



Leaching behaviour of major and trace elements from concrete: Effect of fly ash and GGBS



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ABSTRACT

The leaching of major and trace elements from concrete made with Portland cement, fly ash and GGBS (ground granulated blast-furnace slag) was studied using pH static availability and tank leach tests. The release of substances during the tank leach test occurs by surface dissolution of phases at the concrete surface and diffusion inside the concrete, the amounts depending on the phases controlling solubility and concrete porosity. Alkali release is controlled by diffusion and is thus reduced by lower water/binder ratios and the replacement of Portland cement by fly ash. Ca, Al and S release occurs mainly by surface dissolution of portlandite and AFt/AFm, respectively. The release of V is determined by surface dissolution of V substituted ettringite and/or calcium vanadate. Although fly ash can increase the total V content of concrete, enhancing release, only 2% of the total V content in concrete was available for release.

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1. Introduction

The current trend towards the use of secondary fuels in cement production and more secondary materials in concrete production has focussed attention on the leaching of environmentally relevant substances, in particular toxic trace metals, from concrete construction products in contact with water [1]. In Germany, assessment criteria for the release of environmentally relevant substances during service are currently under consideration for the authorization of building materials [2]. A tank leach test [3] lasting 56 days is specified for the simulation of leaching during service conditions. The test is required if limiting amounts of environmentally relevant substances are exceeded by the concrete components and their eluate in the standard German DEV S4 test [4]. However, incorrect assessment of the leaching of environmentally relevant substances by the tank leach test could lead to the exclusion of otherwise useful materials. Thus the emission of toxic trace metals from concrete in contact with water is a matter of economical as well as ecological interest.

In principle, trace metals are directly leached into water adjacent to concrete surfaces by dissolution of surface phases and/or by dissolution and diffusion in the pore solution, the latter being

affected by concrete porosity and pore structure. The solubility of trace metals depends on their bonding in the hydration products and other concrete constituents as well as their ability to form aqueous complexes and precipitate solubility-controlling phases. Thus, as observed by van der Sloot [5] for Cr, Mo, Pb and Zn in mortar, no general correlation exists between the total amount of trace metals and their availability as defined by the Dutch availability test NEN 7341 [6], i.e. the dissolvable amount per kilogramme dry powdered material. Consequently, an evaluation of environmental risks cannot be based on the total amounts of trace metals contained in concrete alone.

Trace metal dissolution and transport both depend on binder and concrete composition. The replacement of cement by fly ash, for example, can result in larger amounts of trace metals such as V, Cr, and Pb, in concrete. Especially when firing petroleum coke, the amount of V in fly ash can increase dramatically. On the other hand, fly ash imparts concrete a denser long-term microstructure which should hinder the release of trace metals by diffusion. Observation of the release of trace metals at the concrete surface in a tank leach test, e.g. over 56 days with 100 mm concrete cubes according to [3], yields information on the combined effect of the controlling mechanisms.

In this study the effect of fly ash, GGBS and water/binder ratio w/b on leaching is investigated using tank leach and the availability tests. The leaching behaviour of major (Al, Ca, Na, K, S and Si) and trace elements (Ba, Co, Cr, Cu, Ni, Pb, Sr, V and Zn) for concrete made with Portland cement, coal fly ash and ground granulated blast-furnace slag GGBS is considered.

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Table 1
Concretes used in the experiments.

Concrete	Cement <i>c</i>		Fly ash <i>f</i>		GGBS <i>g</i>		Sp ^a (kg/m ³)	Aggregate ^b			Water <i>w</i> (kg/m ³)
	kg/m ³	%	kg/m ³	%	kg/m ³	%		kg/m ³	<i>w/b</i> _{eq} ^c	<i>w/b</i> ^d	
B1	450	0	0	0	0	0	0	1551	0.60	0.60	270
B2	427.5	5	22.5	0	0	0	0	1566		0.58	262
B3	405	10	45	0	0	0	0	1581		0.56	254
B4	360	20	90	0	0	0	0	1611		0.53	238
B5	270	40	180	0	0	0	0	1730		0.41	183
B6	135	0	0	70	315	0	0	1511		0.60	270
B7	450	0	0	0	0	0	0	1800	0.40	0.40	180
B8	427.5	5	22.5	0	0	0.3	1.4	1805		0.39	175
B9	405	10	45	0	0	0.5	2.3	1811		0.38	169
B10	360	20	90	0	0	0.9	4.1	1822		0.35	158
B11	135	0	0	70	315	0	0	1774		0.40	180

^a Superplasticizer.

^b 68% 0/4 mm sand, 32% 4/8 mm gravel.

^c Equivalent *w/b* ratio for mixes with fly ash, $w/b_{eq} = c + k f$, $k = 0.4$, $f/c \leq 0.33$ according to DIN 1045-2 [3].

^d $w/(c + f)$.

2. Materials and methods

Table 1 shows the composition of the mixes used in the experiments. The concretes were prepared using Portland cement (CEM I 42.5 R), fly ash and GGBS at a total binder content of 450 kg/m³ and equivalent water binder ratios *w/b*_{eq} of 0.4 and 0.6. The equivalent ratio is used in German regulations for mixes with fly ash [7] where a factor *k* accounts for the amount of fly ash considered as binder, the rest being considered as filler, for more information

Table 2
Chemical composition of the initial materials.

Substance	Unit	Cement	Fly ash	GGBS	Sand	Gravel
SiO ₂	wt.%	13.95	50.40	36.17	26.66	14.34
Al ₂ O ₃		5.35	27.31	13.36	1.76	1.31
Fe ₂ O ₃		4.88	4.79	1.47	1.00	0.74
TiO ₂		–	1.50	0.66	0.09	0.07
CaO		61.44	7.29	38.05	30.85	36.24
MgO		1.20	1.49	7.12	6.89	8.59
MnO		0.55	0.06	0.70	0.04	0.03
SO ₃		2.95	0.46	0.07	0.01	0.01
Sulfide		–	–	0.98	–	–
K ₂ O		0.78	1.52	0.65	0.24	0.10
Na ₂ O		0.22	0.28	0.30	0.22	0.14
Na ₂ O _{eq}		0.73	1.28	0.73	0.38	0.20
LOI		1.21	2.48	0.00	32.11	38.31
P ₂ O ₅		0.17	1.06	0.07	0.00	0.00
As	mg/kg	10.0	48.7	69.2	4.1	4.8
Se		0.5	1.5	44.5	0.6	1.2
Sb		0.2	8.8	29.7	0.2	0.2
Cd		1.8	0.5	3.8	0.0	0.0
Cr		97.7	343.6	282.0	31.0	44.0
Cu		120.3	106.0	39.0	2.2	3.8
Co		8.4	55.0	3.39	0.4	1.2
Mo		2.3	20.0	8.4	0.2	0.5
Ni		29.0	128.0	25.0	2.2	4.1
Pb		40.0	73.0	0.0	1.6	3.3
V		41.0	336.0	69.0	5.0	6.0
Zn		401.0	219.0	35.0	6.0	7.0
Ba		217.0	1645.0	1053.0	17.0	23.0
Sr		218.0	1723.0	514.0	127.0	129.0

Table 3
Transport relevant properties of the concretes, age 90 days.

Concrete	B1	B2	B3	B4	B5	B6	B7	B8	B9	B10	B11
<i>D</i> _{RCM} (10 ⁻¹² m ² /s)	13.37	14.15	9.07	3.62	2.48	0.59	4.18	4.21	1.39	–	1.45
Porosity (%)	17.59	16.19	15.88	12.59	14.91	17.01	10.40	9.19	9.44	8.77	11.89

see [8]. The aggregate comprised Munich 0/4 mm sand and 4/8 mm gravel graded at A8/B8 [9]. Concretes B8 to B11 were mixed with the help of a polycarboxylate superplasticizer. All the initial materials were characterized chemically by ICP-OES after digesting in nitric acid, Table 2.

Concrete cubes (100 mm) were cast from moulds lined with PTFE sheeting thus avoiding contamination of the concrete by a release agent. The cubes were demoulded after 24 h in a climatic cabinet at 20 °C/98% RH, tightly wrapped in polyethylene foil and stored until testing at 20 °C/98% RH in an argon atmosphere to minimize carbonation. The tank leach test according to [3] commenced at an age of 90 days to reduce the effect of ongoing hydration during the test as observed in earlier investigations [10]. Two concrete cubes were used for each composition; average values are presented in the results. Each cube was immersed in distilled water (90 L/m²) in a polyethylene container, the space above the water flooded with argon and the container sealed. After consecutive elution steps each lasting 1, 2, 4, 9, 16 and 24 days, i.e. total time 56 days, eluate specimens were taken for chemical analysis by ICP-OES and pH measurement. The eluent was renewed after each step, the containers flooded with argon and resealed.

Parallel specimens were prepared in 0.5 L polyethylene bottles. On reaching an age of 90 days, the specimens were demoulded and pore solutions expressed under compressive load. The solutions were analysed chemically by ICP-OES and the pH determined by titration. Other specimens were used for the determination of availability following NEN 7341. Here, pieces of concrete were

Table 4
Pore solution composition [mmol/L], age 90 days. Omission of data for insufficient solution.

Concrete	B1	B2	B3	B4	B5	B6
pH	13.50	13.45	13.44	13.30	13.38	13.07
Al	0.039	0.072	0.064	0.228	0.100	0.315
Ca	–	1.227	1.250	0.881	1.132	0.124
K	233.5	221.5	200.2	142.4	171.4	55.50
Na	99.30	92.50	86.00	70.50	78.50	51.10
S	0.451	0.440	0.408	0.243	0.394	0.151

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