Comparison of ground waste glass with other supplementary cementitious materials

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1. Introduction

Due to the rising demand of infrastructures in industrialised and developing Countries and to the high environmental impact of the cement and concrete industries, the recycling of industrial wastes in the concrete manufacturing is of increasing interest worldwide [1–8]. Of particular relevance are those materials that have pozzolanic properties. Used as a partial replacement of Portland cement clinker, waste materials with hydraulic or pozzolanic properties can contribute to the hydration of the cement paste and, after proper curing, may lead to beneficial refinement of the pore structure [9]. Consequently, these mineral additions delay the penetration of ionic species through the concrete, thus preventing effects of corrosion of embedded steel induced by chlorides or degradation of concrete due to sulphate ions [10].

Interest in recycling waste glass in the production of concrete has recently increased and a significant number of research works have been published showing advantages and side effects. Waste glass has been considered for the use as recycled aggregate for concrete [11–19]. As far as durability is concerned, it was suggested that, due to low absorption capacity, recycled glass aggregate is potentially able to improve resistance to freeze–thaw attack, drying shrinkage [15] and abrasion [19].

Since glass is essentially made by amorphous silica, it has some analogies with traditional pozzolanic materials and, when it is finely ground, it may be used also as a supplementary cementitious material [20–33]. Several studies have shown that, in this case, expansive disruptive action due to ASR is not observed [12,28,29,33] but, conversely, particles finer than 100 μm may even oppose to expansion of coarser glass particles [17]. Conversely, it was shown that the use of finely ground glass may contribute to the strength of mortar or concrete due to the pozzolanic reaction with lime produced by the hydration of Portland cement clinker [20–29]. Some works, which have also investigated possible effects on durability of concrete structures, suggested that, recycled ground glass leads to the refinement of the concrete pore structure (e.g. indirectly evaluated by sorptivity [31,32] or electrical resistivity [30,32] measurements), and delay in the penetration of aggressive ionic species [31–33].

These results encourage recycling of glass as a supplementary cementitious material. Nevertheless, in order to evaluate the real advantages of this addition in relation to the life cycle of reinforced concrete structures, the performance of ground glass should be compared to that of currently most used pozzolanic materials. Furthermore, long-term behaviour of cement materials incorporating glass powder should be investigated. Some researches in literature have characterized the durability properties of ground glass, used as replacement of cement, especially in relation to ASR risks and taking on account as a reference only one traditional mineral addition (typically fly ash or silica fume) [21–23,31,33]. Conversely, the other papers, which concern a broad comparison of glass powder

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with different traditional mineral additions, are limited to the study of pozzolanic reactivity and strength-related properties [20,24].

This paper describes the results of a study aimed at comparing the effects of ground waste glass and traditional types of pozzolanic materials (natural pozzolana, coal fly ash, silica fume) as well as an inert siliceous addition (quartz sand) on the strength and durability properties of mortars. A comparative evaluation is made on the basis of short-term and long-term compressive strength of cement and lime mortars and the resistance to chloride and sulphate ions penetration.

2. Experimental

Waste green bottles were crushed and ground in order to obtain powders with target specific surfaces of about 400 and 600 m^2/kg (respectively indicated with *G*4 and *G*6). Fig. 1 shows particles of *G*6 ground glass observed at the scanning electron microscope and X-ray EDS analysis of the particles.

2.1. Materials and characterization

For comparison, Portland cement (CE) and traditional mineral additions were also considered: a natural pozzolana from the centre of Italy (*PZ*), a silica fume (*SF*) and a coal fly ash (*FA*). Ground quartz sand (*QS*) was also studied, in order to consider an inert addition. Fig. 2 shows the X-ray diffraction patterns of the mineral additions. The fineness of mineral additions was evaluated by means of grading curves obtained by laser granulometry. The specific surface was then calculated from the grading curve. Fig. 3 shows the particle size distribution and the specific surface of the mineral additions and shows that fly ash, natural pozzolana and ground quartz were comparable in terms of fineness to the glass powder *G*4.

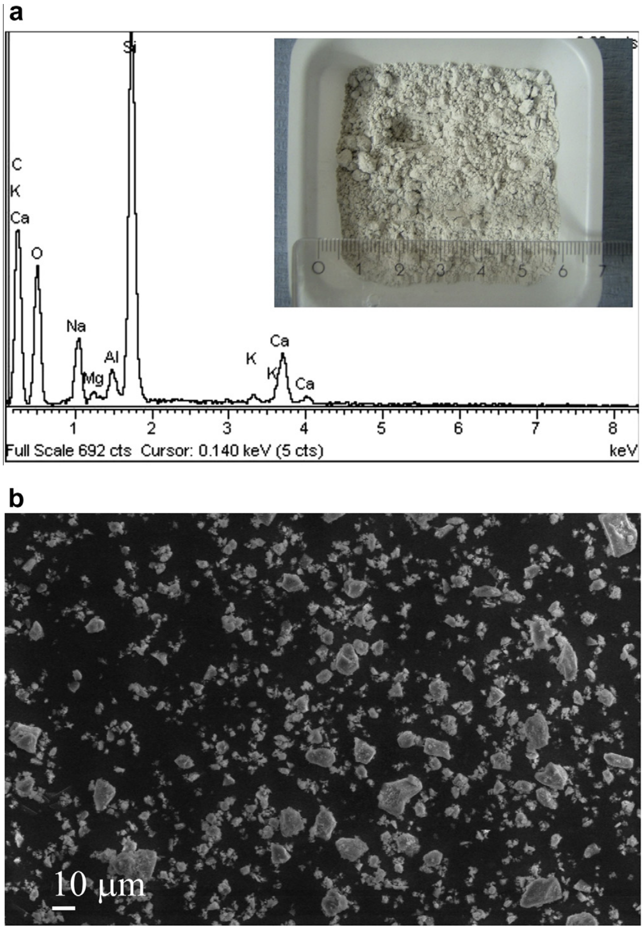

Fig. 1. EDS analysis (*a*) and SEM micrograph (*b*) of ground glass particles (*G*6).

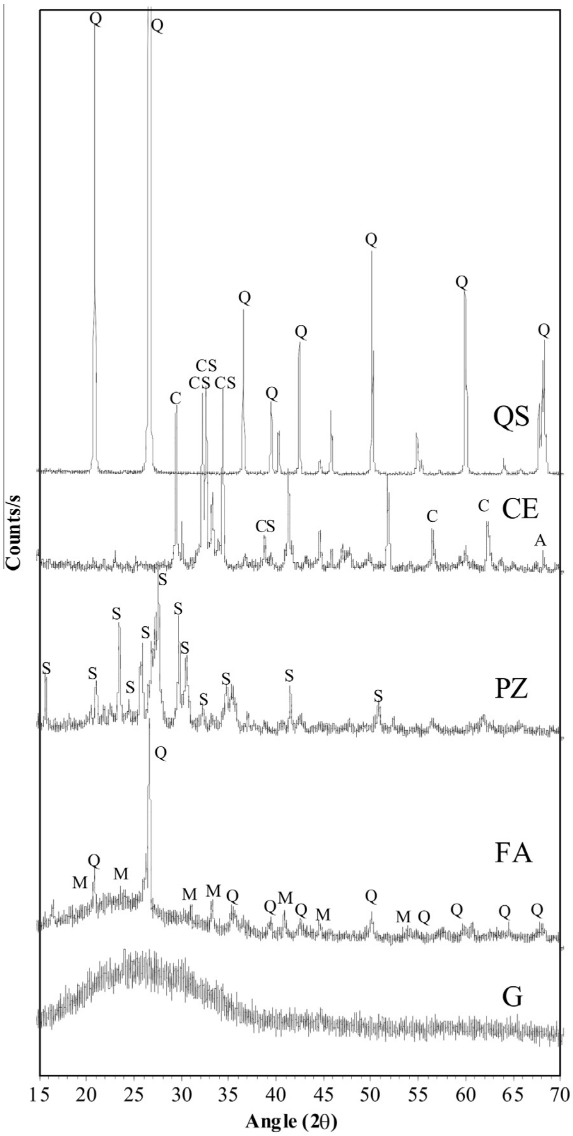

Fig. 2. X-ray diffraction pattern of mineral additions (M = mullite, Q = quartz, S = sanidine, C = calcite, CS = calcium silicate oxide and A = aluminium oxide).

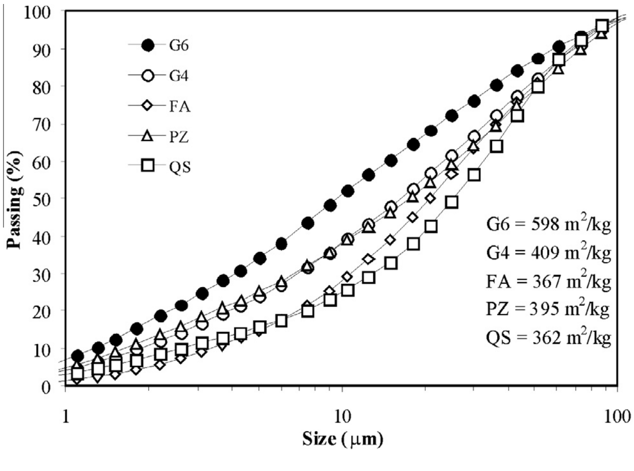

Fig. 3. Particle size distribution and specific surface of mineral additions.

Chemical composition of the glass powder and other mineral additions was studied by means of X-ray fluorescence spectrography (XRF) and results were expressed as percentage by mass of oxides.

The pozzolanicity test (also named Fratini test [34]) included in EN 196-5 European standard for pozzolanic cements was carried out. This test was performed on powder samples obtained by mixing 70% of a reference cement and 30% of each mineral addition (except for silica fume used in replacement of 10% of cement). Twenty grams of powder were immersed in an aqueous solution for 8 days and then the solution was vacuum filtered. The filtrate was analysed for hydroxyl ions (OH^-), by titration against diluted HCl with methyl orange indicator, and Ca^{2+} ions, by titration with EDTA solution after pH adjustment to 13.

2.2. Testing of mortar specimens

Compressive strength was measured on standard mortars made with glass powder and other mineral additions (in partial replacement of 30% of cement, except for silica fume used at 10%). Reference cement was a Portland cement type CEM I 52.5R (according to EN 197-1) identified by *OPC*. Lime-glass mortars were also cast by using a binder obtained by blending 70% of hydrated lime ($Ca(OH)_2$) and 30% of glass powder. For both cement-pozzolana mortars and lime-glass mortars, the water/binder ratio was 0.5 and sand/binder ratio of 3 was used.

Electrical resistivity of cement mortars was monitored during curing in water by measuring the electrical conductance between two external copper plates. An experimentally-determined cell constant was used to convert conductance values into resistivity values.

The resistance to the penetration of chloride ions of the cement–pozzolana mortars was studied using cylinder specimens with height of 60 mm and diameter of 100 mm, following the procedure proposed by NT Build 443 standard. After 28 days of wet curing, the side surface and one of the flat surfaces of the cylinders were masked with epoxy. Then the specimens were immersed for 35 days in a solution with 165 g/L of NaCl. Chloride profiles were measured on powder samples ground at different depths, which were digested in nitric acid and analysed by potentiometric titration with silver nitrate. Chloride profiles were interpolated with the “erf function” obtained from Fick’s second law:

$$C_x = C_s \quad 1 - \mathrm{erf}\,\frac{x}{2\sqrt{D_{\mathrm{app}}t}}\Bigg)$$

where: C_x = chloride content (% by mass of cement) at time t (s) and depth x (m); D_{app} = apparent diffusion coefficient (m^2/s); C_s = surface content (%). Results obtained from different specimens of the same mortar where fitted together to calculate values of D_{app}.

Expansion due to sulphate attack was evaluated on mortar prism specimens ($25 \times 25 \times 280\ mm^3$) immersed in 5% Na_2SO_4 according to ASTM C1012 standard. The length was monitored with a precision of 1 μm throughout the immersion period, in order to detect the expansion in time of the specimens.

3. Results and discussion

Results of tests on ground glass and other mineral additions are summarized in Table 1, which shows the chemical composition, the results of Fratini test and average values of properties measured on mortar specimens at various curing times (more details can be found in Bertolini et al. [35]). In order to compare the relative performance of the mineral additions, Table 2 reports the value of each property as a percentage with respect to the value of the same property measured on the reference mortar (*OPC*). Relative performances of ground glass powders (gray shadowed) and other mineral additions are ranked from the worst (on the left) to the best performance (on the right), whilst the relative position of the reference Portland cement mortar (*OPC*) is reported in bold.

3.1. Pozzolanic reactivity

Table 1 shows the chemical composition of the mineral additions. The prerequisites for a mineral addition to have a pozzo-

Table 1
Results of tests.

Test	*Measured property*	*PZ*	*FA*	*SF*	*G4*	*G6*	*QS*	*OPC*
XRF	SiO_2 (%)	53.75	56.18	90.83	68.67		96.65	19.76
	Al_2O_3 (%)	20.68	27.45	1.59	2.73		0.21	5.19
	Fe_2O_3 (%)	5.84	2.62	0.01	0.66		0.86	2.65
	CaO (%)	4.19	7.4	3.7	11.35		1.08	62.19
	MgO (%)	1.2	1.13	0.43	1.41		0.15	1.7
	SO_3 (%)	0.18	0.48	0.53	0.13		-	3.11
	K_2O (%)	6.63	2.43	0.64	1.11		0.03	0.93
	Na_2O (%)	3.55	0.73	0.11	13.25		0.03	0.31
Fratini	mmol/L CaO	4.62	4.53	11.12	2.53	1.48	10.97	-
	mmol/L OH	68.06	66.78	45.18	85.43	89.91	63.89	-
	Index	0.70	0.67	0.96	0.51	0.32	1.54	-
Compressive strength (standard mortars)	$f_{c,7days}$ (MPa)	42.7	40.6	46.2	41	40.4	43.3	57.1
	$f_{c,28days}$ (MPa)	52.4	49.7	60.2	53	57	49.9	65.9
	$f_{c,90days}$ (MPa)	55.2	62.1	76	68.7	70.7	50.3	73.2
	$f_{c,180days}$ (MPa)	61.6	65.9	76.3	73.2	77.4	52.3	71.7
	$f_{c,7\ years}$ (MPa)	74.7	86.1	96.1	83.9	84.2	70.8	85.3
Resistivity	$\rho_{,7days}$ (Ω·m)	16	25	53	34	40	22	28
	$\rho_{,28days}$ (Ω·m)	42	74	250	86	128	31	42
	$\rho_{,90days}$ (Ω·m)	78	200	379	276	281	36	47
	$\rho_{,180days}$ (Ω·m)	107	345	339	417	392	44	55
Chloride penetration	$D_{,app,28days}$ ($10^{-12}\ m^2/s$)	5.13	1.10	0.25	1.83	1.56	19.71	3.67
Sulphate expansion	Expansion (%) after two months	0.009	0.011	0.005	0.007	0.002	0.1	0.017
	Time to reach 0.1% expansion $^{(*)}$ (days)	388	560	$600^{(0.003\%)}$	$472^{(0.04\%)}$	$472^{(0.04\%)}$	75	250

(*) Duration of tests and maximum expansion (within brackets) is indicated for mortars that did not reach the threshold value.

Table 2
Comparison of performances relative to OPC (%) for mortars with ground glass and other mineral additions

Property		**Relative performance** ← **worst**						**best** →
Compressive strength	7 days	G6 71	FA 71	G4 72	PZ 75	QS 76	SF 81	**OPC** **100**
	28 days	FA 75	QS 76	PZ 80	G4 80	G6 86	SF 91	**OPC** **100**
	3 months	QS 69	PZ 75	FA 85	G4 94	G6 97	**OPC** **100**	SF 104
	6 months	QS 72	PZ 85	FA 92	**OPC** **100**	G4 102	SF 106	G6 108
	7 years	QS 83	PZ 88	G4 98	G6 99	**OPC** **100**	FA 101	SF 113
Electrical resistivity	7 days	PZ 57	QS 78	FA 90	**OPC** **100**	G4 121	G6 141	SF 186
	28 days	QS 73	**OPC** **100**	PZ 101	FA 179	G4 206	G6 308	SF 601
	3 months	QS 77	**OPC** **100**	PZ 167	FA 426	G4 589	G6 600	SF 807
	6 months	QS 80	**OPC** **100**	PZ 194	SF 596	FA 627	G6 713	G4 758
Chloride penetration (D_{app})		QS 538	PZ 140	**OPC** **100**	G-4 50	G-6 42	FA 30	SF 7
Sulphate attack	$t_{0.1\%}$	QS 30	**OPC** **100**	PZ 155	G4 >188,8	G6 >188,8	FA 224	SF >240
	$\Delta L_{2months}$	QS 468	**OPC** **100**	FA 65	PZ 53	G4 41	SF 28	G6 13

lanic behaviour are the presence of amorphous silico-aluminates compounds [9]. Although glass has a significant presence of silico-aluminates, Fig. 4a shows that the overall content of SiO_2, Al_2O_3 and Fe_2O_3 is lower than that of the others mineral additions. Nevertheless, glass differs from the other additions in that it has a high alkali content (Table 1) and it is fully amorphous (Fig. 2). To study pozzolanic activity, the traditional Fratini test, which is aimed at evaluating the pozzolanic behaviour of cement–pozzolana blends [34], was carried out. According to this test, the higher is the quantity of lime consumed by the pozzolanic reaction the lower is the concentration of hydroxyl ions (OH^-) found in the testing solution. Table 1 reports the results of Fratini test in terms of amount of lime (CaO) and hydroxyl ions (OH^-). Ground glass consumed a higher quantity of lime than other pozzolanic additions; in fact, the amount of lime (CaO) found in the solutions was only 1.48–2.53 mmol/L, depending on the fineness.

These results can be compared with the saturation curve for Portland cement, proposed by EN 196-5 standard, to calculate an index that describes the pozzolanic reactivity (the lower is the index below 1 and the higher is the expected reactivity). A Fratini index higher than 1 was calculated only for ground sand quartz, confirming its inert nature (Table 1).

Fig. 4*b* compares Fratini index of the different mineral additions. Results have been ordered from the highest value (corresponding to the worst performance) to the lowest value (best performance). It can be observed that ground glass showed the best performance, corresponding to the lowest values, regardless of the fineness of grinding (*G4* and *G6*).

3.2. Strength

To study the role of ground glass and other additions on the strength of mortar, compressive tests were carried out at early and long curing (Table 1). The reference mortar had 28-day compressive strength of about 66 MPa, while the substitution of cement with 30% of waste glass powder led to a strength of 53–57 MPa (depending on the fineness of the addition). The compressing strength of mortars with glass reached at later age high values, comparable or even higher than those of the reference mortar and mortar with fly ash.

Table 2 shows the strength activity calculated considering compressive strength measured after 7, 28, 90, 180 days and 7 years of curing of mortars cast with mineral additions used as replacement of 30% by mass of cement with the only exception of silica fume (10% by mass).

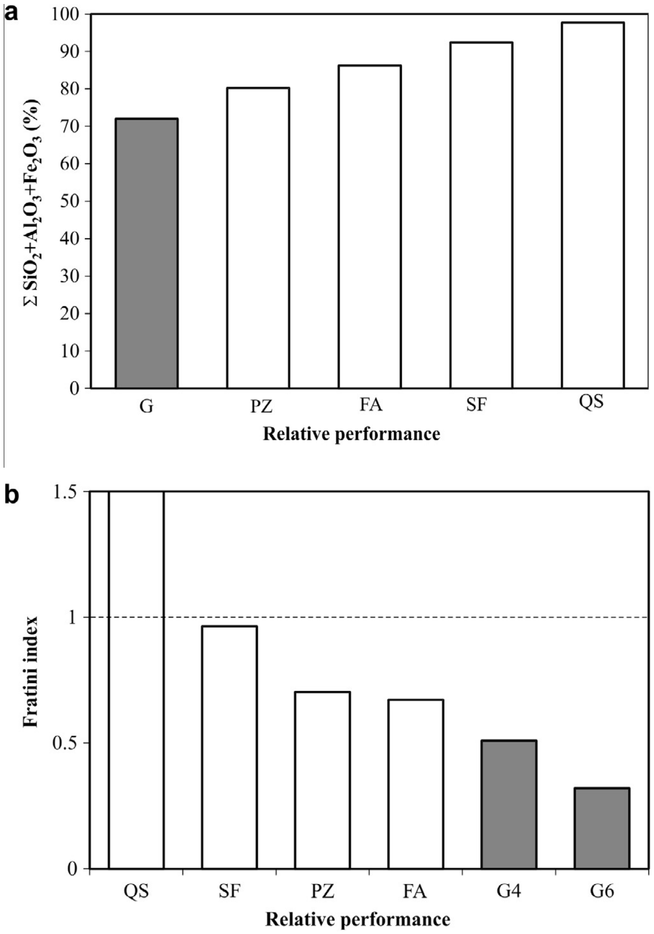

Fig. 4. Relative performance of ground glass compared to other mineral additions in terms of SiO_2, Al_2O_3 and Fe_2O_3 contents and Fratini index.

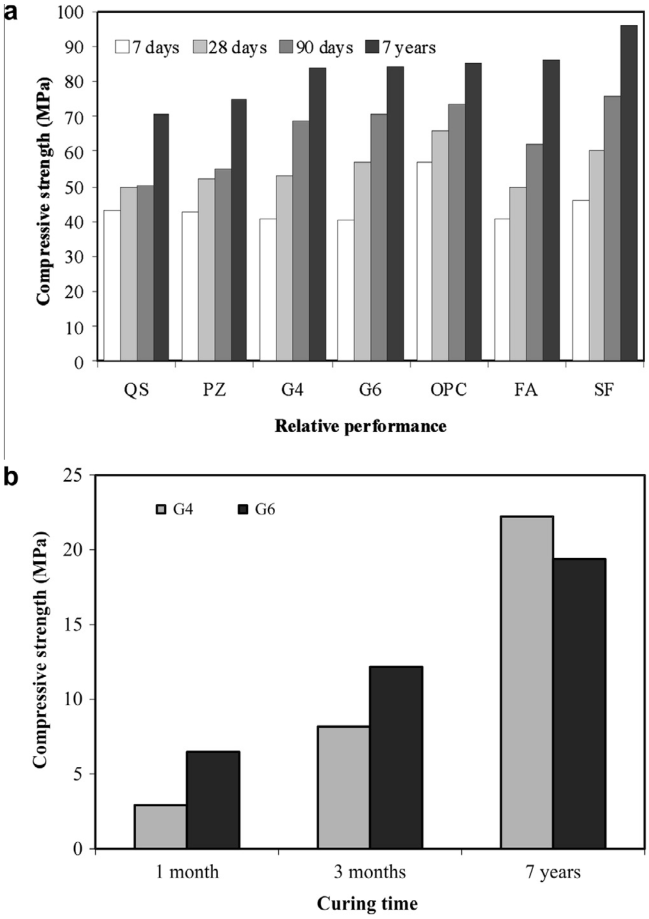

Fig. 5. Compressive strength measured after 7, 28 and 90 days and 7 years of curing on cement mortars with different mineral additions (*a*) and compressive strength of lime-glass mortars as a function of time (*b*).

All additions show a comparable behaviour in the first 7 days of wet curing. After 28 days of curing, values slightly higher than 90% were reached by the mortar with 10% of silica fume (*SF*), according to the well known high reactivity as well as possible filler effect of this extremely fine addition [9,36–37]. Mortars with ground glass (Table 2) had higher values (respectively equal to 80% for *G*4 and 86% for *G*6) than those of mortars with natural pozzolana and fly ash (whose values were in the range 75–79.5%). Only a slight influence of the fineness of glass powder on mortar performance could be observed at early ages, whilst at longer curing time a positive role of a higher fineness could be observed. While mortars with natural pozzolana showed only a slight increase in strength, a prolonged curing allowed a remarkable increase in mortars with ground glass, which reached values of the strength activity index higher than 100%. In particular, a good behaviour could be observed for the mortars with glass powder more finely ground (*G*6) that showed a index of 108% at 6 months of curing, even slightly higher compared to that obtained on the mortar with 10% silica fume (Table 2).

In order to study the long-term effect of the pozzolanic reaction, compressive strength tests were carried out also after 7 years of wet curing. Fig. 5*a* compares short-term and long-term compressive resistance of the different mortars studied; it shows that mortars with ground glass *G*4 and *G*6 reached a 7-year compressive strength of about 80 MPa, which was comparable to that of *OPC* and *FA* mortars.

The effect of ground glass on the compressive strength was also evaluated on lime mortars which were cast by using a binder obtained by blending 70% of slaked lime ($Ca(OH)_2$) and 30% of glass powder. Fig. 5*b* shows that the slow pozzolanic reaction of glass with lime determined an increase in strength even after 7 years of wet curing, when a strength value of about 20 MPa was reached, although the role of fineness of glass powder became thus less significant with the progress of curing.

3.3. Durability

The increase in compressive strength observed until 7 years of curing in cement- and lime-based mortars with the addition of ground glass may be attributed to the decrease in capillary porosity brought about by the pozzolanic reaction. This result suggests that beneficial effects of the pozzolanic activity of ground glass are maintained in time. In the meantime, it shows that no deleterious effects due to possible adverse reactions of glass particles that might take place under wet exposure conditions (e.g. alkali silica reaction) occurred during 7 years of immersion, confirming the results of short-term ASR tests described by other authors in previous works [17,28,29,33].

The variation in time of electrical resistivity of mortar specimens immersed in water may help in understanding the evolution of the microstructure of the hydrated cement paste and the consequent reduction in permeability. Changes in the chemical composition of the pore solution of concrete in the presence of mineral additions [10] and especially in the presence of glass powder (due to the high content of alkali) may affect resistivity measurements. In spite of this, however, the evolution in time of electrical resistivity of mortars during hydration may be used as an indirect measure of the combined effect of the refinement of the pore structure and the ability of the hydrated cement paste to oppose to the penetration of aggressive ions [10,30,38–40].

Table 1 shows that specimens of the reference mortar (*OPC*) and those of mortars with the inert addition (*QS*) reached values of electrical resistivity respectively around 55 Ω m and 44 Ω m after six months of curing. Conversely, mortars with silica fume, ground glass and fly ash, reached values exceeding 300 Ω m. Results of mortars made with natural pozzolana were in intermediate position. The relative performance of each addition is reported in Table 2 as the percentage ratio between the electrical resistivity of mortar with each mineral addition and the electrical resistivity of the reference mortar. At early age (Table 2) only finely ground glass *G*6 and silica fume showed values higher than *OPC*. With the progress of curing all mortars with pozzolanic additions remarkably improved their performance. In particular, mortars with ground glass reached relative values greater than 700% in 6 months of curing (Table 2).

To assess the beneficial effects of hydration of glass powder in relation to durability of reinforced concrete structures, resistance to chloride penetration and sulphate attack were considered. An apparent diffusion coefficient (D_{app}) was calculated by fitting chloride profiles obtained with ponding tests on mortar specimens exposed to a solution of 165 g/L NaCl for 35 days (Table 1). Low values of D_{app} were obtained for mortars with fly ash (*FA*), ground glass (*G*4 and *G*6) and silica fume (*SF*), in the range 0.25–1.8×10^{-12} m^2/s.

The relative performance of mortars in relation to penetration of chlorides is evaluated in Table 2 as the ratio between D_{app} of mortars with mineral additions and D_{app} of the reference mortar. The worst performance was measured for mortars with ground quartz sand (D_{app} was more than five times higher than that of the reference mortar). Also the mortar with natural pozzolana had a diffusion coefficient higher than the reference mortar. Ground glass mortars had a relative performance only slightly lower than that of mortars with fly ash, showing that this addition may significantly improve the resistance of concrete to chloride penetration. The highest resistance to chloride penetration was shown by mortars with 10% silica fume.

A good performance of ground glass was observed also in relation to the resistance to sulphate attack. Table 1 shows the expansion after two months of in concentrated sodium sulphate solution and the time required to produce expansion of 0.1% (which is often assumed as a limit value for blended cements, for instance in ASTM standards C595 or C1157). Mortars with ground quartz sand reached an expansion of 0.1% just after two months of tests and the reference mortar (*OPC*) after about eight months. Much longer times were required by mortars with pozzolanic additions to reach 0.1% expansion. Mortars with ground glass showed a negligible expansion (0.04%) even after more than 1 year of immersion in the sulphate solution (when tests were interrupted). Only the mortar with silica fume showed lower values. Table 2 compares the behaviour of different mineral additions by considering the time required to archive the limit value of expansion of 0.1% relative to that of the reference mortar ($t_{0.1\%}$). The worst behaviour was shown by the mortar with ground quartz sand that reached the limit after only about 30% of time compared to the reference mortar (*OPC*). All pozzolanic additions showed a higher resistance to sulphate penetration with values of the parameter much higher than 100%; this parameter could not be calculated for mortars with ground glass and silica fume, because the threshold value was not reached even after one year when the exposure was interrupted. In order to calculate a relative performance index related to sulphate attack, the expansion value reached by different mortars after 2 months of testing (i.e. the time taken by the mortar with ground quartz sand to achieve an expansion of 0.1%) is also reported in Table 2 ($\Delta L_{2months}$). It can be observed that lower expansion was measured after two months on mortars with ground glass and silica fume compared to the other pozzolanic additions (fly ash and natural pozzolana). Indeed, the expansion was negligible even after more than 1 year of immersion in the sulphate solution both on mortar with glass powder (0.04%) and silica fume (0.003%).

4. Conclusions

The pozzolanic behaviour of waste glass powders with specific surface of 400 m^2/kg and 600 m^2/kg was studied and compared to that of a natural pozzolana, coal fly ash and silica fume.

Ground glass showed the best reactivity by means of the Fratini pozzolanicity test. Blended in percentage of 30% with both Portland cement and lime, it showed to improve strength of mortars. The long-term strength activity index measured on cement-based mortars with ground glass was comparable to that of mortars with fly ash and higher than that of mortars with natural pozzolana.

Mortars with ground glass immersed in water for seven years did not show any sign of degradation and, conversely, increased their compressive strength. Ground glass could improve the resistance to chloride penetration and sulphate attack of mortars more than natural pozzolana and similarly to fly ash.

Although the increase in fineness of glass powder improved strength and durability performances of mortar, the ranking of ground glass with respect to the other mineral additions was not affected by the range of fineness studied in this work.

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