

Innovative hydraulic lime-based finishes with unconventional aggregates and TiO₂ for the improvement of indoor air quality

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Abstract. This paper reports a study on 8 unconventional hydraulic lime-based mortars able to improve indoor air quality by acting as passive systems. Mortars have been prepared with commercial sand or highly adsorbent materials as aggregates with/without TiO₂ as photocatalytic agent, to test also the decomposition of airborne pollutants. Mechanical properties, hygrometric behavior, inhibition of growth of molds and depollution properties have been tested. Despite using porous materials (zeolite and activated carbon), in mortars with unconventional aggregates, compressive strength is higher than in sand-based ones, with a more than double higher water vapor permeability. Zeolite-based mortars have the highest moisture buffering capacity followed by silica gel- and activated carbon-based mortars (1.5–2 times higher than reference, respectively, because of the high porosity of unconventional aggregates). Sand-based mortars show optimum inhibitory capacity against fungal growth. Concerning unconventional aggregates, silica gel mortars have good inhibitory capacity, whereas zeolite and activated carbon give to mortars an optimum substrate for molds. Mortars with unconventional aggregates as silica gel remove more than 80% of tracer pollutant after 2 h of test, whereas zeolite-based mortars remove the 65% of it after 120 min. TiO₂ enhances depollution properties as photocatalytic oxidation agent when the mortar is close to saturation.

Keywords: Indoor air quality / mortar / adsorbent aggregates / titanium dioxide TiO₂ / mechanical properties / microstructure / hygroscopic behavior / inhibition of moulds / depollution properties

1 Introduction

Buildings materials are strictly related to indoor air quality (IAQ) of the environments where they are placed. People are spending up to 90% of their time indoor, where concentration of airborne pollutants, due to insulating necessity, could be higher than outdoor [1,2]. Occupants are potentially exposed to airborne pollutants, non-adequate levels of Relative Humidity (RH) [3] and microorganisms that can be serious health hazards to occupants because of the production of airborne particles such as spores, allergens, toxins and other metabolites [4].

This paper explores the possibility of improving IAQ using building materials able to positively interact with the indoor environment due to their adsorption capacity and/or to their photocatalytic oxidation (PCO) activity:

this capacity is provided by a mortar that can be applied as finishing layer directly on walls or on pre-casted partitions [5].

Highly adsorbent materials are used as aggregates (substituted to conventional calcareous sand) to prepare unconventional mortars able to improve IAQ. In literature, low quantity of activated carbon/zeolite in a photocatalytic cementitious matrix or superadsorbent materials are used in cementitious matrix to guarantee the decomposition of NO_x or Volatile Organic Compounds (VOCs) [6–9].

However, in literature it has been found that by using adsorbent materials (zeolite, silica gel and activated carbon), the adsorption process of the aggregate predominates on the photocatalytic action of TiO₂ [3].

In this study, hydraulic lime is used as binder, which is more sustainable [10] than cement in terms of footprints and more suitable for restoration purpose [11]. Adsorbent materials are used as unconventional aggregates, which are

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characterized by high adsorption properties and currently are not used in the building sector but in chromatography or filters for water/air depuration processes [9], in particular the selected materials are zeolite, silica gel and activated carbon.

TiO₂ is used as photocatalytic agent, to test also the decomposition of airborne pollutants. In fact, the possibility to obtain mortars with photocatalytic activity, is investigated by the addition of 5% of titanium dioxide [12], in order to guarantee the decomposition of airborne pollutants.

In particular, 8 different types of mortar have been manufactured with traditional commercial sand, as reference aggregate, or three different adsorbent aggregates and hydraulic lime as binder, with and without TiO₂ in order to investigate the combined effect of adsorption and photocatalysis.

Mortars have been compared in terms of mechanical strength, morphology and microstructure, permeability, moisture buffering ability, de-pollution properties and inhibition of molds growth.

2 Materials

The hydraulic binder (HL) is the commercial Plastocem[®], produced by Italcementi. HL used belongs to LIC 3.0 according to UNI EN 15368:2010 and the measured density is 2.65 g/cm³.

Conventional aggregate is calcareous sand with a purity of 98%. Aggregates are added during the cast at saturated surface dry (ssd) condition, since all the voids of aggregates are filled by water but the surface is dry. This is the condition when the mix is not influenced by the humidity of the aggregates which can not exchange water during the cast, keeping constant the water to binder ratio (w/b). In case of calcareous sand, ssd condition is reached by adding 5% of water at least 24 h prior to the cast, density in ssd condition is 2.65 g/cm³. Grain size distribution curves of aggregates are shown in Figure 1.

Adsorption process is guaranteed on the mortar by highly porous [4] unconventional aggregates such as zeolite, silica gel and activated carbon.

Zeolite is mainly characterized by a structure with channels, channel intersections and/or cages with dimensions from 0.2 to 1 nm. The negative framework charge that is present inside the voids is compensated by water molecules and cations [13]. The double functionality of zeolite as adsorbent/catalyst medium is well-known in literature [14]: this mineral is usually employed in fluid filters and purifiers as molecular sieving but zeolite is also used in lime based mortars to improve the pozzolanicity of the mix [15]. Zeolite was successfully used previously as aggregates, not only to improve mechanical properties but also to enhance the adsorption of VOC [9]. The used zeolite is a natural clinoptilolite zeolite, commercially available, provided by Samore S.r.l. with a maximum diameter of 250 μm. Low maximum diameter and high specific surface area of the zeolite (evaluated by literature data of about 600 m²/g [9]) imply about 20% in weight of water to reach ssd condition. Density in ssd condition is evaluated as 1.6 g/cm³.

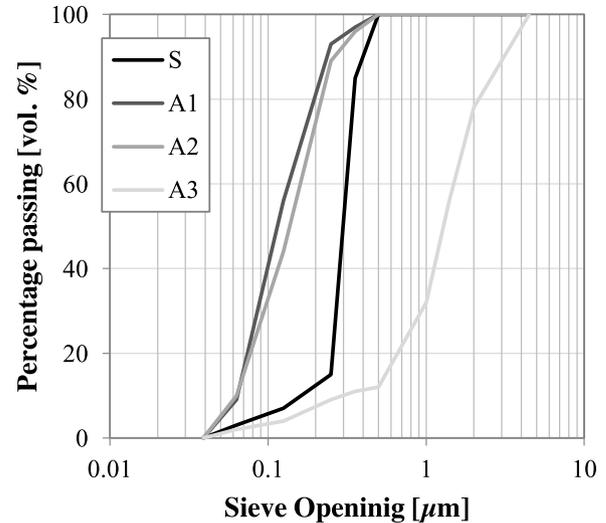


Fig. 1. Grain size distribution of the aggregates, S: sand, A1: zeolite, A2 silica gel, A3 activated carbon.

Colloidal silica is a compound made by variable units of SiO₂, which has high specific surface and high porosity. Silica gel is a non-toxic adsorbent material usually employed to uptake high quantities of moisture from the environment. For this experimentation, a commercial product, Inodorina, distributed by PetVillage S.r.l., is used as aggregate with maximum diameter of 300 μm. Ssd condition is reached when about 86% of water is added (by weight) and, in this condition density is 1.31 g/cm³.

Activated carbon is an adsorbent material used in filter and air/water purifying process. Activated carbon is microcrystalline, non-graphitic form of carbon that has been processed in order to develop the internal porosity [16]. BMD S.p.A. provides the activated carbon used in this experimentation, the commercial name of the product is 205E. This product is in form of cylindrical granules with maximum shape of 4 mm, according to the data sheet density is 0.53 g/cm³ with a specific surface area of 900 m²/g. Although activated carbon is a hydrophobic material, the practical experience suggested to add 30% of water during the cast to reach the same workability of other mortars.

Commercially available titanium dioxides (TiO₂) P-25 Aeroxide[®] by Evonik is used as photocatalytic agent. This TiO₂ is a mixture of anatase-rutile-amorphous phases, 78–14–8% in weight, respectively. Particles have nano-size of about 20–50 nm with a specific surface, measured by BET, is 35–65 m²/g. The pH value is 3.5–4.5 in 4% dispersion and density is evaluated as 3.1 g/cm³.

2.1 Mix design

The mix proportions of mortars are reported in Table 1. All mortars have same water to binder ratio (w/b), equal to 0.6. TiO₂ is substituted by weight to HL and calcareous sand is substituted by volume with zeolite, silica gel and activated carbon. All aggregates are added to the mix in ssd condition (Tab. 1).

Table 1. Mix proportions of mortars (g/L).

Mix	Water	Hydraulic Lime HL	Sand S	Zeolite A1	Silica Gel A2	Activated Carbon A3	TiO ₂
HL-S	255	440	1535	–	–	–	–
HL-S TiO ₂	255	414	1535	–	–	–	26
HL-A1	255	440	–	927	–	–	–
HL-A1 TiO ₂	255	414	–	927	–	–	26
HL-A2	255	440	–	–	759	–	–
HL-A2 TiO ₂	255	414	–	–	759	–	26
HL-A3	255	440	–	–	–	683	–
HL-A3 TiO ₂	255	414	–	–	–	683	26

3 Methods

The workability of mortars was measured with a truncated conical mold and a jolting table according to the standard UNI EN 1015-3:2007.

After 28 days of curing and after exposure to different temperatures, the density (ρ , in kg/m³) of hardened mortars was calculated. The reported values are the averages of three measurements.

Compressive strength is performed according to the standard UNI EN 1015-11:2007. Specimens are casted and cured for 7 days at $20 \pm 2^\circ\text{C}$ and RH $95 \pm 3\%$, for the following 21 days specimens are kept at the same temperature but exposed to a RH = $65 \pm 3\%$. A ‘Galdabini’ hydraulic press with a precision of 1% was used for the compressive strength tests. For the mechanical tests, reported data are the average values of three specimens.

The porosity of different mixes is evaluated by means of mercury intrusion porosimetry (MIP) with a PASCAL 240 mercury porosimeter (Thermo Fisher Scientific, Waltham, MA, USA) with a measuring pressure range from 0.01 to 200 MPa. For each mortar type, three small mortar fragments of about 1 cm³ in volume were tested after 28 days of curing.

Water vapor permeability measurements are carried out according to the UNI EN 1015-19:2007 and data processed according to UNI EN ISO 12572:2007 in order to quantify the water vapor resistance factor, so the permeability to facilitate the drying process of the masonry assemblage can be evaluated, as well as the disposal of water vapor produced inside buildings.

The moisture buffering capacity (MBC) is the capacity of a material to absorb and release moisture from/to the environment where it is placed. In this paper, the influence of unconventional aggregates on MBC of mortars is assessed by a simplified version of the NORDTEST method as it has been shown in a previous work [3].

The study of molds growth on the tested paints was performed according to UNI EN 15457:2014 using *Aspergillus niger*, fungi able to cause health problems including allergies and asthma especially during prolonged indoor exposure. The methodology and the quantification of the inoculum was performed as described in a previous

work [4]. In the current experimentation, the main difference is represented by the sterilization methodology of the sample. To guarantee sterile conditions all ingredients are weighted and mixed in dry condition then powders are put in an oven at 150°C to sterilize them. The powders are mixed with distilled and sterilized water under a chemical laboratory fume hood. The cast is performed on sterile filter papers (surface exposed 6.5×6.5 cm). The specimens are inserted in petri boxes to maintain sterility outside the hood. Samples are inoculated after checking the pH in order to ensure that the initial basicity of the mortar was lost, reaching a range compatible with the *Aspergillus niger* growth. Then, photos of the specimens are taken and images are elaborated with two different software, ImageJ and GIMP2. Pixels, corresponding to the percent of area colonized with molds, are counted. Images are divided in 3 different zones: zone 1: completely colonized; zone 2: boundary zone, where the molds are growing and zone 3: not colonized. The sum of pixels in zone 1 (Z1) and 2 (Z2) gives the percentage of the colonized area.

Depolluting properties are quantified with a gas chromatographer (GC): the depolluting rate of mortar samples are evaluated under different test conditions, with and without UV radiation. In such cases, pure adsorption [9] and adsorption and photocatalytic oxidation if irradiated by UV radiation [3] is evaluated. The methodology is the same illustrated in [3], performed on cylindrical specimens with 8 cm diameter and 0.8 cm height. The depolluting efficiency is evaluated as percentage of C_i/C_0 where C_i is the concentration detected inside the box and C_0 is the theoretical initial concentration equal to 2402 mg/m³. To study the behavior of specimens under saturation conditions, when only mortars containing TiO₂ are tested, MEK is injected more than one time:

- injection of MEK and monitoring for 120 min;
- injection of a re-load of MEK after 120 min and successive monitoring.

Tests are repeated in dark condition and under UV radiation. The initial concentrations (C_0) of the successive loads are considered equal to the sum of the MEK residual concentration of the previous cycle and the new concentration inserted in the box.

Table 2. Mechanical tests and hygrometric behavior results: slump (mm), hardened density (ρ), compressive strength (R_c), water vapor resistance factor (μ) and moisture buffering value (MBV) of hydraulic lime (HL)-based mortars. S: sand, A1: zeolite, A2 silica gel, A3 activated carbon.

Mix	Slump (mm)	ρ (kg/m ³)	R_c (MPa)	μ (-)	MBV (g/m ² *RH)
HL-S	127	1800	9.76	15.53	0.12
HL-S TiO ₂	118	1870	10.87	14.93	0.13
HL-A1	110	1390	11.10	11.45	0.42
HL-A1 TiO ₂	110	1340	8.87	11.05	0.53
HL-A2	123	910	1.00	5.46	0.49
HL-A2 TiO ₂	118	900	0.80	5.56	0.44
HL-A3	105	1220	15.28	6.47	0.26
HL-A3 TiO ₂	111	1210	12.57	5.55	0.39

4 Results and discussion

4.1 Workability

The addition of different amounts of water permits the same workability range to be achieved for different mixes. Slump values fell between 110 and 127 mm (Tab. 2), which correspond to the range of stiff consistence according to the standard UNI EN 1015-6:2007.

4.2 Mechanical properties

Mechanical strength test is evaluated after 28 days from the casting. In this case, the low mechanical strength is due to the use of an hydraulic binder instead of cement [3]. From the results (Tab. 2) it is shown that the addition of TiO₂ does not imply substantial differences in compressive strength results probably due to pore-refining and accelerating effect on hydration, as indicated in [17], despite other findings in literature, where it is shown that TiO₂ could decrease [18] mechanical behavior if the amount of binder decreases.

Zeolite (A1) based mortars show values slightly higher (about 5%) than the reference mortar. With zeolite (A1) the mechanical resistance is enhanced by its pozzolanic activity that forms additional hydration products [19,15].

When mortars prepared with silica gel (A2) are tested, they show the lowest value in terms of mechanical strength: maximum load is 90% lower than sand-based mortars. This is mainly due to the poor adherence between aggregate and hydraulic lime.

The best results are recorded in activated carbon (A3) based mortars, which have mechanical strength 35% higher than the value of the reference mortar, probably due to the good adherence (good interfacial transition zone) between the aggregate and the hydraulic paste [3].

According to the current standard UNI EN 998-1:2010, the maximum value of density for indoor mortar to be classified as lightweight is 1300 kg/m³. Thanks to the total replacement of calcareous sand with silica gel (A2) and

activated carbon (A3), it is possible to obtain a lightweight finishes, this property is highly appreciated for non-structural materials [20,21].

4.3 Microstructure

The microstructure of mortars was studied in terms of cumulative pore size distribution (Fig. 2a) and total porosity (Fig. 2b). In cementitious matrices, the porosity can be divided into: (a) gel pores-nanopores inside the hydration products, with pore diameters of about 0.5–10 nm; (b) capillary pores-micropores between the hydration products, with pore diameters between 10 nm and 5 μ m, strongly dependent on the hydration degree and the w/b ratio; (c) macropores-pores due to entrained air with spherical microbubbles, and (d) pores within the aggregate [22,23].

HL-S and HL-S TiO₂ have a unimodal pore size distribution. Peaks correspond to a pore diameter of 1.20 and 0.73 μ m, respectively. The threshold pore diameter is 2.55 μ m, the same for both mortars. The use of hydraulic lime introduces into the matrix a porosity with higher threshold pores diameter compared to that of cementitious mortar, as elsewhere discussed [12]. Due to similar results between conventional aggregate-based mortars without and with the PCO agent, only mortars without TiO₂ are tested.

In case of zeolite (A2) and activated carbon (A3) based mortars, the pore distribution is bi-modal. Peaks are at 0.13 and 0.02 μ m for zeolite mortar and 2.55 and 0.13 μ m for activated carbon mortar. Threshold diameters are 1.20 μ m for zeolite mortar and 6.94 μ m for activated carbon mortar.

Silica gel-based mortar also shows a bimodal distribution, with peaks at 0.73 μ m and threshold diameters of 1.98 μ m. The main finding is that silica gel-based mortar has the highest percentage of accessible porosity, up to 47%, that is the maximum registered between these specimens, followed by zeolite-based mortar (about 40%). Reference mortar and activated carbon mortar have the lowest values of total percentage porosity, which is around 30%.

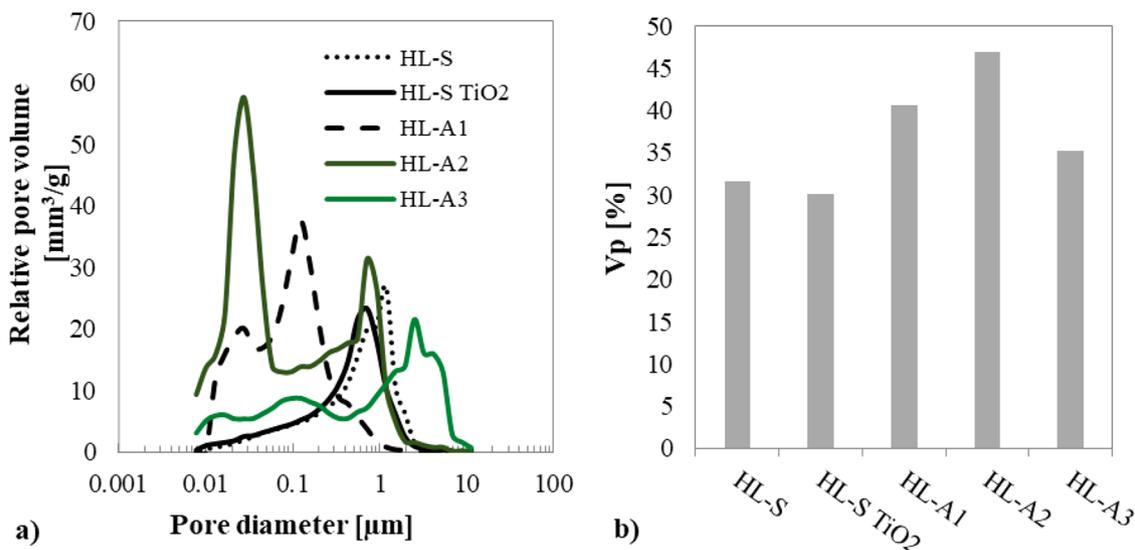


Fig. 2. Porosity of mortar in terms of (a) relative pore volume distribution of different hydraulic lime based mortars and (b) percentage of total open porosity. HL: hydraulic lime, S: sand, A1: zeolite, A2 silica gel, A3 activated carbon.

4.4 Hygroscopic behavior

Water vapor permeability expressed in terms of μ factor is shown in Table 2. The lower the value of μ , the higher the transpirability of mortars. The use of TiO_2 does not show different behavior of mortars, the main differences are related to the used aggregates.

As expected, the lower the porosity (Fig. 2b), the higher the μ factor, in fact sand (S) based mortars have the highest μ .

Zeolite (A1) based mortars shows μ value of about 27% lower than the reference mortar. The lowest value is obtained by activated carbon (A3) and silica gel (A2) based mortars, that are about 61 and 65% lower than sand based mortars, respectively.

However, if compared to commercially available products, all mortars in this experimentation have very high permeability to water vapor: permeable and hygroscopic structures can significantly reduce RH peak values and daily changes in RH, improving the IAQ perception [24].

As already discussed, the permeability to water vapor is proportional to the porosity percentage. This behavior can explain results of silica gel mortars.

Low value of activated carbon based mortars are well explained thanks to Kats and Thompson relation [25]: the lower the pore radius, the lower the transpirability [26].

The interaction between mortars with the humidity of indoor environments is also studied by means of moisture buffering capacity (MBC), in terms of moisture buffering value (MBV).

As in other testes TiO_2 does not change significantly the results of MBV. Sand based mortars have the lowest value, indicating the lowest capacity to be a moisture buffer for indoor applications.

Activated carbon shows a MBV about 1.5 times higher than that of reference mortar, whereas zeolite and silica gel

based mortars have 2.5 and 3 time higher MBV than the reference, respectively.

The differences in results are related to the pore size network of mortars. The higher the quantity of pores, the higher the ability of the material to uptake/release water vapor. However, the origin of this similar behavior are different in case of zeolite and silica gel based mortar. In case of zeolite based mortars, the results are more related to the total specific surface exposed. Instead silica gel based mortars have probably the highest percentage of porosity and show the best behavior in terms of MBV.

This situation can be a problem related to hysteresis phenomena, since the water vapor is absorbed during high humidity exposition but not completely released during the desorption phase [27]. During the test this behavior is slightly detected.

4.5 Inhibition of molds growth test

The inhibition of molds growth is evaluated in order to compare the behavior of different mortars exposed to biological colonization.

Before the test, pH is checked in order to be sure the initial basicity of mortar is lost achieving a range compatible to the *Aspergillus* growth. Figure 3 shows the stereomicroscope images showing the vitality of the used conidia in order to validate the results obtained by visual observation at 28 days of molds growth.

Figure 4 shows the average results obtained by the elaboration of the specimens pictures taken after 21 and 28 days from the inoculum.

If cracks are present, the colonization of the sample is not considered valuable. This is the case of the reference mortar where after 28 days only about 8% of

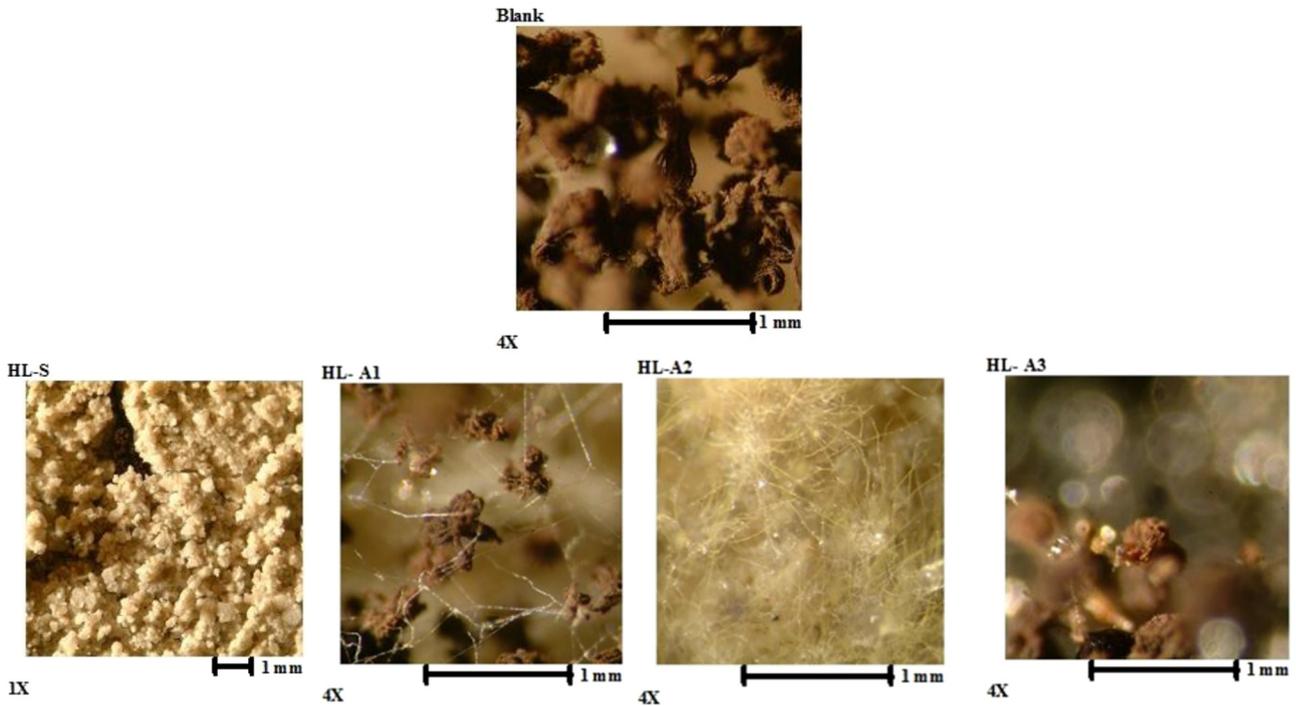


Fig. 3. Stereomicroscope image of inoculated conidia on blank and on different samples.

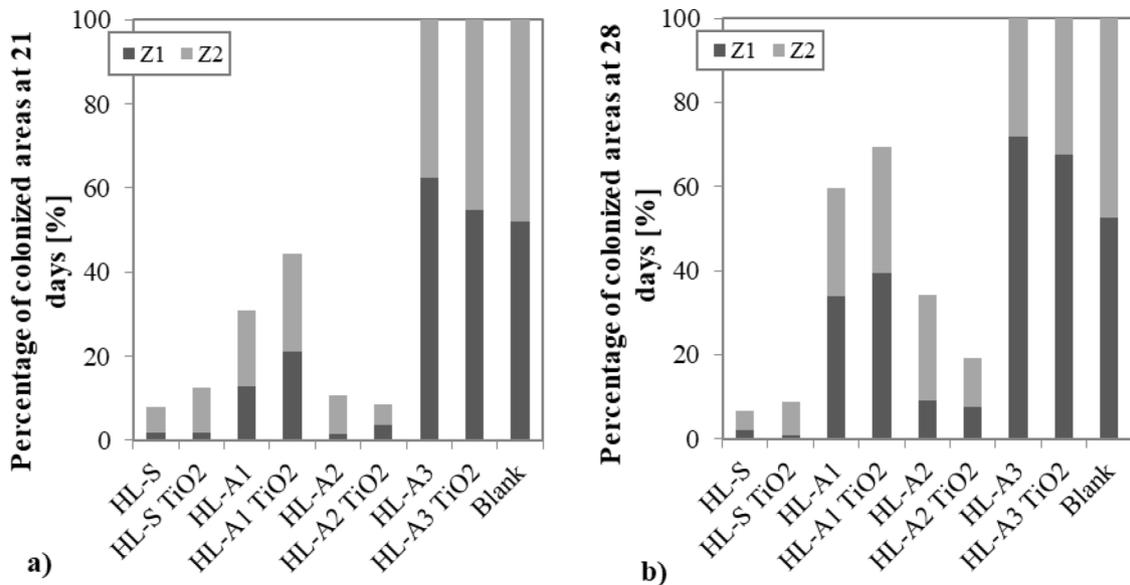


Fig. 4. Average results from the elaborated photos. (a) after 21 days, (b) after 28 days. Z1 and Z2 are the colonized zones by moulds of the specimens, in particular zone 1: completely colonized; zone 2: boundary zone. HL: hydraulic lime, S: sand, A1: zeolite, A2 silica gel, A3 activated carbon.

the area of the specimens is considered colonized. This is confirmed also from the stereomicroscope images (Fig. 3). Sand based mortars have the ability to inhibit the molds growth, which are growing only on the agar. Sand based mortars confirm this behavior also after 28 days from the inoculum: the colonized area is measured and it is lower than 10%.

Zeolite (A1) based mortars specimens are colonized for about 30–40% at 21 days and more than 60% after 28 days. These specimens have a favorable environment molds growth and proliferation, which is vital, with lots of conidia and hyphae (Fig. 3). This can be due to the presence of elements such as potassium that provides additional feedings to molds.

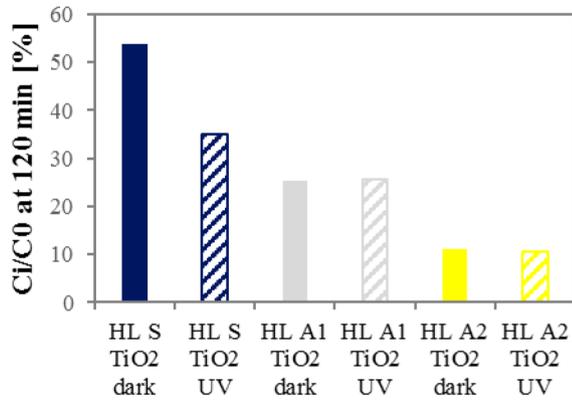


Fig. 5. Depollution efficiency of mortars at 120 min. HL: hydraulic lime, S: sand, A1: zeolite, A2 silica gel.

Silica gel (A2) based mortars show a colonization only where the molds were inoculated. Molds have colonized about 10% of the specimen's surface at 21 days. This percentage increases till 30% after 28 days. However, in case of silica gel mortars, molds are growing with high difficulties since mainly hyphae are detected but only few quantities of conidia, which are strictly related to vitality of molds itself, as shown by Figure 3.

Activated carbon (A3) based mortars have the highest percentage of colonized area, that can be considered close to the total surface area of the sample (100%). In this case, microorganisms such as bacteria preferentially adhere to solid supports made of carbon materials, indicating high biocompatibility. Bacteria may multiply on the surface of the activated carbon [28].

TiO₂ is not influencing the molds growth because it has not been activated before. It is well known that if TiO₂ is activated it has also antibacterial properties [29,30]. The differences detected in zeolite and silica gel based mortars in the presence of TiO₂ after 28 days are probably due only to agar rise in mortar porosity.

4.6 Depollution test

This test is conducted in 2 different conditions: under dark condition and under UVA radiation (10 W/m² on the surface of the specimens) in order to evaluate the pure adsorption and the combined action of adsorption and photocatalytic action. Figures 5 and 6 show the depolluting efficiencies of the mortar after 1 load of MEK and after 6 loads of MEK. In this case, for brevity, due to very similar results obtained by silica gel mortar, results of activated carbon based mortar are not reported.

By analyzing the data obtained from the single load test it is possible to note that, in dark condition, sand-based mortar removes 50%, zeolite mortar 65%, and silica gel mortar up to 80% of initial MEK at 120 min. The higher the MEK loads, the lower the depolluting capacity of all tested mortars. This means that mortars are saturating by increasing the successive MEK loads. However, the main finding is that whereas sand based mortars, without and with the addition of TiO₂ agent, decrease their efficiency and are saturated after 3 MEK

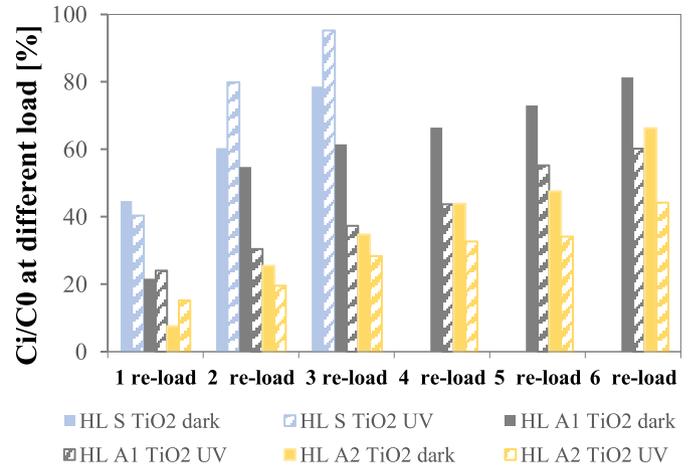


Fig. 6. Depollution efficiency of mortars at different MEK loads. HL: hydraulic lime, S: sand, A1: zeolite, A2 silica gel.

loads, the depollution efficiency of zeolite based mortars decrease from 65 to 20% after 6 MEK loads, and that of silica gel mortars from 80 to 40% after 6 loads. The progressive saturation of the active sites by MEK on the surface of the specimens brings to low the adsorption capacity of the specimen, despite high adsorptive materials are used.

As found in a previous study [3], the system is generally not able to appreciate the single contribution of each process implied in the depolluting activity: adsorption and PCO.

Under UVA radiation, after the first MEK load, all sand based mortars remove 40% of MEK in 120 min of test. When zeolite and silica gel are used, at the first MEK load the depollution efficiency is not increased if compared to dark condition, because active sites are still available.

After the second MEK load, under UVA radiation, sand based mortars do not increase the removal efficiency. On the other hand, in zeolite and silica gel mortars it increases of about 10 and 20% respectively.

PCO increases, becoming the prevalent process, only when the specimen, due to the quantity of pollutant already adsorbed, is close to saturation. This is visible only on unconventional aggregates-based mortars probably because, in case of sand mortars, the quantity of MEK injected within the 1st load is too high, also for the PCO process.

PCO is a process that occurs on the surface. In case of zeolite and silica gel based mortars, it can not occur as long as pollutants are adsorbed by mortar pores. PCO results are hidden when MEK concentration is low (Fig. 5) and the adsorption process is more influential [3]. Only when the specimen is saturated, the pollutants stop at the surface and become available for PCO process.

5 Conclusions

In conclusions, by replacing reference sand with unconventional lightweight aggregates in mortars for indoor applications, the obtained results can be summarized as follows:

- Mortars have the same workability, that is stiff.
- Mortars with activated carbon with and without TiO₂ have the highest compressive strength, 35% higher than sand based mortars. Mortars with silica gel have the lowest mechanical compressive strength, 90% lower than reference mortars. Zeolite based mortars have 5% higher mechanical resistance than sand based mortars. The values of mechanical properties remain acceptable and mortars with silica gel and activated carbon can be classified as lightweight mortars.
- Mortars with sand have the lowest pore volume, followed by activated carbon mortars (with highest pores diameters) and zeolite (with smallest pores diameters).
- Mortars with silica gel and activated carbon have the highest permeability to water vapor, more than double of sand based mortars.
- Mortars with zeolite have the highest moisture buffering value, 3 times higher than conventional based mortars. Silica gel and activated carbon based mortars adsorb and desorb 1.5–2 times more water vapor than sand based mortars, respectively.
- Mortars with conventional sand show optimum inhibitory capacity against fungal growth. Silica gel mortars show a colonization 4 times higher than sand based mortar. Zeolite and activated carbon give to mortar an optimum substrate where mold can grow.

Mortars with unconventional aggregates as silica gel remove more than 80% of MEK after 2 h of test. The use of TiO₂ enhances depollution properties in terms of PCO improvement when, due to the quantity of pollutant already adsorbed, the specimen is close to saturation.

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