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Use of municipal solid waste incineration fly ash in concrete

J.E. Aubert*, B. Husson, A. Vaquier

Laboratoire Matériaux et Durabilité des Constructions (L.M.D.C.) INSA-UPS, 135 avenue de Rangueil, 31077 Toulouse cedex 4, France

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Abstract

With a view of reducing the quantities to be landfilled, the Solvay Company has been working on the development of a new physicochemical treatment for municipal solid waste incineration (MSWI) fly ashes: the Revasol process. This process allows reducing the soluble fraction, fixing heavy metals and eliminating dioxins. This article reports on the characteristics of a treated ash and on its use in concrete. For the latter point, three characteristics were chosen: the compressive strength and the durability of the hardened concrete and its behavior to leaching. From mechanical and durable points of view, the ash incorporated in the concrete behaves like ordinary sand. The leaching tests carried out on the concrete confirm that the process makes it possible to obtain materials without major risks for the environment. Also, these results as a whole suggest that the use of waste in concrete constitutes a potential means of adding value.

Keywords: Waste management; Municipal solid waste incineration; Fly ash; Heavy metals; Concrete

1. Introduction

Regulations on waste elimination make waste management a significant problem for the public authorities today. For a long time, the majority of waste was disposed of in tips but this is now becoming impossible. In France, for example, the law of July 13, 1992, forbids the discharge of municipal solid waste (MSW) in tips as from July 01, 2002 [1]. Replacement solutions must therefore be found. Among these is incineration, which currently treats 35% of MSW in France [2]. Associated with selective sorting and energy recovery, it makes it possible to significantly reduce the volume and weight of waste. However, the main problem with incineration is that it produces waste in its turn, although in smaller quantities. This waste includes bottom ashes (250–300 kg/m^3 of MSW), fly ashes and air pollution control (APC) residues (25–50 kg/m^3 of MSW) [3]. The bottom ashes are already reused, mostly as material for roads, but the fly ashes and the APC residues are considered as final residues for which the only lawful destination is the landfill.

2. The problem

With a view of reducing the quantities to be landfilled, the Solvay Company, in collaboration with the Université Libre de Bruxelles, has been working on development of a new physicochemical treatment for municipal solid waste incineration (MSWI) fly ashes, the Revasol process.

This process was divided into three successive steps:

– Water dissolution of ash: during the washing, pH is maintained below 10.5 to prevent the dissolution of heavy metals. In fact, the washing solution containing soluble salts (especially chlorides KCl and NaCl) is treated to recover the sodium that is used in technical soda plants,
– Phosphation with phosphoric acid to stabilize heavy metals,
– Calcination to eliminate organic compounds (especially dioxins), at a temperature higher than 600 °C. This step also enables the crystalline structure of the final product to be stabilized. [4,5].

This article reports on tests incorporating a treated MSWI fly ash (TFA) in concrete. Studied ash, coming from a single French incinerator, was chosen among 10 fly ashes from European incinerators because it was very rich in heavy metals. This ash was treated according to

* Corresponding author. Tel.: +33-05-6155-7757; fax: +33-05-6155-9949.

E-mail address: aubert@insa-tlse.fr (J.E. Aubert).

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the process presented above. In all cases, the criterion for acceptance was that the formulation suggested should not give the concrete too high a pollution potential so it was subjected to the legal leaching tests applicable to final special industrial wastes intended for dumping. To reduce the number of cases to be studied related to the variety of concrete currently manufactured, we defined two limit formulations:

- a concrete for building of medium compressive strength, which for this reason contains a low proportion of TFA,
- a concrete of low compressive strength, which includes a high quantity of TFA.

To evaluate the influence of TFA introduction in concrete, three characteristics were chosen: compressive strength and physical properties (gas permeability, porosity accessible to water and total porosity) of hardened concrete (which control the durability of concrete) and its behavior to leaching.

3. Experiments

3.1. Materials

3.1.1. MSWI-treated fly ash

Ash was treated in a pilot unit of 25 kg/h capacity. The calcination was carried out in rotary kiln at a temperature of 750 °C. The physical characteristics of TFA are presented in Table 1. For the size distribution of TFA, the fraction coarser than 80 μm was analyzed by sieving and the finer fraction by means of a laser particle size analyzer.

Elemental composition of TFA is presented in Table 2. Major oxides were measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES) and the minor elements by inductively coupled plasma-mass spectrometry (ICP-MS). The water content (drying at 100 °C) and the loss on ignition (calcination at 1000 °C) were also measured. The chemical composition of TFA highlights the high percentage of heavy metals.

The crystalline phases were identified using a Siemens D5000 powder X-ray diffractometer equipped with a monochromator using a Kα ($\lambda = 1789$ Å) cobalt anticathode. The minerals in order of decreasing content were minerals of gehlenite group ($Ca_2Al_2SiO_7$), calcium phosphates including apatite [$Ca_5(PO_4)_5(OH,F,Cl)$] and whitlockite [β-$Ca_3(PO_4)_2$], anhydrite ($CaSO_4$), quartz (SiO_2), titanium oxides including titanite ($CaTiSiO_5$) and perovskite ($CaTiO_3$) and hematite (Fe_2O_3). These minerals were accompanied by an amorphous phase (calcium aluminosilicate).

Table 1
Physical characteristics of TFA

Size distribution (μm)	
D10	6
D50	38
D90	200
Density	
Bulk	2.81
Absolute	2.95
BET specific area (m^2/g)	2.26

Table 2
Chemical composition of TFA

Major oxides content (%)		Minor elements content (mg/kg)			
CaO	25.23	Zn	24,046	Co	79
SiO_2	20.67	Pb	8816	Mo	49
P_2O_5	13.56	Sn	2883	Rb	34
SO_3	11.60	Cr	2078	Bi	30
Al_2O_3	10.01	Ni	1889	V	30
MgO	2.74	Cu	1714	Ce	30
Fe_2O_3	2.73	Ba	1521	Ga	21
TiO_2	1.73	Sb	1457	La	15
Na_2O	1.35	Cd	586	Nd	11
K_2O	1.35	Sr	399	Nb	11
MnO	0.2	W	227		
Water content	0.2	Zr	149		
Ignition loss	6.5	As	120		

Usually, the introduction of untreated MSWI fly ashes into cement-based materials leads to many problems such as delayed setting and loss of workability. A study on pastes and mortars showed that these disadvantages were negligible with TFA, which should make interesting its incorporation in concrete [6].

3.1.2. Concrete constituents

The binder used was an ordinary Portland cement containing more than 95% of clinker (CPA-CEM I 52.5R). Two types of aggregates were used: an alluvial siliceous sand (0/6.3) and an alluvial siliceous gravel (2.5/16). The size distributions of these materials are presented in Fig. 1.

3.2. Concrete compositions

Five mixtures were studied:

- a reference concrete (R),
- two compositions A12 and A50 in which cement was replaced by TFA in the proportions of 12.5% and 50%, respectively,
- and finally, two compositions S12 and S50 in which 12.5% and 50% of cement were replaced by sand.

It was thus possible to compare TFA with cement (comparison between A and R) and with sand (comparison between A and S). The calculated concrete compositions are presented in Table 3.

3.3. Experimental procedures

The samples used for all the tests were stored at 20 °C and 100% relative humidity (RH). The measurements of

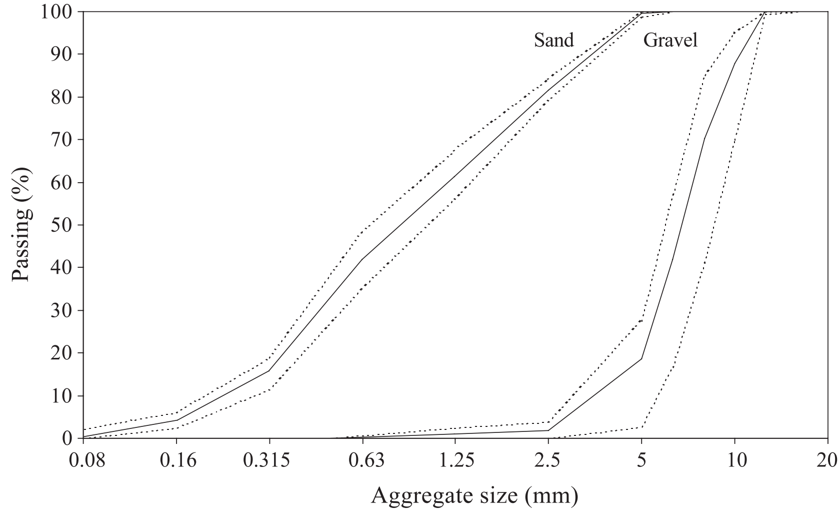

Fig. 1. Size distribution of aggregates used.

physical properties and the leaching tests were performed on 28-day-old samples.

3.3.1. Compressive strength

Compressive strength was measured according to the NF P 18-406 standard on cylinders (11 cm in diameter and 22 cm in height) [7]. For each test, several samples were tested (from 3 to 6 according to the compositions and tests).

3.3.2. Determination of physical properties of concrete

3.3.2.1. Gas permeability measurement. The samples tested were cylinders (15 cm in diameter and 20 cm in height). After 28 days of curing (20 °C, 100% RH), samples were sawn from the original specimen and the first 25 mm of both sides were removed to avoid skin effects. Since the measurement of the permeability depends on the state of saturation of the material, the measurements were made on dried samples. Drying and tests were carried out according to AFPC-AFREM recommendation [8]. The permeability was measured with a Cembureau permeameter. The pressure head was set at 1 bar and the measurements were made when the steady state had been reached.

After the permeability measurement, the samples were sawn into two halves, one being used to measure the porosity accessible to water and the other for total porosity.

3.3.2.2. Measurement of porosity accessible to water. The procedure for this test was that recommended by AFPC-AFREM [8]. The porosity of the concrete was obtained from the difference between the mass of the same sample dry and saturated with water. The total volume of the sample was determined by hydrostatic weighing, and it was thus possible to calculate the porosity accessible to water.

3.3.2.3. Measurement of total porosity. The total porosity of the concrete was calculated starting from the bulk and absolute densities measured on concrete. The density measurements were the same (hydrostatic weighing) and only the

Table 3
Computed composition of concrete

Constituent	Mixtures				
	R	S12	A12	S50	A50
Cement (kg/m^3)	300	262	262	150	150
TFA (kg/m^3)	0	0	38	0	150
Sand (kg/m^3)	724	762	724	874	724
Gravel (kg/m^3)	1087	1087	1087	1087	1087
Water (l/m^3)	195	195	195	200	200

Table 4
Experimental composition and concrete characteristics

	R	S12	A12	S50	A50
Fresh concrete (kg/m^3)	2360 –2400	2360 –2400	2340 –2380	2340 –2410	2350 –2390
Cement (kg/m^3)	310	271	268	155	154
TFA (kg/m^3)	0	0	38	0	154
Gravel (kg/m^3)	1127	1129	1114	1120	1115
Sand (kg/m^3)	741	779	732	900	742
Efficient water (l/m^3)	175	173	172	177	179
Total water (l/m^3)	201	200	198	206	205
Air void (l/m^3)	14	16	16	18	21
Slump (cm)	12	6.5	6.5	5	7.5

Table 5
Physical properties of concrete

	Gas permeability (10^{-16} m^2)	Water porosity (%)	Total porosity (%)
R	3.3–4.6	14.2–15.1	14.2–16.7
S12	1.9–5.1	12.7–14.2	14.5–16.2
A12	1.6–4.6	13.4–16.9	15.9–17.2
S50	35.8–68.0	18.5–20.7	21.5–25.4
A50	17.9–35.6	18.1–22.5	22.7–25.0

state of the material changed: monolithic for the bulk density and crushed for the absolute density. The material was ground until it all passed through the 40-μm sieve, then the powder was ground during 30 more seconds.

3.3.3. Leaching tests

There are currently two tests in the French standards: NF P X31-210 on crushed material [9] and NF P X 31-211 on monolithic material [10]. We have chosen to use both tests and compare the results because neither of the tests taken separately are able to give a simultaneous account of:

- the pollution potential of the concrete in the event of an extraordinary situation (accidental explosion, demolition),
- the retention capacity exerted in normal situations by the monolithic character of hardened cement paste.

The test on crushed material was carried out on a sample crushed to less than 4 mm. Three serial extractions (each during 16 h) are carried out, at a 10:1 liquid to solid ratio, using distilled water.

On the other hand, some modifications were made for the test on monolithic material. The test in its original form is difficult to apply to concrete as it requires a sample obtained by dry coring. Even if it were possible, dry coring would cause significant rises in temperature, which would lead to damage of the concrete, localized on the leached surfaces. To avoid this problem, it was decided to break the samples tested into several pieces; then, a piece from the heart of the concrete was taken as test sample. Of course, this technique prevented us from measuring the surface area in contact with the solution, but we considered that this was less of a problem than the damage caused by dry coring. Moreover, the shape of the pieces taken was comparable for all samples; thus, the transfer areas of the various samples were similar. The conditions of extraction for this test are the same than those used for the test on crushed material.

Various tests were done on the leachates: measurement of the pH, of the soluble fraction, of sulfate content and minor elements content by ICP-MS.

4. Experimental results and discussion

4.1. Properties of fresh concrete

Table 4 indicates the values of slump, density and air content for the fresh concrete and the actual amounts of the various components.

The incorporation of TFA in concrete leads to a slight reduction of workability for identical water/cement (w/c) ratios (A12 and R). It should, however, be noted that sand has the same effect as TFA on the reduction of slump (S12). For the mixtures with high substitution rates (S50 and A50), it was decided to increase the w/c ratio in order to preserve a workability comparable with that of the other mixtures (between 5 and 7.5 cm with the slump test). The quantity

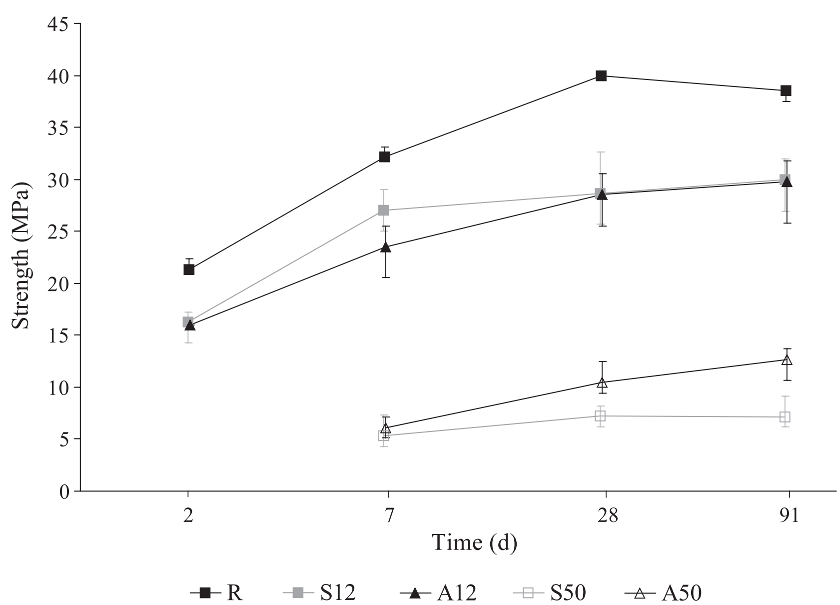

Fig. 2. Hardening of concrete.

Table 6
Comparisons between experimental strength and computed strength (Bolomey) at 28 days

		R	S12	A12	S50	A50
Experimental strength	Minimal	40	26	26	6	9
	Maximal	40	33	31	8	12
Computed strength	with total water	31	25	24	6	6
	with efficient water	37	30	30	10	9

of air content increased with the percentage of substitution; however, for this property too, TFA behaved in a similar way to sand.

4.2. Physical properties of hardened concrete

The physical properties of the hardened concrete are given in Table 5. Three tests were carried out on three different samples and the values presented in the table are the minimum and maximum values.

To clearly distinguish the influence of TFA on these properties, we propose to analyze the results starting from the concrete lowest in cement and containing no TFA, S50. In fact, the only comparisons that isolate the role of cement compared to that of TFA by excluding any other influence (i.e., sand content) are R/S50, A50/S50 and A12/S50.

We note that introducing 150 kg of TFA into one cubic meter of concrete (A50) to replace 150 kg of sand (S50) had no significant effect on the total porosity and the porosity accessible to water, which went from 21.5–25.4% to 22.7–25.0% and from 18.5–20.7% to 18.1–22.5%, respectively. In terms of gas permeability, the introduction of TFA caused a considerable improvement [permeability reduced by half (A50/S50)], the introduction of cement causing a much more marked reduction of permeability [approximately 90% (S50/R)].

A mixed addition (TFA + cement) of 150 kg (A12) was practically as effective in reducing porosity as an addition of 150 kg cement (R) and involved the same reduction of gas permeability. This means that the introduction of TFA into a cement-poor concrete does not degrade its structure and a mixed addition (TFA + cement) helps to seal the structure.

4.3. Compressive strength of hardened concrete

The development of the compressive strength between Days 2 and 90 is represented on Fig. 2.

It can be seen, as it was expected, that the smaller the quantity of cement, the lower the compressive strength.

If we apply Bolomey's formula, it can be seen that at 28 days, the increases in strength observed for an addition of TFA and/or cement relative to the strength of the S50 concrete are practically due to the sole contribution of the cement. Table 6 shows that the measured values fall below the limits calculated using Bolomey's formula [11]:

$$R_c = f_c k\left(\frac{C}{E+v} - 0.5\right)$$

In this relation, f_c represents the characteristic strength of the cement (65 MPa for CPA-CEM I 52.5R cement) and k is a coefficient, which depends on the aggregates (0.5 for siliceous rolled aggregates). Coefficients C, E and v represent the mass of cement, the volume of water and the volume of air voids, respectively. Results differ according to whether the quantity of total water or efficient water is chosen for the calculation of strength. Efficient water is defined as moisture of aggregates + water added + admixture water − absorbed water by aggregates.

In conclusion, from a practical point of view, TFA behaves like fine sand. Furthermore, for high rate of replacement, TFA has a favorable effect on compressive strength.

4.4. Leaching tests

Results of the leaching tests are given in Table 7 for the tests on crushed concrete and Table 8 for those on monolithic concrete. These results are the average of two tests on different samples. The absolute (or relative) accuracy (acc.) of measurements is also given in the tables. The concentrations of the solutions were calculated versus the mass of

Table 7
Concentrations of leached elements (crushed concrete)

Unit	Acc.	R				A12				A50			
		1	2	3	Total	1	2	3	Total	1	2	3	Total
Cr (μg/kg)	2%	138	142	155	435	211	246	352	809	550	867	887	2304
Ni (μg/kg)	2%	149	132	90	371	217	144	95	456	84	86	47	217
Zn (μg/kg)	2%	138	78	56	272	203	107	83	393	199	105	69	373
Cu (μg/kg)	6%	16	27	9	52	20	15	12	47	22	16	11	49
Pb (μg/kg)	2%	2	11	2	15	22	8	5	35	30	11	6	47
Cd (μg/kg)	30%	0.4	9.2	1.4	11	3.0	8.3	4.3	16	1.3	1.2	0.8	3.3
Sn (μg/kg)	7%	0.2	0.9	4.6	6	1.4	0.9	0.8	3	4.2	2.4	0.6	7.2
As (μg/kg)	17%	1.0	0.2	< 0.1	< 1.3	1.2	0.6	0.7	2.5	1.9	2.9	4.5	9.3
pH	0.1	12.5	12.4	12.4		12.5	12.3	12.3		12.1	12.0	11.8	
FS (G/kg)	5%	13.8	10.0	6.8	31	14.0	9.7	7.1	31	6.3	4.4	3.3	14
SO_4 (Mg/kg)	50	< 100	< 100	< 100	< 300	< 100	< 100	148	< 348	209	236	331	776

Table 8
Concentrations of leached elements (monolithic concrete)

Unit	Acc.	R				A12				A50			
		1	2	3	Total	1	2	3	Total	1	2	3	Total
Cr (μg/kg)	2%	43	14	12	69	65	12	13	90	121	35	26	182
Ni (μg/kg)	2%	23	23	12	58	40	13	11	64	49	7	7	63
Zn (μg/kg)	2%	21	153	77	251	48	57	56	161	74	123	92	289
Cu (μg/kg)	6%	3.4	5.3	2.3	11.0	7.8	2.2	2.9	12.9	14.3	7.1	4.6	26.0
Pb (μg/kg)	2%	1.0	1.6	0.7	3.3	3.4	0.8	0.4	4.6	6.6	2.6	1.1	10.3
Cd (μg/kg)	30%	0.2	0.4	0.1	0.7	0.1	0.1	0.2	0.4	0.2	0.2	0.1	0.5
Sn (μg/kg)	7%	0.1	0.0	1.5	1.6	1.2	0.4	0.3	1.9	1.3	1.0	2.3	4.6
As (μg/kg)	17%	1.2	1.0	0.6	2.8	1.2	0.9	0.7	2.8	6.5	8.0	7.5	22.0
pH	0.1	11.5	11.3	11.2		11.5	11.2	10.9		11.2	10.9	10.6	
FS (G/kg)	5%	1.7	1.2	0.6	3.5	1.3	0.9	0.5	2.7	1.0	0.7	0.5	2.2
SO_4 (Mg/kg)	50	< 100	< 100	< 100	< 300	< 100	< 100	< 100	< 300	226	138	148	512

solid (μg/kg). For each leaching, the results can be analyzed in two ways:

- the variation of the leached element concentration for the same composition in the three successive leachates,
- the influence of TFA quantity in concrete on the contents of the leached species.

4.4.1. Crushed concrete leaching

4.4.1.1. Variation of the leached element concentration during the test. For all the mixtures, the leached element concentration decreased or remained constant over time. This was confirmed for the pH and the soluble fraction (FS). Only two species did not follow this variation: chromium and sulfates.

4.4.1.2. Variation according to the percentage of substitution. An increase in the quantity of TFA in the concrete involved more significant leaching of chromium, lead, arsenic and sulfates. On the other hand, the concentrations of the other elements were comparable from one mixture to another. It can be noted that the soluble fraction decreased strongly with the reduction in the quantity of cement. This is explained by the large amount of soluble calcium contained in cement compared with the few soluble species present in TFA (except sulfates, which reacted with the constituents of the cement).

4.4.2. Monolithic concrete leaching

4.4.2.1. Variation of the leached element concentration during the test. The concentrations of leached elements being very weak, it was difficult to bring out trends in their variation. They remained roughly constant or decreased slightly.

4.4.2.2. Variation according to the percentage of substitution. As for the tests on crushed concrete, the higher the percentage of substitution, the higher the concentrations of chromium, lead, arsenic and sulfates leached. In this case, tin also followed the same trend. For the other elements, the results obtained for the various compositions were very similar to each other.

4.4.3. Comparison between leaching behavior of crushed and monolithic concrete

Table 9 shows the total concentrations of the most harmful elements leached. These results are compared with threshold values allowed for leachates from MSWI bottom ashes to be reused in road works in France [12].

Several comments can be made on these results:

- the stabilization process was very effective since the concentrations of leached elements found in the leaching tests were very low compared to those contained in TFA. This was checked in particular for Zn and Pb,
- except for Cr and As, the results obtained on the TFA-containing concrete were very similar to those of the reference,
- only chromium exceeded the legal thresholds in the case of the crushed concrete; all concentrations were satisfactory for the monolithic concrete,
- the concentrations of heavy metals obtained on monolithic concrete were always much lower than those obtained on crushed concrete except in the case of arsenic. The leaching of arsenic is influenced by the pH of solution what already was observed by Van der Sloot [13]: the lower the pH (10.6–11.5 in monolithic

Table 9
Comparison of total concentration leached for crushed and monolithic concrete

	Crushed concrete			Monolithic concrete			Threshold
	R	A12	A50	R	A12	A50	Bottom ashes
Cr (μg/kg)	435	809	2304	69	90	182	1500
Zn (μg/kg)	272	393	373	251	161	289	
Pb (μg/kg)	15	35	47	3	5	10	10,000
Cd (μg/kg)	11	15.6	3.3	0.7	0.4	0.5	1000
As (μg/kg)	< 1.3	2.5	9.3	2.8	2.8	22	2000
FS (G/kg)	30.6	30.8	14.0	3.5	2.7	2.2	50
SO_4 (Mg/kg)	< 300	< 348	776	< 300	< 300	512	1000

concrete vs. 11.8–12 in crushed one), the higher the amount of arsenic leached.

5. Conclusion

The substitution of TFA in place of cement in concrete does not involve a loss of mechanical strength greater than that caused by the reduction in the quantity of cement. TFA behaves like inert fine sand.

Moreover, the physical properties as a whole of fresh and hardened concrete are not deteriorated by the presence of TFA. The physical properties of hardened concrete (gas permeability, porosity accessible to water and total porosity) control the transport properties of concrete and so its durability. These properties are not significantly modified by the introduction of TFA, but the question of long-term durability has not been addressed in this work.

Lastly, the leaching tests carried out on concrete confirm that the process makes it possible to obtain materials with a pollutant potential lower than that characterizing the MSWI bottom ashes, which are accepted for use in roads.

Also, an overall view of these results suggests that the use of treated ashes in hydraulic concrete is potentially profitable. This is all the more true for high proportions of substitution because it provides an interesting alternative to surface dumping.

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