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Harmonisation of leaching test methods in support of EU regulations controlling the beneficial use of industrial slag in construction

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Abstract

The environmental issues related to the beneficial use of steel slag are entering a new phase with the development of regulatory requirements for a decision on End-of-Waste and use of steel slag in construction applications. This has led to the need for testing tools to assess environmental impacts properly. For the characterisation of the release behaviour from soil, sediments, sludge, waste and a wide range of construction products a limited set of basic leaching tests addressing specific aspects of release behaviour are available as standards (technical specification). The robustness validation and intercomparison validation to obtain performance characteristics of the methods are still needed. In contrast to the mostly applied single step leaching tests, the characterisation tests allow chemical speciation modelling to identify release controlling phases and processes. This approach opens new possibilities for waste management and product improvement, when called for. Collecting data from different sources will allow defining a benchmark leaching character for steel slag to be used as reference for QC testing. This approach also provides the justification for reduced testing of regulated substances as well as test frequency based on a risk of non-compliance with set criteria.

Introduction

Steel slag resulting from different steelmaking processes have been used in construction applications, such as coastal protection,[1,2] concrete[3-5] and road base applications.[6-8] The increased interest in beneficial use of alternative materials in construction has also led to an increased use of steel slag and a renewed interest in applying a wider spectrum of steel slag types in beneficial use applications. Such applications have not always been environmentally acceptable,[9] as the emphasis in complying with regulatory requirements was focussed on trace elements and not on aspects such as reducing properties and the high pH of fine grained slag. A few

mistakes in applying unbound fine grained steel slag as sub-base in a parking lot and application of bulk slag in a too small water body with almost stagnant water have resulted in more scrutiny in the judgments of these aspects, for which no criteria exist in any current regulation.[10-13] From several studies in recent years the leaching behaviour of steel slag is reasonably well established,[1,14-17] although for some types of steel slag proper leaching characterisation is still rather scarce. In this work the test methods available for evaluating environmental aspects of steel slag use are addressed in the present regulatory context. In addition, the developments in evaluating test data with respect to testing frequency of relevant substances and practical tools for such evaluation are discussed.

Characterisation leaching tests

Below a short description of the characterisation leaching tests is given with an indication how simplified testing can be carried out, when a material's release behaviour has been established sufficiently.

pH dependence leaching test

This test provides information of the pH sensitivity on the leaching behaviour of the material.[18-21] The listed methods are very similar in nature and, therefore, it is expected that they lead to very comparable results. The leaching test consists of a number of parallel extractions of a material at liquid-to-solid ratio (L/S) = 10 l/kg during 48 hours at a series of pre-set pH values. The pH is adjusted at the start of the experiment with HNO_3 or NaOH (or KOH). After 48 hours of equilibration by end-over-end rotation in PE containers, the suspensions are filtered (0.45 µm) and analysed. The test provides the response of a material to imposed pH changes and an acid-base titration curve to understand the response of the material to acid or base reactions (*i.e.* pH buffer capacity) under environmental scenarios (*e.g.* carbonation, infiltration, sulphur oxidation, soil interfaces).

Percolation leaching test

The column leaching test provides information on the leaching behaviour of the material as a function of the liquid-to-solid ratio (L/S in l/kg).[22-26] The listed methods are essentially similar in nature and, therefore, it is expected that they lead to very comparable results. Multiple eluate fractions are collected over the L/S range 0.1 – 10 l/kg, with the total test duration being approximately 21 days. The leachant is demineralised water or a 0,001 M $CaCl_2$ solution.[24] The test material is applied as received and up-flow (14 ml/h, is applied through a column with a waste height of about 25 cm and a diameter of 5 or 10 cm. The more recently developed DIN 19528[27] deviates from the other listed procedures, as it runs to L/S = 4 l/kg at a much faster flow rate. The L/S ratio can be related to a time-scale via the infiltration rate, density

and height of the application.[28] In this way, the leaching test results can be translated to an emission scenario for a specific application.

Monolith leach test

The monolith leach test provides information on the release per unit surface as a function of time. The method is performed on regular shaped product samples according to standardised procedures.[29-32] The listed methods are essentially similar in nature and, therefore, it is expected that they lead to very comparable results. The specimen is subjected to leaching in a closed tank. Demineralised water is used as the leaching solution at a leachant-to-product volume ratio (L/V) of approximately 5. The leaching solution is renewed at specified time intervals up to 36 or 64 days. The pH, electrical conductivity (EC) and, occasionally, Eh are measured in all eluates before filtration (0.45 µm) and chemical analysis. From this method the predominant release mechanism can be obtained provided the leachant renewal schedule of TS 15863[30] is applied.

Compacted granular leach test

For fine grained materials such as clays, that are granular, but have a very low permeability, the release is based on exposed surface area rather than percolation. The compacted granular leach test has been developed for such materials.[31-33] With the exception of the inner vessel with compacted material, the test is performed very much the same as the monolith leach test. Recently, the possible applicability of this test to coarse granular material, which releases mainly through diffusion, is subject of discussion.

Redox capacity test

The redox behaviour and redox capacity test is not yet standardised at international level, but it is available as a national standard in the Netherlands.[34] It allows identification of whether a material has the potential to impose reducing properties on its leachate. Since the release behaviour under reducing conditions can be very different from the materials behaviour under oxidised conditions, it is important to be aware of the oxidation state of materials.

Sorptive phase parameters for geochemical modelling of the leaching behaviour

The quantities of “reactive” organic carbon in the solid phase (*i.e.* humic acid – HA and fulvic acid – FA) can be estimated by a batch procedure.[35] In short, the procedure is based on the solubility behaviour of HA (flocculation at pH < 1) and the adsorption of FA to a polymer resin (DAX-8). The amount of amorphous and crystalline iron (hydr)oxides in the waste mixture can be estimated by an ascorbic acid (amorphous Fe) and dithionite extraction (amorphous + crystalline Fe).[36] The

amount of amorphous aluminium (hydr)oxides can be estimated by an oxalate extraction.[37] The extracted amounts of Fe and Al were summed and used as a surrogate for hydrous ferric oxides (HFO) in the model.[38] The methods are now standardised in ISO/TC 190 (Soil) under series ISO/DIS 12782 parts 1-5.[39]

Tiered approach for leach testing

As indicated in Kosson *et al.*[40] a tiered approach allows limited testing when sufficient knowledge on materials leaching characteristics is gained. An efficient way of achieving this is by selecting one optimal step from the pH dependence test or a combination of the first few leaching steps from either the percolation or the monolith leach test. The advantage of such an approach is the direct comparability of the data obtained from the full characterisation tests, which can be less straightforward, when applying a single step batch up to a given L/S. By presenting the results in combination with prior characterisation data, the value of judgement of the limited single step increases considerably, as the data is now placed in context.

Analysis of eluates

The eluates from laboratory tests and leachates from field scale studies are preferably analysed for major, minor and trace elements by ICP-AES (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sn, Sr, Tl, V, Zn). Dissolved organic carbon (DOC) and total inorganic carbon (TIC) can be analysed by a Shimadzu TOC 5000a analyzer or a similar instrument. Chloride, fluoride, ammonium and sulphate can be analysed by ion-chromatography. The multi-element methods are highly preferred, as the major elements dictate the chemical environment that controls release of many substances.

Regulations applicable to steel slag

Several regulations may apply to steel slag depending on whether the material is destined for landfill, beneficial use in construction or in agricultural applications. Examples of applicable regulations are the EU Landfill Directive,[41] when not all of the slag produced can be marketed. Article 6 on End-of-Waste criteria under Waste Framework Directive[42] is under development to classify certain alternative material streams as suitable for specific beneficial use applications and thus reduce the burden of handling requirements associated with waste materials. The Construction Products Directive[43] applies in all case where the material is used in construction, which may cover a wide range of applications, such as the use as an added component in concrete, a constituent in brick production or bound and unbound use in road construction. The Construction Products Regulation[44] is in still development, but will go beyond the service life, to which the CPD is restricted. This implies that sustainability aspects and end of life issues are considered more prominently. The

position of steel slag in REACH[45] is not entirely clear.[3] The contribution of Bialucha *et al.* "European environmental policy and its influence on the use of slag products", elsewhere in these Proceedings, gives an update on this.

Evaluation of leaching behaviour of steel slag

In this work we are building on the experience gained in leaching studies on a wide range of materials,[40,46-48] which has shown that there is generally no correlation between leaching and total content. The same characterisation leaching tests (pH dependence, percolation test, monolith leach test and compacted granular leach test) are applicable for waste, soil and soil like materials and construction products (very wide spectrum of products covered in various references).[26,49-53] Below the values of the different characterisation tests for steel slag are provided.

Steel slag types

The following steel slag types can be distinguished:

- Primary steel making slag
    - GBS granulated or ground blast furnace slag
    - ABS air cooled blast furnace slag
    - BOF Basic Oxygen Furnace slag
- Secondary steel making slag
    - EAF C Electric Arc Furnace carbon steelmaking process
    - EAF A Electric Arc Furnace stainless steelmaking slag
    - SMS such as Argon Oxygen Decarburisation (AOD) slag

The amount of characterisation leaching data on the different slag types is limited, which makes generalisations at this point difficult. For the range of slag types, single point test data may be available, but that type of data does not allow conclusions on generic behaviour. As indicated before a main distinction in testing relates to the physical shape of the slag to be tested.

Granulated slag

For granulated slag, the combination of the pH dependence test (*cf.* section 2.1) and the percolation test (*cf.* section 2.2) are the main tools for assessment of the expected emissions at the long-term. The combination of these test methods cover a wide pH range that the material might experience during application as a result of weathering and carbonation reaction. Depending on the application scenario, a wide L/S range might also apply as a result of infiltration water The L/S ratio is related to a time scale through the infiltration rate[28] of the anticipated application scenario.

Monolithic bulk slag

For monolithic bulk slag both pH dependence test (*cf.* section 2.1), the percolation test (only first fraction to simulate pore water composition, *cf.* section 2.2) and the dynamic monolith leach test (*cf.* section 2.3) are the relevant tools.

Coarse granular slag

For non-porous coarse granular slag the compacted granular leach test[31,32] is a tool to address release. This is currently subject of study in the Robustness evaluation program launched by CEN/TC351.

Lysimeter studies and field leachate collection

For verification purposes and to assess the validity of the laboratory testing data, lysimeter studies and field leachate collection are indispensable, as there are various aspects of leaching that cannot be addressed well at laboratory scale. This relates for instance to carbonation by atmospheric or soil derived CO_2, to preferential flow aspects under field exposure conditions (uncovered, partially covered or fully covered), to slow transformation of minerals and biological processes (*e.g.* sulphide oxidation). Comparisons between suitable laboratory test results and field data on the same material are available.[1,2,14,15]

Steel slag type leaching behaviour

The combination of characterisation test results can help understanding the dominant release mechanisms and to predict the resulting long term emission level from the material.[9] In LeachXS[54,55] leaching data on a wide range of materials and products including steel slag have been gathered. The data have been brought together in an overview of which here only a small selection of substances is shown. Figures 1 and 2 give the leaching behaviour of Al, Mn, sulphate, Ba, Cr, Ni, V, Zn from different steel slag types (BOF, EAF, BFS, LD slag) as a function of pH (left) and L/S ratio (right).

The release behaviour as obtained by the pH dependence test is following systematic patterns, which may indicate that the same release controlling phases are active, which needs to be confirmed by modelling. A few samples have been subjected to carbonation and thus show different release behaviour. This is reflected in a wider variation in the cumulative release as a function of L/S. The single points are quality control test results, which generally fall in the range of the characterisation leaching data. Although the data is still limited and not all slag types are represented to the same degree, it appears that for several elements the general leaching behaviour of the different slag types does not seem to be very different. For other elements substantial differences in release are noted. The levels of release may vary

depending on the nature of the slag and this could lead to a benchmark for leaching behaviour of slag types.

Geochemical speciation modelling

Starting from the pH dependence test data, a chemical speciation fingerprint can be generated describing the multi-element release behaviour of industrial slag.

Geochemical modelling

Chemical speciation of the solutions has been calculated with the ORCHESTRA modelling framework[56] embedded in LeachXS.[54,57] The main reason for using this code as opposed to other codes like VISUALMINTEQ or PHREEQC is that LeachXS handles the input for and output from the predefined models runs in ORCHESTRA automatically. In addition, the object oriented nature of ORCHESTRA (flexibility in defining the chemical system) and its speed of calculation are noticeable differences. The input to the multi-element model consists of element availabilities, selected possible solubility controlling minerals, concentrations of reactive Fe- and Al-(hydr)oxides, particulate organic matter and a description of the DOC concentration as a function of pH or L/S (polynomial curve fitting procedure). The mineral phases that were allowed to precipitate were selected after calculation of their respective saturation indices (SI) in the original pH dependence leaching test eluates. Saturation indices were calculated for all > 650 minerals in the thermodynamic database and a selection of the most likely and relevant phases was made based on the degree of fit over a wider pH range, the closeness of the SI value to 0 and expert judgment on the likeliness of possible minerals to occur in the material at hand.

Chemical speciation fingerprint

The results of the pH dependence test are used in mechanistic modelling to quantify the chemical phases (minerals and sorptive phases) that are controlling release. This information is needed to make any success in the prediction of long term release behaviour from any of the slag types. The basic chemical speciation approach is the same for any material, as in all cases mineral dissolution/precipitation, sorption on Fe, Al or Mn (hydr)oxides, and interaction with dissolved (DOC) and particulate organic matter (POM) play a role. It is only the proportions in which specific release controlling phases contribute to the overall release behaviour differ among materials. In Figure 3 an example is given of the chemical speciation modelling results for Ni from a mildly carbonated steel slag. In view of space only Ni is shown, while for all 25 substances simultaneously modelled similar partitioning information is available. The chemical speciation modelling also holds the key for the development of treatment options that will provide a durable solution rather than a

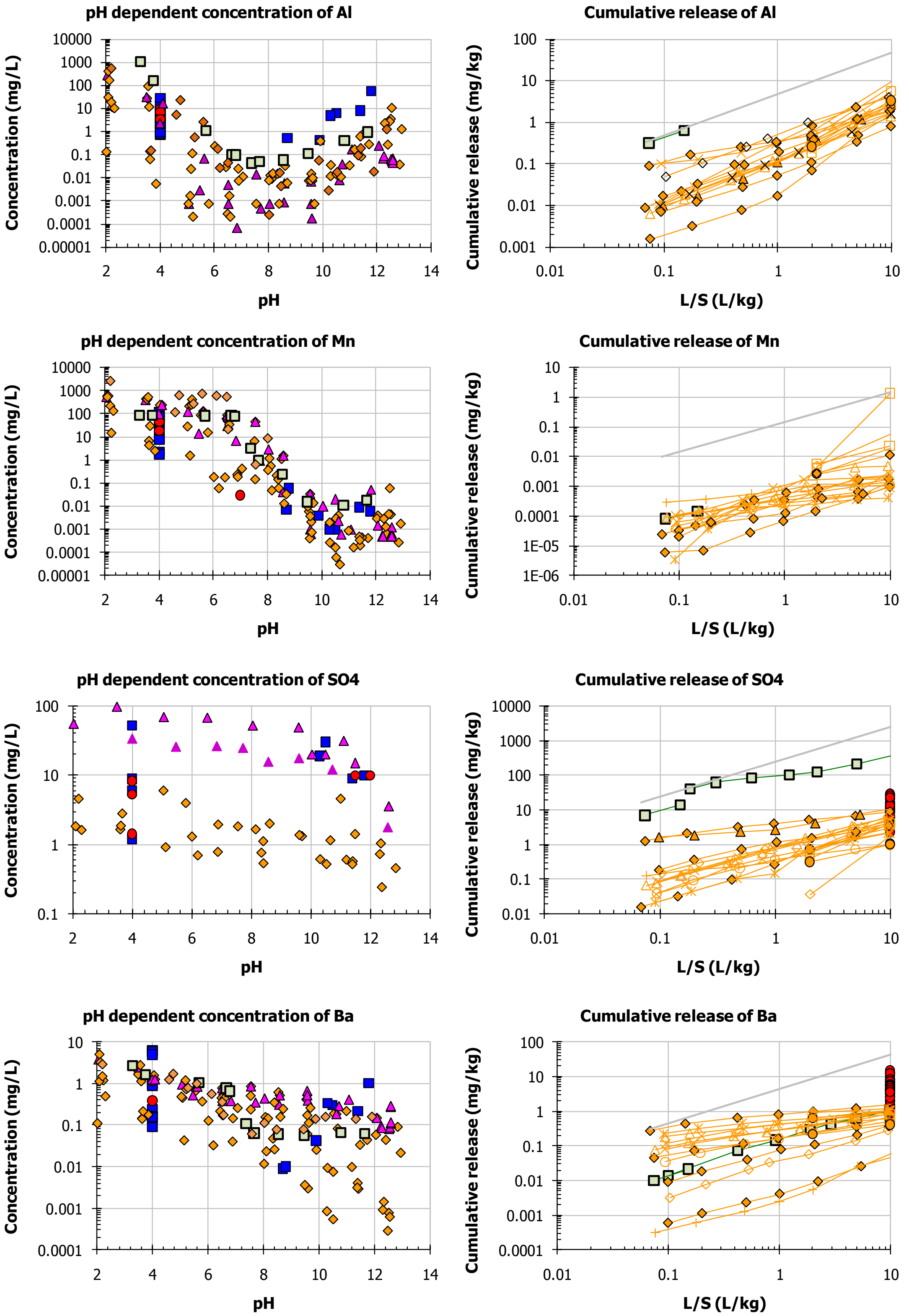

Figure 1: Characterisation leaching behaviour of Al, Mn, sulphate and Ba from steel slag types BFS (light square), EAF (dark square) and BOFS (diamond), LD slag (dot), type not specified (triangle) by the pH dependence test (left) and the percolation (right) test[17,58,59]

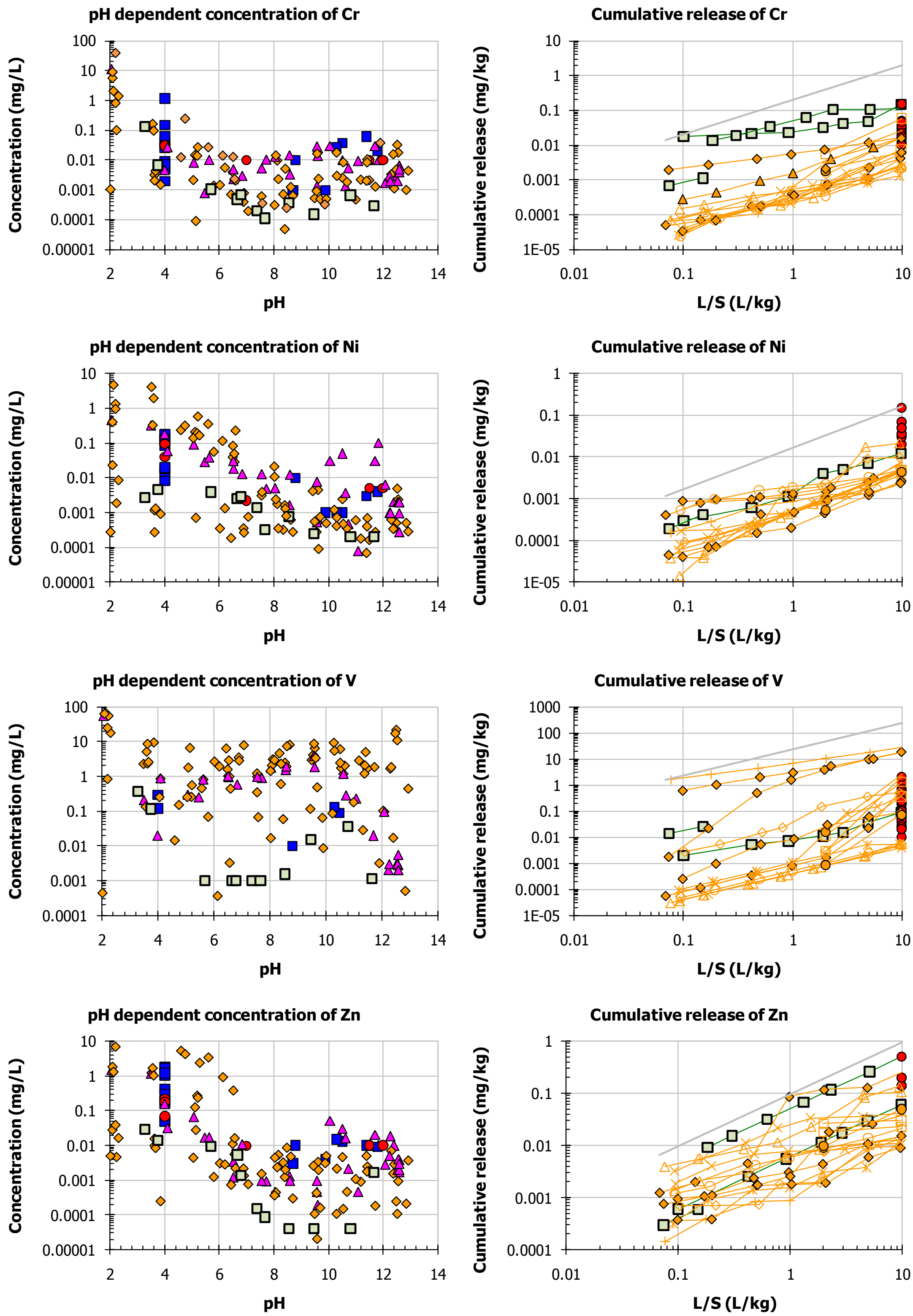

Figure 2: Characterisation leaching behaviour of Cr, Ni, V and Zn from steel slag types BFS (light square), EAF (dark square) and BOF (diamond), LD slag (dot), type not specified (triangle) by the pH dependence test (left) and percolation (right) test[17,58,59]

temporary improvement. Certainly the future CPR will require an assessment of long term release behaviour including recycling stages of the products. Current modelling capabilities hold potential to explore material treatment options prior to costly testing.

Impact modeling and scenarios

The general approach to impact modelling is to define in more detail the application considered and based on that assess the potential impact of release of substances to a point of compliance (*e.g.* a drinking water well). This type of modelling[60,61] has been the basis for the development of the criteria in the EU Landfill Directive,[41] as well as the Dutch regulations for construction products.[10,11]

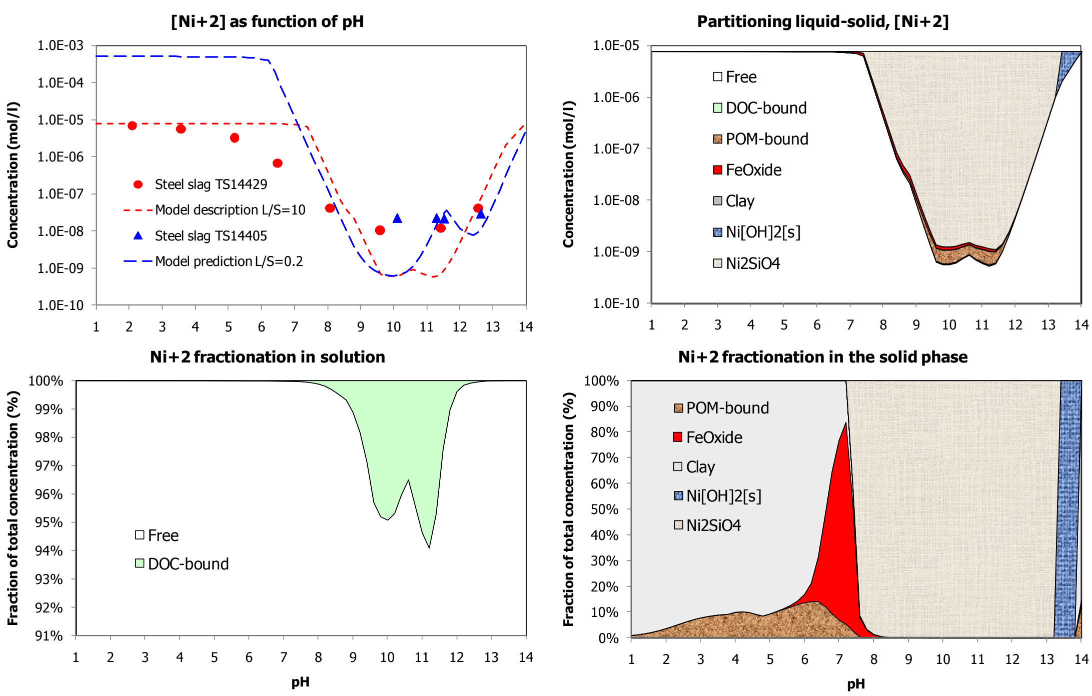

Figure 3: pH dependent release behaviour of Ni from steel slag. Circles – pH dependence test data at L/S = 10; triangles – percolation test data from L/S = 0.1 – 10; line with short dashes – model description for L/S = 10 (total dissolved concentration including DOC bound elements); line with long dashes – model prediction for L/S = 0.2 (same chemical speciation fingerprint used); All concentrations expressed in mol/l (unit volume of solid and liquid with a given L/S). Partitioning of Ni between dissolved and particulate phases for steel slag (top right) and % distribution in liquid and solid phase (bottom left and right).

Approach to address multiple questions related to a material (in line with EN 12920 Methodology):[62]

- Evaluation of the question to be answered (scenario of intended use):
    - Perform the proper testing to assess the relevant properties
    - Evaluate possible critical substances
    - Model the release in the scenario under consideration
    - Verify the outcome against field observations
    - Adjust model when needed;
- Develop criteria by comparison of impact against target objectives or identify compliance with regulatory targets;
- Identify key controlling factors and based on the understanding select a suitable test condition for compliance purposes;
- Draw conclusion on acceptance or rejection.

Cases

Application of LD steel slag as filling material (NL). At the end of 2004, a total amount of 105416 tonnes of granular LD steel slag (0-40 mm) was used as a filling and hardening/paving material in an industrial park (over an area of 80000 m^2). In January 2005, increased pH levels were found in drained (discharge) water and in the nearby surface water, thereby killing fish. When the groundwater below the application was monitored from early February 2005, high pH values (up to pH 13.4) were found.[9] Although the material conformed the 0-40 mm gradation, the main portion of the material was closer to the lower than to the higher range of this particle size range, which resulted in the observed problems.

Application of LD steel slag on the bottom of a pond in a residential area (NL). In 2001, the bottom of a pond in the middle of a small residential area was reinforced and made less permeable with LD steel slag (7-32 mm). This was done to prevent upward “seepage” of groundwater (a common problem in the Netherlands). The pond was I-shaped and had an area of about 46000 m^2. An amount of about 110000 m^3 steel slag was used on the bottom of this pond, a layer with a thickness of about 2.40 metres. All the fish in the pond died only shortly after the application of the slag in the pond. A combination of high pH and low redox potential was measured. The problem here was the small volume of water that was not refreshed regularly (like in a canal or big lake).[9]

The experience gained in these cases can be used as guidance for end users. Clearly, pH and redox conditions resulting from use of an alternative material were not regulated.

QC testing and database

In relation to judgment of slag leaching behaviour and compliance with regulatory criteria questions arise with respect to the variability in production (between facilities of the same type and in time within one facility) and variability between facilities of the same type in different countries. There is also the question of variability between different types of facilities of steelmaking. Finally, slag may be used as produced or after incorporation in a mixture of materials (*e.g.* blended cement, road base mix).

Once a reference base of test data is available from extended characterisation, new test data can be judged against the reference data set to verify compliance of the leaching behaviour. This provides a cost effective means of complying with regulation and at the same time providing detailed information on the behaviour of a material. In Figure 4, new test data (characterisation and compliance test data) are compared with the reference data from earlier work (expressed as the average emission and the 90% confidence interval, n = 13).

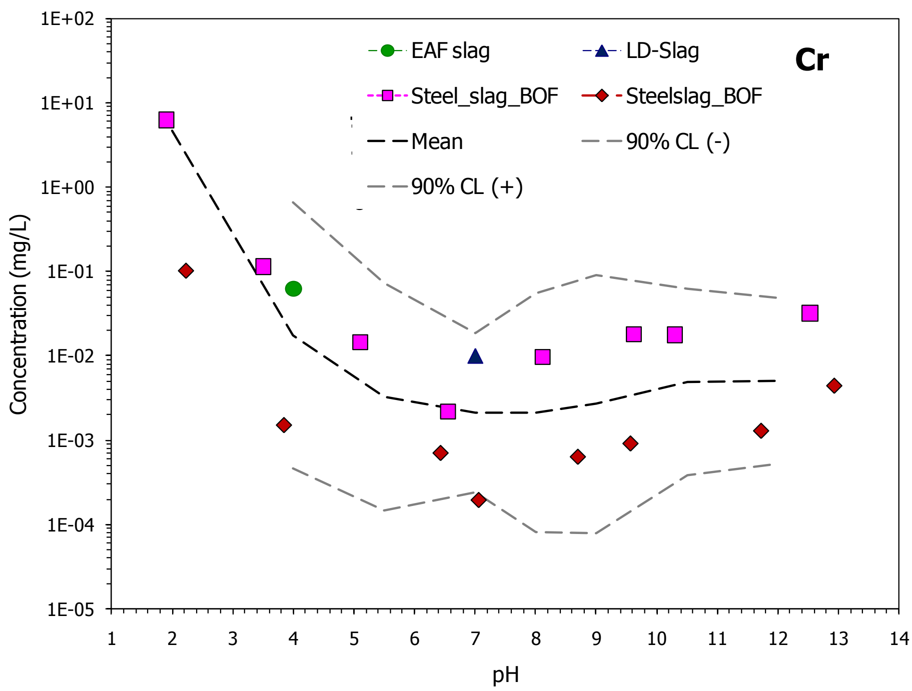

Figure 4: Comparison of a full characterisation of two slag samples against the current reference base on steel slag in the LeachXS database. The single data points (triangle and dot) reflects the possible compliance once the behaviour of a material stream has been sufficiently established.

Both the characterisation test data and the compliance test data fall within the calculated 90% confidence interval of the average emissions. The detailed characterisation data allow choices to be made for quality control and compliance testing with respect to substances of relevance and frequency of testing in case testing is needed. A practical approach is to select one test condition from a full characterisation test for its use in compliance testing as that reduces the need for separate validation of several compliance tests like EN12457.[63-65] The results of such compliance testing shall always be reported in relation to the reference base. LeachXS Lite[47,58] will be a database and data comparison tool, that can provide this functionality.

The experiences with the leaching behaviour of cement mortars from worldwide origin[4,5] and the leaching behaviour of municipal solid waste incinerator bottom ash from worldwide origin,[55] both indicate that leaching behaviour of a material produced in similar installations at different locations in the world produces rather comparable results as far as leaching is concerned. The preliminary results presented above do also suggest that comparable results from characterisation leaching tests are obtained for steel slag. It is concluded that it would be highly beneficial to industry, regulators and end-users, if leaching data for steel slag was brought together to prepare such a benchmark for reference purposes. The context of ferrous slag recycling is described in more detail in the contribution of Bialucha *et al.* "European environmental policy and its influence on the use of slag products", elsewhere in these Proceedings.

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