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Influence of leachate composition on the leaching behaviour of concrete

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ABSTRACT

In this work the leaching of heavy metals from concrete in contact with different leachates was investigated to quantify the influence of the leachate composition. Four different types of ground water, two synthetic waters as well as deionised water were selected and tank leaching tests were carried out using the standardised horizontal dynamic surface leaching test (DSLT). Four concrete mixtures with varying binder compositions and water/binder ratios were tested. The results show that the leachability of trace elements and heavy metals strongly depends on the water hardness respectively the calcium concentration and the availability of hydro carbonate in the leachate. The impact of these parameters is presented. It is pointed out, that the use of deionised water delivers overrated element releases and that by the use of ground water as leachate more realistic evaluations of the environmental compatibility of concrete are achievable.

1. Introduction

A main task in modern days is to reduce the environmental impact of building materials during their production and their application. In order to lower the CO_2-footprint and the consumption of natural resources secondary fuels and raw materials, like ashes, slags and alternative aggregates, are increasingly used in cement and concrete production. Despite all the benefits of this recovery of wastes and by-products it has to be considered that these materials may contain potentially harmful substances like heavy metals and trace elements. Accumulated they lead to higher contents of environmental relevant substances in cement and concrete [1–3].

Being aware of that problem, different countries in Europe developed testing regimes for the leaching of heavy metals and trace elements from concrete and other building materials. The European commission assigned the working group CEN/TC 351/WG1 to develop standardised assessment methods for harmonised approaches relating to dangerous substances. After about a decade of work a technical specification for the so-called "Horizontal Dynamic Surface Leaching Test" (DSLT) was published in 2014 (CEN/TS 16637-2) [4]. The test is to be used for monolithic building materials like concrete. Compact specimens are leached in deionised water over a period of 64 days including 8 leaching steps. The evaluation of the test results is regulated individually in the different member states of the EU.

In Germany a guideline published by the Centre of Competence for Construction *(Deutsches Institut für Bautechnik,* DIBt) is used to evaluate the leaching behaviour of products and raw materials that need a technical approval. The concept is based on threshold values for environmental relevant inorganic and organic parameters in ground water defined by the Federal Working Group on Water Issues in Germany (LAWA) [5,6].

Mass transport finite element simulations are used to predict the concentration of leached substances in the ground water in contact with a concrete building. Using the threshold values for ground water, limit values for the release of heavy metals and trace elements in the DSLT were derived. Two assumptions respectively constraints have to be considered when dealing with the guideline:

a) The release of each element at the concrete surface is assumed to be a diffusion controlled process,
b) Real conditions (e.g. buffer systems, ground water composition, adsorption in the soil) are not considered in the simulation.

So far, the guideline does not consider the impact of using deionised water for the leaching experiments. The dissolving attack due to deionised water on concrete as well as the higher leachability of trace elements and heavy metals are not taken into account. The leachability of the critical elements depends on the pH of the leachate [7–9]. By using tap water or ground water instead of deionised water the buffering capacity in the leachate is vastly increased due to dissolved carbonates in the natural waters, leading to lower pH values. The change in pH can alter the solubility by several orders of magnitude [10]. For many heavy metals the leachability from cementitious materials increases as the pH decreases.

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On the other hand it has to be considered that, due to the low ion concentration of the deionised water, the concrete samples are exposed to an aggressive environment during the experiments, by which the leaching process is accelerated and may consequently be overrated. As deionised water doesn't emulate real leaching conditions for materials exposed to the environment, studies with ground water or synthetic mineralised waters are necessary. In an approach of Lu et al. [11] leaching experiments with simulated ground water were carried out, showing a slightly lower release of heavy metals.

In this work several types of real ground water - considering the geological landscape in Germany - are investigated to quantify their impact on the leaching behaviour of concrete following the technical specification DIN CEN/TS 16637-2, tending to a basis for an improved evaluation concept for the leachability of environmental relevant substances.

2. Materials and methods

2.1. Raw materials and mix design

For the investigations two different ordinary Portland cements and one fly ash were used. A CEM I 32.5 R and a CEM I 52.5 N-SR3/NA for investigations with ground water containing high amounts of sulphates were selected. The chemical composition was measured with an ignited sample via X-Ray fluorescence analysis. The heavy metal and trace elements contents were determined with aqua regia digestion and subsequent use of inductively coupled plasma mass spectrometry (ICP-MS). The SO_3 content was determined using a carbon/sulphur analyser (CS 2000, Elektra). Chloride contents were analysed via potentiometric titration with a silver nitrate solution. The results are stated in Tables 1 and 2.

Four different concrete mixtures were used. The mixtures differ in binder composition and water/binderratio (w/b). For concrete A an ordinary Portland cement (CEM I) was used, concrete B contains fly ash and has a slightly lower w/b ratio. Concretes C and D were designed according to DIN 1045-2 and EN 206-1 as sulphate resistant concretes for the use in a ground water with a sulphate content of 1500 mg/l (exposure class XA2). The mix design of the concretes is shown in Table 3.

The concrete for the leaching experiments was prepared under standard conditions (20 °C/65% relative humidity) in a bin mixer in a batch size of 8 l of fresh concrete. The mixing time was 3 min. The fresh concrete was poured into hard plastic cylinder-moulds with a diameter of 100 mm and a height of 150 mm. The filled moulds were covered with a polyethylene foil, stored at a relative humidity of 95% and a temperature of 20 °C and demoulded after 24 h. Afterwards they were immediately wrapped airtight in three layers of polyethylene foil to protect them from carbonation and loss of moisture. The sealed samples were stored in a climate chamber (20 °C/65% RH) for 56 days and afterwards used for the leaching tests. Compressive strength of the mixtures was tested on three concrete cubes with an edge length of 150 mm per mixture. Dynamic modulus of elasticity was tested on three cylinders with a diameter of 150 mm and a height of 300 mm. After casting the moulds they were stored at 20 °C and 95% relative humidity for one day. Subsequently the samples were demoulded and stored under water for 6 days, afterwards at standard conditions to an age of 28 days. The mechanical properties are shown in Table 3.

Additionally, the pore solution was analysed after 56 days. For this purpose 1000 ml of cement paste with the binder composition and w/b ratio stated in Table 3 were produced and subsequently filled into 250 ml polyethylene bottles to the top to prevent air voids. The bottles were closed tightly to prevent carbonation of the samples. The samples were rotated with 6 rpm (revolutions per minute) for 24 h to prevent sedimentation. After 56 days of storage in the bottle at 20 °C and 65% RH the hardened cement paste samples were demoulded by cutting the PE bottles. The sample was inserted into a high pressure device to extract pore solution with a pressure of up to 520 MPa with an acceleration of 0.5 MPa/s. About 2 ml of the pore solution was used to analyse pH value, redox potential and electrical conductivity immediately after extraction. The rest was weighted and diluted up to 100 ml. Half of the sample was acidified with 5 vol.-% supra pure nitric acid for heavy metal and trace element analysis. The other half was used to determine sodium, potassium, calcium, chloride and sulphate concentrations. Na, K and Ca were analysed by flame photometry (ELEX 6361, Eppendorf) and Cl^- and SO_4^{2-} by ion chromatography (ICS

Table 1
Chemical composition and loss on ignition (LOI).

Parameter	CEM I 32,5 R	CEM I 52.5 N-SR3/NA	Fly ash
	wt%		
LOI	2.01	2.13	1.30
Insoluble residue in HCl	0.62	0.32	85.85
Al_2O_3	5.15	3.39	25.76
SiO_2	19.61	21.00	56.74
CaO	66.67	66.10	3.95
Fe_2O_3	2.51	5.25	5.53
MgO	1.28	1.09	1.21
P_2O_5	0.09	0.05	1.05
TiO_2	0.27	0.18	1.41
MnO	0.07	0.12	0.04
SO_3	3.19	2.77	0.38
Chloride	0.079	0.03	0.017
Na_2O	0.10	0.01	0.67
K_2O	0.98	0.48	2.01

Table 2
Contents of heavy metals and trace elements.

Parameter	CEM I 32,5 R	CEM I 52,5 N-SR3/NA	Fly ash
	mg/kg		
Antimony	5.21	3.28	3.32
Arsenic	3.85	6.83	29.6
Barium	233	123	599
Lead	4.63	7.36	21.4
Boron	64.2	51.1	133
Cadmium	0.137	0.274	0.465
Chromium	50.2	213	42.2
Cobalt	6.14	6.29	15.7
Copper	88.2	112	55.3
Molybdenum	0.95	7.53	13.8
Nickel	23.1	35.7	40.9
Mercury	0.004	0.006	0.063
Selenium	1.24	0.575	8.06
Thallium	0.01	0.01	0.65
Vanadium	62.5	26.6	100
Zinc	267	130	70.8

Table 3
Mix design and properties of the investigated concretes.

Component/ parameter	Unit	Concrete A	Concrete B	Concrete C	Concrete D
CEM I 32.5 R	kg/m^3	300	240	–	240
CEM I 52.5 N-SR3/ NA		–	–	320	–
Fly ash		–	120	–	120
Water (tap water)		180	194	144	146
Aggregate		1850	1719	1927	1847
Water/binder ratio	–	0.6	0.54	0.45	0.41
Compressive strength	MPa	41.9	29.1	58.8	47.5
Dynamic modulus of elasticity		44,000	37,700	54,000	46,700