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MSWI ashes as mineral additions in concrete

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Abstract

The paper describes the results of a research aimed at studying the effect of replacing part of portland cement with fly ash and bottom ash, both from municipal solid waste incinerators (MSWIs). Fly ash was subjected to a washing treatment to reduce the chloride content, while bottom ash was subjected to dry or wet grinding underwater. Concretes with addition of different types of ashes, including a traditional coal fly ash (FA), were manufactured. Fresh and hardened properties of the concretes were compared in order to study the advantages and the side effects of each type of addition. Results showed that MSWI bottom ash is potentially attractive as mineral addition for the production of concrete, provided that the risk of entrapment of hydrogen bubbles produced by corrosion of aluminium metallic particles in the fresh concrete is prevented. This could be achieved by wet grinding the bottom ash so that reactions leading to gas development exhaust within the slurry before this is added to the concrete mixture. However, by considering bottom ashes from different incinerators, a great variability was observed in the time required to complete the hydrogen gas production. Nevertheless, when the hydrogen development in the fresh concrete could be avoided, wet ground MSWI bottom showed a good pozzolanic behavior and proved to give a significant contribution to the development of the strength and impermeability of concrete.

Keywords: MSWI ashes; Blended cement; Pozzolan; Waste management

1. Introduction

The recycling of industrial wastes in the concrete manufacturing is of increasing interest worldwide, due to the high environmental impact of the cement and concrete industries and to the rising demand of infrastructures, both in industrialised and developing countries. Indeed, concrete technology can use great amounts of industrial residues as secondary raw materials [1–5]. For instance, blended cements are normally produced by adding to Portland cement clinker different quantities of coal fly ash (FA), silica fume (SF) or ground granulated blast furnace slag (GGBS). These mineral additions, that have been initially simply used to recycle wastes, turned out to be beneficial with regards to the properties of concrete, especially in relation to its resistance to aggressive species [6,7]. Furthermore, by actively replacing part of the clinker, these additions also allow to save energy and raw materials for cement manufacturing and reduce atmospheric pollution [1].

Ashes from incineration of municipal solid wastes (MSW) have some analogy with the abovementioned traditional mineral additions. Incineration of MSW allows a great reduction in the quantity of the wastes of 65–80% in mass and 85–90% in volume. Nevertheless, several residues, with different characteristics of hazard, remain after the incineration [8]. They can be broadly divided into *bottom ashes* and *fly ashes* [the latter is about 1–3% of the total municipal solid waste incinerator (MSWI) residues]. MSWI FAs are fine and are normally characterized by a high content of chlorides (even higher than 10%) and significant amounts of dangerous substances (such as heavy metals or organic compounds). Frequently, they are treated in a cementitious, organic or vitreous matrix and then disposed of in landfills. MSWI bottom ashes have coarser dimensions (particles can reach several tens of millimeters in size), and the amount of chlorides and hazardous chemicals is usually much lower than that of MSWI FAs.

Several researches have studied the possibility of recycling fly and bottom ashes in the cement and concrete

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manufacturing, both as aggregates or mineral additions [9–18]. Industrial processes have been developed to use these residues as raw materials for the production of Portland cement clinker [9,10]. Bottom ashes have been used as aggregates in concrete, but expansive reactions, often related to the presence of metallic aluminium or aluminium compounds, have been reported to progressively damage the concrete [11–16]. Some treatments have been proposed to reduce expansive phenomena [11,12].

MSWI bottom ashes possess a chemical composition that is not dissimilar from that of FAs. In fact, they are mainly composed of amorphous silica (usually more than 50%), alumina, iron oxide and calcium oxide. This suggests that MSWI bottom ash, once finely grounded, could have pozzolanic or hydraulic behavior and its addition to the concrete mix could have a beneficial role in the development of the microstructure of the hydrated cement paste. Hence, a great advantage in the sustainability of the concrete industry would be achieved if ground MSWI bottom ashes could actually be used as mineral additions. In fact, residues such as MSWI bottom ashes, which are available in great quantities throughout the world, could be converted into a resource able to produce quality concretes.

Some researches have actually shown the pozzolanic activity of ground MSWI bottom ashes showing their reactivity with lime or Portland cement clinker [17,18]. Nevertheless, no successful use of MSWI bottom ashes as mineral addition in concrete has been reported, because of the side effects of this addition. The main side effect is related to the evolution of hydrogen gas after mixing due to the presence of metallic aluminium. In the alkaline environment produced by the hydration of Portland cement (pH around 13), corrosion of some metals (mainly aluminium) produces a great amount of gaseous hydrogen. After placing and compaction of concrete, this gas is entrapped in the fresh material, producing a network of bubbles that leads to significant reduction in the strength and increase in the permeability of the hardened concrete.

This paper summarizes the results of a research aimed at studying the use of MSWI FAs and bottom ashes as mineral additions for the production of structural concrete.

2. Experimental procedure

Bottom ashes and FAs from different municipal waste incinerators in Northern Italy were considered. In order to be suitable as mineral additions in concrete mixes, FAs were submitted to a washing treatment to reduce the chloride and heavy metal contents, while bottom ashes were ground in order to reduce their particle size. Washing of FA was carried out with distilled water, until chloride content lower than 0.4% with respect to the dry mass was obtained. Grinding of bottom ashes was carried out both under dry conditions in a ball mill and under wet conditions in a microsphere mill (a slurry of solid/water ratio of 1:1 was obtained).

A control concrete was cast with 440 kg/m^3 of Portland cement type CEM I 52.5R, according to European Standard EN 197-1, water/cement ratio of 0.50 and 1700 kg/m^3 of crushed limestone aggregate (maximum size=14 mm). In order to study the effect of MSWI FA and bottom ash on the properties of concrete in the fresh and hardened states, mixes with the same composition as the previous one were cast, with the exception of 30% replacement (by mass) of Portland cement by washed MSWI FA, dry ground MSWI bottom ash or wet ground MSWI bottom ash (added as slurry to the mix). In other words, all the concrete mixtures were manufactured at the same water/binder ratio (w/b=0.5). Two concrete mixtures with replacement of 30% by mass of cement with traditional coal FA and ground limestone were also cast. In some mixtures, an adequate amount of acrylic-type superplasticiser was added in order to improve the workability without changing the w/c.

Chemical compositions of the ashes, cement and other additions were determined with inductively coupled plasma (ICP) technique and were expressed as oxide percentage content. XRD analyses were also carried out on the ashes.

Workability of fresh concretes was measured by means of slump and VeBe tests. Compressive strength was measured on 100-mm cubes after various times of wet curing (at about 95% RH). The evolution in time of resistivity of wet concrete was monitored in specific prisms of $50 \times 50 \times 100$ mm by measuring the electrical conductance between two embedded stainless steel wires. An experimentally determined cell constant was used to convert conductance values into resistivity values.

To study the resistance to chloride penetration, 150-mm cubes were exposed to drying–wetting cycles with a 3.5% by mass sodium chloride solution. Only one surface of the cube was exposed to the chloride solution, because the other faces of the cube were covered by an epoxy coating, and chloride profiles were analysed after different times of exposure. Cores, 30 mm in diameter, were taken from the exposed surface and they were cut at depth increments of 5 or 10 mm. The chloride content was determined by potentiometric titration after grinding and digestion in nitric acid, and it was expressed as percentage of the dry mass of the sample.

The corrosion rate of aluminium in alkaline solutions was studied by weight loss tests on sheets of commercial grade aluminium immersed in solutions of pH ranging from 11 to 13.5.

3. Results and discussion

A first series of tests was carried out using MSWI fly and bottom ashes from the incineration plant of Udine (North-east of Italy). The oxide composition of these ashes is shown in Table 1. A chloride content of about 15% by mass was measured in the FA. After several washing treat-

Table 1
Chemical composition of cementitious materials and MSWI ashes (percentage of major elements, not including Cl, has been calculated in terms of oxides)

Oxide (%)	Cement CEM I 52.5R	FA	Udine MSWI fly ash	Udine MSWI bottom ash	Desio MSWI bottom ash
Al_2O_3	4.71	6.15	10.72	10.29	6.36
Na_2O	0.2	0.19	11.34	2.46	1.72
K_2O	0.84	0.19	6.94	0.71	0.4
SO_3	3.48	0.79	8.49	1.21	3.43
CaO	62.7	6.53	37.32	13.25	15.89
Fe_2O_3	1.93	4.49	2.6	14.17	6.53
MgO	1.99	1.7	3.3	2.02	1.99
MnO_2	0.07	0.05	0.05	0.06	0.16
P_2O_5	0.15	1.07	1.55	1.08	0.77
TiO_2	0.19	0.39	–	0.38	0.85
SiO_2	23.74	78.45	14.71	53.41	61.9

ments with distilled water, the chloride content was reduced to 0.4%.

For comparative purpose, Table 1 also shows the chemical composition of another MSWI bottom ash (Desio, in the suburbs of Milan), which will be examined later in the Additional tests and further research section. Fig. 1 shows the XRD analyses of these incinerator ashes.

Fig. 2 shows the particle size distribution of the Udine MSWI bottom ashes after dry and wet grinding. The size corresponding to 50% of passing (d_{50}) was around 15 μm after dry grinding and it decreased to about 3 μm after the wet grinding.

Table 2 shows some properties of the concretes obtained with different mineral additions, including the MSWI bottom ash from Udine. The control concrete, with 440 kg/m^3 of cement and w/c ratio of 0.5 had a slump of 60 mm, a density around 2400 kg/m^3 and a 28-day compressive strength on cube of about 64 MPa. Similar values of workability and density were obtained with the concretes where 30% of cement was replaced with FA and ground limestone. The substitution of cement with 30% of limestone (i.e., an inert addition) led to an average 28-day cube strength of only 43.5 MPa, while the replacement of 30% of cement with FA (i.e., a traditional pozzolanic addition) led to 53.4 MPa (Table 2). Concrete, with the addition of MSWI FA, led to a remarkable reduction in the workability (no slump concrete with VeBe time of 13 s). Nevertheless, after

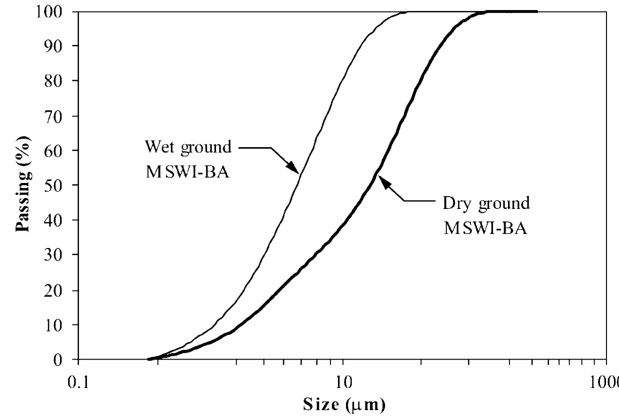

Fig. 2. Particle size distribution of Udine MSWI bottom ash after dry and wet grinding.

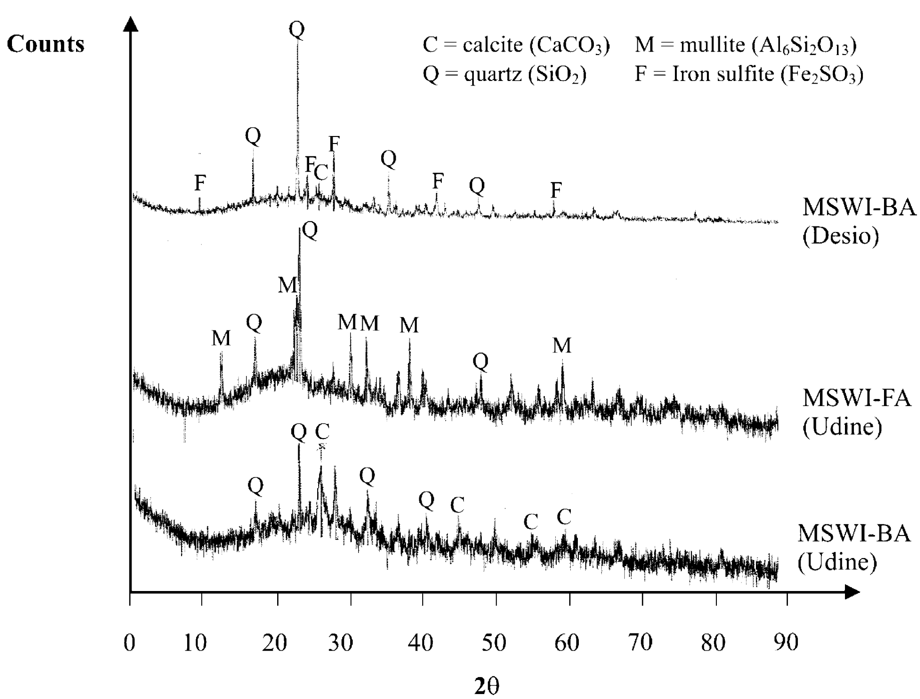

Fig. 1. XRD analysis of the MSWI ashes reported in Table 1.

Table 2
Properties of the fresh and hardened concretes with Udine MSWI ashes

Concrete	Slump (mm)	VeBe (s)	Fresh density (kg/m^3)	28-day density (kg/m^3)	28-day compressive strength (MPa)
100% cement	60	–	2450	2440	63.5
30% coal FA	80	–	2415	2450	53.4
30% ground limestone	90	–	2450	2400	43.5
30% MSWI fly ash	0	13	2450	2450	51.7
30% Dry ground MSWI bottom ash	110	–	2400	2260	20.1
30% Wet ground MSWI bottom ash	0	22	2500	2530	64.5

compaction on a vibrating table, the concrete density approached that of the reference concrete. The 28-day strength was slightly lower than that obtained on concrete with coal FA, showing a certain hydraulic behavior of the MSWI FA.

The concrete with the addition of dry ground MSWI bottom ash did not show any workability problem (its slump was the highest) and its fresh density was comparable with that of the other concretes. Nevertheless, this concrete experienced a remarkable expansion during setting. Expansion was visible to the naked eye on the cube specimens after demoulding (Fig. 3). Because of such expansion, the (water-saturated) density of the hardened concrete decreased to 2260 kg/m^3 (Table 2).

Expansion during setting was due to the development of hydrogen gas, after casting of concrete, in the form of bubbles (several millimeters in diameter), as shown in Fig. 3b. Hydrogen is produced by the cathodic reaction of the electrochemical process of corrosion of some metals [20]. Although, from a thermodynamic point of view, several metals may promote the cathodic process of hydrogen evolution in alkaline solutions (as the fresh concrete pore phase is), due to kinetic reasons, the attention was focused on aluminium. The presence of metallic traces of aluminium in the MSWI bottom ashes

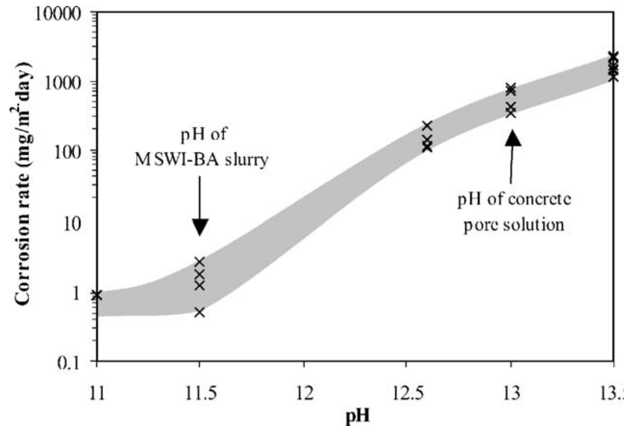

Fig. 4. Corrosion rate of commercial grade aluminium as function of pH, determined from weight loss tests at 20 °C.

was detected by X-ray diffraction analysis. Corrosion of aluminium in alkaline environments is described by the following processes [20]:

– anodic process: $Al + 2H_2O \rightarrow AlO_2^- + 4H^+ + 3e^-$
– cathodic process: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$

Fig. 4 shows the corrosion rate measured by weight loss on sheet of commercial aluminium for domestic use, as a function of pH. It can be observed that aluminium has a very high corrosion rate when it comes in contact with the solution of pH 13 which is formed due to the hydration reaction of portland cement. Under these conditions, aluminium is indeed able to produce a remarkable volume of hydrogen, which is entrapped within the concrete before setting takes place (Fig. 3b). Volumetric tests carried out by immersing a few grams of the MSWI bottom ash in a NaOH solution with pH 14 until the reaction was exhausted showed that 1 g of dry ground bottom ash could develop about 0.15 l of gas (at atmospheric pressure and 20 °C).

Surprisingly, no expansion was observed in the concrete in which 30% of the cement was replaced with wet ground MSWI bottom ash. In this case, a slurry with a solid/liquid ratio of 1:1 was produced by the wet grinding and it was

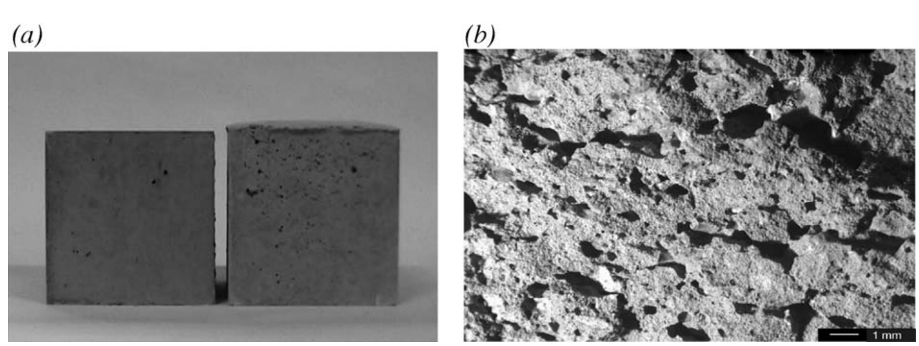

Fig. 3. (a) Comparison of the shape of the cubes after demoulding for the concrete with addition of MSWI bottom ash wet ground (left) and dry ground (right). (b) Example of entrapped voids on the fracture surface of the concrete made with dry ground MSWI bottom ash.

added to the mix (the water in the slurry was considered in the mixing water). Workability of this concrete was very low (Table 2), probably because of the higher fineness of the wet ground MSWI bottom ash (Fig. 2); nevertheless, after proper vibration, a density of 2500 kg/m^3 was achieved. Cube specimens did not expand (Fig. 3) and the density of the hardened concrete was the same as that measured in the fresh concrete (Table 2).

The wet grinding process played a primary role with regards to the absence of expansion. Since the beginning of the grinding process, the slurry showed a remarkable production of (hydrogen) gas bubbles. This can be again attributed to the corrosion process on the newly formed surface of aluminium particles embedded in the bottom ash particles, which are broken by grinding. The pH of the slurry, in fact, spontaneously reached a value around 11.5. This is enough to promote corrosion of aluminium, although at a lower rate than that promoted by the liquid phase of the hydrated cement paste (Fig. 4). Depending on the size of the aluminium particles after grinding, the actual corrosion rate of aluminium in the slurry and the time elapsed, the metallic aluminium particles can be depleted (or mostly depleted) before the slurry is added to the concrete mix. If this occurs, no further expansion can take place in the fresh concrete. Results obtained in this work showed that a few days of rest after grinding could be enough to exhaust the chemical reactions leading to gas evolution. In fact, the results previously described were obtained with a slurry used for concreting only 48 h after grinding.

As far as properties of hardened concrete are concerned, Table 2 shows that the 28-day compressive strength of concrete with 30% wet ground MSWI bottom ash was similar to that of the concrete with 100% Portland cement. It was even higher than that of the concrete with coal FA. This shows that wet ground MSWI bottom ash has hydraulic properties. The beneficial effect of MSWI bottom ash can be observed in Fig. 5 where the compressive strength of concretes is plotted against time. Even at longer curing, the concrete with wet ground MSWI bottom ash has a strength fairly higher than that of the concrete made with only

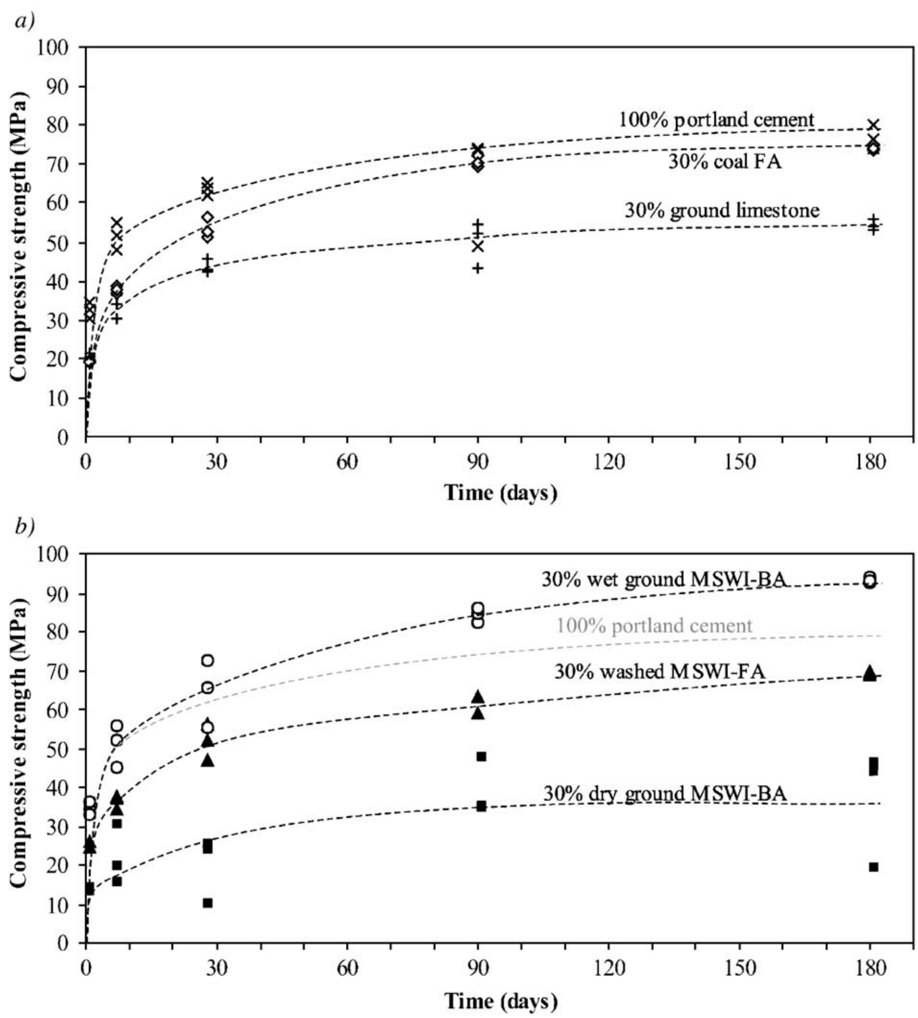

Fig. 5. Compressive strength as a function of time of reference concretes (a) and concretes with Udine MSWI ashes (b).

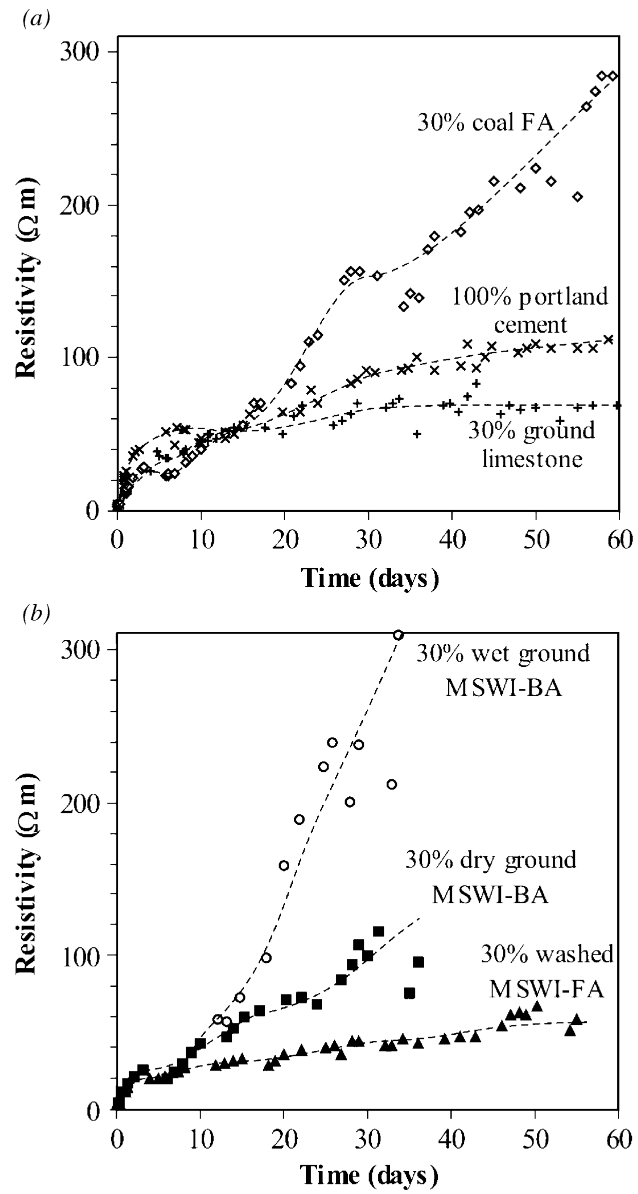

Fig. 6. Electrical resistivity of reference concretes (a) and concretes with Udine MSWI ashes (b) as a function of time after mixing in specimens exposed in the curing room.

Portland cement or with the use of FA. For instance, after 6 months of curing, the former showed a compressive strength slightly higher than 90 MPa, while the latter had a strength around 75 MPa.

The progressive hydration of cement pastes with addition of wet ground MSWI bottom ash could also be indirectly detected by means of electrical resistivity measurements. Fig. 6 shows the increase of the concrete resistivity as a function of time for different mixes, measured on wet specimens (exposed in the curing room at 23 °C). All the specimens show an increase in resistivity as a function of time, due to the microstructural changes in the hydrated cement paste and the consequent reduction in porosity of the cement paste.

Initially, all of the concretes showed the same trend; nevertheless, after about 20 days, the resistivity of the concrete with addition of FA showed a typical sharp increase that can be associated with the pore refinement induced by the pozzolanic reaction [19]. The same behavior could be observed in the concrete with addition of wet ground MSWI bottom ash, supporting the hypothesis that a pozzolanic reaction also occurred in this concrete. Even values of resistivity higher than those measured on FA concrete were reached (Fig. 6).

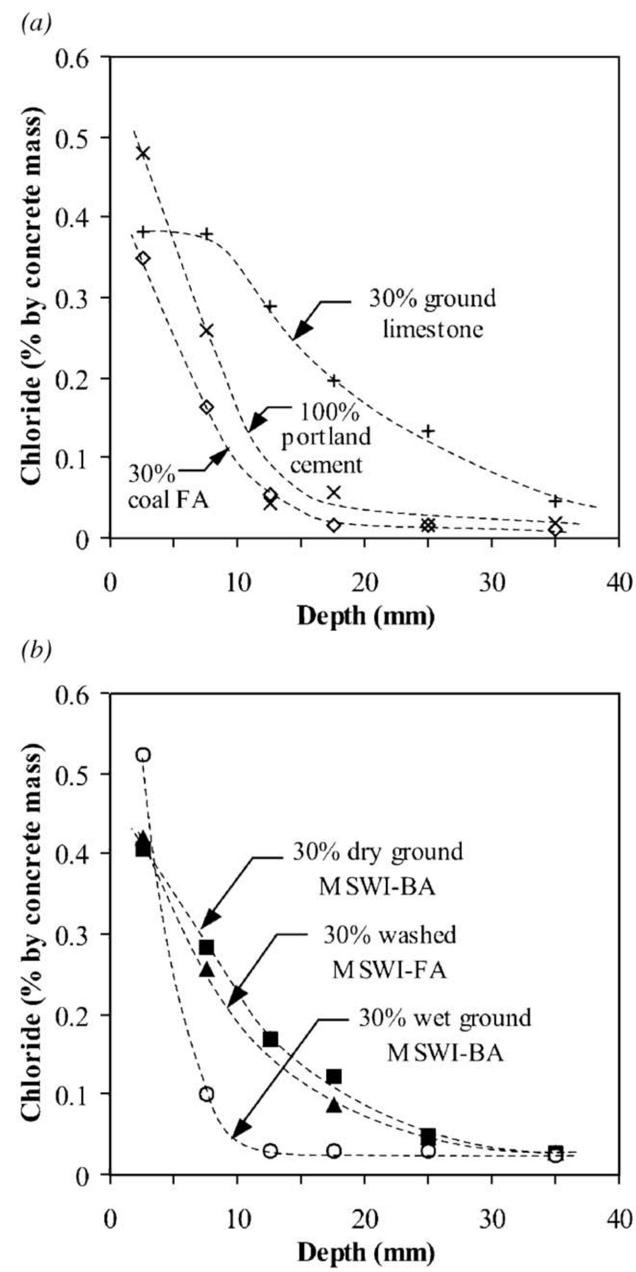

Fig. 7. Chloride profiles after 6 months of exposure to chloride cycles (1-day immersion in 3.5% NaCl solution and 2-day drying) of reference concretes (a) and concretes with Udine MSWI ashes (b).

The beneficial effect of hydration of MSWI bottom ashes could also be observed in relation to chloride penetration. Fig. 7 shows chloride profiles measured on cube specimens after 6 months of exposure to chloride wetting and drying cycles. The concrete with the addition of wet ground MSWI bottom ash, together with the concrete with the addition of PFA, showed the lowest penetration of chloride ions. This behavior can again be related to the well-known pore refinement and decrease in permeability produced by pozzolanic reaction, although some filler effect of finer particles cannot be excluded.

The results discussed above show that the ground MSWI bottom ash from Udine may act as a true cementitious material able to increase strength and durability of concrete, provided that the bottom ash is ground in the presence of water and a fineness comparable to that typical of cement is reached. Accordingly, bottom ash from MSWI treated by a wet grinding process can be advantageously used as an additional ingredient in the production of cement mixes with improved properties of strength, elastic modulus, resistance to aggressive agents and water penetration. The water used for wet grinding is introduced in the concrete mix (as part of the mixing water) and thus no residues are produced.

MSWI bottom ash is more attractive as a potential mineral addition for concrete than MSWI FA. The latter, in fact, during these tests, did not induce such remarkable improvement of the concrete properties and, furthermore, it requires onerous washing treatments to reduce the content of chlorides and hazardous species (with the consequent necessity of proper treatment and disposal of liquid residues).

4. Additional tests and further research

To confirm the results discussed above, further tests were carried out using both the same ashes and MSWI bottom ashes from other incinerators located in Northern Italy. All the tests made by using the same bottom ashes as before (i.e., from the same incinerator and the same batch) confirmed the previous results. It was also shown that, by simply adding a superplasticiser in appropriate dosage, it was possible to increase the workability to a slump higher than 200 mm without changing the properties of the hardened concrete. Even a concrete with self-compacting properties (slump flow of about 700 mm) could be obtained with suitable dosages of superplasticiser admixture and viscosity-modifying agent.

Subsequent tests with MSWI bottom ashes from other incineration plants gave much more erratic results with respect to the Udine bottom ash, as far as the effect of hydrogen evolution is concerned. For instance, no expansion was observed in a concrete made by using bottom ash from Desio incineration plant (whose composition is shown in Table 1) when this material was used 2 days after wet grinding. Measurements of strength and resistivity on this concrete had trends similar to those described above for the Udine bottom ash and thus confirming the pozzolanic behavior of MSWI bottom ashes after wet grinding. However, by using a second batch of bottom ashes from the Desio incinerator, expansion of concrete related to the hydrogen development occurred even after 3 months from the wet grinding. On the other hand, a third batch did not give any expansion 2 days after wet grinding. Similar variability was observed by using ashes from other incinerators. This shows a remarkable variability for the time required to deplete aluminium (or any other metals responsible of hydrogen evolution) contained in the slurry.

Further tests are being carried out in order to investigate the effect of possible factors that can influence the time for the exhaustion of the hydrogen evolution reaction within the slurry. The aim of these tests is to define possible treatments that can reduce this time to acceptable values (e.g., few days) regardless of the provenience and the batch of the bottom ashes. Factors being studied are as follows: (a) the fineness of MSWI bottom ash particles in the slurry produced by wet grinding; (b) the temperature of the slurry during grinding and the subsequent rest period (i.e., from the end of grinding to the time it is added to the concrete mix); (c) the stirring conditions of the slurry during the rest period; (d) the possible acceleration of the hydrogen evolution reaction in the slurry by increasing its pH; and (e) the reduction of nonferrous metals in the bottom ash before wet grinding. The understanding of the role of these factors could give the basis for the development of an industrial process that, by using the wet ground MSWI bottom ash to produce structural concrete, could give a remarkable contribution to the sustainability of the concrete industry.

Of course, a second important step towards a possible use of this type of concrete in real structures will be the evaluation of risks related to possible leaching of dangerous substances. Normally, MSWI bottom ashes have a negligible content of chlorides and heavy metals and these are expected to be bound in the dense cement matrix of a structural concrete. Nevertheless, any possible risk should be evaluated taking into account the exposure condition of the structure, its destination, the effect of degradation or ageing phenomena (e.g., carbonation, cracking), etc. Moreover, possible risks related to the demolition of the structures at the end of their service life should be considered. As a first step, traditional leaching tests are being carried out on alkaline and carbonated concretes in order to get a first picture of possible risks.

5. Conclusions

Tests described in this paper showed that bottom ashes from MSW incineration are potentially attractive as mineral additions for the production of concrete.

When MSWI bottom ashes were added to the concrete mix after being dry ground, strength and durability of concrete were negatively affected by entrapment of gas bubbles. This was the consequence of hydrogen produced by the cathodic reaction of corrosion of particles of aluminium (and possibly other nonferrous metals) that were contained in the bottom ashes. Conversely, wet grinding could avoid problems related to the evolution of hydrogen in the fresh concrete. In this case, the reactions leading to the development of gas began in the slurry produced by wet grinding and they continued afterwards until, after a certain time, they exhausted, so that the slurry could be safely added to the concrete mix. Nevertheless, a remarkable variability was observed for the time required to terminate these reactions. For some ashes, a couple of days were enough, while in same cases a much longer time, of the order of magnitude of months, was not sufficient to end the gas development. Further tests are being carried out to study the factors controlling this phenomenon.

When the negative effects of hydrogen evolution in the fresh concrete could be avoided, the wet ground MSWI bottom ashes showed to behave as a pozzolanic addition. Replacement of 30% of portland cement with this ash led to a remarkable improvement in the strength and to a significant reduction in permeability of concrete, even more than that obtained when the same quantity of FA was used.

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