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Waste Stabilization/Solidification (S/S) of EAF dust using fly ash-based geopolymers. Influence of carbonation on the stabilized solids

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A B S T R A C T

The effect of the curing in a carbonated ambient of the solids obtained after the stabilization/solidification (S/S) of metallurgical waste using geopolymerization technology is described in this paper. The electric arc furnace (EAF) dust to stabilize contains hazardous metals such as Pb, Cd, Cr, or Zn. Different geopolymeric agents such as potassium hydroxide, potassium silicate, metakaolin, and blast furnace slag have been used. Mixtures of EAF waste with these geopolymeric materials and class F fly ash as main silica and alumina source have been processed. Samples were submitted to an accelerated carbonation test. Compressive strength tests and different leaching tests for determining the efficiency of heavy metal immobilization have been carried out. Comparison of fly ash-based geopolymer systems with classical Portland cement (OPC) stabilization methods has also been accomplished. Compressive strength values far better than those achieved by hydraulic S/S methods were easily obtained by geopolymer solids at 28 days. Carbonation produced a great increment of compressive strength in samples containing a higher K_2O proportion. The geopolymer S/S solids also manifested better leachability behavior in general, showing very promising results. Carbonation effects on pH and on Zn and Pb leachabilities are more marked for OPC than for geopolymer S/S solids.

1. Introduction

EAF dust is generally classified as hazardous waste by many regulations because of its heavy metal content. Stabilization/solidification (S/S) techniques have been used worldwide to treat this and other similar residues. Although EAF dust is described as a waste product that is difficult to stabilize, the use of different S/S systems containing Type F coal fly ash, lime, dolomitic lime, Type I Portland cement, lime, blast furnace slag, silica fume, sodium silicate, low-grade MgO, or combinations of them may be found in the literature (Olmo and Irabien, 2002; Fuessle and Taylor, 2004; Fernández Pereira et al., 2001; Fernández et al., 2003; Caldwell et al., 1999).

Geopolymer technology has recently received attention in waste stabilization/solidification (S/S) systems of heavy metal-containing wastes. In the immobilization of toxic metals, geopolymer systems function similarly to cement binders in terms of encapsulation. They have, however, improved chemical and physical properties, such as structural integrity, low permeability, high compressive strength, and durability (Davidovits and Comrie, 1988; Van Jaarsveld et al., 1997; Davidovits, 1994). Some authors have studied the potential use of geopolymer systems as S/S agents of EAF dust, a waste that presents a challenge for both recovery, as many elements are difficult to separate, or disposal, because many of the elements are hazardous (Stegemann et al., 2000; Fernández Pereira et al., 2009). Cured conditions have been studied in order to establish better conditions in a landfill (Fernández Pereira et al., 2009). In general, the best results for 28-days-cured S/S solids were obtained when samples were cured at room temperature. But, it is

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Table 1

Main elements of Fly ash (FA), Kaolin, Blast Furnace Slag (BFS), Portland cement (OPC) and Electric Arc Furnace (EAF) dust.

	Moisture 105 °C	LOI 750 °C	Main Elements (wt %)						
			Fe_2O_3	CaO	MgO	SiO_2	Al_2O_3	Na_2O	K_2O
FA	0.05	3.32	5.86	3.94	1.84	63.91	21.51	0.68	1.67
Kaolin	1.16	12.17	0.51	<0.03	0.17	65.23	31.97	0.05	1.28
BFS	0.24	0.31	0.41	43.49	7.60	41.08	10.70	0.26	0.32
OPC	0.15	–	2.15	61.7	2.76	21.2	5.00	0.13	0.16
EAF	0.42	12.77	36.32	8.60	3.05	6.21	2.00	1.13	0.48

necessary to study the evolution with time of properties, such as compressive strength or leachability, when the S/S solids are landfilled. In the long term, many processes that could affect the S/S solid properties, such as the carbonation, are produced.

Much research relating to carbonation has been performed on cementitious forms. Carbonation occurs when Portlandite and other calcium bearing phases in the waste form react with CO_2 to form calcite ($CaCO_3$). It has been reported that carbonation may directly alter the chemical and physical properties of solidified waste forms (Cocke and Mollah, 1993), but contradictory results regarding metal leachability have been published. According to Lange et al. (1996, 1997), the effects of carbonation appear to be beneficial on the leachate production of cement-solidified hazardous wastes, showing a significant reduction in leachable metals extracted. However, other studies show that carbonation produces the opposite effect. In an evaluation of the role of carbon dioxide on the long-term performance of cementitious waste forms, carbonation increased the release of Cd, Pb, and Co (Bin Shafique et al., 1998). For the S/S of municipal solid waste incineration residues, the pastes were exposed to an accelerated carbonation procedure, increasing the leachability of heavy metals such as Zn and Cr (Alba et al., 2001).

In this paper and to the best of our knowledge for the first time, a carbonation accelerated process has been carried out in fly ash based geopolymer S/S solids, with the aim of studying its influence in mechanical and leaching properties.

2. Experimental

2.1. Materials

Low-calcium fly ash (ASTM class F) from the combustion of high quality pulverized coal in one of the largest coal power plants in the South of Spain, Los Barrios (550 MW), was used as the main silicoaluminate agent. Blast furnace slag (BFS) as a lime and silicon source and kaolinite as a secondary silicoaluminate source were also used. Ordinary Portland cement (OPC, type II) used in order to compare geopolymer stabilization/solidification (S/S) process with a conventional S/S process using OPC lime and fly ash. The main elements of the four S/S agents are detailed in Table 1.

The potassium silicate solution used to prepare the geopolymer samples was named KSil 0,65 (K_2O/SiO_2 weight ratio = 0.649), and was supplied by Industrias Químicas del Ebro (Zaragoza, Spain). The product specifications are listed in Table 2.

Table 2

Specifications of potassium silicate solutions.

	SiO_2, wt %	K_2O, wt %	Density 25°C, g·cm^{-3}
KSil 0,65	23	14.9	1.38

The main elements content in EAF dust is shown in Table 1 and the heavy metals content in the waste is shown in Table 3. Metakaolin (MK) was prepared after Kaolin treatment at 750°C, according to Madani et al. (1990).

2.2. Preparation of S/S solids

Three kinds of geopolymer samples were prepared maintaining the fly ash and EAF dust proportions constant and adding different amounts of potassium silicate, KOH, BFS, and MK with the same methodology used in previous studies (Fernández Pereira et al., 2009). The three compositions were chosen for their good physical and chemical properties obtained in previous investigations (Luna Galiano et al., 2009): KSil 0.8 BFS 0.3, KSil 0.8 KOH BFS 0.3 and KSil 0.8 MK 0.3. Distilled water was used throughout. The water content was adjusted to achieve the same workability in all the mixtures. With the mass obtained, cylindrical plastic moulds (30-mm diameter and 41-mm height) were filled and compacted. Finally, samples were vibrated for 5 minutes in order to release bubbles. On the other hand, hydraulic binders such as Portland cement (OPC), fly ash, and lime have also been used in order to compare geopolymer S/S solids with conventional S/S solids. Compositions of geopolymeric matrices are shown in Table 4. Compositions of hydraulic binder matrices are shown in Table 5.

2.3. Carbonation test

Seven days after the preparation, the samples were de-molded and placed in a plastic tray, and then were introduced into a thermally sealed impermeable plastic bag. CO_2 was introduced into the bag through a valve connected to a CO_2 bottle. A vacuum pump was used to release air from the bag while CO_2 entered the bag using another valve. A moist ambient is necessary to promote the carbonation reaction, so trays were filled with 40 mL of water to maintain a constant ambient moisture content during the process. The CO_2 content was kept constant during the process at a 99% by volume approximately. A general view is shown in Figure 1.

The effect of the carbonation exposing time was studied by removing samples from two different bags at 28 or 84 days. Three monolith samples of each composition were used for compressive strength test and the fragments obtained after breaking the samples were used for leaching tests.

Table 3

Heavy metal content in Electric Arc Furnace (EAF) dust.

	Metal content g·kg^{-1}			
	Zn	Cd	Cr	Pb
EAF	342.9	0.29	2.108	2.29

Table 4
Composition of S/S matrices using geopolymers.

	Proportion by weight							
	EAFD	FA	KSil	KOH	MK	BFS	MS	Water
KSil (0,65) 0.8 BFS 0.3	0.5	1.4	0.8			0.3		0.31
KSil (0,65) 0.8 KOH BFS 0.3	0.5	1.4	0.8	0.42		0.3		
KSil (0,65) 0.8 MK 0.3	0.5	1.4	0.8		0.22			0.30

2.4. Compressive strength

Compressive strength test was performed as per ASTM D-1633-84 using two cylinders of each sample and averaging the experimental values obtained. All samples were tested after 28 days. A Suzpecar MEM-102 / 50 t compressive strength testing apparatus was used.

2.5. Leaching tests

Apart from the compressive strength test, the stabilization/solidification efficacy was assessed by using four leaching tests: UNE-EN 12457 (2003), USEPA TCLP (1986), NEN 7341 (1994) and NEN 7345 test (1995). Both the UNE-EN 12457 and TCLP tests are batch leaching tests using water and an acetic acid solution respectively. The availability test described in NEN 7341 is aimed to indicate the maximum amount of contaminant which can be leached from the material under extreme conditions, in other words, the fraction of the total contaminant potentially available for leaching. The test is performed on finely ground material ($< 125\ \mu m$) at a high dilution ($L/S = 100$) by controlling the pH at 7 for three hours and subsequently at pH 4 for another three hours using $1M\ HNO_3$. The NEN 7345 test (tank leaching test) is a monolith-type leaching test that can be used to assess the leaching potential of stabilized/solidified wastes over the long term. The leaching agent is distilled water acidified at pH 4 with nitric acid and the extraction fluid is renewed in eight extractions at 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days from the test start. The GANC (Generalized Acid Neutralization Capacity) test (Isenburg and Moore, 1992) is, as the ANC, a single-batch procedure that uses from zero to six equivalents of acid (in this case acetic acid, the same acid used in the TCLP test) per kilogram of solids. The GANC test was adapted as described by Vale Parapar et al. (1998). From the data recorded, we obtain the acetic acid consumption for pH values ranging from approximately 12.5 to 5.5 and the corresponding GANC curves.

After the tests, samples were filtered through Whatman membrane filters (pore size 0.45 μm). When the procedures were completed, the metal contents in the solution were determined using Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES).

The aim of S/S technology is to obtain solids that reduce the overall environmental impact of waste disposal. To achieve this goal, the USEPA criteria, the EU Landfill regulations (European Landfill Directive, EULFD) (1999), and the Dutch Building Materials Decree (DBMD) (1995) were followed to establish quality criteria on

Table 5
Composition of S/S matrices using classical hydraulic binders.

	Proportion by weight			
	EAFD	FA	CaO	OPC
OPC CaO	1	1.4	0.2	0.2

metal concentrations limits in leachate from S/S solids after the usual curing time (28 days). The quality criteria applied to the metal leachate concentrations are shown in Table 6.

3. Results and discussion

3.1. Compressive strength (CS)

Figure 2 shows the influence of carbonation on the CS measurements carried out in the samples studied.

As can be seen in Figure 2, the variations of the compressive strength of carbonated samples cannot be explained easily, since samples with KSil+(BFS)+KOH and OPC samples showed the highest values in carbonated conditions, while KSil+metakaolin+KOH samples, showed the highest resistances at high temperatures and carbonation slightly worsens the CS. Samples with KSil and BFS showed a different behavior, since carbonation time improved the CS. Therefore, it is difficult to find a tendency or regularity to explain this behavior.

The high CS of KSil (0,65) 0.8 KOH BFS 0.3 at 84 days of carbonation can be emphasized. This fact could be due to some crystal formation since all samples appear covered with a white layer similar to that formed by the oversaturation of an inorganic salt solution (NaCl, K_2CO_3, etc).

Besides, all the samples kept in the carbonation chamber (bag) showed a water layer on their surface. This phenomenon was probably produced in the course of the CO_2 filling process, when the expanding process from CO_2 (l) to CO_2 (g), led to a temperature reduction (Joule-Thomson effect) that caused the water condensation on the samples as the atmosphere in the bag was moisture saturated. So, carbonation samples had higher water content than samples cured at ambient or at 60°C. To conclude, it is possible that the high quantity of moisture in carbonated samples has counteracted the CS increment potentially produced by the carbonation process. To avoid this phenomenon, a CO_2 expanding chamber to heat the gas to be used in the carbonation chamber could be an option.

3.2. Leaching tests

3.2.1. UNE-EN 12457-4 leaching test

Table 7 shows the pH and Zn, Pb, Cr and Cd concentrations of the EN 12457-4 leachates.

Both for OPC and lime and for geopolymer systems, the pH values in EN 12457-4 leachates at 28 days were between 11 and 13. Carbonation lowers the leachate pH considerably to 8.4–9.6, values improving the leaching behavior of all samples, especially the OPC samples.

In general, the lowest leached amounts for the metals studied were obtained for EAF stabilized with geopolymeric activators. Large amounts of Pb (and Cr in some instances) were leached from samples prepared with OPC and lime, probably because of a high leachate pH (12.2–12.3). Apart from this phenomenon, there must be other reasons because the pH differences with KSil (0,65) 0.8

Fig. 1. General view of carbonation test.

BFS 0.3 are not so high. Therefore, the low Pb leachabilities found in geopolymer samples at the same (high) pHs evidence a higher degree of Pb immobilization in geopolymer matrices and must be emphasized.

The lowest leached amounts were found for samples prepared with KSil 0,65 and slag at room temperature. In general, lower S/S solid leachabilities are observed at room temperature instead of 60°C, except for Cr which almost always has lower concentrations in the 60°C-cured samples.

Regarding the carbonation time, pH slightly increased with time, except for KSil (0,65) 0.8 KOH BFS 0.3 which showed the lower pH at 84 days. In this case, Pb concentration increased with the carbonation time from 15.2 at 28 days to 48 mg/kg after 84 days of carbonation, probably as a consequence of the lower pH. Zn leachability increases slightly with the carbonation time in all the samples.

3.2.2. TCLP leaching test

Table 8 shows the pH and the metals concentrations in the TCLP leachates.

After 28 days of curing at room temperature and at 60°C, the leachates of all the samples prepared with OPC and lime were alkaline, with pH values around 11. However, samples prepared with geopolymeric activators showed a neutral or slightly acidic pH. With respect to the carbonation process, a clear neutralization effect can be observed, especially for OPC samples that showed a pH drop from 11 to 6.6. However, in KSil (0,65) 0.8 MK 0.3 samples, a pH increment is produced due to the low pH of samples

at ambient and 60°C (2.94 and 3.07, respectively), possibly caused by some contamination. For the rest of samples, no significant differences exist in pH with the curing conditions.

In OPC samples, carbonation reduces pH and consequently Zn, Pb, and Cd leachabilities decrease. For samples prepared with geopolymer agents, in general carbonation improves Zn, Pb, and Cd leachabilities. Carbonation time also produces a decrease in metal concentrations. The best values are obtained for KSil (0,65) 0.8 KOH BFS 0.3 samples independently of the curing condition.

3.2.3. NEN 7341 leaching test

The calculated leached fractions, that is, the percentage ratio between the measured leached metal concentration and the total concentration of the same metal in EAF dust for samples cured at room temperature, at 60°C and in a carbonated atmosphere are depicted in Table 9.

Low extraction percentages were found for Zn and Cr, so these two metals were easily immobilized in almost all mixtures. Practically, almost 100% of Cr is immobilized using both geopolymeric and hydraulic binders. Conversely, Pb and Cd showed the highest availabilities in all samples, always being better immobilized using geopolymeric agents (KSil (0,65) 0.8 KOH BFS 0.3). Samples stabilized by hydraulic binders cured at ambient temperature and at 60°C produced a Pb leached fraction of nearly 100%. Carbonation in OPC samples reduced the lead leachability considerably (leached fractions from 100 to 37.5%) as well as KSil (0,65) 0.8 BFS 0.3 samples (from 18.8 to 1%). Carbonation led to a pH reduction, and therefore, in general, metal retention is increased.

3.2.4. NEN 7345 leaching test

The leachate pH remained constant throughout the test time in all the (sub)-samples. In all the cases carbonation reduced the leachate pH. In OPC samples the carbonation produced pH drops of around 4 units while in geopolymeric samples the pH dropped 1.5–2 units.

Conversely, the leachate conductivity dropped with time in all the (sub)-samples. Carbonation reduced leachate conductivity for KSil+ KOH+BFS and OPC samples, but in the other cases there were no relevant differences between carbonated and non carbonated samples.

Table 6

Quality criteria.

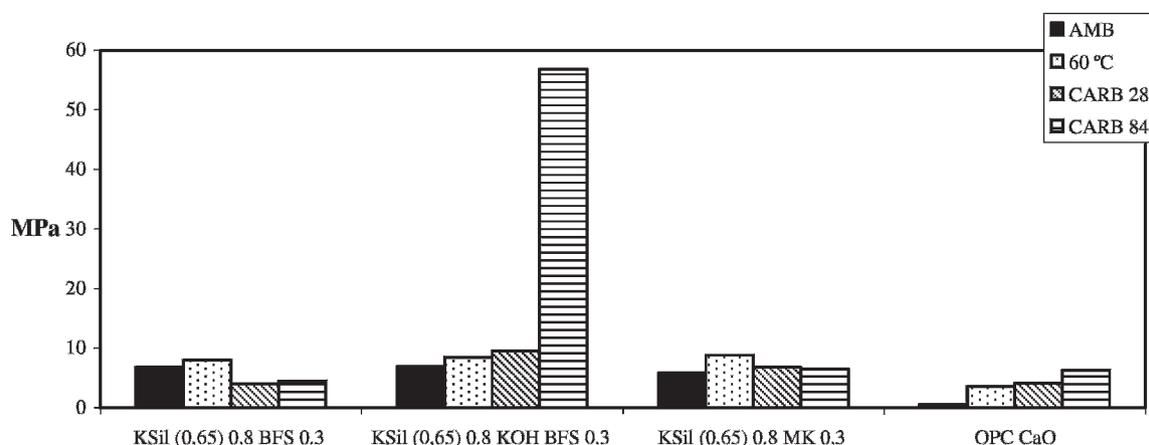| Parameter	Range/Value			
	2–12.5			
pH	Cd	Cr total	Pb	Zn
USEPA				
Metal concentrations in TCLP leachate (mg·L^{-1})	0.5	5	5	300
Inert Waste (mg·kg^{-1}) EULFD	0.04	0.5	0.5	4
Non Hazardous Waste (mg·kg^{-1}) EULFD	1	10	10	50
Hazardous Waste (mg·kg^{-1}) EULFD	5	70	50	200
U1 (mg·m^{-2}) DBMD	1	150	100	200

Fig. 2. Compressive strength at 28 days.

Table 10 shows the cumulative concentrations at 64 days of tank leaching test leachates.

When non carbonated geopolymer and OPC S/S solids are compared, in general the leaching benefit of geopolymeric techniques is clear, especially for Pb and Cr, while Zn concentrations are lower using both conventional and geopolymer methods. Cd concentrations are under the detection limits in all samples, so it is impossible to draw any conclusion. Carbonation reduces pH. This reduction makes the Zn and Pb retention of KSil BFS samples and KSil MK samples worsen. Besides, Zn is increased in OPC samples. However, the pH drop produced a reduction in Pb concentration in OPC samples, improving its leachability. Regarding Cr, carbonation caused a clear reduction of the metal concentration for geopolymeric samples, but the effect in OPC samples is the opposite.

All non carbonated samples showed leachate concentrations for Zn, Cr and Pb under the DBMD U1 limits, except OPC samples in the case of Pb. In relation to Cd concentrations, the high Cd detection limit, does not strictly allow drawing any conclusions on the accomplishment of the U1 limit.

Table 7
pH and metal concentrations in UNE-EN 12457-4 leachates at 28 days and EULFD limits.

		Metal concentration (mg·kg^{-1})			
	pH	Zn	Pb	Cr	Cd
KSil (0,65) 0.8 BFS 0.3 AMB	11.43	0.9	<2	3.6	≤0.2
KSil (0,65) 0.8 BFS 0.3 60°C	11.23	5.7	8	6.5	≤0.2
KSil (0,65) 0.8 BFS 0.3 CARB 28	9.57	0.8	5.4	≤0.5	≤0.2
KSil (0,65) 0.8 BFS 0.3 CARB 84	9.85	1.3	3.7	1	≤0.2
KSil (0,65) 0.8 KOH BFS 0.3 AMB	12.68	17.5	17	8	≤0.2
KSil (0,65) 0.8 KOH BFS 0.3 60°C	12.8	41.2	16.1	6.4	≤0.2
KSil (0,65) 0.8 KOH BFS 0.3 CARB 28	10.73	0.7	15.2	1.6	0.2
KSil (0,65) 0.8 KOH BFS 0.3 CARB 84	10.37	4.1	48	2.9	≤0.2
KSil (0,65) 0.8 MK 0.3 AMB	10.9	1.4	2.1	15.2	≤0.2
KSil (0,65) 0.8 MK 0.3 60°C	11	5.5	8	8.3	0.2
KSil (0,65) 0.8 MK 0.3 CARB 28	9.41	1.2	8.6	2.1	≤0.2
KSil (0,65) 0.8 MK 0.3 CARB 84	9.53	3.5	7.6	1	≤0.2
OPC CaO AMB	12.21	12.6	3760	≤0.5	≤0.2
OPC CaO 60°C	12.17	14.7	950	2	≤0.2
OPC CaO CARB 28	8.36	2.3	2.2	2.2	≤0.2
OPC CaO CARB 84	8.55	5.7	<2	1.2	≤0.2
Inert waste		4	0.5	0.5	0.04
Non hazardous waste		50	10	10	1
Hazardous waste		200	50	70	5

3.2.5. GANC Leaching Test

Figure 3 shows the GANC curves for samples prepared using potassium silicate and blast furnace slag cured in carbonated and non-carbonated ambient.

As can be seen, there are not significant differences between both curves, although a slight difference between carbonated and non-carbonated ambient is noticed in the range of 0 to 1 eq/kg. This is possibly due to the pH fall led by the carbonation test, as it was observed in the UNE-EN leaching test.

Figure 4 shows the Zn, Pb, Cr, and Cd leachabilities for the samples studied in carbonated and non-carbonated ambient.

As can be seen, carbonation does not affect the possible Zn, Pb, Cr, and Cd chemical speciations in the studied S/S solids. Zn and Pb seem to exhibit the characteristic hydroxide amphoteric behavior, with a pH of a minimum of solubility. Cd solubility has a different behavior, with a step-shaped curve. Cd solubility reduces at a minimum in a pH range between 8 and 12 (possibly $Cd(OH)_3$ controlled solubility). Cd concentration increase rapidly when pH reduce in acidic medium, to reach a constant value at pH less than 6. Cr leachability does not show a relationship with pH as clear as other metals do; possibly due to the existence of both Cr(VI) and Cr(III) in the EAF dust and in the S/S solids in different proportion,

Table 8
pH and metal concentrations in TCLP leachates at 28 days and USEPA limits.

		Metal concentration (mg·L^{-1})			
	pH	Zn	Pb	Cr	Cd
KSil (0,65) 0.8 BFS 0.3 AMB	6.5	362	4.15	<0.05	1.22
KSil (0,65) 0.8 BFS 0.3 60°C	6.27	553	14.6	<0.05	1.63
KSil (0,65) 0.8 BFS 0.3 CARB 28	6.36	417	2.99	<0.05	1.41
KSil (0,65) 0.8 BFS 0.3 CARB 84	6.42	426	0.15	≤0.05	1.21
KSil (0,65) 0.8 KOH BFS 0.3 AMB	8.49	137	<0.2	<0.05	0.03
KSil (0,65) 0.8 KOH BFS 0.3 60°C	5.84	105	0.99	<0.05	0.22
KSil (0,65) 0.8 KOH BFS 0.3 CARB 28	7.25	135	0.29	<0.05	0.56
KSil (0,65) 0.8 KOH BFS 0.3 CARB 84	7.11	18	0.14	0.07	0.54
KSil (0,65) 0.8 MK 0.3 AMB	2.94	1544	3.38	2.24	4.51
KSil (0,65) 0.8 MK 0.3 60°C	3.07	1640	3.69	2	4.58
KSil (0,65) 0.8 MK 0.3 CARB 28	6.79	804	7.61	<0.05	2.44
KSil (0,65) 0.8 MK 0.3 CARB 84	6.3	782	4.21	≤0.05	2.04
OPC CaO AMB	10.97	0.05	0.9	0.31	<0.2
OPC CaO 60°C	11.24	0.09	0.3	0.48	<0.2
OPC CaO CARB 28	6.5	415	1.47	0.06	1.6
OPC CaO CARB 84	6.72	364	0.43	≤0.05	1.74
USEPA		300	5	5	0.5

Table 9
Leached fractions in availability test leachates (NEN 7341).

	Leached fraction (%)			
	Zn	Pb	Cr	Cd
KSil (0,65) 0.8 BFS 0.3 AMB	7.89	10.2	0.25	24.4
KSil (0,65) 0.8 BFS 0.3 60°C	8.56	18.78	0.25	25
KSil (0,65) 0.8 BFS 0.3 CARB 28	6.85	1.03	0.28	23.1
KSil (0,65) 0.8 KOH BFS 0.3 AMB	0.47	0.98	0.59	5.8
KSil (0,65) 0.8 KOH BFS 0.3 60°C	1.3	0.94	0.26	4.8
KSil (0,65) 0.8 KOH BFS 0.3 CARB 28	0.16	1	0.27	5.1
KSil (0,65) 0.8 MK 0.3 AMB	7.11	0.91	0.74	28.4
KSil (0,65) 0.8 MK 0.3 60°C	8.94	0.9	0.39	35.1
KSil (0,65) 0.8 MK 0.3 CARB 28	6.86	1.01	0.27	32.3
OPC CaO AMB	9.79	100	0.25	43.7
OPC CaO 60°C	15.46	100	0.25	64.1
OPC CaO CARB 28	13.73	37.47	0.27	53.7

two Cr forms which have a very different response to pH (Fernández Pereira et al. 2001).

4. Conclusions

Although the variations of the compressive strength of carbonated samples cannot be explained easily, it is clear that this mechanical property is slightly higher in carbonated samples than in samples cured at room temperature.

In general, in almost all the leaching tests conducted, the carbonation effect is more noticeable in OPC samples than in geopolymer samples. Carbonation considerably reduces the pH and worsens leachability. In the TCLP test, curing the S/S solids at 60°C during geopolymer preparation lowers the pH and worsens leachability in most cases. In many cases, heating (without carbonation) lowers the pH of TCLP leachates even more than carbonation. When all the cases were compared, the best metal immobilization results were obtained with a geopolymeric mixture containing KSil and BFS.

Table 10
Results of the NEN 7345 tank leaching test in S/S solids at 64 days and Category 1 (U1) limit according to Dutch Building Materials Decree (DBMD).

	Cumulative concentration (mg·m^{-2})			
	Zn	Pb	Cr	Cd
KSil (0,65) 0.8 BFS 0.3 AMB	11.54	36.92	33	<3.69
KSil (0,65) 0.8 BFS 0.3 60°C	18.27	39.94	55.87	<3.4
KSil (0,65) 0.8 BFS 0.3 CARB 28	34.04	73.6	10.17	<3.54
KSil (0,65) 0.8 BFS 0.3 CARB 64	45.81	72.15	8.85	<3.54
KSil (0,65) 0.8 KOH BFS 0.3 60°C	49.5	82.54	72.87	<3.77
KSil (0,65) 0.8 KOH BFS 0.3 CARB 28	37.21	39.02	11.28	<3.61
KSil (0,65) 0.8 KOH BFS 0.3 CARB 64	56.22	98.08	9.03	<3.28
KSil (0,65) 0.8 MK 0.3 AMB	49.85	73.26	143	<3.47
KSil (0,65) 0.8 MK 0.3 60°C	26.32	41.94	70.39	<3.42
KSil (0,65) 0.8 MK 0.3 CARB 28	74.99	143.27	30.56	<3.47
KSil (0,65) 0.8 MK 0.3 CARB 64	48.08	183.7	8.22	<3.29
OPC CaO AMB	109	15503	8.98	<3.59
OPC CaO 60°C	85.18	3970	17.35	<3.56
OPC CaO CARB 28	122	33.46	38.91	<3.5
OPC CaO CARB 84	245	38.91	14.4	<4.72
DBMD U$_1$	200	100	150	1

The pH reduction noted above is important regarding the stability of the S/S solids with time. The better response of the geopolymer S/S solids regarding this property implies a clear advantage for geopolymer S/S solids over OPC S/S solids.
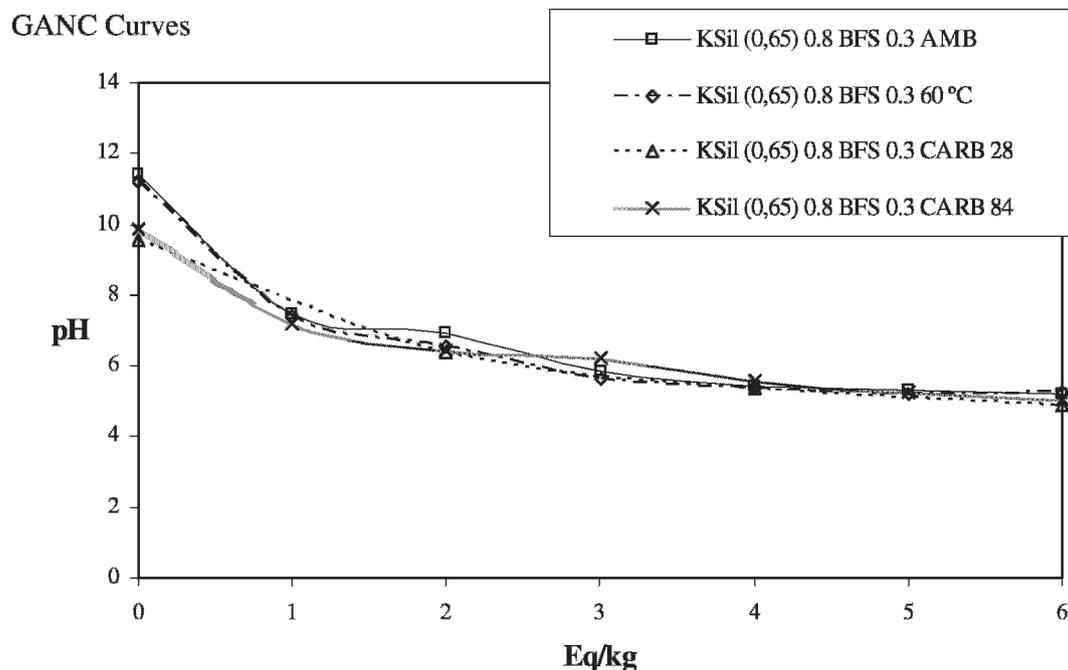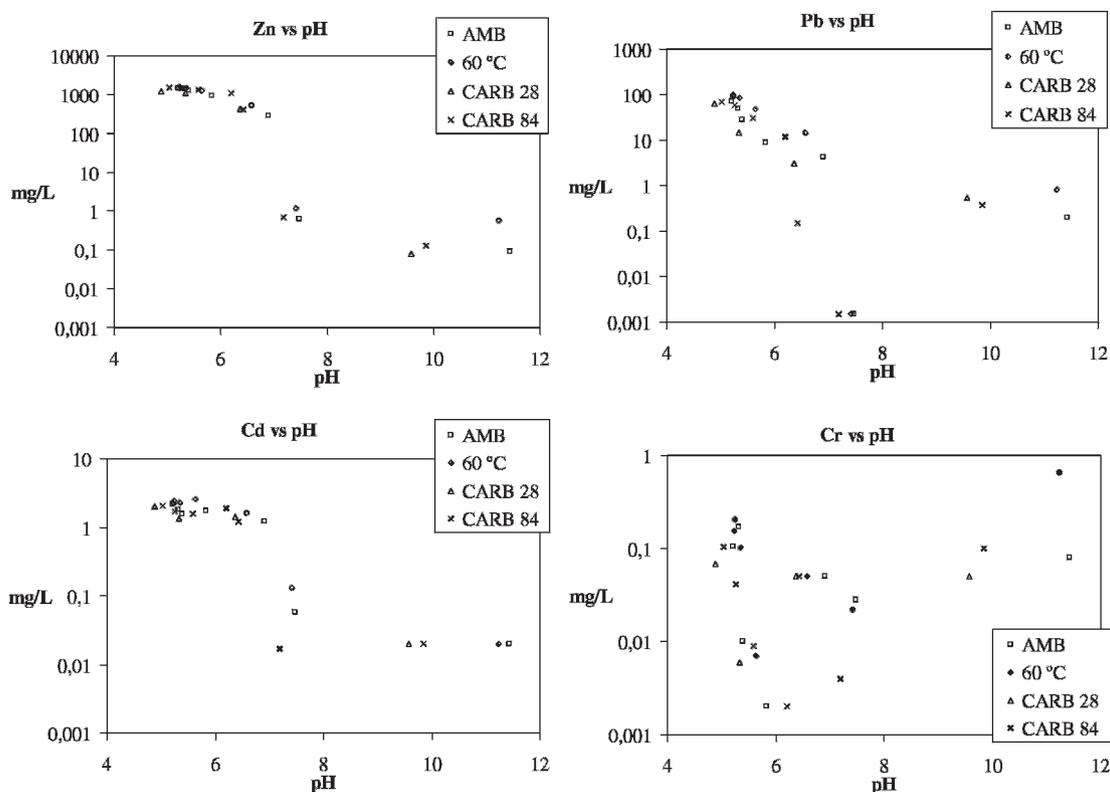
Fig. 3. GANC Curves.

Fig. 4. Zn, Pb, Cr and Cd concentrations versus pH in sample KSil (0,65) BFS 0.3.

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