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PROBLEMS OF ASSESSMENT OF HEAVY METALS LEACHING FROM CONSTRUCTION MATERIALS TO THE ENVIRONMENT

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Abstract

The civil engineering structures and elements, wherein concrete makes the principal component, have been in common use and can be found all around us. Since numerous waste materials and industrial by-products have been used in the production of cement and concrete, attention should be paid to the environmental impacts of contemporary construction materials. Within that area, it is especially important to evaluate the release of heavy metals to the aquatic environment and/or to the soil. The paper presents the test methods which are applicable in the assessment of the leaching level of heavy metals from the construction materials.

Streszczenie

Konstrukcje i elementy budowlane, których podstawowym składnikiem jest beton, są powszechnie stosowane i wszechobecne w środowisku. W związku ze stosowaniem w produkcji cementu i betonu odpadowych oraz ubocznych produktów przemysłowych należy zwrócić uwagę na ocenę wpływu współczesnych materiałów budowlanych na środowisko. Szczególnie ważna w tym kontekście jest ocena uwalniania metali ciężkich do środowiska wodnego bądź glebowego. W artykule przedstawiono problematykę oceny uwalniania metali ciężkich z materiałów budowlanych do środowiska.

Keywords: **Cement; Concrete; Leaching; Heavy metals; Environment.**

1. INTRODUCTION

The scientific circles have, for many years, been involved in the development of a system which would be capable of evaluating the environmental impacts of the construction materials. The work in that field has been initiated by the public concerns within two areas. The first of them is undoubtedly connected with the need to satisfy "the basic requirements" as specified in the Construction Products Directive [1]. That document imposes the obligation to produce the construction materials, and also to use the structures which have been made of those materials, in the way which prevents said materials from influencing adversely human health and the environment.

The other area of concern results from the more and more frequent use of alternative and/or waste materials as substitutes in the construction material manufacturing process, which is especially the case in the production of concrete. The properties of e.g. silica-containing fly ash or granulated blast-furnace slag and their effects on the quality of the cement or concrete products leave hardly any room for doubts. Nevertheless, the environmental impacts of the products in which these have been used are incomplete, and they are still under discussion [2, 3]. The applicable test procedures are needed for long-term evaluation of hazardous substances, including heavy metals, which are released to the environment from composite materials in which secondary raw materials and/or wastes have been used.

The problems of environmental evaluation become a burning question since cement composites have been used more and more frequently in the disposal of hazardous wastes by solidification, if those wastes contain heavy metals [4-9]. Hence, when considering the environmental assessment system for construction materials, one may not disregard those materials/items which have been "produced" in the solidification process.

2. TEST METHODS FOR THE LEACHING ASSESSMENT OF HAZARDOUS SUBSTANCES (HEAVY METALS) FROM CONSTRUCTION MATERIALS

The whole system of tests should first of all give consideration to different environmental conditions to which given construction materials and/or concrete structures are exposed. The author is of the opinion that the assessment methods for the environmental release of hazardous substances should take into account the whole product life-cycle of the construction materials, starting from their production, going through their useful service life, and until their end-of-life stage, with the possibility of recycling or re-use [10].

The key question is whether there is any acceptable level of emissions of pollutants (heavy metals) from the construction products to the soil, surface and/or ground waters, and whether there are versatile methods to establish those levels. The answers to those questions should take a few viewpoints into consideration [2, 3, 11]:

- The construction products are used in various configurations, under various exposure conditions, i.e. at various "application scenarios". However, a limited set of chemical and/or physical factors only will control the release of heavy metals from the construction items, and only a few factors may be dominant in practice.
- Which factor and/or scenario may be of practical importance for every individual product or group of products?
- Which leaching test(s) should be employed for individual products under different "application scenarios" to learn the potential release level of heavy metals?

When selecting a method for evaluation of leaching, one should consider the form of the construction material under investigation: different heavy metal release mechanisms may be involved. In case of monolithic elements, for example, heavy metals will be released as the result of surface wash off, diffusion and/or dissolution processes [12].

Various factors need to be considered under test conditions since they will affect the performance of individual heavy metals. Structural changes due to external impacts (changes in temperature, in pH, contact with water) may produce escalation in the release of heavy metals to the environment [13]. The oxidation and carbonation processes, as well as other corrosive impacts of aggressive media, involve the need of understanding the effects of individual factors on the heavy metal release mechanisms, with consideration given to the whole service cycle of the construction material (civil structure). Hence, the Dutch TANK tests, for example, make allowance for the impacts of changing pH of the extracting liquid on the tested material (pH from 4 to 12) [14]. The external conditions also include the service conditions of the material (structure), the liquid-to-solid ratio (L/S), time of exposure to the extracting medium, type of exposure, pH value, temperature, and mechanical impacts (abrasion, erosion, frost penetration) [15-17].

The internal factors which may be specific to the test construction material comprise: porosity, thermal conductivity, shape, extended surface, size of element and reactivity of its material (susceptibility to carbonation, alkalinity), and the length of its service.

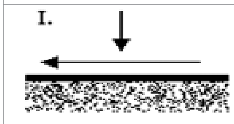
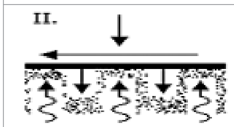
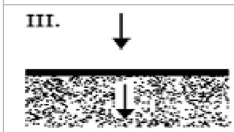
Hence, various possible application scenarios (and their combinations) for the construction materials have been reported in the professional literature [2, 3, 10, 18]:

- granular products which are placed in soil or on its surface,
- monolithic products which are placed in soil or on its surface,
- monolithic products which are alternately subjected to wetting and drying processes (e.g. elements of civil structures which are exposed to weather conditions – rain and sun),
- unbound granular materials e.g. construction debris with varying particle size
- pipes (e.g. drinking water transfer pipes) which are placed underground (leaching of heavy metals is possible both to the transferred water and to the surrounding soil),
- monolithic products which are placed in water (e.g. coastal structures).

A different approach to the application scenarios (Table 1) has been provided in the draft standard, in the documents developed by the European Technical Committee TC 351 [15-17]:

- scenario I – recommended for impermeable products which are placed underground or underwater, and/or the surface of which is washed by moving water, e.g. metal plates, metal strips, roofing-tiles, glass elements, bituminous products;
- scenario II – that is specific for low permeability products, where water is transferred inside their matrices through capillary pores. The soluble substances are transported outside the matrix due to advection and diffusion, e.g. bricks, concrete ele-

Table 1.
The leaching scenarios for dangerous substances from construction materials, and suggested test methods [15-17]

Scenario		Suggested test methods		Examples of materials
	Impermeable materials	Dynamic Surface Leaching Test (DSLT)	Method which tests leaching of dangerous substances versus pH value (pH – test dependence)	Metal plates, glass items, etc.
	Low permeability materials	DSLT method. A method for broken up material should additionally be used.		Concrete items, bricks, mortar, etc.
	Permeable materials	Percolation method (column test)		Soil, broken up material e.g. construction and demolition debris, etc.

ments, mortar, concrete tubes;

- scenario III – permeable (porous) products, through which water finds it easy to penetrate just by gravity, e.g. soil, high porosity materials, construction and demolition debris.

Various test methods can be used in practice to evaluate how much heavy metals are released from the construction materials (concrete, construction materials, construction debris) to the environment [19]. Those methods may be classified from the viewpoint of:

- time of leaching test: long-term methods – like the TANK tests [12] as mentioned above, and short-term methods – e.g. method as per EN 12457-4 [20], which can be used to characterise granular wastes and sediments with regard to their leachability of heavy metals,
- leaching dynamics: static methods – which make it possible to predict the performance of the hardened concrete under static conditions, as for example TANK tests, and which are based principally on EA NEN 7375 [12]; and dynamic methods – these prefer leaching tests under dynamic conditions, as for example the method which is provided in EN 12457-4 [20]. The extracting liquid is contacted with a high surface area of disintegrated waste materials, which makes that method applicable e.g. for construction and demolition debris samples,
- sample pre-treatment – the samples may have the following forms: intact test piece, broken up sample, sample which was cut out from a monolithic block and then broken up. The TANK tests investigate monolithic test pieces which are placed in containers filled up with a leaching liquid (demineralised water) and kept there for a specified period of time. Figure 1 presents a sample in the test container. According to EN 12457-4 [20], on the other hand, a sample of 100 g is broken up to obtain the grain size below 10 mm, it is covered with a suitable volume of water (liquid-to-solid ratio L/S = 10), and it is shaken over 24 h. That standard may be employed to evaluate leaching of heavy metals from construction and demolition debris.

  The column tests in which the test material is subjected to comminution (grain size < 2 mm) and packed into a column make us of the percolation process to leach out heavy metals from the construction material samples [21]. The diagram of an exemplary stand for column tests is presented in Figure 2.
- reaction of the leaching medium – neutral or acidic. According to EA NEN 7375 [12], a liquid (distilled water) is employed with neutral pH which contacts a monolithic test piece, while according to the documents of the Technical Committee CEN PrCEN/TS 14429 [14] the test piece is subjected to the impact of a liquid at different pH levels (from 4 to 12).

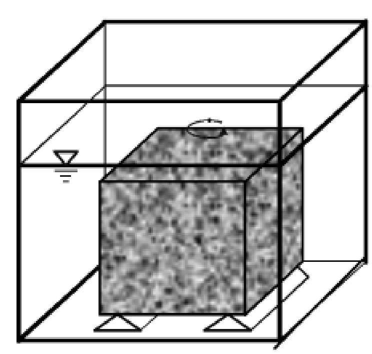


Figure 1.
Principle of placing concrete test pieces in a test container, in accordance with EA NEN 7375 [12]

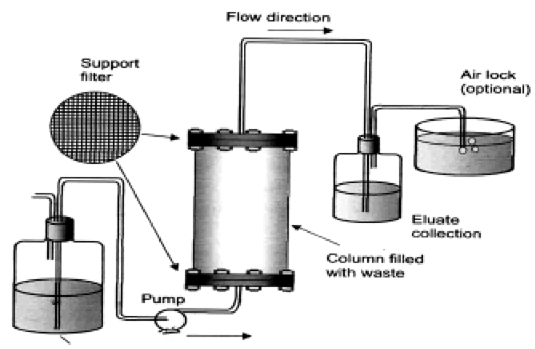


Figure 2.
Experimental stand for up-flow percolation tests

It is also an important issue to establish priorities for the use of leaching tests. Three basic levels are distinguished in the testing hierarchy [22]:

1. Characterisation tests – used for basic characterisation of the release from the product. The collected data allows classification of the materials by categories. One category may cover those materials for which the heavy metals leaching mechanism is similar.
2. Compliance tests – whose purpose is to verify, with the use of other examination methods, whether a material complies with the previous characterisation, and then with the assumed criteria. Once the leaching behaviour has been investigated and characterisation has been established, a simple test which measures the same property (e.g. leaching at a certain pH level) suffices. A close relationship between characterisation and compliance should be ensured.
3. On-site verification/quality control tests – they are quick tests to see whether a material complies with the behaviour as determined earlier or as expected, in its practical application. In general, a simple chemical measurement (e.g. pH, conductivity) or a visual check may be done in this case. In order to be sure, however, at least a compliance test is required.

The main advantage that such a testing hierarchy has, is that once a characterisation step has been done, much more simplified testing on compliance level can be chosen to verify the consistency of subsequent data with the characterisation test results. Examples and detailed descriptions of test methods and how they can be used for various purposes were presented by van der Sloot et al. [22]. The exemplary applications of individual tests at adequate characterisation levels were summarized in Table 2.

3. SUMMARY

The purposefulness and importance of the research programmes to be taken up are confirmed by the fact that the Technical Committee TC 351 has been established within the European Committee for Standardization (CEN), and its aim is to develop applicable regulations in this area which will be adhered to by all EU member states. At the same time, more and more reports are published not only on the need to evaluate the leaching process of dangerous substances from the construction materials and solidification matrices but also on prediction of that process over a longer time horizon. In order to provide that capacity, it is necessary to develop a model for the leaching process versus time, and also versus other parameters (e.g. pH of extracting liquid) which affect the release of heavy metals [3, 24-27]. One of the conditions was, moreover, announced for the complex and correct evaluation of the emission of heavy metals from matrices in which the mineral binder makes the basic component: the tests must give due consideration to the atmospheric impacts [25, 28].

As can also be seen from the documents which were prepared during the work of the Technical Committee TC 351, the efforts made to develop the environmental assessment system for the construc-

Table 2.
Examples of tests at adequate characterisation levels [14, 20-23]

Test	Brief description	Level
prEN14429	pH-dependence test, on granular or size-reduced products	characterisation
prEN14405	column test, on granular products	characterisation
EN12457	batch test (natural pH of extracting medium), granular products	compliance
WI292010	“TANK test”, monoliths	compliance

tion materials (inclusive in particular of the aspect of leaching of hazardous substances) tend to classify those materials into two groups: those which must be subjected to further testing (FT- Further Testing) and those for which preliminary tests will not be followed by any further testing (WT – Without Testing) [29]. That arrangement would be applicable to:

- construction elements which were produced with the use of one or more materials, e.g. binders, pipes, windows;
- materials from which a product was moulded, and which control the release from that product, e.g. bricks, ceramic roofing-tiles, walls which were made of preserved timber; or materials which were directly used in the construction work, e.g. concrete, timber, unbound/bulk aggregates for embankments and road foundations;
- components which are used for the production of material(s), e.g. cement, lime, concrete aggregates;
- elements of a construction product, e.g. plaster-cardboard panels and complete windows which were installed within prefabricated wall units, window frames and window units with double glazing.

The construction materials and their components contain hazardous substances. However, how those materials are used in a construction product and the intended service conditions for that product will determine how those hazardous substances are released from the product, if any. The information of hazardous substances which can potentially be released from a construction product should be based on standard tests as referred to adequate application scenarios, and it should be provided in the label, together with the CE mark.

If the materials were grouped as those for which further tests are required for the release of hazardous substances, and those for which no tests are needed since the mechanism and level of emission is already known, it would make the control procedures easier for the producers; it would also enable implementation of the all-European classification of materials. The document of the Technical Committee contains a provision, however, that the member states will have the right to specify their domestic limit values within the release of hazardous substances to the soil, ground water and surface water, or even within atmospheric indoor emissions [29].

REFERENCES

[1] (89/106/EEC) Construction Products Directive.

[2] *van der Sloot H.A.*; Comparison of the characteristic leaching behaviour of cements using standard (EN 196-1) cement mortar and an assessment of their long-term environmental behaviour in construction products during service life and recycling. Cement and Concrete Researches, Vol.30, 2000; p.1079-1096

[3] *Dijkstra J.J., van der Sloot H.A., Spanka G., Thielen G.*; How to judge release of dangerous substances from construction products to soil and groundwater. ECN-C-05-045.2005

[4] *Hills C.D., Pollard S.J.T.*; The influence of interference effects on the mechanical, microstructural and fixation characteristics of cement solidified hazardous wastes forms. Journal of Hazardous Materials, Vol.52, 1997; p.171-191

[5] *Luz C.A., Rocha J.C., Cheriaf M., Pera J.*; Use of sulfoaluminate cement and bottom ash in the solidification/stabilization of galvanic sludge. Journal of Hazardous Materials, Vol.B136, 2006; p.837-845

[6] *Batchelor B.*; Overview of waste stabilization with cement. Waste Management, Vol.26, 2006; p.689-698

[7] *Kosson D. S., van der Sloot H. A., Eighmy T. T.*; An approach for estimation of contaminant release during utilization and disposal of municipal waste combustion residues. Journal of Hazardous Materials, Vol.47, Issues 1-3, 1996; p.43-75

[8] *Król A., Giergiczny Z.*; Immobilization of heavy metals (Pb, Cu, Cr, Zn, Cd, Mn) in the mineral additions containing concrete composites. Journal of Hazardous Materials. Vol.160, 2008; p.247-255

[9] *Król A.*; Heavy metals leaching from composites solidifying galvanic sewage sludge. Polish Journal of Environmental Studies, Vol.16, No. 2A, Part III, 2007

[10] *Ehrnsperger R., Misch W.*; Implementation of Health and Environmental Criteria in Technical Specifications for Construction Products. Deutsches Institut für Bautechnik (DIBt). Research Report 200 62 311/2006

[11] Schemes for testing either "new"/unapproved constituents of concrete and mortar or production concrete and mortar for release of regulated dangerous substances into soil, groundwater or surface water. CEN/TC 51/WG12/TG6/2005

[12] EA NEN 7375:2004 Leaching characteristics of moulded or monolithic building and waste materials. Determination of leaching of inorganic components with the diffusion test. The Tank Test

[13] *Helms G., Thorneloe S.*; Improved leach testing for evaluating fate of metals from coal ash. 7th International Conference on the Environmental and Technical Implications of Construction with Alternative Materials, Lyon, France, 2009

[14] PrCEN/TS 14429:2004 Characterisation of waste – Leaching behaviour tests – Influence of pH on leaching with initial acid/base addition.

[15] PrCEN/TC 351/WG1 N 117 TS – 2 2009 – 01 – 19

[16] PrCEN/TC 351/WG1 N 118 TS – 3 2009 – 01 – 19

[17] PrCEN/TC 351/WG1 N 142 TS – 1 2009 – 03 – 15

[18] *Bhatty M.S.Y.*; Fixation of metallic ions in Portland cement. Proceedings of the 4th National Conference Hazardous Wastes and Hazardous Materials, Washington 1987; p.140-145

[19] *Nagataki S., Yu Q., Hisada M.*; Effect of leaching conditions and curing time on the leaching of heavy metals in fly ash cement mortars. Advances in Cement Research, Vol.14, No.2, 2002; p.71-38

[20] EN 12457-4:2002 Characterisation of waste – Leaching Compliance test for leaching of granular waste materials and sludges – Part 4: One stage batch at a liquid to solid ratio of 10 l/kg for materials with particle size below 10 mm (without or with size reduction)

[21] DIN 14405:2004 Characterization of Waste. Leaching behaviour tests. Up-flow percolation test

[22] *van der Sloot, H.A., Dijkstra J.J.*; Development of horizontally standardized leaching tests for construction materials: a material based or released based approach? ECN-C--04-060, Energy Research Centre of the Netherlands (ECN): Petten, The Netherlands; 2004

[23] CEN TC 292 WG2 Monolith compliance leaching test WI292010

[24] *Van Herck P., Van der Bruggen B., Vogels G., Vandecasteele C.*; Application of computer modelling to predict the leaching behaviour of heavy metals from MSWI fly ash and comparison with a sequential extraction method. Waste Management, Vol.20, 2000; p.203-210

[25] *van der Sloot H.A., van Zomeren A., Meeussen J.C.L., Seignette P., Bleijerveld R.*; Test method selection, validation against field data, and predictive modelling for impact evaluation of stabilised waste disposal. Journal of Hazardous Materials, Vol.141, 2007, p.354-369

[26] *Beyer Ch., Konrad W., Maier U., Susset B., Park Ch.H., Rügner H., Bauer S., Liedl R., Grathwohl P.*; Assessment of contaminant leaching from secondary materials in road constructions – numerical modelling of mass transfer and attenuation processes. 7th International Conference on the Environmental and Technical Implications of Construction with Alternative Materials, Lyon, France, 2009

[27] *Sakanakura H., Endo K., Osako M., Sakai S.*; Development of an environmental behaviour prediction model incorporating predominant parameters obtained from leaching tests. 7th International Conference on the Environmental and Technical Implications of Construction with Alternative Materials, Lyon, France, 2009

[28] *Steketee J., Timmerije M., van Aalten J.*; Accelerated carbonation and washing of MSWI bottom ash: from laboratory to full scale. 7th International Conference on the Environmental and Technical Implications of Construction with Alternative Materials, Lyon, France, 2009

[29] Technical Report – Final Draft pr CEN/TR 15858. Construction products – Assessment of the release of regulated dangerous substances from construction products based on the WT, WFT/FT procedures; 2008