

RELEASE FROM RECYCLED CEMENT BASED MATERIALS UTILISING THE DATA HANDLING AND MODELLING TOOL LEACH-XS.

Christian J. Engelsen^{1,2}, Hans A. van der Sloot³,
Erik Stoltenberg-Hansson⁴, Gordana Petkovic⁵

¹ SINTEF Building and Infrastructure. P.O. Box. 123 Blindern. N-0314 Oslo. Norway. Phone: +47 22 96 55 55.
Fax: +47 22 96 57 25. E-mail: christian.engelsen@sintef.no.

² University of Oslo. P.O. Box 1033 Blindern. 0315 Oslo.
Phone: +47 22 85 54 46. Fax: +47 22 85 54 41.

³ Energy Research Centre of the Netherlands. P.O. Box. 1. 1755 ZG Petten. Netherland.
Phone: +31 224 564249. Fax +31 224 568163. E-mail: vandersloot@ecm.nl.

⁴ Norcem A.S. 3950 Brevik. Norway.
Phone: +47 35 51 11 72. E-mail: estolt-h@online.no.

⁵ Norwegian Public Roads Administration. P.O. Box 8142 Dep. N - 0033 Oslo Norway. Phone: +47 22 07 35 00.
Fax + 47 22 07 38 66. E-mail: gordana.petkovic@vegvesen.no.

ABSTRACT

In this study leaching properties were studied from a geochemical modelling point of view in order to explain mechanisms such as sorption to iron and aluminium (hydr)oxide surfaces, complexation to organic matter, precipitation and dissolution of mineral phases. Release experiments were conducted for laboratory prepared concrete samples as well as aged samples using a standardized pH dependent leaching method.

Characteristic leaching patterns were obtained for Ca, Mn, Ni and Sr in the different samples of recycled concrete aggregates. The leaching patterns differed significantly between carbonated and uncarbonated samples demonstrating the information that can be gained from such a leaching test. Geochemical speciation modelling was applied over the entire pH range using the speciation and transport modelling framework Leach-XS/ORCHESTRA.

The pH dependent leaching data were used in the model. Good agreement between predicted and measured equilibrium concentrations almost over the entire pH range. Sorption to Al and Fe surfaces, organic complexation and mineral precipitation were identified as controlling leaching processes at certain pH regions.

The data management and modelling system Leach-XS/ORCHESTRA proved to be powerful tool not only for the leaching process identification, but also in connecting national criteria with laboratory and field data obtained in the project. It was easy to evaluate the environmental impact from the materials considered in this study with respect to Mn and Ni for the chosen road scenario.

1. INTRODUCTION

Leaching processes in waste and recycled materials are complex as a large number of chemical reactions are involved, including sorption, ionic exchange, complexation to organic and inorganic substances, precipitation and dissolution of mineral phases [1]. In theory, every reaction could be described by a reaction constant assuming chemical equilibrium.

Calculation made on a chemical system that includes only one process and a few reactants could be carried out easily. However, the chemical systems in the context of leaching are often multi-component systems, with competing leaching processes, and the mathematical calculations need to be solved iteratively. It would therefore be impossible to let the different equilibrium reaction equations work together in concert and to approach mathematically solutions without a computer model. Furthermore, in order to increase the fundamental knowledge of these processes and therefore be more entitled to predict the long term leaching behaviour, such complex chemical systems need to be described.

In this study the characteristic leaching patterns of Ca, Sr, Mn and Ni were obtained for a range of cement based recycled aggregates (crushed concrete and masonry). This included sample batches taken at the recycling facility (aged) and samples prepared in laboratory (fresh), thus comprising carbonated and none carbonated samples. Geochemical speciation modelling was applied on the leaching results over the entire pH range, using the speciation and transport modelling framework ORCHESTRA [2]. This model was integrated in the data management tool Leaching Expert System (Leach-XS) [3]. An extended version of the thermodynamic database MINTQA2 [4] was used in the highly alkaline systems, including the latest thermodynamic modelling developments [5,6]. The model description used in this paper was developed in a recent study [7] for the major oxide elements in the cement system, and promising results were obtained. The main objective for the modelling work in this paper was to compare the computer modelled data with the measured leaching data in order to demonstrate the relevance of the geochemical approach for crushed concrete leachates. A good match between the predictions and the measurements provides a strong basis for identification of the predominant leaching process in a given pH region by use of the speciation calculations (element partitioning) also provided by the model.

Leach-XS was used as the data management tool in order to relate scenario specific criteria with laboratory and field leaching data. The Norwegian Pollution Authorities (NPA) criteria for Mn and Ni in fresh water [8] were included in the Leach-XS criteria database. These criteria were used in the reverse calculation according to the scenario described in [9]. This resulted in pore water concentrations limits (source term) that also were included in the criteria database. In addition, leaching and pH data determined at field site [10] were used in the same context as they provided the actual release and the pH change over a period of 2.5 years. Data handling and interpretation carried out in this study is highly relevant for the ongoing European standardisation work in CEN/TC 351 "Construction products: Assessment of release of dangerous substances".

2. MATERIALS AND METHODS

2.1 Preparation of materials and test samples

The materials studied are shown in Table 1. Sample batch A, B and C, each of approximately 500 kg, were collected from the recycling facility BA Gjenvinning AS in Oslo. They originated from different old concrete and masonry buildings, i.e. they were partly carbonated. Sample batch D originated from a section of the highway E6 (25 km south of Oslo) that was constructed with concrete pavement in the beginning of the 1980's. Due to reconstruction, the concrete pavement was demolished. Cores of the old road pavement were also taken before demolition in order to assess the carbonation level and the concrete quality in terms of compressive strength. Carbonation was insignificant as the concrete layer had been covered with asphalt. Sample batch E was prepared in laboratory using Portland cement (Norcem Industrisement, CEM I), natural Norstone AS aggregate and admixture (super plasticizer SSP 2000). The clinker content in the cement was 94% as gypsum (4.4%), limestone filler (<0.4%), iron sulphate (0.6%) and triethanolamin as grinding aid (0.06%) were interground. The concrete mix was prepared with a water/cement ratio of 0.6, a cement content of 325 kg/m³ and the admixture content was 1.95% of the cement weight. The casting, curing and hardening were carried out in the concrete laboratory of Norcem A.S. Following a hardening period of 150 days, the cubes were size reduced and fractionated into sample batches E1, E2 and E3, with the particle size ranges 0-4 mm, 4-8 mm and 8-16 mm respectively as listed in Table 1 .

All sample batches were prepared into laboratory samples of 1.3-1.4 kg according to the standards EN 932-2 and EN 15002 [11,12]. One laboratory sample from each sample batch was further size reduced to a particle size of less than 1 mm using an automatic agate mortar. For laboratory samples of relatively coarse grain size, i.e. <16 and 8-16 mm, manual pre-crushing with granite blocks was needed in order to have an appropriate size for the agate mortar. Each of the size reduced laboratory samples were thereafter split into series of 15-20 equal test samples (each of 80-100g), again according to the splitting procedure in EN 932-2. The size reduction from a laboratory sample into test samples (<1 mm) was conducted prior to the leaching tests. We refer to [7] for further details concerning the sample preparation.

The material pH in this study was referred to as the equilibrium pH in the leaching suspension where no acid or base has been added, see paragraph 2.2. In Table 1 it can be seen that the material pH for samples A, B and C were significant lower than for samples D and E, in accordance with the origin of the sample batches described above. It also indicates that sample A was slightly more carbonated than sample B and C, whereas the material pH for sample D was found to be the same as for sample E, i.e. insignificant carbonation. Sample D and E will therefore be referred to as *fresh* samples. The carbonation level of the samples was also verified by thermogravimetric analysis (TGA) online coupled with a mass spectrometer (MS), as significant amounts of CaCO₃ (or other carbonates) only were expected in samples A-C.

According to the Norwegian declaration scheme for recycled concrete aggregate, RCA is classified in the two main categories; Type 1 and Type 2 [13] with different requirements for unbound and bound use. For unbound use, the materials of Type 1 are required to contain more than 96% by weight of crushed concrete and/or natural aggregates. In the type 2 category, the materials must consist of more than 90% concrete, masonry and natural aggregates. In both categories the content of recycled asphalt is limited to less than 5%. The sample batches undertaken in this study belonged to both categories as given in Table 1.

Table 1 List of sample batches and sample properties used in the model [7]

	Sample ^a						
	A	B	C	D	E1	E2	E3
Particle size (mm) ^a	0-10	10-20	10-38	20-120	0-4	4-8	8-16
Classification (Type) ^b	2	2	2	1	1	1	1
Material pH ^c	11.6	11.9	12.0	12.6	12.7	12.7	12.8
Total carbon (%)	2.51	1.56	1.78	0.37	0.32	0.25	0.21
Total inorganic carbon (%) ^d	0.92	0.88	0.88	0.13	0.060	0.039	0.027
Total organic carbon (%)	1.59	0.68	0.90	0.24	0.26	0.21	0.18
Solid Humic Acid (g/kg)	1	0.5	0.5	n.r. ^f	n.r. ^f	n.r. ^f	n.r. ^f
DHA fraction (%) ^e	0.02-0.1	0.02-0.1	0.02-0.1	n.r. ^f	n.r. ^f	n.r. ^f	n.r. ^f
HFO (kg/kg) ^g	0.013	0.016	0.010	0.012	0.023	0.016	0.014

^a Sample A, B, C and D was collected at a recycling plant. Samples E1-E3 were prepared in laboratory using standard Portland cement, type CEM I. The original particle sizes are listed.

^b According to Norwegian declaration system [13].

^c pH taken from the eluate of the leaching test with no addition of acid or base.

^d Determined as total inorganic carbon (TIC) by the TOC Shimadzu analyser and verified by TGA-MS. For sample A, the TGA-MS values were used, see text.

^e Fraction of the leached dissolved organic carbon (DOC).

^f Not relevant parameter.

^g Hydrous ferric oxide.

2.2 Leaching test

The pH dependent batch leaching test CEN/TS 14429 was used [14]. The principle of this method is to assess the pH leaching behaviour of inorganic elements in terms of equilibrium concentrations at different pH values. In order to comply with the representative mass in EN 15002 [11], test portions of approximately 60 g with a particle size of <1 mm was used. In each leaching test a minimum of 9 similar suspensions were prepared with deionized water (> 18.2 MΩ cm) in 1000 ml acid cleaned HDPE vessels. Required amounts of 1.8-3.6 M HNO₃ (Suprapur) or 1.0 M NaOH (analytical grade) were added to the suspensions in order to achieve end pH values in regular intervals between 2 and 13.5. The final liquid to solid ratio (L/S) used was 10 ± 0.3 ml/mg and the suspensions were equilibrated for 48 h in an end-over-end roller followed by sedimentation over night. Following this, the pH and the conductivity were determined, before the suspensions were filtrated through 0.45 μm membrane filters.

2.3 Chemical analysis

In this paper the results concerning Ca, Mn, Pb and Sr are discussed. However, a number of constituents were measured with Varian Vista ICP-OES (Varian Ltd, Australia) in the filtrated eluates, including Al, As, B, Ba, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sn, Ti, V and Zn. The anions Cl⁻, Br⁻, SO₄²⁻ and F⁻ were analysed by ion chromatography (Dionex IC25, CA, US) and the dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) were measured by a TOC analyser (Shimadzu 9000a) in the eluates. In eluates not analysed by ion chromatography, Cl⁻ was determined by titration with silver nitrate (Metrohm 686 Titroprocessor). Measurements of pH in the eluates were conducted on a computerized titration system (Metrohm Basic Titrino 794), while the conductivity was measured with an ion analyser (Hach Sension analyzer). The measured concentrations of all constituents in the eluates were used in the model.

The same elements determined in the eluates by ICP-OES were determined in the solid samples by the same technique after the materials were ground to <125 μm in the agate mortar and decomposed by a mixture of concentrated HNO₃ and HClO₄ (9.5 ml + 0.5 ml).

Total Inorganic Carbon (TIC) contents were determined (as CO₂) by Shimadzu SSM TOC analyser by acidification with concentrated H₃PO₄. In addition, thermogravimetric analyses (TGA) were carried out for all samples in order to verify the inorganic carbon content. The outlet gas from the TGA was analysed online using a HPR-20 quadrupole mass spectrometer (Hiden Analytical) with regard to CO₂. The total carbon (TC) contents in all samples were determined by combustion and infrared detection (Eltra CS 800, Neuss, Germany). The total organic contents in the samples were estimated from the difference between TC and TIC analysis. The ICP-OES, ion chromatography, TIC and TOC (eluates) analyses were carried out at Energy Research Centre of the Netherlands (ECN). TC determinations were performed at Norcem A.S. All other measurements were conducted at SINTEF or University of Oslo, Department of Chemistry.

2.4 Data management tool and geochemical modelling

The recently developed data handling and modelling software Leach-XS (Leaching Expert System) version 1.4.0.1 [3] was used. The system, which has been developed at the Energy Research Centre of the Netherlands (ECN), processes the input data needed for geochemical modelling in a systematic way. The data can be compared with existing European regulatory criteria or leaching data from other materials in its leaching database. National criteria could also be included in the database. In addition, the system has incorporated the speciation and transport modelling framework ORCHESTRA, for which mineral saturation, solution speciation and sorption processes can be calculated based on equilibrium models and thermodynamic data [2]. In this study ORCHESTRA was used for geochemical speciation modelling. An extended version of the thermodynamic database MINTQA2 [4] was used in order to apply the model in highly alkaline systems. We refer to [7] for the reaction equilibrium constants used in the model.

To include the contribution from organic matter interactions (complexation) the non-ideal competitive adsorption (NICA)-Donnan model [15,16] was integrated in the ORCHESTRA framework. In general, these interactions were not believed to be important due to the nature of the cement samples, as reactive humic substances are absent to a large extent. For the samples of old origin (A-C), however, organic matter interactions were taken into account. Only small fractions of the solid and dissolved organic carbon determined in these samples (solid and leachates) were assumed to be reactive, see Table 1. The specific fraction of reactive humic substances used in the model was derived from the modelling of Cu concentrations in the leachates, i.e. when the differences between the predicted and the measured concentrations were less than half an order of magnitude in the pH area of 4-9. Ion activity coefficients were calculated by Davies equation. Oxidizing conditions were assumed in the system and $\text{pH} + \text{pe} = 15$ was therefore used in the simulations.

Specific binding of metal cations and (oxy)anions to hydrous ferric oxide (HFO) was calculated by the model using the generalised two-layer model of Dzombak and Morel [17]. To determine the amount of sorbent mineral applied as input in this model, selective chemical extractions were carried out for samples E1-E3. The content of amorphous and crystalline iron (hydr)oxides was determined by dithionite extraction, whereas the amorphous part was estimated by ascorbate extraction according to [18]. The aluminium (hydr)oxides in the samples were determined by oxalate extractions [19] as the amorphous aluminium hydroxide was treated as a sorbent mineral in the same way as amorphous iron oxide. Leached concentrations of Fe and Al at $\text{pH} < 2$ were also used to calculate the HFO and compared with

the values derived from the extractions. The values used in the model, shown in Table 1, are based on this comparison and an optimisation procedure is explained in [7].

3. RESULTS AND DISCUSSION

3.1 Characteristic leaching patters

The characteristic pH dependent leaching patters can provide important information. In a recent study [7] a description of the leaching mechanisms was given for the major elements (Al, Ca, Mg, Fe, Si and S). The carbonation effects were identified, described and quantified. In Figure 1 the Ca release patterns from [7] are shown. At the material pH it was demonstrated, by means of geochemical modelling, that calcite was predominating for the aged materials (A-C) whereas the cement hydrate phases (C-S-H, CH, AFt and AFm)¹ were predominating for the *fresh* materials (D and E2) with respect to Ca partitioning among the solid mineral phases.

The release patterns for Sr are also included in Figure 1 and the same leaching patterns as for Ca were found with respect to the carbonation. Such aging effects can clearly be identified on the basis of the pH dependent release data. However, to identify the predominating leaching mechanism, geochemical modelling can provide adequate predictions. In the case of Sr the model predictions showed interaction with strontianite and ettringite for the aged and the fresh materials respectively.

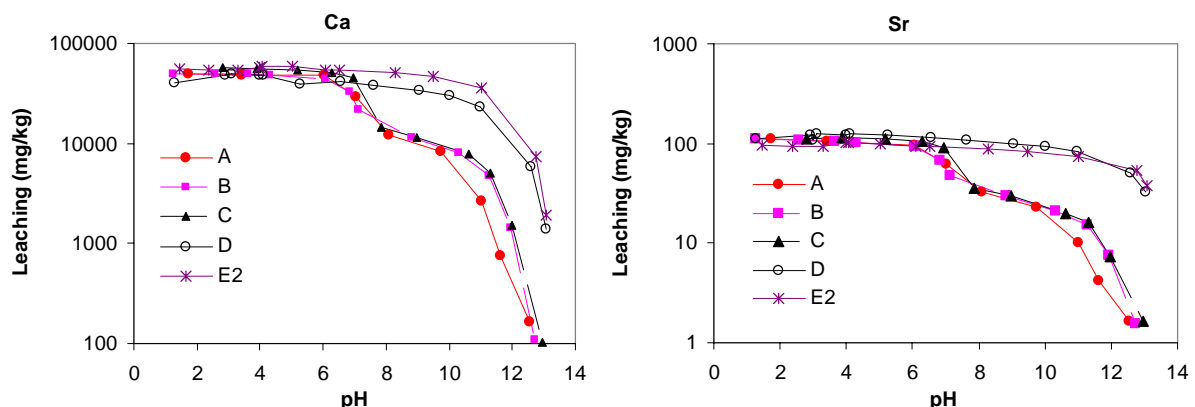


Figure 1 Leaching of Ca and Sr as function of pH for carbonated (A-C) and none carbonated (D and E2) samples. Material pH is the second last data point on each leaching curve.

The release patterns for Mn and Ni are shown in Figure 2. In general, the released quantities increase as the pH decreases over a large concentration range. However, differences can be seen when the release patterns for the aged and fresh materials are compared. Around the material pH (second last data point) the released concentrations were higher for samples A-C compared to samples D and E2. This was more pronounced for the Ni release. Both elements were seemingly more bound in the more *fresh* cement matrix. In the next section model predictions will be used in order to identify the interactions.

¹ Cement chemistry notation: C = CaO; S = SiO₂; A = Al₂O₃; m = mono; F = Fe₂O₃; t = tri; H = H₂O

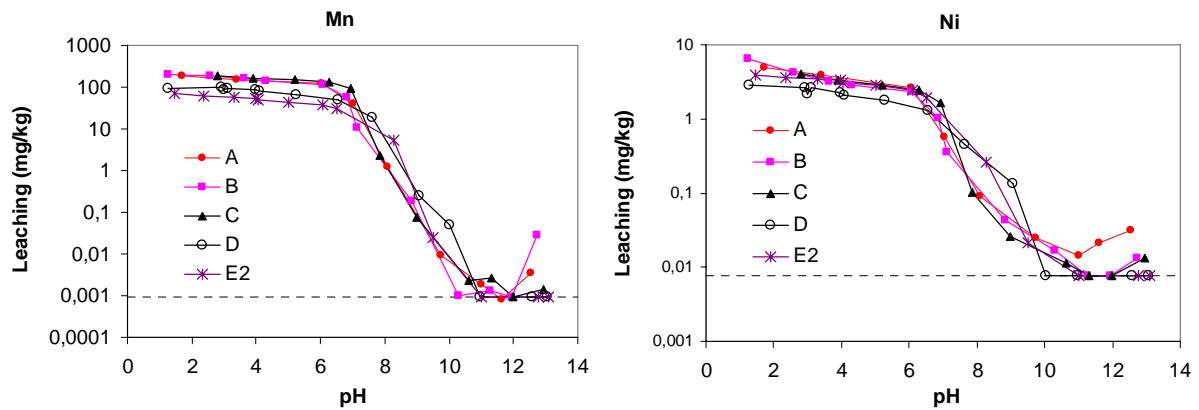


Figure 2 Leaching of Mn and Ni as function of pH for carbonated (A-C) and none carbonated (D and E2) samples. Material pH is the second last data point on each leaching curve. Material pH is the second last data point on each leaching curve. Dashed lines indicate the limit of detection.

3.2 Geochemical modelling

In order to achieve acceptable agreement with measured concentrations and predicted concentrations for the elements of environmental concern, an adequate model description for the major elements are required, as obtained in [7]. The simulated and the measured equilibrium concentrations for Mn and Ni are shown in Figure 3. The concentrations are well simulated almost over the entire pH range for Ni and to a certain extent for Mn. In the most alkaline pH region deviations between prediction and measurements occurred and it was most pronounced for sample D (no carbonation). In order to fully understand some of the leaching processes, the mineral and solution speciation behind the solubility predictions were determined. These calculations are carried out by Leach-XS/ORCHESTRA in the same simulations as for the solubility predictions. A short overview of the leaching processes is included in Figure 3 where the dashed lines indicate the pH regions where the different mechanisms were significant.

It can be seen that sorption to aluminium and iron (hydr)oxides surfaces (HFO) describe the leaching processes well for the Ni release in the relevant pH region, as excellent agreement was achieved. Binding of Mn and Ni to solid organic matter (SOM binding) was also significant in sample A. The speciation calculations, however, showed that the SOM binding had minor influence compared to HFO sorption, although the former interaction was significant over a larger pH region than the latter with regard to the release of Mn.

The speciation predictions in the more alkaline pH region showed precipitation of metal hydroxides. For Ni the leaching processes were described quite well using $\text{Ni}(\text{OH})_2$ as the solubility controlling mineral phase up to a certain alkaline pH (~ 11.5). At higher pH disagreement between prediction and measurements was found for sample D. The model results clearly showed that Ni was bound to the cement hydrate phases as it strongly deviated from the $\text{Ni}(\text{OH})_2$ solubility curve. In addition, it can be seen in Figure 3 that such discrepancies were not found for the Ni released from sample A in the alkaline pH region. This is in agreement with the fact that a large quantity of the hydrate phases in this sample were degraded due to carbonation in service life and the elements that are bound to such phases (sorption/incorporation/substitution) therefore become more available.

The release of Mn was described by the solubility of manganite. Some disagreements between predictions and measurements were achieved in the alkaline pH region. However, Mn interactions with the hydrate phases can most likely be seen in sample D based on the shape of the leaching curve. The largest deviations in the predictions were also found for this sample in the most alkaline pH region.

The modelling results for the other samples were consistent with the results shown in Figure 3. This shows that the model is applicable to laboratory samples as well as samples collected at the recycling site. The model also gives an insight to the leaching mechanisms at different pH levels that is realistic, as the overall match between predictions and measurements are close. In a later publication sorption of anions in the slight acid pH region will be described and calculated by the same model tools.

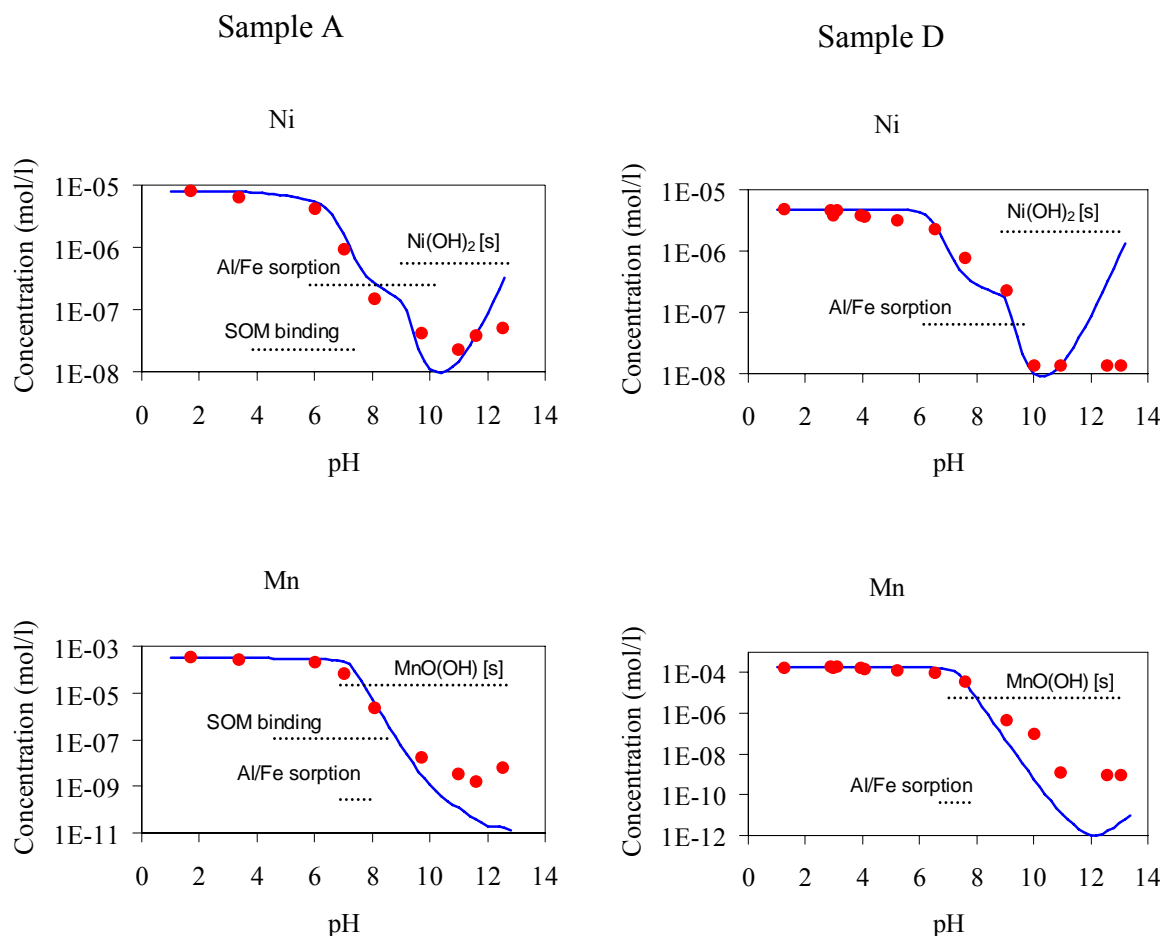


Figure 3 Leached concentrations and model predictions as function of pH for Mn and Ni. Circle points and lines represent the measured concentrations and the model predictions respectively. Dashed lines indicates the pH range of the predicted leaching controlling mechanism, i.e. solubility control, complexation to solid organic matter (SOM) and sorption to aluminium and iron oxide surfaces (Al/Fe).

3.3 Environmental relevance

For the characterisation of the environmental behaviour of construction products and materials, leaching tests alone are not sufficient as the test results need to be related to an impact assessment. This can be done by using the modelling framework Leach-XS/ORCHESTRA.

In the previous section the leaching mechanisms for Mn and Ni in the entire pH region was described in a fresh and an aged material. In addition, a full description of the major oxide element speciation in the cement system is given in [7]. The compliance shown between the pH dependent test data and the data from the geochemical modelling means that it is possible to predict the release from cementitious materials to the water phase under a wide range of exposure conditions. Based on the model results easy and relevant judgements can be done in relation to a release scenario. If for example oxidizing conditions are prevailing, in a percolation scenario (e.g. road scenario), the initial release of SO₄ and Al will be largely dependent on the carbonation degree of the material. Thus, when the material is highly carbonated (final pH ~ 9 in the drainage water), maximum and minimum solubility of SO₄ and Al respectively are expected [7]. At this pH the solubility of Mn and Ni is almost at the minimum as shown in the previous section. In contrast, the solubility will approach maximum at this pH for the elements that tend to form oxyanions under such conditions (CrO₄²⁻, MoO₄²⁻ etc.).

In order to systemise all relevant information and relate the test data to a release scenario in a rational way, the limit values for fresh water class II (moderate polluted) [8] issued by Norwegian Pollution State Authorities (NPA) were included in the Leach-XS criteria database. Furthermore, these values were recalculated and also added to the database according to the road scenario developed in [9] in order to be valid as pore water concentration limits. Finally, we also included the field site measurements from the road scenario specifically described in [10]. These data comprise measurements for almost 3 years with regard to pore water concentrations and field pH. The results are shown in Figure 4 and visualise how laboratory leaching data are used in relation to criteria and field site data. The scenario specific criteria are dependent on how and where the point of compliance (POC) is chosen. In Figure 4 the scenario NPA limit was reverse calculated from a chosen POC to the source term (pore water of the material) at a fixed hydraulic gradient.

The NPA limits show the relevance of an evaluation of the material in relation to an impact assessment scenario. The field site data in Figure 4 show the upper and lower values with respect to pH and the infiltration concentrations. When the data are presented in such a way, it is easy to make the relevant evaluations. From Figure 4 it can be seen that the Mn and Ni release will be complying with the Norwegian criteria for fresh water in the chosen scenario.

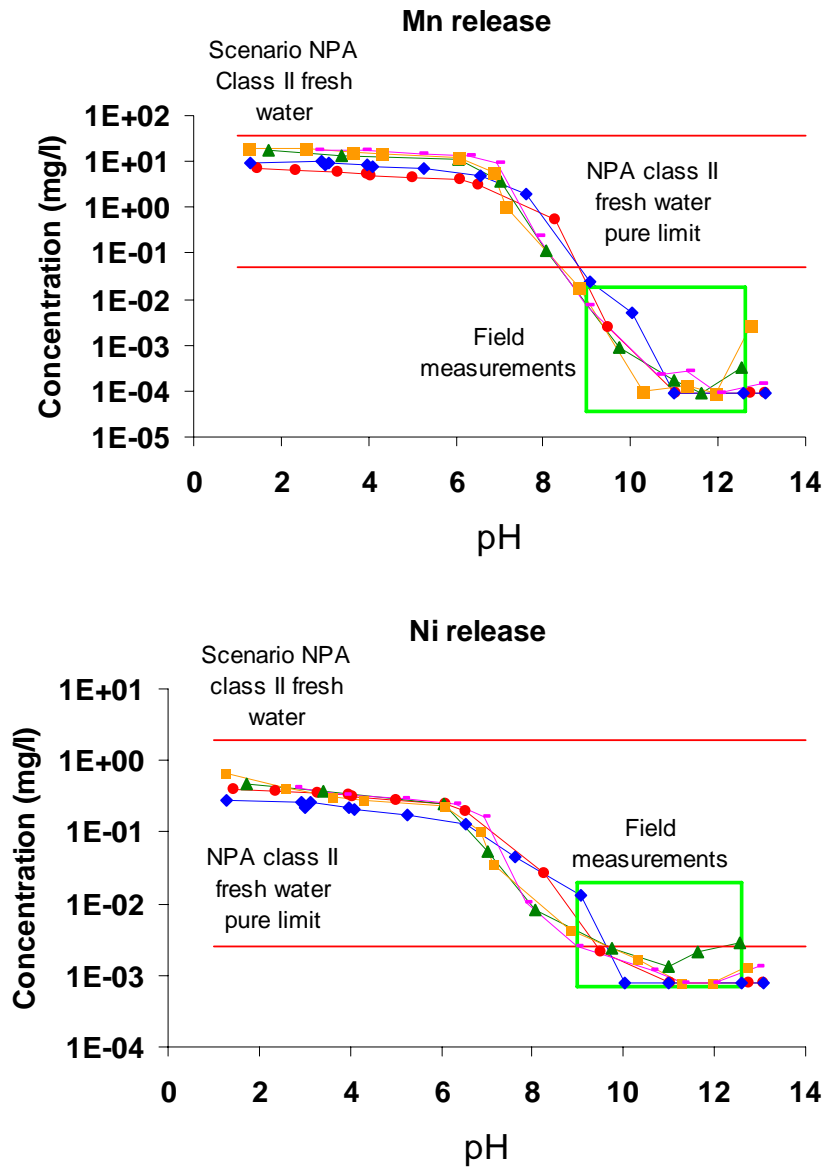


Figure 4 pH dependent leaching data related to an impact assessment scenario. The rectangle shows the upper and lower measured pore water concentrations at field site and also the pH region they appeared in during 2.5 years measurements.

4. CONCLUSION

The pH dependent leaching test provided data that distinguished between aged and *fresh* materials on the basis of the characteristic leaching patterns of Ca, Mn, Ni and Sr. The results in this paper showed how important information could be gained by exploring the pH dependent features of the materials with a standardised test method. By applying such data in the geochemical speciation model Leach-XS/ORCHESTRA, specific insight into the leaching processes at different pH values were possible. In this study good agreement was achieved between model and measured data. Such process identification is of vital importance for making realistic and relevant estimates of the long term release. Furthermore, Leach-XS connected the pH dependent data, scenario based criteria and the field leaching data in a relevant and rational way.

The findings of this study support the Ecricem-project [20] and contribute to the work in CEN/TC 351 “Construction products: Assessment of release of dangerous substances”, which develop horizontal test methods for leaching to soil and groundwater and release to indoor air.

ACKNOWLEDGEMENT

This study has been supported by Norwegian Public Roads Administration, Norcem A.S and Elkem AS Materials. The authors acknowledge Morten Bjerke for assistance in the experimental work.

5. REFERENCES

- [1] F. Glasser, A. Johnson, B. Lothenbach, F. Winnefeld, E. Wieland, A. Wallisch, Mechanisms and modeling of waste/cement interactions - International Workshop, May 8 to 12, 2005, Meiringen, Switzerland, *Waste Management* 26 (7) (2006) 687-688.
- [2] J.C.L. Meeussen, ORCHESTRA: An object-oriented framework for implementing chemical equilibrium models, *Environmental Science & Technology* 37 (6) (2003) 1175-1182.
- [3] www.leachxs.net.
- [4] J.D. Allison, D.S. Brown, K.J. Novo-gradac, MINTEQA2/PRODEFA2, Geochemical assessment model for environmental systems: version 3.11 databases and version 3.0 user's manual; Environmental Research Laboratory, U.S. EPA: Athens, GA, (1991).
- [5] B. Lothenbach, F. Winnefeld, Thermodynamic modelling of the hydration of Portland cement, *Cement and Concrete Research* 36 (2) (2006) 209-226.
- [6] B. Lothenbach, E. Wieland, A thermodynamic approach to the hydration of sulphate-resisting Portland cement, *Waste Management* 26 (7) (2006) 706-719.
- [7] C.J. Engelsens, H.A. van der Sloot, G. Wibetoe, G. Petkovic, E. Stoltenberg-Hansson, W. Lund, Release of major elements from Recycled Concrete Aggregates and geochemical modelling, *Cement and Concrete Research* (2007), submitted.
- [8] Norwegian Pollution State Authorities, Environmental classification of the quality of fresh water, SFT 97:04 (1997).
- [9] G. Petkovic, C.J. Engelsens, A.O. Haoya, G. Breedveld, Environmental impact from the use of recycled materials in road construction: method for decision-making in Norway, *Resources Conservation and Recycling* 42 (3) (2004) 249-264.

- [10] C.J. Engelsen, H.A. van der Sloot, G. Petkovic, E. Stoltenberg-Hansson, G. Wibetoe, W. Lund In *WASCON 2006, 6th International Conference on the Environmental and Technical Implications of Construction with Alternative Materials*; M. Ilic, J.J.J.M. Goumans, S. Miletic, J.J.M. Heynen, J. Senden, Eds.: Belgrade, 2006.
- [11] EN 15002: Characterization of waste - Preparation of test portions from the laboratory sample, (2006).
- [12] EN 932-2: Tests for general properties of aggregates - Part 2: Methods for reducing laboratory samples, (1999).
- [13] J. Karlsen, G. Petkovic, O. Lahus In *Sustainable Building 2002, 3rd International Conference on Sustainable Building*; T.D. Pettersen, Ed.: Oslo, Norway, 2002.
- [14] CEN/TS 14429: pH dependence leaching test (initial acid/base addition), (2005).
- [15] C.J. Milne, D.G. Kinniburgh, W.H. Van Riemsdijk, E. Tipping, Generic NICA-Donnan model parameters for metal-ion binding by humic substances, *Environmental Science & Technology* 37 (5) (2003) 958-971.
- [16] D.G. Kinniburgh, W.H. van Riemsdijk, L.K. Koopal, M. Borkovec, M.F. Benedetti, M.J. Avena, Ion binding to natural organic matter: competition, heterogeneity, stoichiometry and thermodynamic consistency, *Colloids and Surfaces a-Physicochemical and Engineering Aspects* 151 (1-2) (1999) 147-166.
- [17] D.A. Dzombak, F.M.M. Morel *Surface complexation modeling; Hydrous Ferric Oxide*; John Wiley & Sons: New York, 1990.
- [18] J.E. Kostka, G.W. Luther, Partitioning and Speciation of Solid-Phase Iron in Salt-Marsh Sediments, *Geochimica Et Cosmochimica Acta* 58 (7) (1994) 1701-1710.
- [19] L.C. Blakemore, P.L. Searle, B.K. Daly, *Methods for chemical analysis of soils*; sci. rep. 80; NZ soil Bureau; Lower Hutt, New Zealand, (1987).
- [20] H.A. van der Sloot, D. Hoede, R.P.J.J. Rietra, R. Stenger, T. Lang, M. Schneider, G. Spanka, E. Stoltenberg-Hansson, A. Lerat, *Environmental criteria for cement based products - ECRICEM - Phase 1: Ordinary Portland Cements-ECN-C--01-069 (2001), Phase II: Blended Cements (under publication).*