca**
Effects of the particle size distribution on leaching in a single batch test (L/S=10, T=24 hrs) for MSWI incinerator bottom ash.
pH dependence test data for P slag from previous work [16] and this work.

- **As**
- **Al**
- **Cu**
- **Si**
- **Cr**
- **Mn**
Effects of the particle size distribution on leaching in a single batch test (L/S=10, T=24 hrs) for P slag.
pH dependence test data for cement based products from previous work [16] and this work.
Effects of the particle size distribution on leaching in a single batch test (L/S=10, T=24 hrs) for cement based product (Stabilized MSWI fly ash at 10 % addition to a regular mortar mix; CS : 39 N/mm²).

[Graph showing leaching of various elements such as Si, Al, SO₄ as S, Pb, Mo, Li, Na, and K across different size ranges.]