



Use of admixtures in organic-contaminated cement–clay pastes

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ABSTRACT

In this work microstructure, porosity and hydration degree of cement-based solidified/stabilized waste-forms were studied before assessing their leaching behaviour. 2-Chloroaniline was chosen as a model liquid organic pollutant and included into cement pastes, which were also modified with different admixtures for concrete: a superplasticizer based on acrylic-modified polymer, a synthetic rubber latex and a waterproofing agent. An organoclay, modified with an ammonium quaternary salt (benzyl-dimethyl-tallowammonium, BDMTA), was added to the pastes as pre-sorbent agent of the organic matter.

All the samples were dried up to constant weight in order to stop the hydration process at different times during the first 28 days of curing, typically, after 1 day (1 d), 7 days (7 d) and 28 days. Then, the microstructure of the hardened cement–clay pastes was investigated by powder X-ray diffraction (XRD). The hydration degree and porosity were studied by thermal analysis (TG/DTA) and mercury intrusion porosimetry (MIP), respectively. For samples cured for 28 days a short-term leach test set by Italian regulation for industrial waste recycling (D.M. 5 February 1998) was performed. The best results showed a 5% release of the total initial amount of organic pollutant.

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1. Introduction

A wide range of hazardous wastes is stabilized and solidified using cementitious materials [1]. Usually, heavy metal ions are chemically bound to the cement matrix [2,3], while organic contaminants are only physically separated from the outside environment [4–6] by the hardened cement paste. Indeed, in toluene/cement mixtures Butler et al. [4] found (using synchrotron X-ray microtomography, electron probe microanalysis and ²⁹Si MAS NMR) that toluene is almost uniformly dispersed within the matrix and is not chemically bound to any cement phase, but remains highly mobile; moreover, a small portion of toluene forms large spherical vesicles. The findings of Nestle et al. [5] confirmed the formation of liquid pockets in cement pastes contaminated with apolar aliphatic and polar organic liquids, while the solvent phase, in the case of aromatic apolar compounds, was finely dispersed. The absence of chemical interactions between organic contaminants and the cement matrix was also observed by Natali Sora et al. [6] who studied the leaching behaviour of cement pastes admixed with 2-CA and found that the aromatic amine was almost completely released within 6 months. Then, the total inability of the

cement matrix itself to immobilize organic pollutants was unequivocally proven; the use of pre-sorbent agents, then, is the only likely way making the use of a cement-based stabilization and solidification process (S/S), in presence of organic pollutants, possible.

Nonetheless, there is wide experimental evidence that the presence of organic compounds in hydrating cement pastes might adversely affect the hydration reactions [7–10]. Among others, Gussoni et al. [10] observed a pronounced delay in the cement hydration when 2-chloroaniline was admixed with fresh cement paste using ¹H NMR spin–spin relaxation and imaging.

Furthermore, whenever incineration is not applicable or too expensive, S/S process of mixed organic/inorganic wastes seems to be the only alternative for safe and low-cost disposal. Moreover, it was suggested that adverse effects of organic contaminants could be overcome by the use of pre-solidification adsorbent materials, like activated carbons and organophilic clays [11]. Unfortunately, just a few experimental studies, aimed at demonstrating the effectiveness of the organophilic clays as pre-adsorbents in the cement-based S/S technology, can be found in the literature since the 1990s [7–8,12–16]. In a recent work [16] some of the present authors found that, using organophilic clays as pre-adsorbent, the amount of pollutant released from solidified wastes during a dynamic leaching test could be markedly reduced, even though there was yet no evidence of chemical binding between the organic

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substance and the matrix. Obviously, the reversibility of this sorption has to be considered in the risk assessment of the possible reuse or disposal of these solidified wastes. Then, in this work we used three different commercial admixtures, which should have reduced the porosity of the matrix (a rubber latex and a superplasticizer) or repels water (waterproofing agent) to improve the performance of cement pastes admixed with organophilic clays and organic pollutants. To our knowledge, few studies have been addressing the use of organic admixtures in cement-based S/S technology, for instance a couple of interesting applications of rubber latex in the immobilization of heavy metal ions into a cement paste [3–17]. A recent issue of this journal (vol. 141, issue 2) was devoted to cover the state-of-the-art advances on S/S, thus showing the international interest in the development of this technology, as demonstrated by papers dealing with technical sustainability and project lifecycle of in situ S/S [18,19]. In particular, most of the papers were dedicated to the treatment of inorganic wastes (trace metals [20], As and Pb [21]), or petroleum contamination [22,23].

The aim of the present work was to study the combined influence of admixtures (i.e. waterproofing agent, rubber latex, and superplasticizer) and an organic pollutant (2-chloroaniline pre-adsorbed onto an organophilic clay) on the hydration process of an ordinary Portland cement paste after the first 28 days of curing. The pollutant mobility in the solidified cement matrix was assessed by leaching test. The phase composition was determined by X-ray powder diffraction; a quantitative determination of the two main crystalline phases, i.e. portlandite and calcite, was performed by thermogravimetric analysis; the solidified sample microstructure was observed by scanning electron microscopy (SEM) and supported by EDXS.

2. Experiments

A schematic representation of the line of reasoning is reported in Fig. 1; two sets of cement paste samples were prepared: in the former one the contaminated pastes were mixed with 2-CA and organoclay without a pre-adsorption process of 2-CA onto the organoclay, in the latter the pastes were prepared after a 24-h pre-adsorption process of 2-CA onto the organoclay. For each set of samples different admixtures for concrete were used: superplasticizer, rubber latex and waterproofing agent (see Section 2.2).

2.1. Materials

All the cement pastes were prepared using Ordinary Portland Cement (OPC) Type I 52.5 R (CementiRossi Piacenza, Italy), an organic-modified montmorillonite (provided by Laviosa Chimica Mineraria s.r.l., Italy) as pre-solidification adsorbent of the organic liquid pollutant and three different types of commercial admixtures for concrete, in particular a superplasticizer based on acrylic-modified polymer, a synthetic rubber latex and a waterproofing agent (all products were provided by Mapei, Italy).

The model organic pollutant chosen was 2-chloroaniline (2-CA) Fluka (99.5% purity). Waters Milli Q[®] water was used throughout this work. The organoclay (OC3) [24], used as supplied by the producer, was a montmorillonite modified with an ammonium quaternary salt (benzyl-dimethyl-tallow-ammonium, BDMTA); the basal spacing, determined by XRD analysis, was $22.7 \pm 0.2 \text{ \AA}$ and the amount of organic matter (om), estimated from the weight loss measured by thermogravimetric analysis, was 0.32 kg of organic matter per kg of solid ($\text{kg}_{\text{om}}/\text{kg}$).

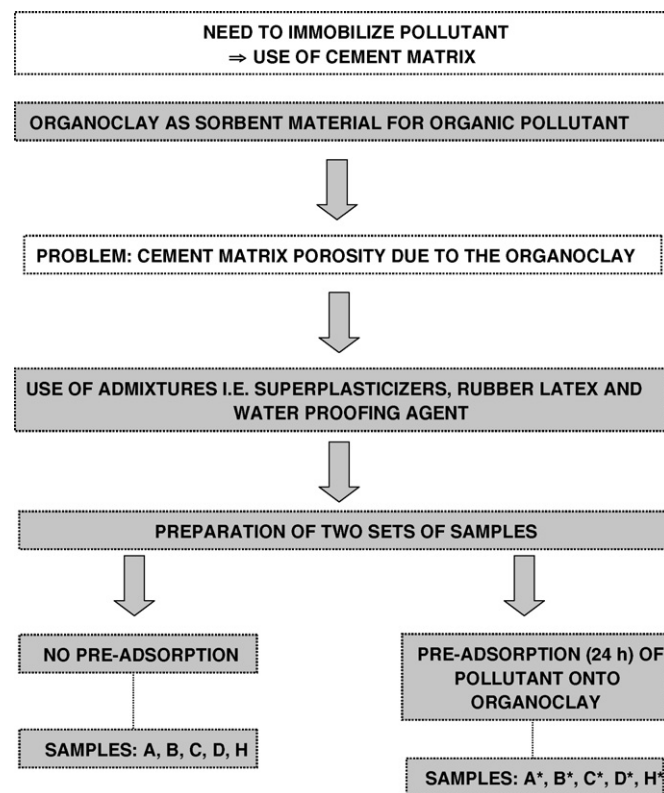


Fig. 1. Flowchart of the work.

2.2. Sample preparation

Fifteen series of samples were prepared: series Ap, Bp, Cp, Dp and Hp (contaminated without pre-adsorption process) and Ap*, Bp*, Cp*, Dp* and Hp* (contaminated with pre-adsorption process) contained a model organic pollutant and the organophilic clay; the other five series (uncontaminated) only the organoclay (series A–D and H).

Different types of organic admixtures, chosen among those typically applied in the concrete mix-design, were added: a superplasticizer based on acrylic-modified polymer in the series B/Bp/Bp*, a synthetic rubber latex in the series C/Cp/Cp*, a waterproofing agent in the series D/Dp/Dp* and a last one prepared with the combined use of superplasticizer and latex H/Hp/Hp*. Series A/Ap/Ap* were prepared without admixtures as references.

The procedure for preparation of the cement pastes is represented in Fig. 2. The fresh pastes were cast and sealed into polyethylene cylindrical moulds (3.2 cm height and 2.2 cm diameter), only partially filled. All samples were cured in an air-conditioned room at $23 \pm 1 \text{ }^\circ\text{C}$.

One specimen for each group was demoulded at 28 days and oven-dried up to constant weight in order to stop hydration. Then the solidified monolith was cut: a fraction of the sample was ground in a grinding mill, to obtain the powders to be used for XRD and thermogravimetric analysis (TGA); while some fragments were used for SEM-EDX analysis and mercury intrusion porosimetry (MIP). A second series of specimens was demoulded after 28 days and used to perform the leaching test.

A second set of contaminated samples was prepared; in this case the organoclay and 2-chloroaniline suspended in water were maintained in contact for 24 h on a rotating arm; then the slurry obtained was admixed with cement powder, the remaining water and the admixtures (samples marked with letter –p*).

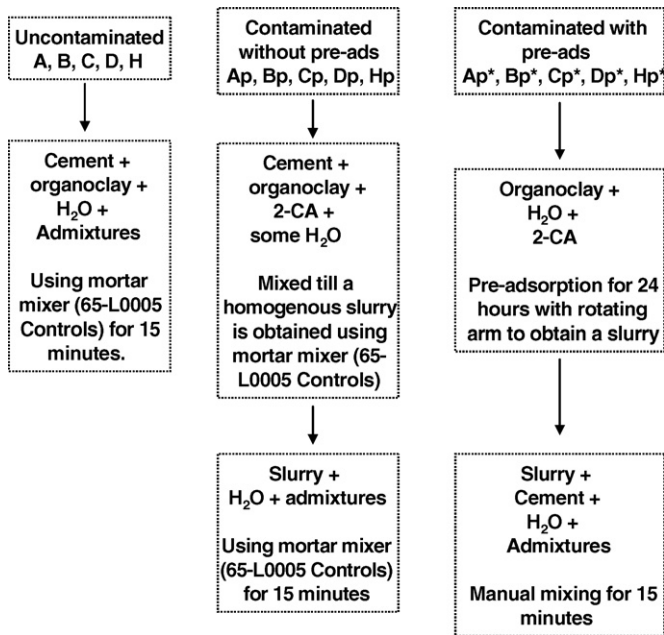


Fig. 2. Flowchart of the different sample preparations: non-polluted samples (left), polluted samples without pre-adsorption of 2-CA onto organoclay (middle), polluted samples with pre-adsorption (right).

Table 1 reports the compositions of all the samples.

2.3. Leaching tests

The leaching test adopted in the present work was indicated by the Italian regulations [25] in order to assess whether a waste can be considered hazardous or not. It is a dynamic leach test which has to be performed on monolithic samples using deionized water as leachant.

The leaching experiment started after 28-day curing: the solidified samples were smoothed, cleaned and each of them hung in a 60-mL jar filled with 35 mL of water, completely immersed and maintained at 24 °C without agitation; the liquid to solid ratio was of 5:1. Water was periodically renewed: at 2, 8, 24, 48, 72, 102, 168 and 384 h (16 days). Concentration of 2-CA in the leachates was determined at each leachant renewal, according to the test procedure; the amount of 2-CA was gas-chromatographically determined after iso-octane extraction. GC analyses were performed with a Carlo Erba Mega mod. 5160 instrument, equipped with an on-column injector, flame ionization detector and HP-fused silica capillary column 0.32 mm internal diameter and 50 m length,

Table 1
Composition of cement pastes

Sample	w/c	w/sm	sp/c	rl/c	wp/c	af/l	s/c	p/c
A	0.85	0.65	–	–	–	–	0.30	–
Ap	0.85	0.65	–	–	–	–	0.30	0.08
B	0.67	0.52	0.016	–	–	–	0.30	–
Bp	0.67	0.52	0.016	–	–	–	0.30	0.08
C	0.85	0.65	–	0.05	–	–	0.30	–
Cp	0.85	0.65	–	0.05	–	–	0.30	0.08
D	0.85	0.65	–	–	0.01	–	0.30	–
Dp	0.85	0.65	–	–	0.01	–	0.30	0.08
H	0.56	0.46	0.010	0.15	–	0.002	0.30	–
Hp	0.56	0.46	0.010	0.14	–	0.002	0.30	0.08

w: water; c: cement; sm: solid matter = cement + organoclay; sp: superplasticizer; rl: rubber latex; wp: waterproofing agent; af: anti-foaming agent; l: liquids; s: slurry = water + 2-CA + organoclay; p: pollutant = 2-chloroaniline.

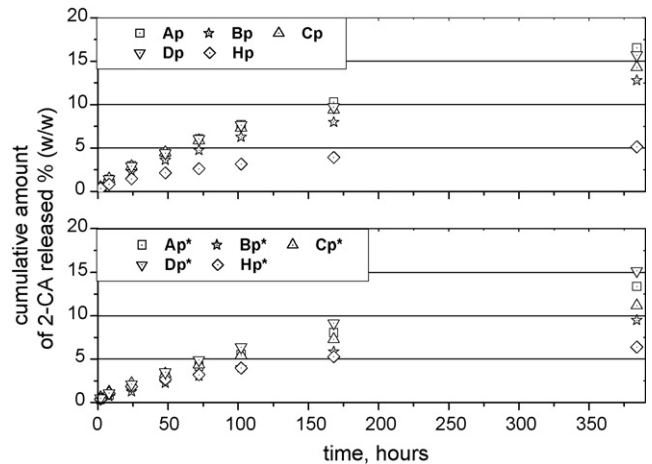


Fig. 3. Result of leaching test, after 28 days of curing, for all contaminated samples with (–p*) and without (–p) pre-adsorption.

coated with 5% phenylmethylsilicone rubber, 0.5 μm thickness. The temperature was linearly raised from 70 to 130 °C at 4 °C/min; then to 250 °C at 10 °C/min. A final isothermal time of 5 min at 250 °C was maintained.

Three gas-chromatography (GC) analyses were performed for each point and the results, reported in Fig. 3, are the mean value of such analyses. The standard deviation estimated was about 10% in the worst case (low concentration leachates) and less than 0.5% in the best one (high-concentration leachates).

2.4. Thermogravimetric analysis

Thermal measurements were performed with a DTA-TG SEIKO 6300; TG analyses were carried out in nitrogen from room temperature up to 1000 °C, at 10 °C/min rate. Powdered samples, previously dried to prevent hydration, were about 20 mg. The amount of portlandite (Ca(OH)₂) and of calcite (CaCO₃) was determined from weight-loss between 400 and 500 °C and 500 and 800 °C, respectively; all values are reported as grams per 100 g of dried sample. Data relative to Ca(OH)₂ amount in series C and H were corrected to account for latex thermal loss.

2.5. X-ray powder diffraction (XRPD)

The X-ray diffraction (XRD) measurements were carried out with a Philips diffractometer at room temperature over the 2θ range from 2° to 80° using graphite monochromated Cu Kα radiation. The step scan was 0.02° and the measuring time 10 s per step. The phase analysis was made using the Diffrac Plus Evaluation software (Bruker AXS).

2.6. Scanning electron microscopy

A Cambridge Stereoscan 360 scanning electron microscope equipped with a Oxford Inca Energy 200 Link energy dispersive spectrometer (EDS) was used for the morphological and chemical analysis of the hydrated samples. SEM-EDX analysis were carried out on both polished and fractured samples. All the samples were gold coated to prevent charging effects.

2.7. Mercury intrusion porosimetry

The pore structure of the different systems was determined by means of mercury intrusion porosimetry (Carlo Erba 4000

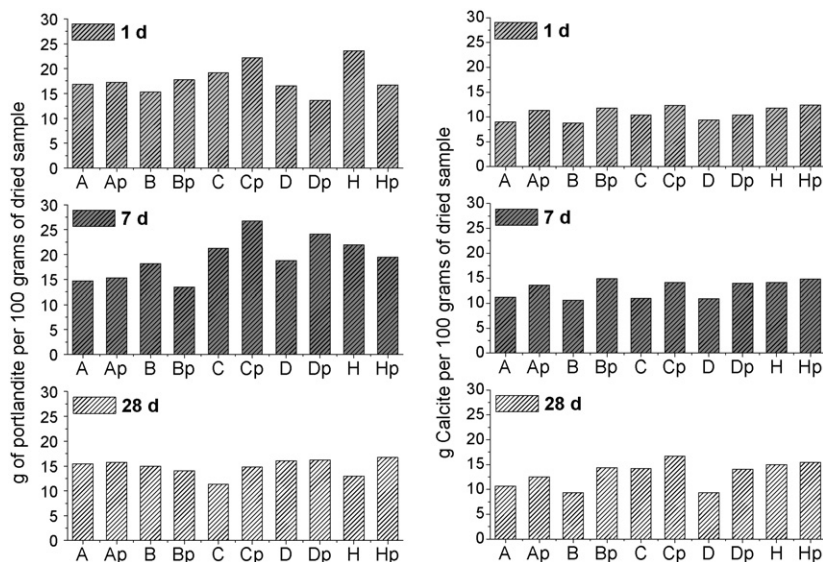


Fig. 4. Amounts of portlandite and calcite estimated by means of TG/DTA for uncontaminated and contaminated sample at different times of curing (1, 7 and 28 days).

porosimeter) for pressures up to 4000 bar ($r=0.2$ nm according to Washburn model). The samples (fragments of 1.5–2 g) were previously dried in oven and then put in a dryer-vessel until the constant weight was reached.

2.8. Permeability test

Two different tests were performed: gas-permeability (i.e. N_2) and liquid-permeability (i.e. water). Gas-permeability was measured fluxing N_2 through the samples (cylinder with area = 5 cm^2 and length = 2.495 cm), setting flux-pressure with a pressure reducer and measuring the gas-flow rate (in the range 0.25–2.5 atm). A water-permeability test with Hoek triaxial cell was performed on a system opportunely treated (diameter = 38 mm , length = 74 mm). Growing pressures up to 8 bar were applied and the flow rate measured.

3. Results and discussion

Since the interactions between the organoclay and the organic pollutant are relatively weak and reversible [24], it is vital to reduce the porosity/permeability of the matrix to decrease the release of the pollutant; so different organic admixtures were used. Indeed, it is known that the superplasticizer drastically lowers the water amount (and consequently the porosity), whereas rubber latex develops a polymeric film around cement particles that should hinder the pollutant mobility.

3.1. Leaching behaviour

Fig. 3 shows the cumulative amount of 2-chloroaniline, released after 16 days, for both the samples with (–p*) and without (–p) pre-adsorption. At the end of the leaching test the sample without any admixture (Ap) released 16.5% of 2-CA, while those prepared using the superplasticizers (Bp), the rubber latex (Cp), the waterproofing agent (Dp) released 12.8, 14.3 and 15.7%, respectively. The use of the admixtures shows a slight improvement (about 3.7, 2.2 and 0.8%). The process of pre-adsorption yielded better results: 13.4, 9.5, 11.2 and 15.1% of 2-CA released in samples Ap*, Bp*, Cp* and Dp*, respectively. If compared with sample Ap, there was a further

improvement of 3.2, 7.1, 5.4 and 1.4%, respectively. The combined use of superplasticizer and latex evidenced a release of 5.1 and 6.4% for samples Hp and Hp*, respectively. These results were more encouraging having reduced the release by 11.4% (Hp) and 10.2% (Hp*) with respect to the reference (Ap). The lower w/c ratio of the pastes treated with superplasticizer (series B and H) was vital to obtain better results due to its positive effects on reducing the total open porosity. The better leachability results of the so-called pre-adsorption series can be ascribed to the more effective adsorption process. In a previous study [24] it was assessed that the adsorption process onto organoclays is not instantaneous and 24 h could have been a careful balance between reaching thermodynamic equilibrium and technological requirements. Even if a shorter time, maybe 1 h or somewhat less, could be more than enough for an effective time/efficient process, it is quite clear from our results that in 15 min there is a non-negligible amount of pollutant that did not yet enter the clay galleries. As a direct consequence the fraction of pollutant adsorbed onto the external clay platelets surface is likely to be more easily leached and this accounts for the slightly better performance of the series with the pre-adsorption. Within a family of samples, i.e. A–D and H series, there is a clear link between w/c ratio, porosity and leaching behaviour. It is well-known that the lower the w/c ratio the lower the porosity; indeed in samples A, C and D ($w/c=0.85$) the porosity exceeds 40% while for sample B ($w/c=0.67$) it is about 37% and for H ($w/c=0.56$) about 32%. For the last two samples the great improvement in porosity is due to the use of superplasticizers which enables a lowering of the w/c ratio. As a consequence the better leaching behaviour is observed in B and H samples and the worst one in A, C and D samples.

3.2. Thermal analysis

A quantitative determination of the amounts of portlandite and calcite was performed by TGA and results were reported in Fig. 4 as grams of substance per 100 g of dried sample. From the experimental data it can be observed that neither the pollutant nor the admixture greatly influenced the hydration process. In particular, in 1-day-old samples, contaminated and uncontaminated, large amounts of portlandite were detected. By comparing the amounts of portlandite in series A and B, a similar trend was observed: in

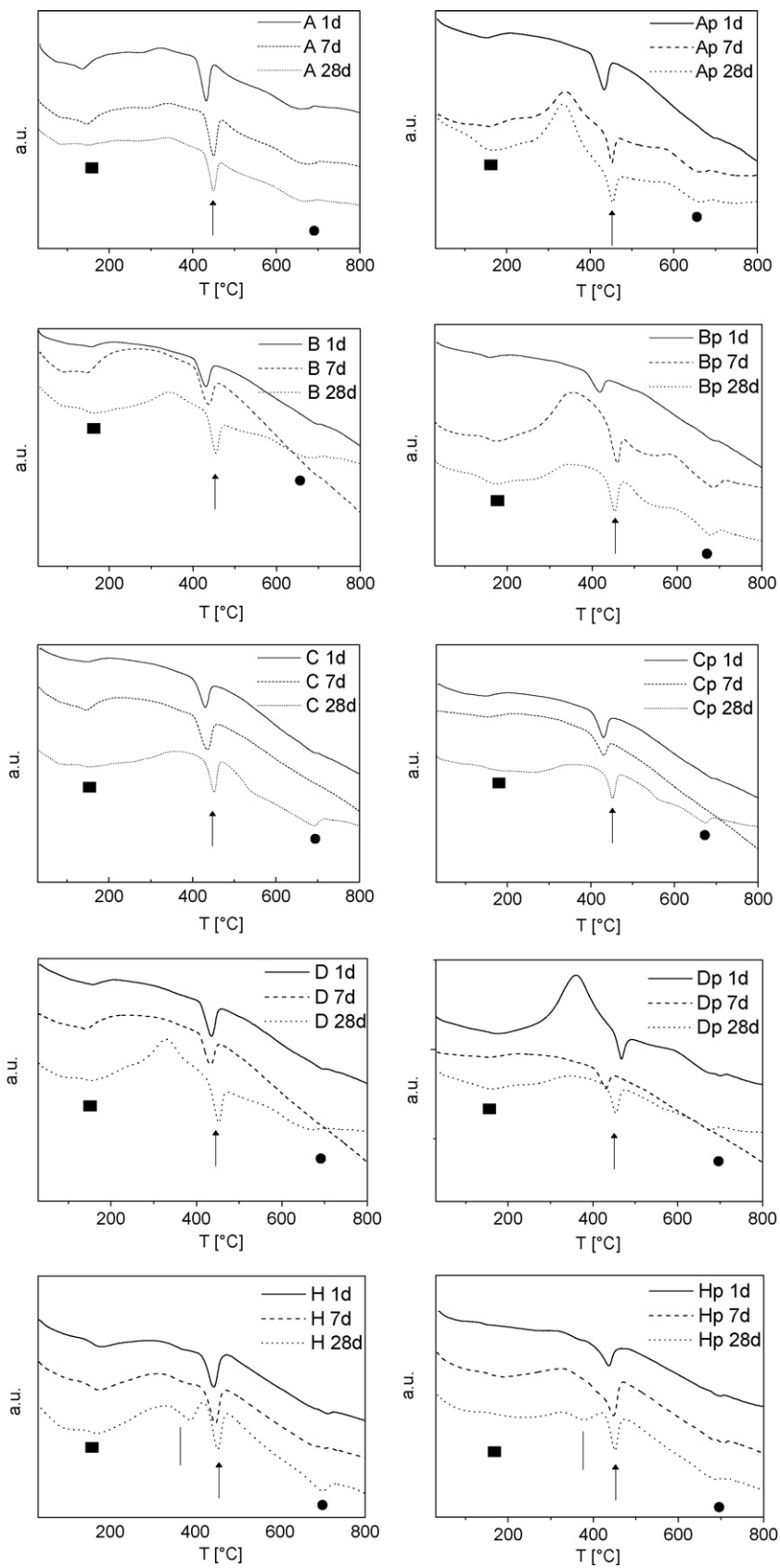


Fig. 5. Comparison of DTA curves of all samples, uncontaminated and contaminated at 1, 7 and 28 days.

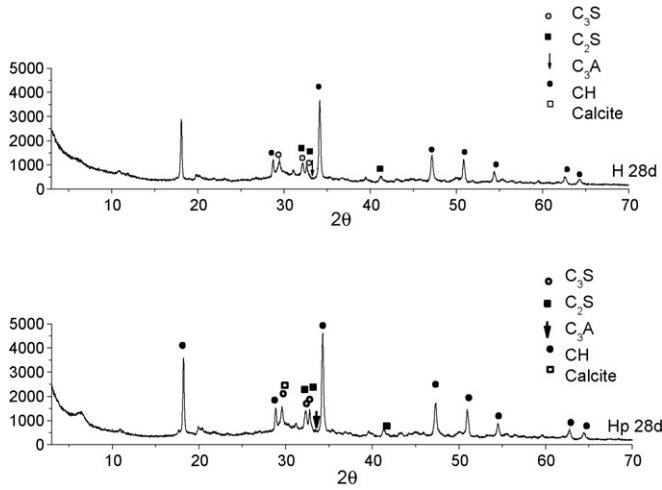


Fig. 6. XRD pattern for samples H and Hp at 28 days of curing.

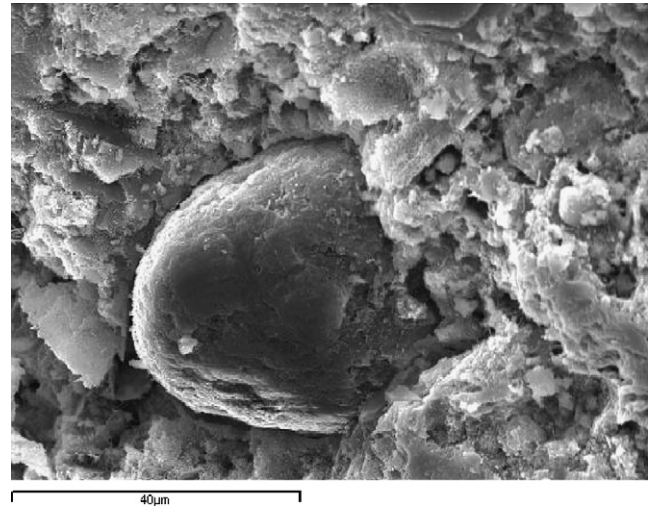


Fig. 7. SEM pictures, on fracture, of a grain of organoclay inside the cement matrix of sample A28d.

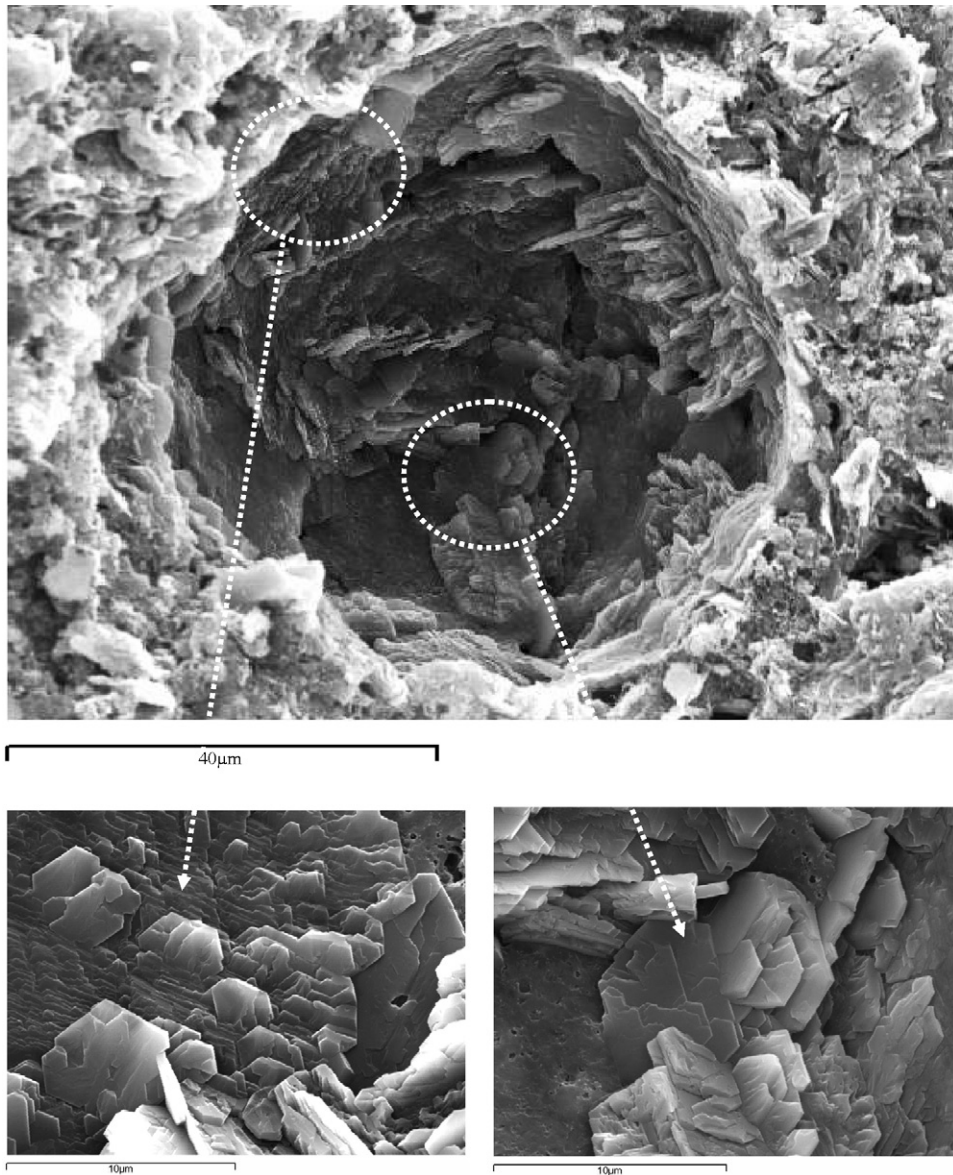


Fig. 8. SEM pictures, on fracture of sample Cp at 28 days, of a large cavity where portlandite was detected.

fact, portlandite slightly decreased in the first 28 days of hydration as a consequence of the normal carbonation process. Some anomalies were observed in the samples containing latex (series C and H) and waterproofing agent (series D), rising some doubts on the usual way adopted to estimate the amount of portlandite, i.e. from the weight loss in the range of 400–500 °C. Indeed, the calcium hydroxide interacts with the polymer [26] causing the dehydration process to be difficult; as a consequence, it was reported that two endothermic peaks appeared in the usual dehydration area of portlandite [27].

Typical DTA curves are shown in Fig. 5: all the samples in the first 28 days of hydration are reported. The broad endothermic peak (■) at low temperature (around 200 °C) is due to the decomposition of calcium silicate hydrates, while the sharp one at about 450 °C (†) corresponds to the dehydration of calcium hydroxide, and at about 700 °C it is due to the decomposition of calcite (●) [28].

By comparing the DTA curves it can be noticed that series A and B show the same trend; on the contrary, some anomalies can be observed in series containing latex (series C and H) and waterproofing agent (series D). The greatest differences can be noticed in series H, where the amount of latex is higher than in series C: after 28 days of curing it is clearly visible a second peak at about 400 °C (I) which probably originates from the calcium hydroxide bound to the polymer, as suggested in literature [26]. Probably there is a similar effect in series C and D, but it does not appear from DTA curves. Indeed, high amounts of portlandite, comparable to those found in the H series, are present also in C and D samples, at 1 and 7 days of curing (Fig. 4). On the contrary amounts in 28-day-old samples are more or less the same for all series.

In all the samples the carbonation was not negligible; indeed, the amount of calcite ranged from 9.0 to 16.7 g of calcite per 100 g of dried sample (Fig. 4), but about 1.3 g came from the clinker. The carbonation was consistently larger in polluted samples (from 10.4 to 16.7 g per grams of dried sample) with respect to the corresponding monolith without 2-CA (from 9 to 15 per grams of dried sample); probably the presence of the pollutant may favour the carbonation of the polluted samples.

3.3. XRD analysis

Fig. 6 shows a typical XRD powder pattern for the 28-day-old paste of series Hp. Tricalcium silicate, β -dicalcium silicate, portlandite, calcite, small amounts of dolomite and quartz, modified montmorillonite and amorphous hydrated calcium silicate (C–S–H) were found in pastes of series A–D and H, respectively. The patterns of 7-day-old pastes are similar to the 28-day-old pastes.

In contaminated pastes carbonate phases, mainly $\text{Ca}_4\text{Al}_2\text{O}_6(\text{CO}_3)0.5(\text{OH})\cdot 11.5\text{H}_2\text{O}$ (PDF # 410221) were found as products of the hydration reactions. These carbonate phases might have been present in very small quantities also in uncontaminated samples, but due to the overlapping of the strongest peaks in the range $10^\circ < 2\theta < 12^\circ$, their identification was difficult. Crystalline ettringite was not detected as a consequence of the drying treatment performed to stop hydration reactions after curing, which might have damaged the ettringite crystals.

The OPC hydration was roughly estimated from the XRD patterns, by measuring the integrated intensity of the portlandite peaks at $d = 4.89$, 2.62 and 1.79 Å, respectively. The comparison of the integrated intensities of portlandite peaks highlighted that the lower water-to-cement ratio due to the addition of the superplasticizer, led to some delay in the hydration kinetics, while the other

series showed similar behaviour. No significant differences were found between contaminated and uncontaminated samples of a same series except for series C.

3.4. SEM

SEM images were used to characterise all the samples and allowed to identify their microstructure. Large organoclay grains, with a diameter of about 30–50 μm , were found in the matrix, as shown in Fig. 7, in all the samples. It was noted that the organoclay grains were not strongly bound to the cement matrix, thus causing a weakness zone around the site of incorporation of the grain inside the cement paste.

Furthermore, large cavities were observed in the samples containing the rubber latex (C and Cp), as shown in Fig. 8. The inner part of these pores was characterised by the growth of euhedral crystals of portlandite; it was interesting to observe that the portlandite crystal size was much larger in the polluted sample (Cp) than in the uncontaminated (C).

Also in samples belonging to series H large cavities were detected as reported in Fig. 9. It was possible to identify euhedral crystals of portlandite and few needles of crystalline ettringite, not detected by XRD analysis.

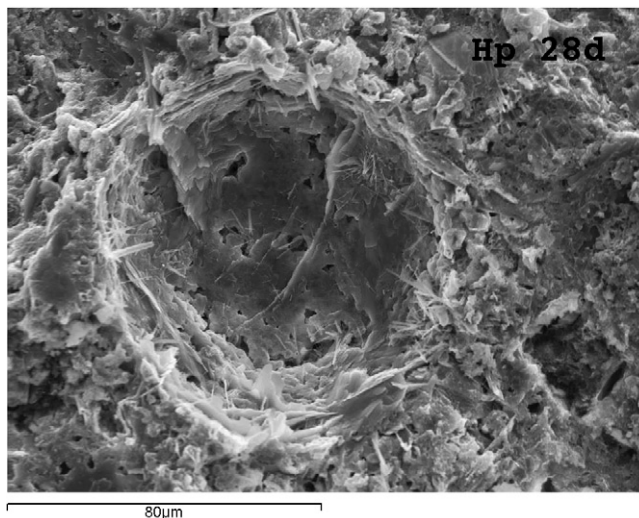
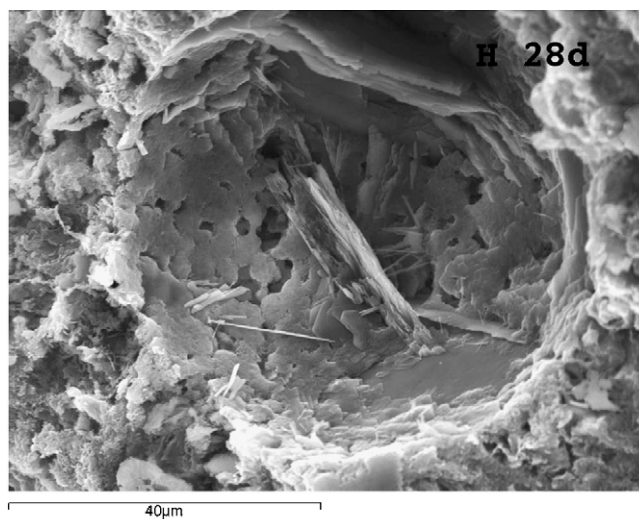
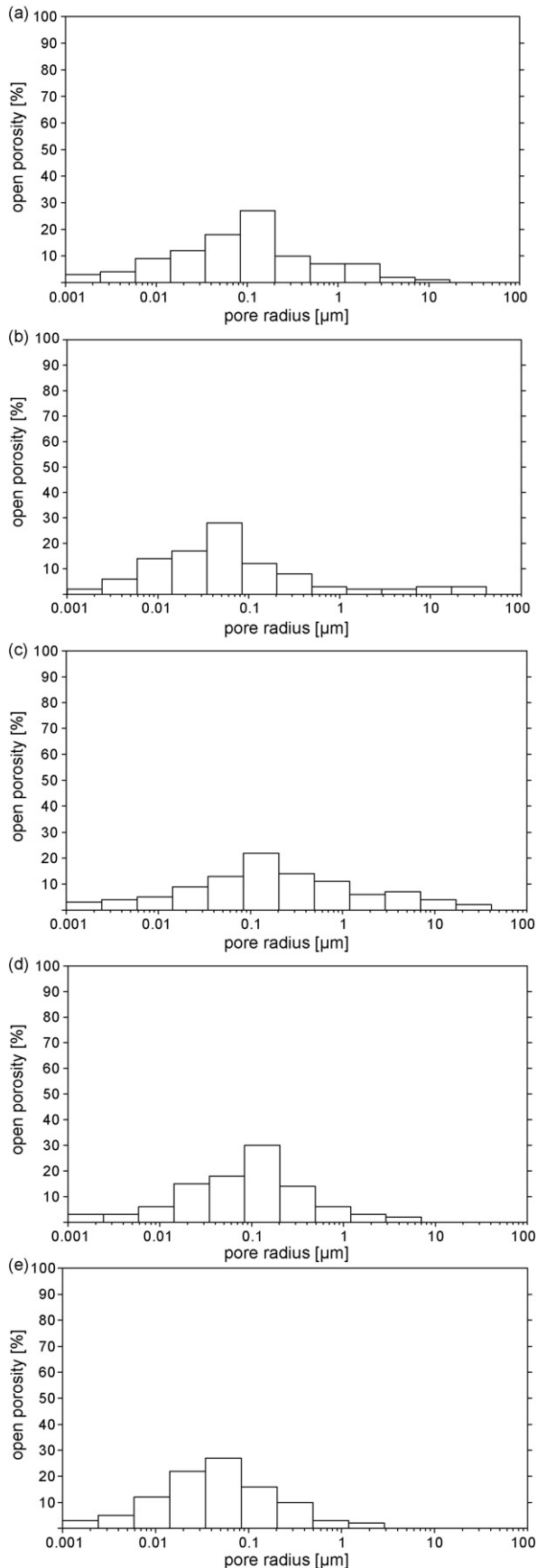


Fig. 9. SEM pictures, on fracture of sample H and Hp at 28 days, of large cavities where portlandite and ettringite were detected.



3.5. Porosimetric investigations

The investigated samples generally exhibit a great value of total porosity (32–45%) related to a high w/c ratio and to the presence of the clay. Fig. 10a shows the pore size distribution for the mixture Ap (cement + organoclay + 2-CA) used as a blank sample. It shows the highest value of open porosity (45%) with a modal class centered at 0.15 μm . Sample Bp having a lower w/c ratio than Ap, thanks to the superplasticizer, exhibits a total open porosity equal to 37% and a modal class lightly shifted to smaller pore sizes (Fig. 10b). The mixtures Cp and Dp do not show significant differences in the pore sizes distribution; their open porosity is 40 and 42%, respectively (Fig. 10c and d).

The Hp system (Fig. 10e), with superplasticizer and rubber latex, gives the best results in the leaching test, exhibiting the lowest value of open porosity (32%) resulting from the mix-design adopted which permitted the w/c ratio to be reduced.

Therefore, the use of superplasticizer shows by itself to be suitable to producing significant improvements of the microstructure of the cement matrices. Although the samples maintain high porosity values, their pore size distribution shifts towards extremely small pore size classes whenever the w/c ratio is reduced.

There is a close relationship between the porous microstructure of a cement paste and its permeability: the permeability of a cement paste mainly depends on the large capillary pores. The permeability test, performed on A and Bp samples, gave values lower than 0.001 mD, corresponding to “impervious permeability” [29], suggesting a highly not interconnected porosity.

Moreover, it is known that permeability to water may not be detectable for pores having a diameter lower than 100 nm [30]. It must be considered, also, that a part of the porosity in the samples might not be interconnected, thus not influencing the permeability. Last, it has to be considered that permeability tests were performed on samples after the 28-day curing, but before starting the leaching process. It is well known that the leaching process changes the microstructure of the monoliths [31]; as a consequence, it also alters the pore microstructure and probably the permeability.

The low permeability values, measured before starting the leaching test, explains the good leaching behaviour in the first 8 h of all the samples, no matter if and what admixture was used. Indeed, the microstructure of the samples, probably, is not yet much altered by the action of water during the first 8 h. On the contrary, after 1 day leaching, the DLT curves of the different samples spread apart (Fig. 3): the ones showing higher porosity, due to higher w/c ratios, probably undergo more important microstructural modifications than the ones having lower w/c ratio (and porosity); so justifying the leaching behaviour observed.

4. Conclusions

The addition of the admixtures did not affect the phase composition of the hydration products, as confirmed by XRD analysis. However, DTA revealed the presence of an interaction between portlandite and the latex in the series H, which alters the normal dehydration behaviour of portlandite.

Moreover, the admixtures seem to enhance the $\text{Ca}(\text{OH})_2$ formation in the first week. As a consequence, a more severe carbonation is observed after 28 days in series C, D and H.

Fig. 10. Pore size distribution of contaminated samples: (a) Ap* used as a reference system, open porosity = 45%; (b) Bp* system with superplasticizer, open porosity = 37%; (c) Cp* system with rubber latex, open porosity = 40%; (d) Dp* system with waterproofing agent, open porosity = 42%; (e) Hp* with rubber latex superplasticizer, open porosity = 32%.

The leaching test pointed out that the addition of the admixtures improves the leaching behaviour of the solidified monoliths, if compared with the reference sample (A). Indeed, primary importance in determining the results of leaching test is covered by the w/c ratio: the lower the w/c ratio, the lower the porosity and consequently the amount of organic pollutant released by the cement matrix. Anyway, the amount of 2-CA released is quite low for all the series; indeed, the permeability to water resulted very low both in reference and polluted samples, thus suggesting a non-interconnected porosity.

Although the pre-adsorption process improves the leaching behaviour, the gain is probably not significant enough to justify a time-consuming step (24 h vs 15 min) from a technological point of view.

The combined use of admixtures, i.e. superplasticizers and latex, is even more encouraging because it reduces the release by about 10% with respect to the reference sample (Ap).

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