# LEACHING BEHAVIOUR OF ARTIFICIAL AGGREGATES EU project BRST-CT98-5234

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## Acknowledgement/Preface

This work has been carried out in co-operation with the University of Leeds (UK) as co-ordinator of EU CRAFT project registered under contract number BRST-CT98-5234.

## Abstract

In the framework of EU project "Utilising innovative kiln technology to recycle waste into synthetic aggregate" (BRST-CT98-5234), the leaching behaviour of artificial aggregates has been studied to assess its environmental compatibility in the various stages of its use. Since the conditions are very different for the different applications, the assessment calls for a variety of different leaching test conditions. The pH dependence test is used to cover important differences in pH environment to which the materials are exposed to as well as for an assessment of the acid buffering capacity of the material. It is found that artificial aggregate has a low buffer capacity, which makes it sensitive to externally imposed pH conditions. Utilisation and storage exposed to acidic conditions needs to be avoided. The results of the pH dependence test and column leaching test are mutually consistent while the CEN TC 154 method systematically provides lower values.

The artificial aggregates studied here will not adversely affect the concrete in its service life. The main issue for aggregate use is the recycling and the "end of life" condition, when the material becomes construction debris. Not metals, but oxyanions, such as Cr VI and Mo are most relevant under the recycling, reuse and "end-of life" condition. This is based on the criteria set by the Dutch Building Materials Decree as this is the only legislation available to make such an environmental judgement.

A concise test has been applied to assess crucial aspects of leaching for different production mixes. Also concluded is that results from the CEN TC 154 test are different from more elaborate characterisation tests. Alternative compliance tests that do give consistent results with the more elaborate characterisation tests are available.

Keywords: Leaching, aggregates, modelling, long term behaviour, trace elements.

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## 1. INTRODUCTION

In the framework of EU project Utilising Innovative Kiln Technology to Recycle Waste into Synthetic Aggregate (BRST-CT98-5234), a new kiln design (Trefoil) with a more efficient energy use is applied, which has a high flexibility to process different waste materials as feedstock [1,2]. The starting materials and the artificial aggregates were produced at the School of Civil Engineering (Leeds) and at the plant of RMC Tilbury Aggregates, a facility in Tilbury (Essex, UK). The artificial aggregates have been tested for their leaching behaviour to assess their environmental compatibility in the various stages of their use.

Therefore emphasis is placed on the following stages of the aggregate life cycle:

- Exposure of artificial aggregate to leaching in temporary storage
- Exposure of concrete containing artificial aggregate ("primary" application or service life).
- Recycling of construction debris as aggregates in new concrete ("primary" application or service life)
- Reuse of construction debris in road construction, dam fillings etc. ("secondary" applications).
- Disposal of demolished concrete with artificial aggregate in landfills (,,end-of-life" application).

In all of these phases of use, the release of constituents from the material needs to be addressed to ensure environmental compatibility. Since the conditions are different for the different uses, the assessment calls for different leaching conditions [3].

The assessment of the environmental quality of aggregate and aggregate in concrete is commonly based on the determination of the leaching characteristics of these materials, i.e. the potential release of toxic constituents such as heavy metals or organic compounds to groundwater or soil.

A variety of laboratory leaching tests has been developed world-wide, of which a few are already used for regulatory control purposes. This is a major cause for confusion as the basis of reference is not the same (see Figure 1.1), and different tests may lead to different results and different interpretations [4]. An important factor that explains a large part of the differences between test results of different tests is the pH at which the test is performed. In Fig 1.1 an illustration is given to show that the pH range in which the different tests are performed explains the different limit values set by various governments. For instance, the US-EPA has set a high limit value and uses a test with a rather low pH while other governments use lower limit values and use tests without acidifying the material. For most materials the solubility of heavy metals is rather high at low pH values while the solubility is low at neutral pH values.



Figure 1.1 Leachability of lead from MSWI bottom ash as a function of the pH value in relation to various regulatory limits (Horizontal lines reflect limits as defined by: US-Environmental Protection Agency; Swiss Federal regulation; NWRF – German regulation in North Rhine Westfalia; NL – Dutch disposal act; French disposal regulation) and standard tests (TCLP -Toxicity characteristic leaching procedure (USA); TVA – Swiss CO2 test; DIN – German batch leach test (S4); AFNOR - French batch leaching test; NEN – Dutch column leaching test). Vertical lines reflect most common working range of pH.

For a sound understanding of the short and long term leaching behaviour of materials, single leaching tests are generally of limited use. Therefore, characterisation tests focussing at the understanding of the long-term behaviour are needed. In addition, parameters influencing the principal leaching mechanisms have to be determined.

Monolith leaching tests apply to bound applications (monolithic materials) [5,6] and percolation tests [7,8] to most other applications. The pH dependence test [9] is used to cover important differences in pH of the environment to which the materials are exposed to as well as for an assessment of the acid buffering capacity of the material. The acid (or base) buffering capacity provides insight in the sensitivity of the materials to externally imposed conditions. In the framework of CEN TC 154, a compliance test for aggregates has been drafted [10]. This method has been tested as well to verify its performance relative to the more elaborate leaching tests.

In this work the following questions in relation to environmental properties of artificial aggregates are addressed:

What are the leaching characteristics of artificial aggregate?

Which constituents are of relevant from an environmental point of view?

How do raw materials translate into environmental quality of aggregate?

What is the relationship between physical properties and leaching?

How variable or constant is the quality of aggregate prepared from different raw materials?

How to assess the different exposure scenarios of aggregate -stock pile - loose filler - use in concrete (service life, "end of life")?

How can the environmental quality of aggregate be assessed quickly?

In the Construction Products Directive (CPD [11]) a requirement is stated that addresses the need to include "Dangerous Substances" in all CEN standards for construction products. This Craft project "Utilising innovative kiln technology to recycle waste into synthetic aggregate", the ECRICEM project on the leaching studies on cement mortars [12,13,14], and the work carried out in the framework of the EU Project Harmonisation of Leaching Extraction Tests

[15,16] can provide useful information to draft harmonised European Technical environmental specifications for the construction industry.

In this part of the research (BRST-CT98-5234) we do not evaluate the total environmental impact of the new technology, such as emissions during the process in the kiln via the flue gas cleaning system to the atmosphere.

## 2. EXPERIMENTAL

According to the work programme of the Craft project, the experimental section consisted of the following individual tasks:

- Collection of raw materials
- Study of pore structure of artificial aggregates
- Rough mass balance evaluation based analysis of aggregate composition
- Evaluation of leaching properties of artificial aggregate in its life cycle
- Evaluation of test methods
- Comparison with regulation

## 2.1 Materials

## 2.1.1 Raw materials

A range of raw materials which have potential for use as feedstock for the aggregate production is listed in Table 2.1. The suitability of these materials as raw materials for the preparation of artificial aggregates, which meet the necessary technical specifications, is studied at the School of Civil Engineering (Leeds)[17,18]. A key aspect is the water uptake of the artificial aggregates (see section 2.2). The raw materials have been tested using the pH dependence test to identify the main leaching features of the starting materials for later comparison with the behaviour of artificial aggregates produced.

Sample Designation	remarks
Glensanda washings	Granite rich waste from mining
Paper sludge	Waste product from the paper industry
Shredder waste	Car shredder residue
Harbour sediment	Contaminated harbour sediment (Rhine)
MSWI Bottom ash	MSWI bottom ash
Sewage sludge	Municipal sewage sludge
PFA	Coal fly ash
RMC clay	Clay
Fly ash	MSWI Fly ash

Table 2.1 Raw materials used to produce Artificial Aggregates

## 2.1.2 Artificial aggregates

Artificial aggregates produced by firing in an innovative kiln (figure 2.1) at lab-scale from several combinations of starting materials (Leeds group), and aggregates resulting from full scale aggregate production have been obtained (Table 2.2). Artificial aggregates were tested using the pH dependence test [9] and the concise leaching test [19,20]. On a limited number of aggregates (due to the large amount of material required) the TC 154 method [10] has been applied. One aggregate sample from granite washings was subjected to all tests (all leaching tests are to be discussed in chapter 2.5) including a column test on the intact pellets has been carried out. From a limited number of aggregate samples concrete specimens have been prepared and tested using the tank leaching test [6], a compliance tank leaching test [21] and the pH dependence test [9]. This allows an evaluation of the material in different stages of use, such as storage prior to use, use as an aggregate in concrete, its recycling as construction debris in the

same application, its reuse in other applications (e.g. road base construction) and its end-of-life situation (ultimate disposal).

Sample Designation	Mix Composition (dry weight)
Tilbury 2	11 % RMA clay – 6 % sewage sludge - 83 % PFA
Glensanda	100 % Glensanda washings
Harbour sediment	90 % Glensanda washings – 10 % harbour sediment
MSWI-Bottom ash	63 % ash – 15 % RMA clay – 22 % water
Car shredder residue	65 % shredder waste – 16 % RMA clay – 22 % water
JKRLWA	20 % Clay – 64 % incinerated sewage sludge – 16 % sewage sludge
Glensanda 01	100 % Glensanda washings
Glensanda 02	95 % Glensanda washings – 5 % RMA clay
Glensanda 07	90 % Glensanda washings – 10 % Arnhem dredgings
Glensanda PP	90 % Glensanda washings – 10 % paper sludge
230321	50 % RMA clay - 50 % PFA
230322	44 % RMA clay - 44 % PFA - 7 % MSWI fly ash – 4 % bottom ash
230323	42 % RMA clay - 42 % PFA - 7 % MSWI fly ash – 8 % bottom ash
230324	36 % RMA clay - 36 % PFA - 7 % MSWI fly ash – 16 % bottom ash
230325	46 % RMA clay - 46 % PFA - 7 % MSWI fly ash
230326	48 % RMA clay - 48 % PFA – 3.5 % MSWI fly ash
230327	43 % RMA clay - 43 % PFA - 14 % MSWI fly ash
Jan12pel	85% Glensanda - 15 % clay

 Table 2.2 Designation of artificial aggregates

## 2.1.3 Concrete cubes with artificial aggregates

In order to evaluate the leaching characteristics of cements containing artificial aggregates test specimen were prepared according to the British standard (BS 1881 part 108:1983) [22] that is related to ISO 1920 but which is not classified as equivalent (Table 2.3). After demoulding at the age of 24 hours, the mortar samples for the leaching tests were wet cured at 20 °C for another 27 days in plastic bags to prevent pre-leaching. DIN sand is used in the preparation of the cubes. The material was tested as monolith and after crushing to less than 2 mm for their recycling and "end-of-life" characteristics.

Sample code	Composition (% volume)
Tilcem	51 % Aggregate 15 % Portland cement 34 % Sand W/C = $0.53$
Glencem	51 % Aggregate 15 % Portland cement 34 % Sand W/C = $0.53$
Glensanda 07	51 % Aggregate 15 % Portland cement 34 % Sand W/C = $0.53$
Gravel	51 % Gravel 15 % Portland cement 34 % Sand W/C = $0.53$
Reference	

Table 2.3 Concrete cubes with aggregates



Figure 2.1 Cross section of the TREFOIL kiln used to generate artificial aggregate [2]

## 2.2 Physical Tests

Characteristic physical properties of the aggregates and standard cubes – i.e. setting time, water demand, soundness, and compressive strength at 1, 2, 7 and 28 days – were determined at Leeds. In addition, microscopic investigations of porosity and microstructure were carried out using the techniques specified in table 2.4. Structural investigations have been carried out on aggregates and sintered bricks (reference) by scanning electron microscopy (SEM).

Table 2.4	Physical	and	Structural	Tests
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Test	Method
Porosity	Water uptake
	Hg porosimetry
Structure/morphology	Scanning electron microscopy

## 2.3 Particle Size Distribution

The artificial aggregate size is determined by using sieves of appropriate size. For different tests material has been size-reduced to meet the specifications. Size reduction was carried out using a jaw crusher.

## 2.4 Chemical Composition

## 2.4.1 Major elements and Trace Elements

In the test cements, major and trace elements were determined by means of Induced Coupled Plasma Emission Spectrometry (ICP) after complete dissolution of the matrix using HF,  $HClO_4$  and  $HNO_3$ . The anions were determined by ion-chromatography.

## 2.5 Leaching Tests

#### 2.5.1 Test Procedures

The following leaching tests have been selected for the study:

#### Characterisation leaching tests

- pH dependence leaching test [9]: This test provides information on the pH sensitivity of leaching behaviour of the material. The test consists of a number of parallel extractions of a material at a liquid/solid ratio (L/S) of 10 (l/kg) for 48 hours at a series of pre-set pH values. Since pH is one of the main leaching controlling parameters, the information can be used to evaluate the repeatability in testing (resulting from measurement at steep concentration - pH slopes) and to provide information on the sensitivity to pH in specific field scenarios. The acid neutralisation capacity (ANC) derived from the test is a useful property in this respect. For material characterisation this has been proven to be a very useful method [15,16]. The method is standardised in two experimental modes by CEN TC 292 Working Group 6[9].

- Column leaching test with similarities to NEN 7343 [7] percolation test is developed at European level in CEN TC 292 WG6 [23]. It is a column test in which 7 eluate fractions are collected within the range of L/S = 0.1-10 l/kg. The total test duration is approximately 21 days. The leachant is demineralised water (DMW). The test material is applied as received (Ø 1 cm) and up-flow (14 ml/h) is applied through a column waste height of 28 cm and a diameter of 10 cm.

- NEN 7345 Tank leach test [6]: In this test the specimen is subjected to leaching in a closed tank. The leachant is renewed after 8 hours and 1, 2.25, 4, 9, 16, 36, 64 days at a leachant to product volume ratio (L/V) of approximately 5. The results are expressed in mg/m<sup>2</sup>. This test is a procedure to evaluate the release from monolithic material by predominantly diffusion control (e.g. exposure of structures to external influences). The distinction is necessary, as the transport limitations set by a solid form result in a significantly lower environmental impact than derived from crushed material. This condition is valid as long as the product retains its integrity. To assess the behaviour after disintegration or demolition of monolithic forms, the information obtained in the pH dependence leaching test is very relevant, as in this situation the pH is likely to change to more neutral conditions. A dynamic leach test for monolithic materials is now subject of standardisation in CEN TC 292[24].

- Compacted granular leaching test NVN 7347 [25]: This test resembles the tank leach test for monolithic materials in the way it is carried out and the data are handled. The method is designed for granular materials that behave as a monolith in the scenario under investigation (e.g. clay lens in a sandy soil).

#### Compliance leaching tests

- EN 12457. CEN TC 292 compliance test for granular materials EN 12457 [26] - has been recently validated [27]. Part 3 consists of two extraction steps at L/S=2 and subsequently at cumulative fraction L/S=10. In the hierarchy of CEN TC 292, this is a compliance test (level 2 test).

- Compliance monolith leach test [28]. Although still in development by CEN TC 292 WG2 the main test features are: the specimen is leached after vacuum saturation with demineralised water at an liquid to area ratio of 5, eluates are produced after 6, 24 and 48 hours and subsequently analysed.

- Leach test for aggregates EN 1744-3 as developed in CEN TC 154 Aggregates [10]. This method consists of a leaching of the intact aggregate at a liquid to solid ratio of 10 (l/kg) for 24 hours. For this purpose the aggregate is placed on a grid above a magnetic stirrer. No particle size requirements are provided to limit the contribution of fines.

#### *Concise leaching tests*

- Concise leaching test [19,20], which has been proposed to address the main factors relevant to leaching from waste. It consists of four extractions for 24 hours at L/S=1 and L/S=10 at the material's own pH and two subsequent extractions at L/S=10 under pH control at pH=4 and neutral pH (or mild alkaline pH, when the material is neutral by itself).

- To evaluate the slow release of constituents by diffusion from the artificial aggregates a time series measurement of intact artificial aggregates was carried out in a batch at L/S = 5 with sampling of the eluate after 24, 48, 72 and 96 hours.

Test name:	NEN 7343	EN 1744-3	PrEN xxx	NEN 7345	NVN 7347	PrEN xxx	EN 12457-3	Concise test	EN 12457-4
	Percolation test	Aggregate leach	pH dependence	Diffusion Test	Compacted	Compliance test	Compliance test	Batch test with	Compliance test
		test	leaching test		Granular Leach	for monolithic	for leaching of	two L/S values	for leaching of
Type of test:	Column	Batch extraction	Batch tests at			material	granular waste	and two	granular waste
	leaching test		specified	Tank leaching	Tank leaching	Tank leaching	materials	controlled pH	materials
	Netherlands		conditions	test		test		conditions	
Country/reference	CEN TC 296 WG6	CEN TC 154	CEN TC 292 WG6	Netherlands	Netherlands	EU CEN TC292	EU CEN TC292	WASCON 1997	EU CEN TC292
Particle size:	<4 mm	< 2mm	< 2 mm	> 40 mm	0 - 20 mm	> 40 mm	< 4 mm (95%)	< 2 mm	< 10 mm (95%)
Leachant:	DMW or HNO <sub>3</sub> at pH 4	DMW with pH control using HNO <sub>3</sub> or NaOH (pH 3-12)	DMW with pH control using HNO or NaOH (pH 3-12)	DMW initially acidified to pH=4 with NHO <sub>3</sub>	DMW	DMW	DMW	DMW (2) and two with pH control using HNO3 or NaOH (pH 4 – 7)	DMW
Amount of solid	0.5-0.7 liter	? g	100 g	> 100 g	> 100 g	> 100 g	$100 \pm 5 \text{ g}$	100 g	$100 \pm 5 \text{ g}$
L/S (l/kg) per step	cum.L/S: 0.1, 0.2, 0.5,1,2,5,10	10	10	L/V = 5 l/l	L/V = 5 l/l	L/V = 1.5 l/l	2 in 1 <sup>st</sup> step 8 in 2 <sup>nd</sup> step	10	10
Maximum accumulated	10	10	10	Not applicable	Not applicable	Not applicable	10	10	10
Number of steps	7	1	8	(8 fractions collected)	(8 fractions collected)	(3 fractions collected)	2	4	1
Contact time per step	total: max 3 weeks	24 hours	48 hours	Varies (64 days for all 8 steps)	Varies (64 days for all 8 steps)	Varies (2 h up to 48 h)	6 h for 1 <sup>st</sup> step and 18 h for 2 <sup>nd</sup> step	48 hours	24 hours
For serial batch tests: Renewal of leachant or solid	Not applicable	Not applicable	Not applicable	New leachant	New leachant	New leachant	New leachant	not applicable	
Method of agitation:	Not applicable	magnetic stirring	ANC mode : End o rotation, 5 - 10 rpm pH stat mode: magnetic stirrer	No agitation	No agitation	Mild stirring	End over end or roller-table rotation (10 rpm)	ANC mode : End o rotation, 5 - 10 rpm pH stat mode: magnetic stirrer	End over end or roller-table rotation (10 rpm)
Filtration/filter size:	0.45 μm	0.45 μm	0.45 μm	0.45 µm	0.45 μm	0.45 μm	0.45 μm	0.45 μm	0.45 μm
Comments:	recently	Published	WG document			Some details not		WASCON paper	
	accepted in EU	standard. No				yet fixed		Own pH same as	
		limitation on						EN 12457-3	
		particle size							
		distribution or							
		on fines							

Table 2.5 Leaching Test Conditions

## 2.5.2 Analytical Methods

The following methods were applied in the analysis of the eluates:

Component	Analytical Method
Metals / Non-metals	ICP-AES, AAS
Anions (Cl, Br, sulfate)	Ion chromatography

#### 2.6 Geochemical modelling

In geochemical modelling the measured concentration of major, minor and trace elements together with pH and, if relevant, redox info is used as input to MINTEQA2 or ECOSAT [29,30] (large number of minerals in the associated database). The output of the program is a list of saturation indices (SI value) for element-mineral combinations. This allows a comparison of the extent to which a given mineral phase may be controlling solubility of a particular element. An SI close to 0 is indicative of possible solubility control. Generally, a range of measured pH values should show SI values approaching 0. A single data point is insufficient. Over the entire pH range from pH 4 to 12 a few mineral phases may be relevant. SI values between +0.5 and – 0.5 (factor 3 in concentration) are considered a good match.

## 2.7 Data Presentation and Interpretation

Data interpretation for leaching from granular materials is related to the behaviour of constituents in terms of leaching controlled by either availability or solubility [31]. Leaching test results may be expressed either as

- Leachate concentration in [mg/l], or as
- Constituent release in [mg/kg of material].

Regulatory test results are expressed both as leachate concentration and in mg/kg for comparison to given limit values, but they do not consider the mechanism responsible for release phenomena observed.

Results expressed as leachate concentrations allow the comparison of constituent solubility reflecting the chemical speciation of the elements and the leachate conditions (i.e. pH).

Transformation of measured leachate concentrations into constituent release is necessary for the comparison of data obtained at different liquid to solid (L/S) ratios, and for the determination of the general mechanism, i.e. availability. Examples (Figure 2.1):

- Chlorine (Cl) represents an "availability"-controlled element. Data from tests at different L/S ratios expressed in [mg/l] lead to apparent differences, while data presented in [mg/kg] show that in all cases the whole fraction available for leaching is released.
- Silicon (Si) represents a solubility-controlled element. Here, presentation of leaching data in [mg/kg] leads to differences, whereas data presented in [mg/l] show the solubility control in the pH region of 3 to 8.

#### **Conversion formula:**

Constituent release [mg/kg] = leachate concentration  $[mg/l] \times L/S$  ratio [l/kg]



Figure 2.2 Distinction between Solubility Control (Si) and Availability Control (Cl) in the Presentation of Leaching Test Results

The pH dependence test data are used as a basis of reference for data from other tests [15]. The data from the pH dependence test are used for geochemical modelling of potential solubility controlling minerals. The data of the column percolation test are given as a function of the L/S. The relevant data from the compliance tests are inserted in these respective graphs. The release from the intact artificial aggregates is addressed by taking into account the diffusion from the interior of the artificial aggregates. For this purpose measurements were extended in one series up to several days of leaching. The release from the concrete specimen is addressed to assess the contribution of aggregates to the overall release from the concrete during service life, recycling and "end of life" conditions.

## 3. RESULTS

After a description of the properties of the raw materials used in the study, the physical and leaching properties of the aggregates are discussed. The leaching characteristics of the main application of artificial aggregate as replacement of natural aggregate is addressed followed by modelling and a description of different scenarios of use of aggregates in the different stages of its life-cycle.

## 3.1 Raw Materials

#### 3.1.1 Total composition

The various raw materials have been analysed for total composition. They have also been characterised in terms of leachability by the pH dependence test. In table 3.1 the composition of the various materials is given.

Element	Glensanda	Paper	Shredder	Harbour	MSWI	Sewage	det.
	washings	sludge	waste	sediment	Bottom ash	sludge	lim.
Al	64311	25298	5916	72782	35438	25912	0.8
As	0	0	6.9	25.6	3.1	10.7	3
В	5	10	308	131	136	62	0.3
Ba	2142	179	1826	954	1193	475	0.07
Ca	34665	74150	12818	87506	114934	52108	4
Cd	0.1	0.1	22.1	0	7.2	5.0	0.1
Co	7.8	0.4	13.4	24.5	10.2	10	0.5
Cr	25.5	2.6	198.8	304.7	170.5	127	0.9
Cu	8	23	4618	169	1669	555	0.6
Fe	21225	2961	48050	44061	48553	13242	1
Κ	28894	1620	1702	25422	5673	6966	13
Li	61	8	11	89	15.7	10	0.1
Mg	8198	3668	2271	13903	14251	6219	0.8
Mn	753	56	403	1263	593	359	0.1
Мо	2	2	33	8	13.8	10	0.8
Na	21652	1047	4013	4627	13710	3025	8
Ni	15	2	94	533	93	93	0.9
Р	808	276	520	3216	5904	22496	0.7
Pb	42	9	2380	209	2053	157	4
S	1178	728	3290	2660	6700	7113	2
Sb	0	0	58.4	22.6	35.5	28	2
Se	0	0.2	0	0.3	1.5	0.00	1.2
Si	165661	74426	22579	208434	29973	65151	48
Sn	0	3.4	281	509	153	44	5
Sr	393	162	112	342	203	272	0.05
Ti	1448	309	998	4104	3236	2403	0.4
V	45	2.7	17	148	30.2	26	0.3
Zn	69	33	4276	1101	2185	1178	0.4

Table 3.1 Total composition of raw materials

The firing process effectively removes all organic contaminants present in the raw materials as the firing temperature is high enough to destroy all organic matter. Metals and oxyanions will largely remain in the matrix and will in general become less leachable [32]. In section 3.6 this aspect will be addressed in more detail.

#### 3.1.2 Leaching of waste versus aggregate

In Appendix I the leaching behaviour of the raw materials is given: paper sludge, MSWI bottom ash, shredder waste, paper sludge, granite washings and harbour sediment.

A short description of the leaching behaviour (pH dependence leaching test) of raw materials is provided below:

*Granite washings* – Granite washings are not critical from a leaching point of view for any of the parameters measured (about 28) according the Dutch Building materials Decree [33].

*Paper sludge* - Paper sludge is characterised by a very high DOC (dissolved organic carbon) and a relatively high TIC (carbonate). The elements Se, Mo, Ba and Cu only exceed critical regulatory limits at pH > 10 [33]. The material has a strong buffering capacity, as the amount of acid needed to increase the pH by one pH unit is quite significant (2 mol/kg).

*Shredder waste* - The buffer capacity of shredder waste is relatively small. This implies that the material is sensitive to both acid and base influences. For instance in case of Zn, a small change in acid addition will lead to a significant increase in Zn leachability (more than 1 order of magnitude for 0.1 mol/kg). The elements Cu, Cd, Mo, Sb, Zn and Pb exceed the regulatory criteria [33] over a large portion of the pH range studied. Cr and Sn become critical at high pH, whereas Ni and Co become critical at low pH. TOC shows a characteristic increase for many materials containing organic matter (increase of TOC with increasing pH).

*Harbour sediment* - A small change in acid addition leads to a quite significant change in Zn leachability. The acid amount required to reach a pH below 5 is substantial. The agreement between CEN test data and the pH dependence test data are generally good. A duplicate analysis also leads to reproducible results, unless the concentrations become low and low analytical detection is a problem. For various elements it is clear that a relatively small change in pH may lead to very significant changes in leachability. Ni, Co and Zn show several orders of magnitude change within one to 2 pH units.

*MSW Incinerator bottom ash* – Much information is available on this material [4], which indicates that Cu, Mo, Cl and Sb are potentially critical with respect to the Dutch Building Materials Decree.

In comparison with leaching data on artificial aggregates, produced from mixtures of wastes, generally significantly lower leachability is observed for many constituents.

In Appendix II a comparison between raw material leachability and aggregate leachability is given for shredder waste and for harbour sludge.

For the shredder waste, a sharp drop in metal leachability (Ni, Cu, Pb, and Zn) after incorporation in aggregate is noted. This is attributed to the removal of the dissolved organic matter controlling metal leachability from shredder waste. The leachability of oxyanions, such as Se, V, Mo and Sb, is increased relative to the leachability of shredder waste. This is attributed to the transformation of part of the reactive Fe, Mn and Al phases capable of retaining the oxyanions into silicate matrix components and thus no longer available as sorption sites for oxyanions. Sulphate, Ca and Ba show an increased leachability. K leachability decreases, which may be attributed to partial incorporation of K in the silicate matrix. This type of behaviour has been noted before in sintering [32, 34].

For the harbour sludge, similar observations are made for the metals (Cu, Ni, Pb). For the oxyanions, however, the leachability decreases or remains equal.

## 3.2 Physical Properties of aggregates

The results of the water uptake in aggregate after vacuum saturation is given in Appendix III. In appendix III also the Hg porosimetry data are given for a few selected samples. The agreement between porosity derived from the water uptake and that from the Hg porosimetry is good (figure 3.1), which implies that for quick quality control of aggregate production this is a simple and useful method. The Hg porosimetry provides much more insight in the pore size distribution, which can show significantly different pore size distributions (see figure 3.2).



Figure 3.1 Porosity by water uptake and by Hg porosimetry correlate well.

The artificial aggregates with a relatively high water uptake are characterised by a significant contribution of pores in the  $100 - 10 \mu m$  range. In the four materials studied, the contribution to porosity in the  $10 - 0.1 \mu m$  range is limited, while the aggregates with high porosity also show a significant contribution of pores in the  $0.1 - 0.01 \mu m$  range. This type of porosity may well be attributed to prolonged degassing at a stage (solidifying), where gas formation should have stopped. It is noted that in particular the aggregates produced from the sediment were observed to be bloating. Since the sediments are reducing, this could be a delayed oxidation effect (with formation of SO<sub>2</sub>). This could be verified in the future by analysing the gas phase in the artificial aggregates shortly after firing.



Figure 3.2 *Hg porosimetry data of artificial aggregates in comparison with traditional gravel and a molten slag.* 

The water uptake can be tested on-site (so-called on site verification tests), as a quick test for water uptake. This quick test by vacuum saturation, and a measurement of conductivity, are easy to perform and they provide useful direct on hands information to control process conditions.

## 3.3 Particle Size Distribution

The particle size of the aggregates is generally rather constant. Particle size range: 5 - 12 mm. For more homogeneous batches particle sizes ranging from 8 - 10 mm have been found. Size may have an effect on leaching behaviour [35].

## 3.4 Morphological Properties of Aggregates

The results of the investigations on the aggregates at a microscopic level by SEM point at an important similarity of the aggregate structure with that of sintered bricks. This is reflected by the smooth rounded surfaces and porous nature of both materials. A coarse aggregate like MSWI bottom ash does not have these characteristic rounded surfaces, which are illustrative of superficial melting of the matrix. The temperature in the process is not aimed at turning the material in a fully vitrified granule. This feature puts the aggregate production in the domain of

sintering processes and brick manufacture. In Appendix IV the SEM micrographs of different aggregates and sintered bricks are given. For comparison a concrete pavement block ash is shown as well.

## 3.5 Chemical Composition

The chemical composition of the aggregates is given in Table 3.2 for major and minor elements and trace constituents. A complete mass balance has not yet been made as information of the flue gas is needed for that. For a mass balance it is important to be aware that analysis should be based on true totals by using HF as destruction agent. Due to the redistribution of elements between mineral and leachable phases, this is crucial to allow proper conclusions to be made.

From an environmental point of view the composition is of little relevance as materials with high concentration may leach very little, whereas materials with low total concentration but a low pH may be highly leachable. Particularly after treatment in a process like aggregate production, the incorporation in the matrix is an important aspect of the process.

	Tilbury2 aggregate	Glensanda aggregate	det. lim.
Al	92424	80057	0.8
As	12.7	0	3
В	304	33	0.3
Ba	900	2336	0.07
Ca	20891	41802	4
Cd	0.5	0.2	0.1
Co	27.0	11.1	0.5
Cr	334.4	162.6	0.9
Cu	131	21	0.6
Fe	54654	23365	1
Κ	17043	34871	13
Li	94	59	0.1
Mg	9803	9222	0.8
Mn	427	823	0.1
Mo	21	6	0.8
Na	7057	24592	8
Ni	735	586	0.9
Р	2393	811	0.7
Pb	144	137	4
S	1148	87	2
Sb	8.4	0	2
Se	12.0	4.1	1.2
Si	238024	250015	48
Sn	22	5.5	5
Sr	370	476	0.05
Ti	5012	2801	0.4
V	234	54	0.3
Zn	431	137	0.4

Table 3.2 Composition of aggregates (mg/kg)

The chemical composition of artificial aggregates in concrete is given in table 3.3 in comparison with the composition of the Portland cement used and in comparison with concrete with natural gravel (reference).

	Tilbury 2 cement	Glensanda	Gravelcem	Portland Cement	det. lim.
		cement	Gravel reference		
Al	41462	44469	19244	26694	0.8
As	14.0	4.0	3.2	11.3	3
В	106	32	49	45	0.3
Ba	423	1036	201	301	0.07
Ca	285787	254423	224508	453879	4
Cd	0.6	0.26	0.73	2.6	0.1
Со	13.8	8.6	7.7	13.3	0.5
Cr	132	76	144	83	0.9
Cu	48	21	17	31	0.6
Fe	28605	20414	14676	22592	1
Κ	7430	15694	4243	4589	13
Li	49	43	28	41	0.1
Mg	8773	8684	5459	9888	0.8
Mn	384	526	288	433	0.1
Мо	4.32	0.94	0.02	0.28	0.8
Na	2835	10105	1919	1653	8
Ni	169	111	451	43	0.9
Р	1279	854	611	1058	0.7
Pb	34	43	21	31	4
S	6643	5537	5224	10593	2
Sb	12.0	5.3	6.3	11.5	2
Se	4.4	0	0	1.7	1.2
Si	118820	135824	67993	92028	48
Sn	6.1	3.4	1.2	3.2	5
Sr	712	710	520	990	0.05
Ti	2805	2283	1055	2020	0.4
V	177	116	97	178	0.3
Zn	205	150	127	220	0.4

Table 3.3 Composition of concrete with artificial and natural aggregate.

The total composition of the artificial aggregates is generally higher than that of the natural aggregate. The question is whether such an increase in total composition is reflected in the environmental performance of the aggregates.

## 3.6 Leaching Characteristics

In this section, the different aspect of leaching of artificial aggregates and artificial aggregates in concrete are addressed. In Appendices I to XVIII all testing data and more extended graphical presentations are given. The different leaching tests address different aspects of leaching. In the following sub sections, the different test results and their mutual relationships are discussed. Table 3.4 provides an overview of all test performed on the different raw materials, aggregates and final products.

Sample Designation	Tests							
	EN 1744-3 (TC 154)	PrEN xxx (ANC mode)	NEN 7345 tank test	NVN 7347 CGLT	EN 12457-3	Concise test two L/S, 3pH	EN 12457-4	NEN 7343 column test
raw materials								
Glensanda washings		I,II						
MSWI Bottom ash		Ι						
Paper sludge		Ι						
Shedder waste		I,II						
Sewage sludge		Ι						
SED(Harbour sediment)		I,II						
PFA						Ι		
RMC clay						Ι		
Fly ash						Ι		
Artificial aggregates								
Tilbury 2	VII#	V,VI,XI,XII	XI	XV	VII			VI*
Glensanda		V,XI, XII	XI					
Harbour sediment		VIII,X			II	VIII		
MSWI-Bottom ash		V,VIII,XI,X	X,XI			VIII		
Car shredder residue		V,VIII,X	Х		II	VIII		
JKRLWA		V	Х					
Glensanda 02		V,X	Х					
Glensanda 07		V,XIII,X	Х					
Glensanda PP		V,X	X					
230321		V,XVIII						
230322		XVIII			V			
230323		XVIII			V			
230324		V, XVIII						
230325		XVIII			V			
230326		XVIII			V			
230327		XVIII						
brick reference	x / X	XI	-	3737	X 774			X 771 X7X7
Jan12pel	VI	VI		XV	VI!			VI!,XX
Artificial aggregates in								
Tilbury cement		XILXIII	XIV.XV					XVIII
Clangendo coment			XIV					
Glensanda cement								
Glensanda cement 07		АШ,АШ						
Gravel reference								

Table 3.4 Organisation of tests on raw material, artificial aggregates, and cement products (mentioned are the Annendices in which the results are given)

\*NEN7343: on intact aggregates and wet/dry cycles during percolation test #time series during EN1744-3 (TC 154)

! 4 and 10 mm

explanation on abbreviations (see also chapter 2.5):

Aggregate leach test (CEN TC 154) EN 1744-3

pH dependency test (CEN TC292 WG 6) prEN xxx:

Batch leach test with two L/S values and two controlled pH conditions Concise test:

EN 12457-4, Compliance test for leaching of granular waste materials.

## 3.6.1 Comparison of Aggregates with pH dependence Test

The pH dependence leaching test (ANC mode) [9] has been used to compare the leaching characteristics of aggregates produced from different source materials. The main objective for using the pH dependence test is to identify changes in leachability, which will imply changes in chemical speciation.

NEN 7343 Percolation test

NEN 7345: diffusion or tank test.

NVN 7347: Compacted Granular test,

EN 12457-3 Compliance test for leaching of granular waste materials

The pH dependence test data cover a wide range of potential exposure conditions in service life (own pH and externally imposed pH), in the recycling stage as aggregate and in the "end of life" conditions after full carbonation.

#### pH Dependence test results

In Appendix V the results of pH dependence test measurements on aggregates are given for the various recipes studied. In some cases, the presence of specific elements in elevated concentration levels is reflected in the leaching behaviour of the resulting artificial aggregates. These data illustrate that the pelletising and sintering process to produce artificial aggregates leads to a product with a leaching behaviour that is relatively constant and shows the same generic leaching character in spite of using different raw materials. Some typical deviations are noted:

The use of high contributions of coal fly ash in the mix to produce aggregates leads to increased leachability for B, Cr, Mo, As, V, Se and Sr relative to all other mix designs. In a mix design with MSWI fly ash significantly increased levels of B, Ca, Mg, Li, Na, Cr (probably as chromate), Mo, V, Se, SO<sub>4</sub>, Ba, Sr, K, Cd, Pb and Zn are observed. In view of the degree to which leaching levels are increased and the number of elements being increased significantly, it is not recommended to apply this material in a mix design. In case of car shredder waste, a few elements deviate from the general pattern: Ca, Mg, B, Li, Mo, Sb, SO<sub>4</sub>, Sr, Ni and Pb. In case of sewage sludge, only P is elevated relative to the other mix designs.



Figure 3.3 Acid neutralisation capacity (ANC) for artificial aggregates and artificial aggregate in concrete.

#### Acid neutralisation capacity

Besides the leaching behaviour as a function of pH, the Acid Neutralisation Capacity (ANC) of artificial aggregates and artificial aggregates in concrete is obtained (figure 3.3). The difference between the ANC for aggregates and aggregate in concrete is quite significant. In the latter case, it is entirely determined by the cement-based matrix. For comparison the ANC for Portland cement mortars is illustrated in figure 3.4. The ANC for the cement mortars is higher than for concrete as the amount of cement used in concrete is less than used in the preparation of standard mortar bars. The ANC of artificial aggregates is relatively low. A slightly alkaline own pH is caused by an incomplete reaction of calciumcarbonate in the raw materials after

conversion to calciumoxide with the silicate and aluminium phases. The low ANC implies that the material is sensitive to external influences, which would imply that in temporary storage the pH is likely to be near neutral even for aggregates that in testing show a pH of around 10.



Figure 3.4 Acid neutralisation capacity for Portland cement mortars.

The ANC of the aggregates is mainly determined by the high ANC from the cement used in making concrete. Therefore, the leachability of aggregates at their own pH is not so meaningful for an assessment of aggregate use in concrete. This aspect of putting the proper question to be answered in relation to aggregate testing is discussed in section 3.7.

The ANC data of the aggregates are used in combination with information on external stresses to evaluate how fast the pH of the material under consideration will change.

As carbonation is the main factor controlling pH change in cement-based materials, an issue to be resolved is to what extent pH control by  $CO_2$  will produce the same or slightly different results in a pH dependence test. In future work, this aspect needs to be addressed.

The results obtained so far suggest that there is a consistent behaviour between aggregates produced from different starting materials. The aggregate production process tends to impose common characteristics in the leaching behaviour of aggregates with only individual deviations depending on the level of certain elements in the starting materials.

Aggregates have a low leachability, even Na and K are incorporated to a large extent in the silicate matrix.

#### 3.6.2 Comparison of pH dependence test data with percolation test

In Appendix VI the test results of two types of artificial aggregates (Tilbury 2 and Janpel, for materials see Table 2.1) are shown, which have been tested using the full pH dependence leaching on size reduced artificial aggregates (< 4mm) and the complete percolation test as described in NEN 7343 on intact (non-size reduced) artificial aggregates. In the case of Tilbury2 pellets also a down flow percolation test was carried out, in which the column was left saturated and completely drained, subsequently left dry for some time and then saturated quickly and left to stand for some time. This cycle was repeated 6 times. In case of the artificial aggregates, a

percolation test was carried out on size reduced material (< 4 mm). In addition the EN 12457 part 3 method was applied.

The agreement between the end point of the percolation test on intact artificial aggregates (L/S=10) and the pH dependence test (L/S=10: d< 4mm) at the pH corresponding with the pH in the percolation test (Tilbury 2, own pH= 9.58; Janpel, own pH= 9.8) is very good across all parameters analysed. In figure 3.5 this is illustrated for Ba, Mo, Zn, Al, Ca and Sr. This observation is valid for both artificial aggregates. This indicates that local equilibrium appears to be established. Particularly, since mobile elements like Na, K, Mo show depletion, it follows that the porosity of the aggregates is such that release of soluble constituents from the artificial aggregates can be expected within reasonably short time frames (days to weeks). Typical solubility controlled elements, such as Al, P and several metals also behave consistently. Similar results, depletion of available Na and K although much higher total amounts of Na and K are retained in matrix [32].

The agreement for the Tilbury pellets between the intermittent wetted pellets and the fully saturated percolation is very good across the wide range of elements analysed. Only for Mn a deviation is observed with a higher release of Mn from the intermittent wetted pellets. This may be attributed partly to a very sensitive Mn leachability in the pH domain around 9.6.

In the comparison between intact and size reduced artificial aggregates tested in the percolation test on Janpel, the agreement between the test results is quite close. This is particularly true for the L/S=10 condition at the end of the test. It is very clear that the size reduction enhances the rate of release of mobile constituents. This is reflected in the higher initial release levels for Na, K, Li, Mo, B, Ca, Mg, SO<sub>4</sub>, V and Sr. The solubility controlled elements show very little difference between intact artificial aggregates and size-reduced artificial aggregates, which is consistent with release controlled by solubility. For some elements (A1, Si) a slight difference is noted, which is at least in part related to the somewhat higher pH in the size-reduced material (pH 10.4-9.8) as compared to the intact artificial aggregates (pH 9.8 - 9.5).

The results for the EN 12457 test part 3 generally match well with the results of both pH dependence test and the percolation test. In several cases the results of EN 12457 Part 4 are below the levels measured in the pH dependence test or the percolation test. This is related to the combination of testing time and diffusion controlled release from larger particles. Diffusion from 10 mm particles is not complete in 24 hours. The EN 1744-3 (TC 154) test results, however, are in all cases well below the pH dependence test data, as well as below the percolation test results. Even those corresponding to intact artificial aggregates. The claimed equilibrium condition aimed for in the latter test is not reached due to diffusion limitations. The time selected for the test is too short to allow release of mobile constituents from the porous artificial aggregates. Since the fraction of mobile constituents is leached relatively fast, at least these elements should be measured properly and should not be underestimated.

	Tilbury2 Aggregate		Glensanda Aggregate			
	Total	max. pH stat	% Available	Total	max. pH stat	% Available
	mg/kg	mg/kg		mg/kg	mg/kg	
Al	92424	57.9	0.06	80057	74.3	0.093
As	12.7	1.51	11.93		0.11	
В	304	11.0	3.63	33.2	0.84	2.52
Ba	900	1.67	0.19	2336.2	3.88	0.17
Ca	20891	848	4.06	41802	489	1.17
Cd	0.5	0.07	14.18	0.2	0.003	1.45
Co	27.0	2.28	8.44	11.1	2.18	19.59
Cr	334	0.09	0.028	163	0.05	0.03
Cu	131	4.22	3.22	20.6	0.37	1.80
Fe	54654	133	0.24	23365	8.6	0.037
Κ	17043	51.46	0.30	34871	61	0.17
Li	93.5	0.42	0.45	59.3	0.16	0.28
Mg	9803	149	1.52	9222	47	0.51
Mn	427	3.29	0.77	823	3.4	0.42
Mo	21.2	1.22	5.78	6.3	0.24	3.89
Na	7057	48.2	0.68	24592	68	0.28
Ni	735	5.64	0.77	586	0.27	0.05
Р	2393	6.69	0.28	811.2	0.91	0.11
Pb	144	0.39	0.27	137.3	3.02	2.20
S	1148	482	42.0	86.7	7.84	9.04
Sb	8.4	0.14	1.73			
Se	12.0	0.65	5.43	4.1	0.11	2.61
Si	238024	196	0.082	250015	227	0.091
Sn	21.7	0.19	0.88	5.5	0.06	1.19
Sr	370	2.11	0.57	475.7	1.36	0.29
Ti	5012	0.06	0.001	2801	0.04	0.001
V	234	0.62	0.27	53.6	0.22	0.41
Zn	431	1.25	0.29	137	0.76	0.56

Table 3.5 Percentage available for leaching in aggregates relative to the total composition.

#### 3.6.3 Time series measurement using EN 1744-3

The test EN 1744-3 has been carried out for a longer time to evaluate the assumption of approaching equilibrium. From the data in Appendix VII, it is clear that the time series points out that equilibrium is not yet established after 24 hours of leaching intact artificial aggregates. In case of solubility controlled elements the differences may not be very large. The release is for most elements increasing significantly between 24 and 96 hours contact time. This is consistent with the observations in the previous section on intact and size – reduced artificial aggregates

A key issue in the discussion on the leaching of artificial aggregates in a leaching test focuses on the particle size and the diffusion of constituents from the interior of the particles into the surrounding solution. The internal porosity is the crucial parameter here. The results from the up-flow column experiment (diameter 10 cm, height 28 cm, flow rate 14 ml/h) on uniform aggregate (diameter around 10 mm, aggregate porosity 15 %) is compared with pH dependence test on the size-reduced aggregate (broken to < 2 mm, 24 hrs, L/S=10, own pH). In Table 3.6 the results are given for the elements Al, B, Ca, K, Li, Mg, Mo, P, S, Si and Sr, which are accepted on the basis of sufficient analytical sensitivity. In the case of Ba (sufficient sensitivity), the pH

difference between column and pH dependence test is important (steep gradient). An interpolation of the pH dependent leaching data to the pH in the column would lead to a better agreement for Ba.

1 /			1	
Parameter	Column on intact	pH dependence test on	Ratio Column/pH	
	aggregate (d=10 mm,	broken aggregate	dependence test	
	uniform size) Cumulative	(d < 2 mm)		
	release at L/S=10 (mg/kg)	L/S=10 (mg/kg)		
pН	9.01	9.58		
Al	3.58	2.99	1.20	
В	4.47	4.69	0.95	
Ca	131	164	0.80	
K	7.9	11.1	0.71	
Li	0.27	0.29	0.94	
Mg	49.0	42.7	1.15	
Mo	0.86	0.88	0.98	
Р	1.98	1.74	1.14	
S	106	125	0.85	
Si	26.7	24.7	1.08	
Sr	0.32	0.35	0.91	
Average			0.98	
Std			0.15	
N			11	

Table 3.6 Comparison of leaching from intact particles (column test) and equilibrium test (pH dependence test) data (Artificial Aggregate: Tilbury 2) release (see also Appendix. VI.

After log transformation (table 3.7) the elements that are accepted before transformation also pass, but in addition elements at relatively low concentrations, such as Co, Cr, Cu, Fe, Mn, Se and Zn show a good agreement. The rational behind the log transformation is that at low concentrations a larger tolerance is acceptable. A factor 2 difference in concentration at a level of 0.01 mg/kg can be considered equal taking the analytical uncertainty into account. This can only be shown after log transformation. The elements Cd, Pb, Ni and Sb are too close to the detection limit to be able to draw conclusions and therefore cannot contribute positively to a conclusion, but do not challenge it either. At the percolation rates applied diffusion from the interior of uniform particles up to 10 mm is fast enough in the way the column is operated (particularly the flow rate of 14 ml/h) to provide the same end results as a batch test with size-reduced material at LS=10. Only, when the porosity of the material is low (e.g. molten slag) a more significant effect of delayed release can be expected and flow rate becomes an important factor. However, under that condition the question arises, if extending the running time of a column test is useful, or if another way of determining the contribution of diffusive release should be addressed (compacted granular leach test [25]).

In a comparison of the release at LS=10 for the column test (10 mm pellets) with the release from the same material broken to less than 2 mm at the corresponding pH (LS=10) in the pH dependence test, no significant difference is noted for several elements.

Element	Log column	Log pH stat	Ratio log column/log pH	
	(mg/kg)	(mg/kg)	stat	
Al	0.554	0.476	1.165	
В	0.651	0.671	0.969	
Ca	2.117	2.215	0.956	
Co	-1.892	-1.699	1.114	
Cr	-1.690	-2.095	0.807	
Cu	-1.767	-1.843	0.959	
Fe	-1.528	-1.572	0.972	
Κ	0.896	1.044	0.858	
Li	-0.568	-0.539	1.054	
Mg	1.690	1.630	1.037	
Mn	-2.645	-2.301	1.149	
Мо	-0.067	-0.057	1.175	
Na	1.390	1.683	0.826	
Р	0.297	0.240	1.234	
S	2.026	2.097	0.966	
Se	-1.114	-0.878	1.270	
Si	1.427	1.393	1.024	
Sr	-0.497	-0.456	1.090	
Zn	-1.906	-1.847	1.032	
		Average	1.032	
		Std	0.128	
		Ν	19	

Table 3.7 Comparison of log transformed data.

By applying a log transformation, it is clear that the agreement even extends to the elements leached in low concentrations. The effect of crushing the aggregate has little effect on the leaching of very soluble elements at higher liquid to solid ratios (L/S > 2 l/kg) suggesting that the porous nature of the aggregate only leads to a relatively small delay in release (see Appendix VI(2)). To address long term environmental impact size reduction is the better alternative for testing than running the test longer to reach a stable end point. This will most likely improve the repeatability of the test results [34].

The pH dependence leaching test data and the column leaching test results are internally consistent, which allows translation of leaching test data from percolation tests to other exposure conditions. The effect of crushing the aggregate has little effect on the leaching of elements at higher liquid to solid ratios (L/S > 2 l/kg) suggesting that the porous nature of the aggregate only leads to a relatively small delay in release. To address long term environmental impact an alternative would be to run the test for a longer period to reach a stable end point but the results show that size reduction is a simple and good choice.



Figure 3.5a Comparison of aggregate leaching of intact pellets (10mm) with size reduced aggregate (<2 mm) in pH dependence test indicating consistency of data.



Figure 3.5b Comparison of aggregate leaching of intact pellets (10mm) with size reduced aggregate (<2 mm) in pH dependence test indicating consistency of data.

## 3.6.4 Comparison of concise test with full pH dependence test

In general, it is desirable to limit the amount of testing that needs to be done. In the limited testing as much relevant information as needed must be derived. The concise test [19] has been developed as a means to reduce the level of testing, but maintain information on key aspects of leaching. These comprise a notion of the pH dependence and a notion of the response to L/S. The latter allows conclusions on the occurrence of solubility control or wash-out of mobile species or fractions of elements. In the hierarchy of testing developed in CEN TC 292 [3] (see also chapter 1), the concise test takes an intermediate position between characterisation and compliance. In Appendix VIII, the results of the concise test (pH dependence test part) are compared with the full pH dependence test. This comparison has been made for three types of artificial aggregates containing harbour sediment, MSWI bottom ash and shredder waste. Besides a comparison between concise test and pH dependence test a comparison between the three types of artificial aggregates is also possible.

The concise test offers a good option for an optimised quick testing protocol for quality control of the aggregate production process. This procedure leads to results within a few days. Several crucial aspects of leaching are addressed: solubility control, wash out, pH sensitivity over a wide pH range.

#### 3.6.5 Comparison between compliance tests, concise test and percolation test

In Appendix IX the results of the comparison of the EN 12457 parts 3 and 4 and the compliance test results of EN 12457, EN 1744-3 and concise tests are compared with the results obtained by the percolation leaching test on intact specimen (HSED, harbour sediment). The latter is the characterisation test reflecting closest the behaviour of artificial aggregates under field conditions. The tests enable a good differentiation of the process responsible for leaching/binding. The soluble components such as K and Na show the same emission in the pH dependence test and in the percolation test (column test). For solubility controlled components the concentrations are controlled by the minerals in the matrix or adsorption processes. These show a constant slope in the percolation test and they often show a pH dependent leaching behaviour (see also examples in chapter 2, figure 2.1).

#### 3.6.6 Influence of raw materials on aggregate leaching behaviour.

In part the relationships between the raw materials and the resulting aggregate has already been addressed in section 3.6.1. In Appendix X for a few elements a direct comparison between leaching behaviour of individual elements and the leaching behaviour of the aggregate is given. The significantly higher leachability of Zn in aggregate from shredder waste is consistent with the high level of Zn in shredder waste. At neutral to low pH, however, the effect is hardly noticeable. The increased B leachability in shredder waste also relates to a high level of B in shredder waste. In case of MSWI bottom ash the B level does not result in increased leachability. In case of Ba, the variable levels of Ba leachability are not reflected in the artificial aggregate. The process of pelletising and sintering creates a new mineral matrix with its own leachability characteristic. Only in case of extreme levels of a given constituent will this raw material related behaviour translate into the aggregate leaching characteristics. It is important to know, which elements are more likely to lead to increased release levels. In general, the metals are reasonable tied up in the mineral matrix provided the levels are not too high. The oxyanions Mo, V, As, Se, Sb and Cr are more likely top cause increased release levels as these elements are converted into their oxides upon sintering and become mobile when in contact with water.

#### 3.6.7 Comparison of artificial aggregate with sintered brick behaviour

In the evaluation of emissions, the pelletising and sintering plant are placed in the domain of incineration of waste. It must be realised that as far as the final product of the sintering process

is concerned, the material has no relationship or resemblance with MSWI bottom ash. In terms of its leaching characteristics it fits better with sintered brick manufacturing. In Appendix XI the comparison of artificial aggregate (Tilbury and Glensanda), sintered brick and MSWI bottom ash is given. For most elements, the leaching behaviour of sintered brick and artificial aggregate is very similar. The concentration level may not be the same at all times, but the shape of the leaching curve can be very similar in those cases (same speciation). The MSWI bottom ash differs significantly in its leaching behaviour on several counts. Cu, Ba, Cr and P are really different. For several elements the leaching levels are very different (K, Ca, Na, Sr, Zn, and SO<sub>4</sub>). So in accordance with the microscopic evaluation, also the leaching points at a close relationship between the artificial aggregate production and sintered brick production.

## 3.6.8 Comparison of aggregate leaching with aggregate in concrete

Artificial aggregate is produced primarily for its use as aggregate in concrete in replacement of natural aggregate. The change in leachability resulting from preparing a concrete cube with artificial aggregate can be assessed by comparing the leachability of aggregate with that of size reduced aggregate in concrete. In Appendix XII the results of this comparison are given. Three combinations of aggregate and the concrete cube containing the same aggregate are available. However, these concrete cubes were not tested for all elements. For comparison, concrete containing natural gravel has been analysed as well.

There are almost no elements, where the leachability of the aggregate exceeds that of the crushed concrete cube. Only Se and Mo have a higher leachability. In the case of Cr one of the concrete-aggregate combinations has a lower Cr leachability. This concrete Glensanda 07 must have some form of reducing properties as no other factor can explain the behaviour of Cr, which generally features high leachability in Portland cement.

In most cases the leachability from the concrete-aggregate mix is higher than that of the aggregate alone. This implies that the own contribution from cement is for many elements rather significant: Ba, Cr, K, V, Li, Ca, B, SO<sub>4</sub>, Mg and Sr.

This implies that the environmental quality of the aggregate will generally meet the requirements for making a suitable substitution for natural aggregate. A key issue however, will be the recycling and end-of-life conditions (see section 3.8.3).

## 3.6.9 Comparison of artificial aggregate concrete with Portland cement mortars

A comparison with regular Portland cement mortars allows one to verify to what extent the artificial aggregate influences the overall leaching behaviour of the mortar mix. In Appendix XIII the comparison between artificial aggregates and regular Portland cement mortars is provided [12]. Zn, Ba, Ni and K with the exception of one of the artificial cements are not significantly different. In some artificial aggregate concrete cubes the leachability of Mo, V and As is higher than that of regular Portland cements. The general leaching behaviour of artificial aggregate concrete and regular Portland cements is very similar, which points at the same solubility controlling phases.

#### 3.6.10 Tank test data of artificial aggregate concrete

In Appendix XIV the results of tank leach tests on artificial aggregate concrete is compared with the tank test results from regular Portland cements. The release as observed for artificial aggregate concrete is quite comparable with regular Portland cement. The release of Pb, Co, Mo, Ni, Sb, V, SO<sub>4</sub> and most likely Se as well is elevated compared to regular Portland cement. The release in comparison with regulatory limits will be addressed in section 3.8.

The incorporation of the aggregate into concrete highlights that the release of elements of with elevated release such as Cr, V and K mainly stem from the cement itself with the aggregate being responsible for the majority of the Mo and Se. The release from concrete with artificial

aggregate generally will not lead to limitations. However, the main concern will be in the recycling and "end of life" situation, where leaching conditions due to carbonation as a result of exposure to the atmosphere will turn neutral, under which condition release of elements differs greatly from that at the high pH normally encountered in cement-based products. Oxyanions (e.g. Mo, V, As, Se are more leachable under such conditions and can current regulatory limits.

The tank leach test has been applied to assess the release from monolithic specimen at the own pH (generally pH > 11 in eluate). However, many cement-bound applications are in contact with an imposed neutral pH. The latter situation is most relevant for cement-based products in contact with natural surface water, where the external neutralisation surmounts the supply of alkalinity from within the matrix. The difference between own pH and imposed neutral pH has been evaluated in another study [12]. In figure 3.6 the difference is illustrated for Cd from Portland cement mortars, which shows a higher leachability at neutral pH than at high pH. Irrespective of the regulatory limit values, the 10 times lower release levels at own pH give a false sense of security for conditions where neutralisation will take place. Similar observations have been made with significant differences in release between own and neutral pH. Not in all cases the leachability increases with decreasing pH. The pH dependence test data indicate the direction and the approximate magnitude of change as a result of neutralisation by carbonation.



Figure 3.6 Cd release from monolithic Portland cement mortars using NEN 7345 at own pH and an controlled pH=8 [12].

The tank test data reflect that release as observed in cement mortars generally appears to be diffusion controlled, although not all elements will show this type of release from all mortars. Release curves may reflect a number of different situations, such as depletion of soluble species, when the combination of specimen size and testing time is wrong. Delayed release may occur, when a solubility controlling phase is limiting the release of a given constituent in the initial phase of leaching. Wash-off, where in the first or first two fractions an increase release is noted. Finally, chemical changes may occur during the testing period, such as a change in redox state in the surface of the matrix or a change in pH such that the leachability of specific components in affected significantly. The latter changes can be anticipated from the pH dependence leaching test. Figure 3.7 gives graphic examples of such different release behaviour.



Figure 3.7 Different types of release behaviour in the tank leach test (examples from NEN 7345[6])

From the tank leach test effective diffusion coefficients for the mortar can be derived, which within certain assumptions allows prediction of release at longer time-scale than measured in the lab. The effective diffusion coefficients (expressed as  $pD_e = -\log D_e$ ;  $D_e$  in m<sup>2</sup>/s) can only be used with the corresponding availability data (in mg/kg).

Using these data as starting point predictions can be made of release in long term. For a given scenario either the own pH test data or the neutral pH test data are used. This depends on the exposure condition of the construction product. The combination of pDe, availability with each

their uncertainty provides a range in the release with time, which can be used to assess acceptable impact.

Here the evaluation of impact is based on a comparison with the regulatory criteria as laid down in the Dutch Building Materials Decree [33], in which the release behaviour is incorporated.

#### 3.6.11 Compacted granular leach test versus tank leach test

In situations where the release from aggregate is diffusion controlled, the compacted granular leach test [25] provides a means to assess the relevant release parameters. In Appendix XV the test data for aggregate in the CGLT are compared with the tank test data for artificial aggregate concrete. The CGLT generally provides more consistent results. For many elements the release from the aggregate is not very different from the release from aggregate in concrete. Ca release from the tank leach tests is low, which relates to the low Ca release at very high pH. K release is higher in the tank leach test than in the CGLT. Mo and sulfate release are reduced after incorporation in the cement matrix.

## 3.7 Modelling

The modelling of geochemical reactions allows identification of possible solubility controlling phases. As such it forms the basis of making predictions in the long-term. In addition, it allows modifications to be made to the mix design of raw materials to reach a better end product.

## 3.7.1 Geochemical Modelling

The geochemical modelling focuses at the phases that are relevant from a leaching point of view. It should be realized that the external conditions of the aggregates or aggregate in concrete products exposed to the environment largely determine the release. Mineral phases in the interior of a solid matrix are of much less relevance. The modelling of cement chemistry has traditionally been focused at the high pH environment relevant for the cement paste. However, in environmental modelling the surrounding is important as well, and is in some cases more determining. In addition, as stated in the previous work [13,14], release in the normal service life of cement-based products may be limited. Most cement-based products will be recycled at some point in their life cycle, at which point they may be size reduced and may be carbonated to a large extent as a result of the higher exposed surface area. The size reduction and neutralization are conditions that reflect changes in the leaching behaviour that are better described with the modelling in the pH range 7 - 12 than high pH cement phases only [13]. E.g. ettringite considered to be active in retaining oxyanions will degrade below pH 10 and thus release the bound oxyanions upon further carbonation.

In Appendix XVI modelling results for aggregates are given. Since the material has undergone a sintering, the matrix is strictly inorganic. Several potential solubility controlling phases have been identified over the entire pH or over only a part of the pH range studied:

Ni – Ni(OH)<sub>2</sub> Zn – Zn2 SiO<sub>4</sub> Cu – Cu(OH)<sub>2</sub> and CuO Mg – Mg(OH)<sub>2</sub> and Clinoenstatite (MgSiO<sub>3</sub>) Al – Al(OH)<sub>3</sub> amorphous and Microcline (KAlSi<sub>3</sub>O<sub>8</sub>) Si – SiO<sub>2</sub> quartz P – AlPO<sub>4</sub> and Ca3(PO<sub>4</sub>)<sub>2</sub> SO4- Ba75%Ca25%SO<sub>4</sub> Cr – Ba(S96%Cr4%)O<sub>4</sub> solid solution Ca - BaCaSO<sub>4</sub> Ba – BaCaSO<sub>4</sub> Pb – Pb(OH)<sub>2</sub> Fe – Am. Fe(OH)<sub>3</sub> In figure 3.8 the modelling results for two elements Ni and Cu are illustrated as examples.


Figure 3.8 *Geochemical modelling of the leaching behaviour of Ni and Cu from aggregate pointing at largely inorganic leachability controls.* 

## 3.8 Comparison with regulatory criteria

The long term environmental behaviour of artificial aggregates is approached using the methodology as proposed in ENV 12920 [3]. This is a scenario type of approach consisting of testing and release prediction by modelling.

At the European level no regulatory criteria have as yet been set for the environmental properties of aggregates and aggregates in cement-based products. Such criteria are important to control the use of undesirable raw materials in cement production, aggregate production and use of fillers. In the Netherlands the Building Materials Decree has been developed [33], which addresses both granular (unbound) materials and monolithic materials. In the appendices V and XIV, the regulatory criteria for granular materials (Category I and Category II of the Building Materials Decree [33]) as well as for monolithic materials are inserted for evaluation of the potentially critical components for the use of artificial aggregate in concrete.

As aggregate may be used in different scenarios, different evaluations apply. In addition, the life-cycle of artificial aggregate needs to be taken into account. In figure 3.9 the life cycle of aggregate is visualised. This element of judgement is not implemented in the Building Materials Decree. For Portland cement mortars a first evaluation of this approach has been made [13]. There are still some uncertainties and choices need to be made as to what is a relevant condition (particle size) to assess to judge long term behaviour. Key aspects are the material morphology (particle size) and the buffer capacity of the material, which leads to a most likely pH condition (carbonated). Below the different stages of the life cycle of aggregate are addressed using the relevant test condition that pertains to that scenario.

# LIFE CYCLE OF (SYNTHETIC) AGGREGATE



Figure 3.9 Life cycle of aggregate in concrete with (temporary) storage, service life – recycling, reuse and end-of life conditions

### 3.8.1 Aggregate in temporary storage

After production aggregate may have to be stored prior to use. The question is to what extent this storage can be outside or that artificial aggregates need to be stored covered. An additional question is whether the leachate, if generated, needs to be collected. A distinction can be made here in the storage of the final product at the site of production and the temporary at the location of use. The relevant conditions to be taken into account relate to a reasonable storage time, the

estimated infiltration and the test to be used. The percolation test reflects best the conditions in a storage depot. Due to the relatively short time turn around time a low liquid solid ratio applies.

### Storage at production plant

In case of storage of aggregate at the production site, continuously fresh aggregate is placed and leached. If an storage height of 3 m is assumed with a density of 1000 kg/m3 and a turn around time of one year, the L/S would be around 0.1. For a 100-year production operation, each year this same amount would be leached. So by assuming 100 times the release at L/S=0.1, as measured in the percolation test an estimate of release can be made. In figure 3.10 the results are shown for Mo and Se. It follows that the long term release from an on-site storage facility may be significant relative to the soil impact as defined by the Building Materials Decree. This implies that a storage pad with leachate collection would be required.

#### Temporary storage on site of utilisation

For the temporary storage on the site of utilisation this time of exposure is rather short and the net infiltration may not even lead to leachate production. If a worst case condition were to be selected, a one year storage at 300 mm/year for a height of 3 m and a density of 1000 kg/m3 leads to an L/S of about 0.1. This assumption ignores water uptake by the aggregate. This latter aspect may be unfavourable from a technical point of view as water in the aggregate increases its weight and possibly affects the water/cement ratio. Such condition would lead to a very low release and not lead to requirements of isolation on site.



Figure 3.10 Evaluation of (temporary) storage of aggregate at the production plant or on the site of Utilisation.

### 3.8.2 Aggregate in concrete (service life)

In figure 3.11 the range of release data for aggregate in concrete is given in comparison with regulatory limit values of the Building Materials Decree [33]. Only Se appears to be potentially critical in relation to the low limit values specified in the BMD. Particularly, antimony can become critical, as it is relatively close to the limit. These aspects need to be looked into, when judging the selection of raw materials in aggregate production.



Figure 3.11 Evaluation of aggregate in concrete during service life on the basis of criteria defined by the Building Materials Decree. Results from tank test, NEN 7345 [6] for cement products with Tilbury and Glensanda aggregates in comparison with cement products from ECRICEM [12].

#### 3.8.3 Recycling of construction debris from artificial aggregate in concrete

In recycling, the material is used again in the same application. This implies that the same judgement as for the first round is applicable (section 3.8.2). An issue to be taken into account is the degree to which alternative raw materials will accumulate, when besides artificial aggregate also alternative materials are used in production of concrete and when special fillers are used. During the recycling verification of this accumulation effect may be required. The test to apply would be the concise test.



Figure 3.12 Judgement of construction debris from artificial aggregate in concrete for reuse in road base or embankment (RMC-clay; PFA- coal fly ash; 21-26 aggregate mix with MSWI bottom ash and MSWI fly ash).

#### 3.8.4 Reuse of construction debris from artificial aggregate in concrete

Reuse means that the material is no longer suited for recycling or there is not sufficient demand for that application. In such a case the material is used in a lower ranking application, such as road base or embankment. The condition relevant for that application would be the pH dependence test covering the range from pH 11-12 (intact larger pieces) to pH 7-8 (fully carbonated material. In figure 3.12 this judgement based on pH dependence test is illustrated. As can be seen Sb, Se and SO4 have a level of leaching that may exceed the limits as set in the Dutch Building Decree. Aggregates produced from 100 % coal fly ash (PFA) will have limitations due to Cd, Cu, Mo, Se, and Sb. In addition, mix designs with MSWI fly ash do not perform well from an environmental point of view. This material should not be used in aggregate production.

#### 3.8.5 Artificial aggregate debris in end-of life (disposal)

The judgement for the end-of-life situation for artificial aggregate that is no longer fit for purpose, which will be largely size reduced to a relatively fine graded material can be judged based on the probable full carbonation of such material using the pH dependence test. The relevant condition for judgement would be an imposed neutral pH after size reduction to < 4 mm to mimic long term disposal conditions. The criteria on which the release is judged will now be different from the one described before (section 3.8.4), as the EU landfill Directive applies. For instance, Pb could fulfil the conditions for inert waste disposal.



Figure 3.13 Life cycle of building materials in relation to different types of testing of environmental material behaviour.

#### 3.8.6 Quality control

The hierarchy in testing as developed by CEN TC 292 is equally valid for the judgement of aggregate in concrete as it is for judgement of waste characterisation. The above discussions

illustrate this. For a proper understanding of release controlling factors and in the development of raw material mixes, the basic characterisation data prove to be very useful. However, for a routine quality control such long tests are not suited. The short tests as described in this work can very well be used for quality control purposes. Particularly, when such data are generated against the background of characterisation information now available from this work. When produced aggregate compliance test results fit with the characterisation background, no further action is needed. If a significant deviation occurs, a first check of the analytical work will follow. If that is consistent, the concise test can be applied to verify level of discrepancy with characterisation data. Only, if that fails a full characterisation would be needed. In the latter situation, it better is known what is causing non-compliance with limit values.

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

Table 3.7 Applications and test use

In table 3.7 the test conditions for the different uses and life stages of aggregate are summarised together with the appropriate compliance test. In a more abstract way is shown graphically in figure 3.13, which illustrates the life cycle of building materials including artificial aggregates. The same basic evaluation would apply to other building materials as well.

For each of these applications the relevant boundary conditions can be identified.

A major point is this context is whether the material will remain intact, as that decides which model to apply and what test data to use. Another crucial factor is the degree of contact with

water. Intermittent wetting will result in net slower release in most cases, unless the wetting results in increased degradation (water-air interface, corrosion by sea spray). The degree of contact with water will also have an influence on the exposure to atmospheric  $CO_2$  or  $CO_2$  derived from biological degradation. Permanent submersion will generally result in slow carbonation. Below typical exposure conditions relevant to an overall assessment are given.

### 3.9 Relevant Data from Other Sources

#### 3.9.1 Pb and Zn Additions

Leaching experiments with PbZn slag as aggregate, as fine ground material and straight addition of PbO and ZnO to cement paste have been carried out as a side study in relation to a study on alternative materials in pavement applications [36]. This work has resulted in levels of Pb and Zn in the concrete cubes far exceeding any of the compositions in this work. The Pb concentrations in the concrete containing PbZn slag as aggregate and concrete containing PbO/ZnO as addition to the cement paste are respectively 8750 and 9190 mg/kg. The Zn concentrations in these two materials are respectively 29800 and 22800 mg/kg. In figure 3.14 the pH dependence test data on size reduced cubes are shown in comparison with cements studied in this work. In the relevant pH domain pH > 7 no significant change in leachability was observed in the tank leach test on concrete with PbZn slag for either Pb or Zn due to the solubility control dictated by the cement matrix. For the PbO and ZnO addition to cement, the 100 fold increase of the Zn concentration results in only a 10-fold increase in the leachability from the tank test, whereas Pb release is not changed. It appears that, Zn incorporated during the clinker production (Weimar tests) does not lead to a different leaching behaviour than direct addition of ZnO to the cement paste. If over the testing period only a limited change in pH occurs, the pH dependence test data can be used the obtain an estimate of the magnitude of change in release from the tank leach test.



Figure 3.14 Leaching behaviour of crushed concrete with different levels of Pb and Zn

# 3.9.2 Comparison with Data from other sources

In the framework of the Mammoet project Lytag has been tested as aggregates. In the table 3.8 the total amount, the extractable amount at L/S=100 and pH 4, and the leached amount in a tank test is given. Also for clarity the extractable amount is given as a percentage of the total amount.

	total	extracted	(% tot.)	tank test		total	extracted (% tot.)	tank test
		L/S=100		64 days			L/S=100	64 days
pH4								
Ca	22521	1695	(7.5)	1530	Cu	165	4.82 (2.9)	< 0.42
Na	4003	66	(1.6)	34.7	F	120	18.7 (15.6)	7.48
Κ	20259	81	(0.4)	32	Mo	14	0.65 (4.6)	3.74
PO4	2912	7.5	(0.3)	<59	Ni	152	8 (5.3)	<5.07
SO4	819	1067	(130)	862	Pb	102	0.025 (0.0)	< 0.13
As	47.9	0.33	(0.7)	15.2	Sb	9.6	0.2 (2.1)	0.32
Ba	1274	21.75	(1.7)	8.01	Se	6.7	0.02 (0.3)	0.32
Cd	0.95	0.014	(1.5)	< 0.01	Sn	6		<9.88
Cl	5000	28.5	(0.6)	6.94	V	307	0.35 (0.1)	4.8
Cr	170			<0.98	Zn	213	2.59 (1.2)	1.3

Table 3.8 *Characterisation of leaching from aggregates from E fly ash (mg/kg) (32).* 

# 4. CONCLUSIONS

# 4.1 Aggregate performance

The leaching characteristics of artificial aggregate and aggregate in concrete have been determined by examining the leaching behaviour as a function of pH in the pH range 4 -13, which has revealed very consistent and systematic leaching patterns. The leachability in the pH range 5 to 13 is most relevant from an environmental point of view, as field exposure conditions may well cover this range. Metals generally show a minimum leachability at neutral pH. Oxyanions feature a maximum leachability at neutral pH and salts show no relation with pH. A consistent behaviour between aggregates produced from different starting materials is noticed. A consistent behaviour between pH dependence leaching test and column leaching test data has been observed for the full range of major, minor and trace elements analysed.

On the basis of this information, a concise test for waste testing suitable for quality control of aggregates has been adopted. The concise test offers a good option for an optimised quick testing protocol for quality control of the aggregate production process. This procedure leads to results within a few days. Several crucial aspects of leaching are addressed: solubility control, wash out, pH sensitivity over a wide pH range.

Similar to the situation on other fields, the pH dependence leaching test forms a solid basis for mutual comparison of aggregate leaching behaviour as it has the possibility to make distinction between aggregate types based on chemical aspects. In addition, it sets other commonly used tests in perspective and it provides information on leachability on the different relevant exposure conditions. Based on the pH dependence leaching test information geochemical modelling has been carried out, in which interesting solubility controlling phases (e.g. Ni(OH)<sub>2</sub>), Zn<sub>2</sub>SiO<sub>4</sub>) have been identified. It can be noted that also in aggregates leachability is for many components to large extent dictated by solubility control. Column results at L/S=10 are very comparable to pH dependence test data (L/S=10) at same pH after size reduction (<2 mm). In other words, artificial aggregates can be size reduced for testing. Column data in many cases show a slope of 1, which means solubility control for several components. Data in mg/l at low L/S are relevant for impact in short term (aggregate storage; relevant elements are As, Mo, V, B, Se).

A comparison has been made between the pH dependence data on size reduced (< 2mm) Tilbury2 and Glensanda aggregates on the one hand, and column tests data of intact Tilbury 2, with pellets in up-flow and in intermittent down-flow mode, on the other hand. The results show that for these materials very similar results are found using the different tests. The column up-flow and intermittent down flow are very similar (slight delay in release in down-flow mode). This indicates that for the relatively porous aggregates a percolation test operated at a flow rate of about 14 ml/hour is slow enough for local equilibration to occur.

A comparison between aggregates and aggregates in concrete indicates that for many elements Portland cement dominates the release. This is particularly the case for Cr, V and soluble salts like Na and K.

To evaluate the environmental impact of artificial aggregates, the different uses (Table 4.1) during its life cycle need to be addressed. This implies storage of aggregates prior to use, use as aggregate in concrete (service-life), recycling as aggregate in concrete, reuse of construction debris (including aggregates) in road-base or embankment, and finally disposal of construction debris, including aggregates, for which no use can be found.

If in the stage of construction debris additional environmental problems are created as a result of using alternative raw materials, in aggregate production, better control over the selection of raw materials may be necessary.

Scenario	Stage	Characterisation	Compliance
Aggregate production	Production and quality	pH dependence test on size reduced material	Compliance batch test at own and imposed neutral pH (L/S=10) after size reduction
Aggregate in temporary storage	Service life	Percolation test	Compliance batch leaching test (L/S=2, own pH)
Aggregate in concrete in contact with surface water and constructions on land (pilars, quays, breakwaters, locks)	Service life	Tank leach test (imposed neutral pH) up to 64 days	Monolith compliance test (imposed neutral pH)
Aggregate in concrete from demolition recycled as aggregate for concrete	Recycling	Tank leach test (imposed neutral pH)	Monolith compliance test up to 2 days (imposed neutral pH)
Aggregate in concrete from demolition reused unbound in roadbase or embankment	Reuse	pH dependence test on crushed material and percolation test	Batch test (L/S=10)at own pH and imposed neutral pH using crushed material
Aggregate in road base from demolition reused in road base or embankment	Recycling	pH dependence test on crushed material and percolation test	Batch test (L/S=10)at own pH and imposed neutral pH using crushed material
Aggregate in concrete or in road base from demolition going to landfill	Disposal End-of-life	pH dependence test on crushed material and percolation test	Batch test (L/S=10)at imposed neutral pH using crushed material

Table 4.1 Applications and test use

In the comparison with regulatory limits (relevant regulations are only present in the Netherlands at this stage), the service life of concrete generally does not pose a problem. Only Sb, Mo, and Se seem potentially critical. However, the analytical sensitivity for Se is at a borderline. A more precise analysis may show that Se is less critical (recommendation). The Cr leachability from the artificial aggregates is low. As Cr is easily oxidised to Cr(VI) at higher temperatures, it appears that oxygen starved conditions prevail in the kiln. This condition is favourable and should be maintained.

Significant differences between small particle size (as required by some tests) and coarser graded material (latter more relevant from construction debris evaluation point of view) has been observed. Further work is needed to address the complex issue of finer, fully carbonated material next to superficially carbonated material with a core of high alkalinity. A scenario approach through modelling, testing separate fractions and combined fractions is needed to identify the ANC balance between sub-fractions to conclude in which circumstance the fine or the coarser fraction dominates release.

Cement has a very significant acid neutralisation capacity (ANC). During service life  $Ca(OH)_2$  is only marginally leached and/or converted to calcite. In a recycling stage as unbound

aggregate (construction debris) or in "end of life" conditions (disposal), this conversion can be quite significant (*role of carbonation*). In this process of neutralisation, a significant pH buffer is formed. The equilibrium dictated by calcite ensures a pH between 7.8 and 8.3 for many exposure conditions. For environmental assessment of concrete with artificial aggregate in its different life cycle stages, this implies that a pH domain between pH 7.8 and 12.6 is the most appropriate. Only in rather acidic environments, the buffer may not be sufficient to maintain stable conditions over long time scales.

Porosity measurements by simple vacuum saturation proved consistent with Hg porosimetry, which makes the former a quick and suitable method for aggregate quality control.

From the point of view of a good quality of aggregate and a sustainable material supply, a good potential for artificial aggregate excists on the basis of contaminated soil and or sediment with sewage sludge. These materials are not too heavily contaminated and the supply is sustainable. The organic contaminants, which are generally most critical, are effectively eliminated.

The Construction Products Directive (CPD) requires that "Dangerous Substances" are addressed in all CEN standards for construction products. Based on the results presented this Craft project "Utilising innovative kiln technology to recycle waste into synthetic aggregate" in combination with the ECRICEM project on the leaching studies on cement mortars and the work carried out in the framework of the EU Project Harmonisation of Leaching Extraction Tests can provide valuable input to draft harmonised European Technical environmental specifications for the construction industry.

The EN 1744-3 test from CEN TC 154, a compliance test for aggregates contains an arbitrary choice of conditions unrelated to potential environmental impact. Time leaching series up to 96 hrs (LS=5) on intact artificial aggregates compares better with size reduced material pH dependence test (LS=10), while the EN 1744-3 test systematically gives lower results. This implies that the EN 1744-3 test will underestimate environmental impact and release in practice because it does not reach equilibrium in 24 hrs. The contact time in EN 1744-3 is not suitable for the specified particle size. Size reduction is a simple alternative to avoid this drawback.

For CEN TC 227, aggregate in concrete, CPD requirements can best be reached following the developments of testing of cement-based products, in which the tank leaching test is used as basis for judgement of release during service-life. A percolation test is used for re-use of construction debris as unbound aggregate. These more detailed tests are used for limit setting. For compliance, short tests are available.

For CEN TC 154, aggregate in roadbase, CPD requirements can be met by judging materials in percolation dictated regime. This situation is relevant for most of the relatively porous aggregates used in this application. For non-porous materials, such as molten slags from industrial processes, the size-reduction may prove to be not consistent with applications due to reducing conditions developing after size reduction. In that case, a type of compacted granular leach test is the most appropriate method. This method has similarities with the current EN 1744-3 procedure. Only a different type of configuration should replace the grid and the testing should be longer for the characterisation mode (level 1). The compliance test can be short and similar to the compliance test for monolith materials and can be completed in 2 days.

Artificial aggregate produced in a Trefoil kiln results is a sintered product that has similar characteristics -both micro morphological and leaching behaviour - as conventional sintered brick produced from clay.

# 4.2 Recommendations

It is suggested for a complete environmental evaluation to determine a mass balance for all components, as the gas phase emissions might be critical for some specific constituents. This may lead to acceptance limits for raw material based on waste composition for: Hg, Cd, Cl, SO<sub>4</sub>, F. For this aspect a full-scale operating kiln is crucial, as prediction of emission from small scale facilities are too complex.

It is also suggested to check Se leachability from artificial aggregates and aggregate in concrete using the most sensitive hydride-AAS analysis.

The acid buffer capacity of the aggregates can be controlled by the raw material selection. There is a significant difference in pH of artificial aggregates. Tilbury: pH 9; Waste artificial aggregates: pH 10.5 - 12. In the wastes apparently more free lime remains. This aspect may be related to the porosity. This aspect is recommended for further work. To produce aggregate with sufficiently low porosity the melting temperature of the raw materials should not be too low, as it will then seal the surface and trap any gas formed. For some applications lightweight aggregate is preferable. In that case Ca levels should be increased to create a lower melting point of the aggregate.

The pH dependence test is carried out with  $HNO_3/NaOH$  addition as needed to reach a certain target pH. In case of cement matrices, pH adjustment with  $CO_2$  may prove more appropriate for some constituents. This implies using  $CO_2$  bubbling with control of  $CO_2$  ratio in gasflow to reach a preset pH.

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# 6. APPENDICES













pH Dependence test












































### **APPENDIX III – Porosity**

E.C.N. KARAKTERISERINGS LABORATORIUM

AUTOPORE II 9220 V3.03

PAGE 1

SAMPLE DIRECTORY/NUMBER: NYQVIST5/334		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: TILBURY2	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 12:00:51 02/01/99	

PENETROMETER NUMBER: 116ADVANCING CONTACT ANGLE: 141.3 degPENETROMETER CONSTANT: 10.79 æL/pFRECEDING CONTACT ANGLE: 141.3 degPENETROMETER WEIGHT: 1.0000 gMERCURY SURFACE TENSION: 480.0 dyn/cmSTEM VOLUME:0.4120 mLMERCURY DENSITY:13.6000 g/mLMAXIMUM HEAD PRESSURE: 0.0323 MPaSAMPLE WEIGHT:0.4750 gPENETROMETER VOLUME:3.0000 mLSAMPLE+PEN+Hg WEIGHT:1.0000 g

LOW PRESSURE: EVACUATION PRESSURE: 50 æmHg EVACUATION TIME: 5 min MERCURY FILLING PRESSURE: 0.0097 MPa EQUILIBRATION TIME: 10 sec

HIGH PRESSURE: EQUILIBRATION TIME: 10 sec

> INTRUSION DATA SUMMARY (FROM DIAMETER 0.0020 TO 420.0000 æm)

TOTAL INTRUSION VOLUME = 0.2448 mL/g TOTAL PORE AREA = 25.950 sq-m/g MEDIAN PORE DIAMETER (VOLUME) = 0.2021 æm MEDIAN PORE DIAMETER (AREA) = 0.0066 æm AVERAGE PORE DIAMETER (4V/A) = 0.0377 æm STEM VOLUME USED = 28 %

AUTOPORE II 9220 V3.03

PAGE 2

SAMPLE DIRECTORY/NUMBER: NYQVIST5/334		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: TILBURY2	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 12:00:51 02/01/99	

#### PORE CUMULATIVE INCREMENTAL DIAMETER VOLUME VOLUME æm mL/g mL/g

	, 8	
153.7206	0.0000	0.0000
97.3195	0.0007	0.0007
84.0212	0.0291	0.0284
74.0822	0.0327	0.0036
64.2022	0.0530	0.0202
55.8062	0.0542	0.0012
48.1917	0.0562	0.0019
42.0498	0.0571	0.0010
36.3542	0.0609	0.0037
31.7247	0.0625	0.0017
27.4445	0.0739	0.0114
23.7866	0.0759	0.0019
20.5959	0.0809	0.0050
17.8934	0.0847	0.0039
15.3198	0.0870	0.0022
13.3052	0.0896	0.0026
11.5461	0.0926	0.0031
10.0144	0.0926	0.0000
8.6848	0.0946	0.0019
7.5048	0.0962	0.0017
6.4325	0.0962	0.0000
5.4878	0.0962	0.0000
4.7224	0.0976	0.0014
4.0957	0.0996	0.0019
3.5315	0.1005	0.0010
3.1065	0.1007	0.0001
2.7242	0.1030	0.0024
2.3511	0.1030	0.0000
2.0294	0.1035	0.0004
1.7425	0.1041	0.0007
1.5163	0.1046	0.0004
1.3169	0.1046	0.0000
1.1439	0.1051	0.0006
0.9948	0.1062	0.0011
0.8637	0.1062	0.0000
0.7350	0.1064	0.0001
0.6462	0.1064	0.0000
0.5617	0.1068	0.0004
0.4841	0.1078	0.0010
0.4169	0.1090	0.0012
0.3663	0.1097	0.0007
0.3103	0.1125	0.0028
0.2727	0.1143	0.0018
0.2329	0.1184	0.0042
0.2026	0.1223	0.0039

AUTOPORE II 9220 V3.03

PAGE 3

SAMPLE DIRECTORY/NUMBER: NYQVIST5/334		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: TILBURY2	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 12:00:51 02/01/99	

æm	mL/g	mL/g
0.1766	0.1273	0.0050
0.1531	0.1329	0.0055
0.1327	0.1395	0.0067
0.1153	0.1460	0.0065
0.0994	0.1537	0.0076
0.0862	0.1613	0.0076
0.0750	0.1692	0.0079
0.0649	0.1765	0.0074
0.0563	0.1838	0.0072
0.0486	0.1894	0.0057
0.0422	0.1940	0.0046
0.0367	0.1983	0.0043
0.0317	0.2034	0.0051
0.0275	0.2055	0.0021
0.0238	0.2073	0.0018
0.0206	0.2105	0.0032
0.0179	0.2116	0.0011
0.0154	0.2138	0.0022
0.0134	0.2161	0.0022
0.0116	0.2181	0.0021
0.0100	0.2204	0.0022
0.0087	0.2231	0.0028
0.0065	0.2294	0.0062
0.0049	0.2360	0.0067
0.0042	0.2410	0.0050
0.0038	0.2448	0.0037
0.0042	0.2448	0.0000
0.0049	0.2448	0.0000
0.0005	0.2448	0.0000
0.0087	0.2448	0.0000
0.0100	0.2440	0.0000
0.0113	0.2440	0.0000
0.0155	0.2440	0.0000
0.0178	0.2448	0.0000
0.0178	0.2448	0.0000
0.0205	0.2448	0.0000
0.0237	0.2448	0.0000
0.0315	0.2448	0.0000
0.0364	0.2448	0.0000
0.0420	0.2448	0.0000
0.0489	0.2448	0.0000
0.0585	0.2448	0.0000
0.0647	0.2448	0.0000
0.0747	0.2448	0.0000

AUTOPORE II 9220 V3.03

#### PAGE 4

SAMPLE DIRECTORY/NUMBER: NYQVIST5/334		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: TILBURY2	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 12:00:51 02/01/99	

æm	mL/g	mL/g
0.0863	0.2448	0.0000
0.0996	0.2448	0.0000
0.1150	0.2448	0.0000
0.1327	0.2448	0.0000
0.1547	0.2448	0.0000
0.2047	0.2448	0.0000
0.2771	0.2448	0.0000
0.3647	0.2448	0.0000
0.4873	0.2448	0.0000
0.6513	0.2448	0.0000
0.8661	0.2438	-0.0010
1.1595	0.2427	-0.0011
1.5496	0.2426	-0.0001
2.0846	0.2419	-0.0007
2.7976	0.2412	-0.0007
3.7886	0.2405	-0.0007
5.1127	0.2396	-0.0008
6.9232	0.2394	-0.0003
9.3674	0.2388	-0.0006
12.4753	0.2381	-0.0007
16.9792	0.2370	-0.0011

AUTOPORE II 9220 V3.03

PAGE 1

SAMPLE DIRECTORY/NUMBER: NYQVIST5/333		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GRIND	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 11:57:08 02/01/99	

PENETROMETER NUMBER: 575ADVANCING CONTACT ANGLE: 141.3 degPENETROMETER CONSTANT: 10.79 æL/pFRECEDING CONTACT ANGLE: 141.3 degPENETROMETER WEIGHT: 1.0000 gMERCURY SURFACE TENSION: 480.0 dyn/cmSTEM VOLUME:0.4120 mLMERCURY DENSITY:13.6000 g/mLMAXIMUM HEAD PRESSURE: 0.0323 MPaSAMPLE WEIGHT:0.7180 gPENETROMETER VOLUME:3.0000 mLSAMPLE+PEN+HgWEIGHT:1.0000 g

LOW PRESSURE: EVACUATION PRESSURE: 50 æmHg EVACUATION TIME: 5 min MERCURY FILLING PRESSURE: 0.0097 MPa EQUILIBRATION TIME: 10 sec

HIGH PRESSURE: EQUILIBRATION TIME: 10 sec

> INTRUSION DATA SUMMARY (FROM DIAMETER 0.0020 TO 420.0000 æm)

TOTAL INTRUSION VOLUME = 0.0091 mL/g TOTAL PORE AREA = 1.947 sq-m/g MEDIAN PORE DIAMETER (VOLUME) = 21.0873 æm MEDIAN PORE DIAMETER (AREA) = 0.0067 æm AVERAGE PORE DIAMETER (4V/A) = 0.0187 æm STEM VOLUME USED = 2 % \*\*\*\*

AUTOPORE II 9220 V3.03

PAGE 2

SAMPLE DIRECTORY/NUMBER: NYQVIST5/333		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GRIND	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 11:57:08 02/01/99	

æm	mL/g	mL/g
153.7206	0.0000	0.0000
97.3195	0.0021	0.0021
84.0212	0.0029	0.0008
74.0822	0.0031	0.0002
64.2022	0.0032	0.0001
55.8062	0.0034	0.0002
48.1917	0.0040	0.0006
42.0498	0.0040	0.0000
36.3542	0.0040	0.0000
31.7247	0.0043	0.0003
27.4445	0.0043	0.0000
23.7866	0.0044	0.0001
20.5959	0.0044	0.0000
17.8934	0.0048	0.0004
15.3198	0.0050	0.0003
13.3052	0.0050	0.0000
11.5461	0.0050	0.0000
10.0144	0.0050	0.0000
8.6848	0.0050	0.0000
7.5048	0.0050	0.0000
6.3408	0.0050	0.0000
5.4212	0.0050	0.0000
4.6720	0.0050	0.0000
4.0570	0.0050	0.0000
3.5024	0.0050	0.0000
3.0839	0.0050	0.0000
2.7063	0.0050	0.0000
2.3378	0.0050	0.0000
2.0196	0.0050	0.0000
1./352	0.0050	0.0000
1.3108	0.0050	0.0000
1.5127	0.0050	0.0000
0.002/	0.0050	0.0000
0.9924	0.0050	0.0000
0.0010	0.0050	0.0000
0.7557	0.0050	0.0000
0.0432	0.0050	0.0000
0.4835	0.0050	0.0000
0.4165	0.0050	0.0000
0.3659	0.0050	0.0000
0.3101	0.0050	0.0000
0.2725	0.0050	0.0000
0.2328	0.0050	0.0000
0.2024	0.0050	0.0000

AUTOPORE II 9220 V3.03

PAGE 3

SAMPLE DIRECTORY/NUMBER: NYQVIST5/333		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GRIND	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 11:57:08 02/01/99	

æm	mL/g	mL/g
0.1765	0.0050	0.0000
0.1530	0.0050	0.0000
0.1326	0.0050	0.0000
0.1153	0.0050	0.0000
0.0994	0.0050	0.0000
0.0862	0.0050	0.0000
0.0750	0.0051	0.0001
0.0649	0.0052	0.0001
0.0563	0.0052	0.0000
0.0486	0.0052	0.0000
0.0422	0.0052	0.0000
0.0367	0.0052	0.0000
0.0317	0.0053	0.0001
0.0275	0.0053	0.0000
0.0238	0.0053	0.0000
0.0206	0.0056	0.0003
0.0179	0.0058	0.0002
0.0154	0.0061	0.0003
0.0134	0.0062	0.0002
0.0116	0.0065	0.0003
0.0100	0.0067	0.0002
0.0087	0.0071	0.0004
0.0065	0.0080	0.0009
0.0049	0.0083	0.0004
0.0042	0.0089	0.0006
0.0038	0.0091	0.0002
0.0042	0.0091	0.0000
0.0049	0.0091	0.0000
0.0003	0.0091	0.0000
0.0007	0.0091	0.0000
0.0100	0.0091	0.0000
0.0113	0.0091	0.0000
0.0154	0.0091	0.0000
0.0178	0.0091	0.0000
0.0205	0.0091	0.0000
0.0237	0.0091	0.0000
0.0273	0.0091	0.0000
0.0315	0.0091	0.0000
0.0364	0.0091	0.0000
0.0420	0.0091	0.0000
0.0489	0.0091	0.0000
0.0585	0.0091	0.0000
0.0646	0.0091	0.0000
0.0747	0.0091	0.0000

AUTOPORE II 9220 V3.03

PAGE 4

SAMPLE DIRECTORY/NUMBER: NYQVIST5/333		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GRIND	HP 12:23:11 01/26/99	
SUBMITTER: HOEDE	REP 11:57:08 02/01/99	

æm	mL/g	mL/g
0.0862	0.0091	0.0000
0.0996	0.0091	0.0000
0.1149	0.0091	0.0000
0.1326	0.0091	0.0000
0.1546	0.0091	0.0000
0.2044	0.0091	0.0000
0.2767	0.0091	0.0000
0.3640	0.0091	0.0000
0.4859	0.0091	0.0000
0.6489	0.0091	0.0000
0.8619	0.0091	0.0000
1.1519	0.0091	0.0000
1.5360	0.0089	-0.0002
2.0603	0.0088	-0.0001
2.7539	0.0083	-0.0006
3.7089	0.0080	-0.0003
4.9687	0.0074	-0.0006
6.6614	0.0071	-0.0004
8.8946	0.0068	-0.0003
11.6516	0.0065	-0.0003
15.4904	0.0060	-0.0006

AUTOPORE II 9220 V3.03 PAGE 1

SAMPLE DIRECTORY/NUMBER: NYQVIST5/335		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GLENSANDA	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:04:41 02/01/99	

PENETROMETER NUMBER: 124ADVANCING CONTACT ANGLE: 141.3 degPENETROMETER CONSTANT: 10.79 æL/pFRECEDING CONTACT ANGLE: 141.3 degPENETROMETER WEIGHT: 1.0000 gMERCURY SURFACE TENSION: 480.0 dyn/cmSTEM VOLUME: 0.4120 mLMERCURY DENSITY: 13.6000 g/mLMAXIMUM HEAD PRESSURE: 0.0323 MPaSAMPLE WEIGHT: 0.9340 gPENETROMETER VOLUME: 3.0000 mLSAMPLE+PEN+Hg WEIGHT: 1.0000 g

LOW PRESSURE: EVACUATION PRESSURE: 50 æmHg EVACUATION TIME: 5 min MERCURY FILLING PRESSURE: 0.0097 MPa EQUILIBRATION TIME: 10 sec

HIGH PRESSURE: EQUILIBRATION TIME: 10 sec

> INTRUSION DATA SUMMARY (FROM DIAMETER 0.0020 TO 420.0000 æm)

TOTAL INTRUSION VOLUME = 0.0935 mL/g TOTAL PORE AREA = 2.419 sq-m/g MEDIAN PORE DIAMETER (VOLUME) = 14.2845 æm MEDIAN PORE DIAMETER (AREA) = 0.0196 æm AVERAGE PORE DIAMETER (4V/A) = 0.1547 æm STEM VOLUME USED = 21 % \*\*\*\*

AUTOPORE II 9220 V3.03

#### PAGE 2

SAMPLE DIRECTORY/NUMBER: NYQVIST5/335		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GLENSANDA	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:04:41 02/01/99	

#### PORE CUMULATIVE INCREMENTAL DIAMETER VOLUME VOLUME æm mL/g mL/g

	U	0
153.7206	0.0000	0.0000
97.3195	0.0002	0.0002
84.0212	0.0002	0.0000
74.0822	0.0002	0.0000
64.2022	0.0004	0.0002
55.8062	0.0004	0.0000
48.1917	0.0004	0.0000
42.0498	0.0008	0.0004
36.3542	0.0008	0.0000
31.7247	0.0008	0.0000
27.4445	0.0008	0.0000
23.7866	0.0008	0.0000
20.5959	0.0013	0.0006
17.8934	0.0034	0.0020
15.3198	0.0432	0.0398
13.3052	0.0499	0.0067
11.5461	0.0548	0.0049
10.0144	0.0576	0.0028
8.6848	0.0583	0.0006
7.5048	0.0588	0.0006
6.3522	0.0588	0.0000
5.5095	0.0591	0.0003
4.7644	0.0591	0.0000
4.1355	0.0594	0.0003
3.5337	0.0594	0.0000
3.0695	0.0595	0.0001
2.6776	0.0595	0.0000
2.3518	0.0596	0.0001
2.0320	0.0596	0.0000
1.7769	0.0596	0.0000
1.5403	0.0596	0.0000
1.3274	0.0596	0.0000
1.1384	0.0596	0.0000
0.9898	0.0596	0.0000
0.8681	0.0596	0.0000
0 7503	0.0596	0.0000
0.6446	0.0596	0.0000
0.5629	0.0596	0.0000
0.4831	0.0598	0.0002
0.4193	0.0598	0.0000
0.4175	0.0570	0.0000
0.3090	0.0614	0.0006
0.2718	0.0625	0.0011
0 2376	0.0638	0.0013
0 2042	0.0659	0.0010
0.2072	0.0000	0.0020

AUTOPORE II 9220 V3.03

PAGE 3

SAMPLE DIRECTORY/NUMBER: NYQVIST5/335		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GLENSANDA	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:04:41 02/01/99	

æm	mL/g	mL/g
0.1764	0.0686	0.0027
0.1530	0.0712	0.0027
0.1333	0.0741	0.0028
0.1155	0.0772	0.0031
0.0994	0.0805	0.0034
0.0861	0.0832	0.0027
0.0750	0.0856	0.0023
0.0648	0.0871	0.0016
0.0563	0.0884	0.0013
0.0487	0.0893	0.0008
0.0421	0.0901	0.0008
0.0366	0.0905	0.0004
0.0317	0.0906	0.0001
0.0275	0.0909	0.0003
0.0238	0.0909	0.0000
0.0206	0.0911	0.0001
0.0178	0.0913	0.0002
0.0154	0.0916	0.0004
0.0134	0.0919	0.0003
0.0116	0.0919	0.0000
0.0100	0.0921	0.0002
0.0087	0.0924	0.0003
0.0065	0.0929	0.0005
0.0049	0.0931	0.0002
0.0042	0.0932	0.0001
0.0038	0.0935	0.0003
0.0042	0.0935	0.0000
0.0049	0.0935	0.0000
0.0065	0.0935	0.0000
0.008/	0.0935	0.0000
0.0100	0.0935	0.0000
0.0115	0.0935	0.0000
0.0153	0.0935	0.0000
0.0134	0.0955	0.0000
0.0178	0.0955	0.0000
0.0203	0.0955	0.0000
0.0257	0.0935	0.0000
0.0273	0.0935	0.0000
0.0313	0.0935	0.0000
0.0304	0.0935	0.0000
0.0420	0.0935	0.0000
0.0585	0.0935	0.0000
0.0646	0.0935	0.0000
0.0746	0.0935	0.0000

AUTOPORE II 9220 V3.03

#### PAGE 4

SAMPLE DIRECTORY/NUMBER: NYQVIST5/335		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: GLENSANDA	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:04:41 02/01/99	

æm	mL/g	mL/g
0.0862	0.0035	0.0000
0.0002	0.0935	0.0000
0.0996	0.0935	0.0000
0.1148	0.0935	0.0000
0.1328	0.0935	0.0000
0.1538	0.0935	0.0000
0.2045	0.0935	0.0000
0.2770	0.0935	0.0000
0.3636	0.0935	0.0000
0.4845	0.0935	0.0000
0.6456	0.0935	0.0000
0.8627	0.0935	0.0000
1.1510	0.0935	0.0000
1.5331	0.0935	0.0000
2.0666	0.0935	0.0000
2.7631	0.0935	0.0000
3.7388	0.0935	0.0000
4.9712	0.0935	0.0000
6.6626	0.0935	0.0000
8.8499	0.0935	0.0000
11.7370	0.0935	0.0000
15.6263	0.0933	-0.0002

AUTOPORE II 9220 V3.03

PAGE 1

SAMPLE DIRECTORY/NUMBER: NYQVIST5/336		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: PbZn SLAK	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:08:24 02/01/99	

PENETROMETER NUMBER: 785ADVANCING CONTACT ANGLE: 141.3 degPENETROMETER CONSTANT: 10.79 æL/pFRECEDING CONTACT ANGLE: 141.3 degPENETROMETER WEIGHT: 1.0000 gMERCURY SURFACE TENSION: 480.0 dyn/cmSTEM VOLUME: 0.4120 mLMERCURY DENSITY: 13.6000 g/mLMAXIMUM HEAD PRESSURE: 0.0323 MPaSAMPLE WEIGHT: 3.6020 gPENETROMETER VOLUME: 3.0000 mLSAMPLE+PEN+Hg WEIGHT: 1.0000 g

LOW PRESSURE: EVACUATION PRESSURE: 50 æmHg EVACUATION TIME: 5 min MERCURY FILLING PRESSURE: 0.0097 MPa EQUILIBRATION TIME: 10 sec

HIGH PRESSURE: EQUILIBRATION TIME: 10 sec

> INTRUSION DATA SUMMARY (FROM DIAMETER 0.0020 TO 420.0000 æm)

TOTAL INTRUSION VOLUME = 0.0055 mL/g TOTAL PORE AREA = 1.901 sq-m/g MEDIAN PORE DIAMETER (VOLUME) = 0.0190 æm MEDIAN PORE DIAMETER (AREA) = 0.0048 æm AVERAGE PORE DIAMETER (4V/A) = 0.0117 æm STEM VOLUME USED = 5 % \*\*\*\*

AUTOPORE II 9220 V3.03

#### PAGE 2

SAMPLE DIRECTORY/NUMBER: NYQVIST5/336		
OPERATOR: RN	LP 11:40:40 01/26/99	
SAMPLE ID: PbZn SLAK	HP 14:50:01 01/26/99	
SUBMITTER: HOEDE	REP 12:08:24 02/01/99	

æm	mL/g	mL/g
153.7206	0.0000	0.0000
97.3195	0.0007	0.0007
84.0212	0.0009	0.0001
74.0822	0.0011	0.0002
64.2022	0.0012	0.0001
55.8062	0.0014	0.0002
48.1917	0.0014	0.0001
42.0498	0.0016	0.0002
36.3542	0.0017	0.0001
31.7247	0.0018	0.0001
27.4445	0.0018	0.0000
23.7866	0.0018	0.0001
20.5959	0.0019	0.0000
17.8934	0.0019	0.0001
15.3198	0.0020	0.0001
13.3052	0.0021	0.0001
11.5461	0.0021	0.0000
10.0144	0.0021	0.0000
8.6848	0.0022	0.0000
7.5048	0.0022	0.0000
6.2543	0.0022	0.0000
5.4347	0.0022	0.0000
4.7086	0.0022	0.0000
4.0933	0.0022	0.0000
3.5029	0.0022	0.0000
3.0462	0.0022	0.0000
2.6599	0.0022	0.0000
2.3381	0.0022	0.0000
2.0218	0.0022	0.0000
1.7691	0.0023	0.0001
1.5345	0.0023	0.0000
1.3230	0.0023	0.0000
1.1352	0.0023	0.0000
0.98/4	0.0023	0.0000
0.8663	0.0023	0.0000
0.7490	0.0023	0.0000
0.6436	0.0023	0.0000
0.5622	0.0023	0.0000
0.4825	0.0023	0.0000
0.4189	0.0024	0.0000
0.3630	0.0024	0.0001
0.308/	0.0024	0.0000
0.2/10	0.0025	0.0000
0.2373	0.0025	0.0000
0.2040	0.0025	0.0000

AUTOPORE II 9220 V3.03

PAGE 3

SAMPLE DIRECTORY/NUMBER: NYO	QVIST5/336
OPERATOR: RN	LP 11:40:40 01/26/99
SAMPLE ID: PbZn SLAK	HP 14:50:01 01/26/99
SUBMITTER: HOEDE	REP 12:08:24 02/01/99

æm	mL/g	mL/g
0.1763	0.0025	0.0000
0.1529	0.0025	0.0000
0.1333	0.0025	0.0000
0.1154	0.0025	0.0000
0.0993	0.0026	0.0000
0.0861	0.0026	0.0000
0.0750	0.0026	0.0000
0.0647	0.0026	0.0000
0.0563	0.0026	0.0000
0.0487	0.0026	0.0000
0.0421	0.0026	0.0000
0.0366	0.0026	0.0000
0.0317	0.0026	0.0000
0.0275	0.0027	0.0001
0.0238	0.0027	0.0000
0.0206	0.0028	0.0001
0.0178	0.0028	0.0000
0.0154	0.0029	0.0001
0.0134	0.0031	0.0001
0.0116	0.0032	0.0001
0.0100	0.0033	0.0001
0.0087	0.0035	0.0003
0.0065	0.0039	0.0003
0.0049	0.0045	0.0006
0.0042	0.0050	0.0005
0.0038	0.0055	0.0005
0.0042	0.0055	0.0000
0.0049	0.0055	0.0000
0.0065	0.0055	0.0000
0.0087	0.0055	0.0000
0.0100	0.0055	0.0000
0.0115	0.0055	0.0000
0.0133	0.0055	0.0000
0.0154	0.0055	0.0000
0.0178	0.0055	0.0000
0.0205	0.0055	0.0000
0.0237	0.0055	0.0000
0.0273	0.0055	0.0000
0.0315	0.0055	0.0000
0.0363	0.0055	0.0000
0.0420	0.0055	0.0000
0.0485	0.0055	0.0000
0.0585	0.0055	0.0000
0.0646	0.0055	0.0000
0.0746	0.0055	0.0000

AUTOPORE II 9220 V3.03

PAGE 4

SAMPLE DIRECTORY/NUMBER: NY	QVIST5/336
OPERATOR: RN	LP 11:40:40 01/26/99
SAMPLE ID: PbZn SLAK	HP 14:50:01 01/26/99
SUBMITTER: HOEDE	REP 12:08:24 02/01/99

æm	mL/g	mL/g
0.0862	0.0055	0.0000
0.0996	0.0055	0.0000
0.1148	0.0055	0.0000
0.1328	0.0055	0.0000
0.1537	0.0055	0.0000
0.2043	0.0055	0.0000
0.2768	0.0055	0.0000
0.3632	0.0055	0.0000
0.4838	0.0055	0.0000
0.6443	0.0055	0.0000
0.8605	0.0055	0.0000
1.1471	0.0055	0.0000
1.5261	0.0055	0.0000
2.0539	0.0055	0.0000
2.7404	0.0055	0.0000
3.6974	0.0055	0.0000
4.8979	0.0055	0.0000
6.5316	0.0055	0.0000
8.6211	0.0055	0.0000
11.3340	0.0055	0.0000
14.9184	0.0055	0.0000

### APPENDIX IV SEM Micrographs of sintered bricks and concrete pavement blocks



Concrete pavement block (irregular edgy structure)



Sintered pavement brick (smoothed surfaces, rounded edges)



Sintered pavement brick 2 (smooth surfaces, rounded edges)



Roof tile (sintered, smooth surfaces and rounded edges).



G22 Aggregate mix 44 % Clay and 44 % Coal fly ash 4 % MSWI bottom ash and 7 % Fly ash



G22 Aggregate mix 44 % Clay and 44 % Coal fly ash 4 % MSWI bottom ash and 7 % Fly ash



G22 Aggregate mix 44 % Clay and 44 % Coal fly ash 4 % MSWI bottom ash and 7 % Fly ash


G23 Aggregate mix 42 % Clay and 42 % Coal fly ash 8 % MSWI bottom ash and 7 % Fly ash



G24 Aggregate mix 38 % Clay and 38 % Coal fly ash 16 % MSWI bottom ash and 7 % Fly ash



G24 Aggregate mix 38 % Clay and 38 % Coal fly ash 16 % MSWI bottom ash and 7 % Fly ash



G25 Aggregate mix 48 % Clay and 48 % Coal fly ash and 3.5 % Fly ash



G26 Aggregate mix 48 % Clay and 48 % Coal fly ash and 7 % Fly ash



G26 Aggregate mix 48 % Clay and 48 % Coal fly ash and 7 % Fly ash



































Appendix VI (1) - 4







Appendix VI (1) - 6









Appendix VI (1) - 7



























APPENDIX VI (2)




B

11

Mg

11

Si

11

S

11

13

13

13

13



















































Own pH Tilbury2 Aggregate : 9.58 Own pH Glensanda Aggregate: 10.73 Own pH Brick: 10.2 Own pH MSWI BA 10.6







Own pH Tilbury2 Aggregate : 9.58Own pH Brick: 10.2Own pH Glensanda Aggregate: 10.73Own pH MSWI BA10.6


















## APPENDIX XIV Results from tank leaching test NEN 7345.

Figure 1. Results from tank test. Samples of Glensanda and Tilbury are indicated. All other samples are Portland cement mortars.



Figure 2. Results from tank test. Samples of Glensanda and Tilbury are indicated. All other samples are Portland cement mortars.



Figure 3. Results from tank test. Samples of Glensanda and Tilbury are indicated. All other samples are Portland cement mortars.. SO4 als S.











