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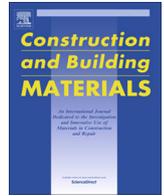
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Hygro-thermal and durability properties of a lightweight mortar made with foamed plastic waste aggregates

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HIGHLIGHTS

- Foamed end-of-waste plastic aggregates were used instead of natural silica sand.
- Increasing artificial aggregates content an increase of macropores was observed, as demonstrated by Mercury Intrusion Porosimetry.
- Pores microstructure variation determined a higher water vapour permeability and a lower capillary water absorption.
- The presence of plastic aggregates reduced both mortar density and thermal conductivity.
- Lightweight mortars containing foamed end-of-waste plastic aggregates are suitable as pavement and/or roofing materials.

GRAPHICAL ABSTRACT



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ABSTRACT

In the present study, hygro-thermal and durability related properties of a cementitious mortar containing highly porous foamed aggregates obtained from polymeric end-of-waste materials were investigated. The evaluation of capillary water absorption, thermal conductivity, water vapour permeability and sulfate attack resistance of samples where natural quartz sand was replaced by 10%, 25% and 50% in volume with foamed aggregates was carried out. Experimental investigations showed that the presence of plastic aggregates decreased mortar density (up to 36%, compared to the reference sample, for the maximum investigated natural sand volume replacement) as well as thermal conductivity (10% for the 50% volume replacement). Moreover, water vapour transmission rate increased at increasing natural sand replacement while capillary water absorption decreased. Finally, after fifteen cycles of sulfate attack test, lightweight mortars evidenced a lower mass loss compared to the reference sample. The results were related to morphological modifications in the mortars bulk porosity, demonstrating by mercury intrusion porosimetry investigations, that polymeric foamed aggregates determine a variation of the pores microstructure, resulting in an increased pores dimension.

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70 **1. Introduction**

71 Replacement of natural aggregates in mortar and concrete pro-
 72 duction is one of the main issue to reduce depletion of natural
 73 resources in production of construction materials. Crushed asphalt
 74 from road rehabilitation [1], cast iron industry by-products [2]
 75 electric arc furnace slag [3,4] and limestone filler recycled from
 76 marble industry [5] have been proposed to manufacture environ-
 77 mentally friendly mortars and concretes. More recently, the use
 78 of polymeric waste based materials as natural aggregates replace-
 79 ment in concrete is a relatively recent idea driven mainly by the
 80 energy and environmental concerns which are becoming remark-
 81 ably relevant in the construction industry. On the other hand, the
 82 use of polymeric lightweight aggregates represent a promising
 83 solution in reducing the specific mass of concrete and mortars.
 84 From the environmental point of view, as plastic waste manage-
 85 ment is one of the most critical drawbacks of such versatile mate-
 86 rials, the use of lightweight aggregates produced from end-of-
 87 waste or post-consumer materials is particularly interesting and
 88 the perspective of their use for new applications represent a chal-
 89 lenge for researchers. To this extent, in the last ten years, several
 90 studies demonstrated the outcomes deriving from the use of recy-
 91 cled plastic wastes as natural aggregates replacement in cementi-
 92 tious materials [6,7]. The incorporation of lightweight aggregates
 93 provides several advantages: reduced specific mass, improved
 94 thermal properties, impact resistance, sound insulation properties,
 95 fire resistance and durability enhancement [8–28]. However, one
 96 of the main disadvantages of using polymeric and lightweight
 97 aggregates is the reduction of mechanical properties. Thus, authors
 98 generally focus the attention on such aspects which are related to
 99 the chemical and physical incompatibility between the polymeric
 100 phase and the cementitious matrix but also to the lower mechan-
 101 ical properties of plastic aggregates compared to natural aggre-
 102 gates. In the literature, plastic aggregates of different polymeric
 103 nature and shape have been investigated but only few studies dealt
 104 with hygro-thermal properties of lightweight mortars containing
 105 plastic aggregates. Moreover, even less authors investigated dura-
 106 bility related problems of such mortars or concretes [8–10]. Hygro-
 107 thermal properties are very important parameters that should be
 108 taken into account in order to provide favorable living conditions
 109 in terms of temperature and relative humidity. Among all plastic
 110 waste materials, better results in terms of interfacial transition
 111 zone (ITZ) and insulating properties were reported when expanded
 112 polymeric aggregates were used, thanks to their porous structure
 113 [11–17,22–24]. Recently, Ramirez-Arreola et al. investigated also
 114 the possibility to produce foamed HDPE nanocomposite aggregates
 115 with different porosity varying nanoclay content [27]. Nanofillers
 116 have also been investigated as consolidating and protective agents
 117 to improve durability of different construction materials [29,30].
 118 Durability related issues negatively affect structures service life,
 119 influencing maintenance costs and structural safety. Thus, it is very
 120 important to investigate durability also of lightweight cementi-
 121 tious materials. Dulsang et al. [10] reported an increased resistance
 122 to sulfuric acid of ethyl vinyl acetate (EVA) waste lightweight con-
 123 crete. Turatsinze et al. [19,20] investigated the effects of rubber
 124 aggregates, obtained from shredded non-reusable tyres, on the
 125 concrete resistance to cracking and toughness increase. Authors
 126 demonstrated that cement-based mortars containing rubber
 127 aggregates are less prone to crack. Moreover, the use of waste plas-
 128 tic materials as mortar or concrete aggregates instead of virgin
 129 polymers, to obtain insulating materials, has not only environmen-
 130 tal but also economic benefits [23]. Finally, as widely reported in
 131 the literature, one of the main drawbacks in the use of plastic
 132 aggregates or fibers in cementitious materials is the weak adhesion
 133 among polymeric materials, especially polyolefin, and the

cementitious matrix [24,31]. In a previous work [24], Coppola
 et al. investigated the possibility to use polymeric aggregates with
 a rough surface, produced from end-of-waste materials by a foam
 extrusion process. The investigations reported an improved bond
 between plastic aggregates and the cement paste but also a good
 dispersion in the specimens cross-section. Moreover, a sharp
 decrease of density was achieved but also consistency decreased
 at increasing natural sand replacement. In this study, hygro-
 thermal and durability properties of lightweight mortars contain-
 ing such foamed end-of-waste aggregates were studied with the
 aim of correlating the results to composite morphology which
 resulted to be strongly influenced by the presence of lightweight
 aggregates.

147 **2. Experimental procedures**

148 **2.1. Materials**

Mortar samples were produced using an ordinary Portland
 cement (CEM I 42.5 N), quartz sand (CEN standard sand according
 to EN 196-1 [32], density of 2610 kg/m³) and lightweight aggre-
 gates (LWAs). LWAs were produced starting from an end-of-
 waste material represented by a polyolefinic blend (polypropylene
 and polyethylene coming from the recycling of flexible packaging
 materials), supplied in densified pellets. A chemical foaming agent
 (Hydrocerol CF) was used to produce by extrusion foamed strands
 that were subsequently grinded to obtain aggregates of four differ-
 ent particle size (2–1.4 mm, 1.4–1.0 mm, 1.0–0.50 mm and 0.50–
 0.18 mm), according to the procedure described in a previous arti-
 cle [24]. Finally, aggregates were sieved and proportioned to par-
 tially reproduce EN 196-1 [32] quartz sand particle size
 distribution.

163 **2.2. Mixtures preparation**

Mortar samples were prepared using dry LWAs to avoid an
 increase of porosity due to the presence of free water released from
 LWAs [24], using a w/c ratio of 0.50 and a cement/sand ratio of
 0.33. Nomenclature of the studied mixtures is reported in Table 1.
 All the mortar samples were prepared according to EN 196-1 [32];
 natural and LWAs were dry mixed in advance to better disperse the
 aggregates in the mixture. Three natural quartz sand volume
 replacements (10%, 25% and 50%) were investigated comparing
 lightweight mortars (LWMs) samples to control specimens, i.e.
 without LWAs. All the samples were wet cured for 28 days in a
 humid chamber at 90% RH and 20 °C.

175 **2.3. Density measurement**

The oven-dry density, ρ_d , of hardened mortars was determined
 on specimens dried at 105 °C until constant mass, according to EN
 12390-7 [33]. Three specimens for each composition were tested
 (experimental data are reported in Ref. [34]).

Table 1
 Investigated mixtures containing three natural sand
 volume replacement by lightweight aggregates (LWAs).

Mortar	LWA (%)
Reference	–
LWM10	10
LWM25	25
LWM50	50

2.4. Capillary water absorption

Lightweight mortars water absorption coefficient due to capillary action, C_w , was determined according to EN 1015-18 [35]:

$$C_w = 0.1(M_2 - M_1) \quad (1)$$

where M_2 and M_1 are the mass of the specimen, in grams, after 90 min and 10 min of immersion, respectively. Three prismatic specimens for each investigated mixture were prepared and tested after 28 days of wet curing. Prior to test, specimens were oven dried up to constant mass at 60 °C. Then, prismatic samples were immersed in deionized water for about 5 mm and the mass change was measured. Considering mass variation, the capillary absorption coefficient was calculated for all the investigated mortars. Experimental data are reported in Ref. [34].

2.5. Water vapour permeability

Water vapour permeability, W_{vp} , of lightweight mortar samples was determined according EN 1015-19 [39] that defines a test method to determine water vapour permeability in steady-state conditions:

$$W_{vp} = \frac{s}{A \Delta p / (\Delta G / \Delta t) - R_A} \quad (2)$$

where s , A and ΔG are sample thickness, area and mass variation; Δt is the time interval; Δp is the gradient of water vapour tension between saturated solution and samples storing chamber and R_A is the resistance to water vapour diffusion in the air between the sample and the KNO_3 saturated solution ($0.048 \cdot 10^9$ Pa m² s/kg, for 10 mm of interspace). Three flat cylindrical specimens (diameter of 75 mm and thickness of about 20 mm), conditioned at relative humidity of 50% and 20 °C, were prepared for each lightweight mortar and sealed on glass containers, in which a saturated solution of KNO_3 was contained. This solution, at 20 °C, provides a relative humidity of 93.2%: the gradient of water vapour pressure between the lower part of the sample (RH = 93.2%) and the environment in which samples are stored (RH = 50%) causes a water vapour flow through the mortar sample. Water vapour resistance factor (μ) was measured according to the following equation:

$$\mu = \frac{\delta_A}{W_{vp}} \quad (3)$$

where δ_A is air permeability ($1.94 \cdot 10^{-10}$ kg/(Pa m s)) in test conditions (20 °C and 50% RH) and W_{vp} is water vapour permeability. Experimental data are reported in Ref. [34].

2.6. Thermal conductivity

Thermal conductivity was measured using the so-called “guarded hot plate technique” (GHP), defined in the ISO 8302

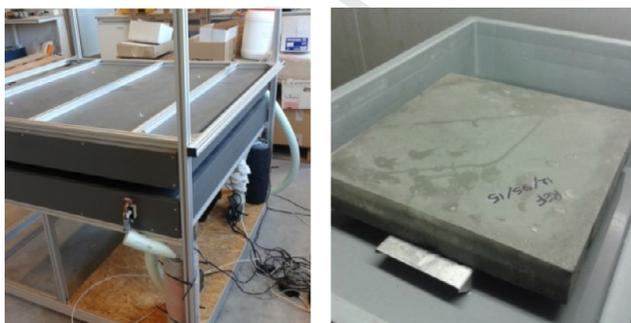


Fig. 1. a) GHP apparatus and b) slab-shaped specimen.

and specified in European standards EN 12664 [36] and EN 12667 [37] (Fig. 1a). The device used in this research was designed, constructed and validated by Dubois and Lebeau [38]. Thermal conductivity measurements were carried out on mortar specimens of size equal to 30 × 30 × 5 cm³ (Fig. 1b), manufactured in wood molds. After 28 days of water curing specimens were conditioned (25 °C and 50% RH) until constant mass before testing. Five tests were performed for each mortar sample to ensure the reproducibility (experimental data are reported in Ref. [34]).

2.7. Sulfate attack

The evaluation of lightweight mortar samples resistance to sulfate attack was evaluated according to the ASTM C88-05 procedure, opportunely modified [40,41]. Mortar specimens were immersed in a saturated solution of sodium-sulfate (Na_2SO_4) at room temperature for not less than 16 h and not more than 18 h, followed by a drying period in an oven at 110 °C up to constant weight. All these steps represent a drying-wetting cycle procedure. The evaluation of mortar behavior to sulfate attack is then made by measuring the weight variation of the lightweight mortar samples after a total of 15 cycles. The volume of the solution was at least five times the volume of the immersed specimens ($75 \times 75 \times 150$ mm³). A visual assessment of specimen deterioration was carried out by taking photos of mortar specimens at the first, eighth and fifteenth cycle.

2.8. Mercury Intrusion Porosimetry

Pore size distribution measurements and cumulative intruded volume were carried out on all the investigated LWMs by a mercury intrusion porosimeter (Pascal 140 and Pascal 240, Thermo Finnigan). The Mercury Intrusion Porosimetry (MIP) measurements were carried out using a contact angle of 141.3°, a Hg surface tension of 0.48 N/m and a pressure ranging from 0 to 200 MPa. Such pressure range covers a pores diameter range between about 0.0075 and 150 μm. Tests were performed on small samples of approximately 0.5 cm³. Samples for porosimetry were dried under vacuum at 60 °C for 12 h and stored in a desiccator until testing.

3. Results and discussion

3.1. Density of lightweight mortars (LWMs)

The possibility to use lighter materials in the building and construction field is very important, thanks to the possibility to reduce the structures dead load. To this extent, using plastic aggregates is possible to reduce mortars density. As expected, at increasing sand replacement, a decrease of mortar density was achieved, considering that artificial aggregates density is 65% lower than mineral sand density and that aggregates generally represent about the 70% of mortar or concrete volume [24]. Dry density values, ρ_d , of hardened mortars are reported in Table 2. A linear relationship exists between sand replacement and mortars dry density, as evident in Fig. 2. Compared to the reference sample, a density reduction of 8, 16 and 36% was obtained for LWM10, LWM25 and LWM50, respectively.

3.2. Capillary water absorption

The water sorption due to capillary suction was evaluated by specimens mass variation measurements and the capillary absorption coefficient C_w was calculated according to Eq. (1) and reported in Table 2. Increasing natural sand volume replacement, capillary

Table 2
Mortars physical and mechanical properties (ρ_d = dry density; W_{vp} = water vapour permeability; μ = resistance to water vapour).

Mortar	Reference	LWM10	LWM25	LWM50
ρ_d [g/cm ³]	2.143	1.981	1.805	1.374
Total open porosity [%]	16	16	17	23
W_{vp} 10 ⁻¹¹ [kg/m s Pa]	2.97	3.60	3.97	5.88
μ	6.56	5.72	4.90	3.38
C_w [kg/(m ² h ^{0.5})]	1.87	1.26	1.02	0.72
Thermal conductivity [W/m K]	1.408	1.365	1.311	1.266

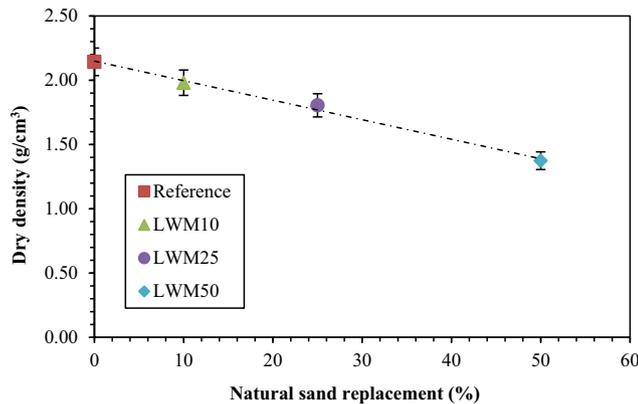


Fig. 2. LWMs dry densities vs. sand replacement.

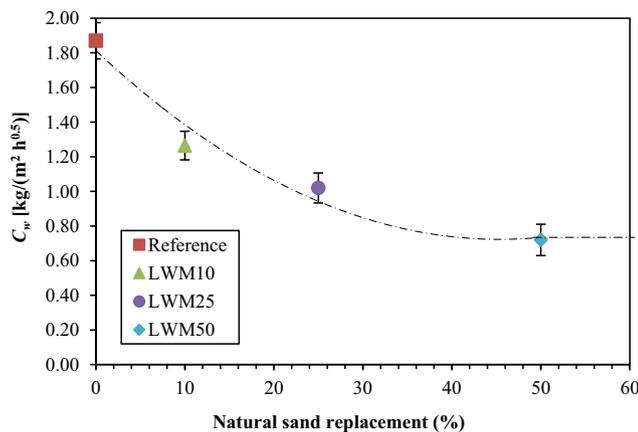


Fig. 3. Capillary water absorption vs. natural sand replacement.

3.3. Water absorption by total immersion

Water absorption is a crucial property for construction materials, as it represents one of the most important indicator of the material durability. The total open porosity was obtained by measuring water absorption and is reported in Table 2. As expected, open porosity increased with sand replacement corresponding to a sharp increase of water absorption occurring with sand replacement higher than 25% (Fig. 4).

As seen before for the capillary water suction, a non-linear relationship exists also between water absorption of the different LWMs and natural sand replacement (Fig. 4). In this case, water absorption of lightweight mortars with more than 25% substitution of sand significantly increased compared to the reference mortar, according to results obtained by other authors [21,43,44]. Such behavior, is in agreement with the aforementioned effect of LWAs on mortar porous structure: the increase of macropores at increasing LWAs content, determined an increase of water absorption and a decrease of capillary suction.

3.4. Water vapour permeability

Hygro-thermal properties are very important to give favourable living conditions and a good water vapour permeability is able to avoid fungi and condensation phenomena. Water vapour permeability of lightweight mortars (LWM10, LWM25 and LWM50) were compared to the reference sample in order to obtain further data about the effect of polymeric foamed aggregates on the transport properties of LWMs. Water vapour permeability (W_{vp}) and resistance (μ) of the investigated samples (calculated according Eqs. (2) and (3), respectively) are reported in Table 2. Vapour permeability increased proportionally to natural sand replacement, corresponding to a reduction of water vapour resistance down to about 50% respect to the reference for lightweight mortars containing 50% of plastic aggregates as recognizable in Fig. 5. Such improvement of vapour transmission capability of LWMs tested samples is related to the overall porosity morphology (i.e. not only

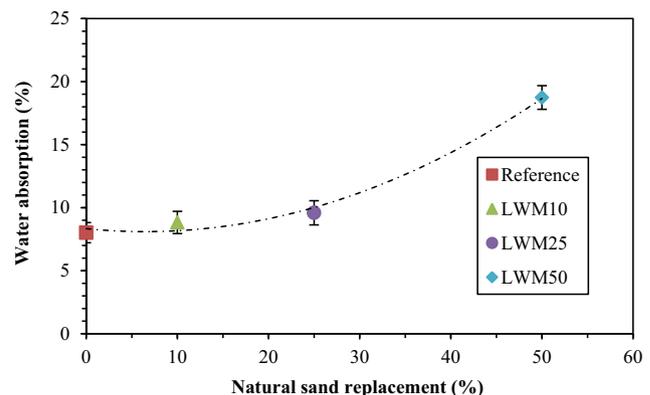


Fig. 4. LWMs water absorption (total immersion).

water suction decreased of 32, 45 and 61%, compared to the reference sample, following a non-linear relationship (as evident from Fig. 3). A plateau in the values of C_w is reached for a sand substitution of approximately 40%, suggesting that at high polymeric aggregates volume content, capillary pores structure is not affected. Such a behavior could be due to an effect of foamed aggregates on the degree of compaction of fresh mortars.

In a previous work, Coppola et al. [24] obtained a decrease of workability at increasing natural sand volume replacement, resulting in a non-linear open porosity increase (Table 2). More specifically, at increasing of polymeric foamed aggregates content samples present a decrease of capillary pores and an increase of macroporous structure in the sample volume. Moreover, as reported also by Marzouk et al. for PET aggregates [42], the hydrophobic nature of polymeric aggregates contributes to slow down the capillary water absorption. As stated previously, such porous structure is advantageous both for water vapour permeability and thermal conductivity, as discussed later.

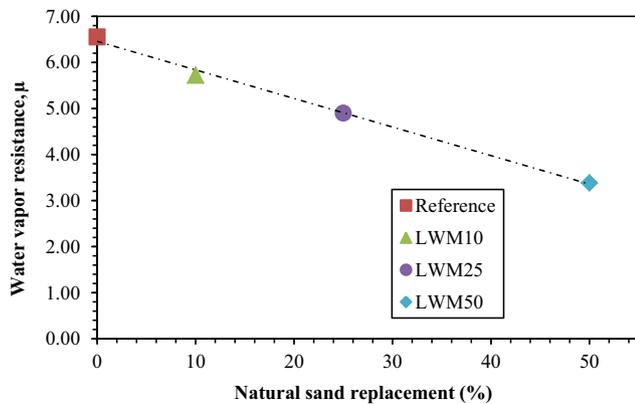


Fig. 5. Water vapour resistance vs. natural sand replacement.

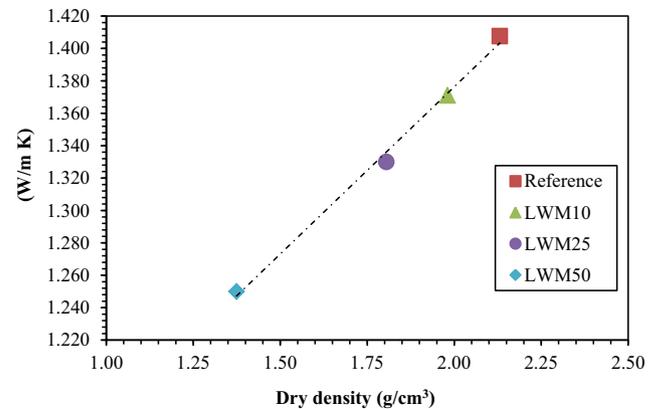


Fig. 7. LWMs thermal conductivity vs. dry densities.

335 to capillary and macropores) and can be explained taking into
 336 account that, in the case of foamed polymeric aggregates, the
 337 intrinsic open porosity of the aggregates themselves is able to cre-
 338 ate a path both for water and water vapour permeation. On this
 339 point, it has also to be considered that no contribution to this result
 340 was given by ITZ around the foamed aggregates, which presented
 341 lower porosity and higher density, as reported previously [24],
 342 unlike other literature results on lightweight mortars containing
 343 plastic wastes [16–18].

344 As well known, LWMs properties are strictly correlated also to
 345 aggregates/matrix ITZ. In Fig. 6 is shown the linear relationship
 346 that exists between capillary water absorption and water vapour
 347 resistance. At increasing natural sand replacement a decrease of
 348 both capillary water absorption and resistance to water vapour
 349 was obtained, confirming what stated before about pores
 350 morphology.

351 3.5. Thermal conductivity

352 The influence of artificial aggregates on lightweight mortars
 353 thermal conductivity was investigated by GHP apparatus, consid-
 354 ering that quartz sand have a very high thermal conductivity com-
 355 pared to the plastic one (about 20 times [45]). As expected, at
 356 increasing natural sand replacement, a decrease of thermal
 357 conductivity was obtained (Table 2). In particular, a reduction of ther-
 358 mal conductivity of 3, 7 and 10% was obtained replacing 10, 25
 359 and 50% of natural quartz sand, respectively. Similarly to other litera-
 360 ture results [16,46] a linear relationship is recognizable between

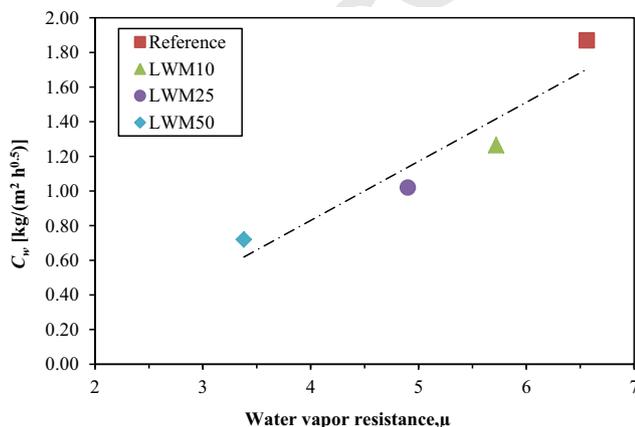


Fig. 6. Capillary water absorption vs. resistance to water vapour.

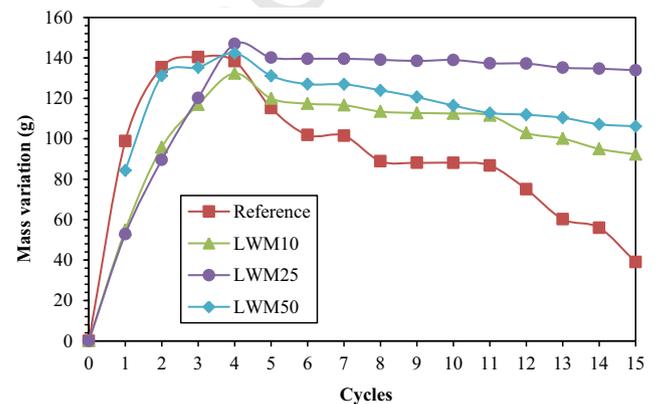


Fig. 8. Mass variation at increasing cycles of the sulfate attack test.

361 thermal conductivity and lightweight mortars dry density which
 362 is, in turn, linearly related to sand replacement (Fig. 7).

363 3.6. Sulfate attack

364 The resistance of LWMs to sulfate attack was evaluated by mass
 365 variation and relative mass (Fig. 8) after 15 wet/dry cycles into a
 366 saturated solution of sodium sulfate.

367 The role of capillary pores is crucial for the resistance to sulfate
 368 attack, resulting in a better behavior of lightweight mortars. As
 369 shown in Fig. 8, the specimens mass initially increases due to the
 370 solution absorption, reaching a peak value that corresponds to the
 371 third cycle for the reference sample and the fourth cycle for the
 372 lightweight mortars regardless the LWAs substitution. Such differ-
 373 ent behavior is mainly due to the different porosity of the
 374 samples and the different time necessary to saturate pores. As
 375 stated before, lightweight mortars have bigger pores that are able
 376 to contrast expansive phenomena associated to ettringite produc-
 377 tion. The presence of bigger pores is also very useful in the case
 378 of freeze/thaw phenomena because also in this case the pressure
 379 exerted on pores surface can lead to cracking and/or disintegration.
 380 In Fig. 9 is reported the relative mass loss (i.e. the ratio between the
 381 mass loss of the specimen and the mass loss of the reference sam-
 382 ple, in percentage). For the previously stated reasons, lightweight
 383 mortar samples reported a lower mass loss than the reference sam-
 384 ple. In particular, LWM25 showed the lowest relative mass loss
 385 while LWM10 and LWM50 have approximately the same value
 386 despite a progressive decrease of relative mass loss could be
 387 expected. However, it should be considered that the lightweight
 388 mortar with the highest artificial aggregates content is more prone

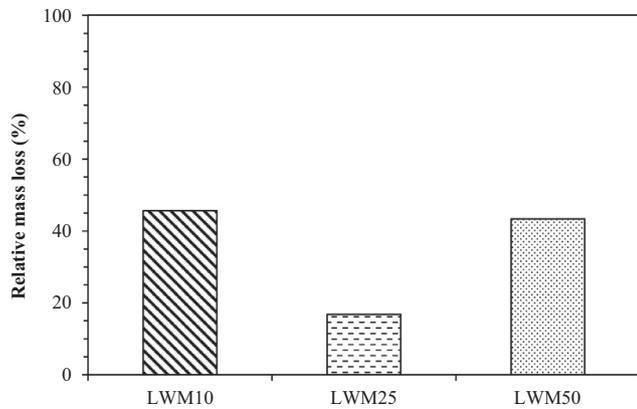


Fig. 9. Relative mass loss after fifteen cycles of sulfate attack.

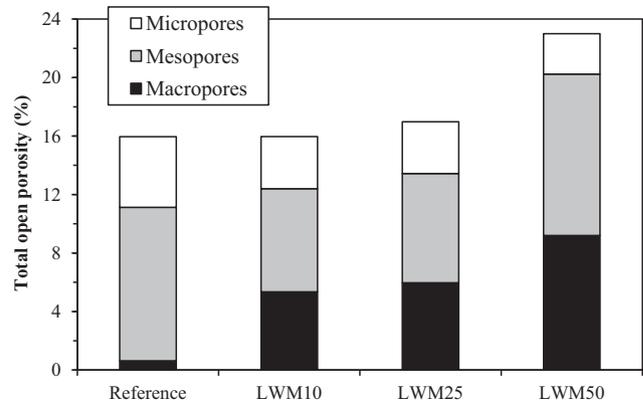


Fig. 11. Mercury Intrusion Porosimetry results for the investigated mortars.

to disintegration due to the low compaction level compared to the other samples. Thus, LWM50 specimens degradation is due both to ettringite formation and disintegration caused by the low cohesion of the sample.

Such different behavior is also clearly visible in Fig. 10 where a visual assessment of mortar specimens deterioration is reported at increasing test cycle (at the first, eighth and fifteenth cycle). The deterioration clearly appears already after 8 cycles as samples are affected by cracking, corners spalling and edge fragmentation. Visual observations confirm mass loss results: the extent of attack of lightweight mortar samples is lower in comparison to the reference sample. In the latter case degradation was more depth while in the formers only the skin was attacked.

3.7. Mercury Intrusion Porosimetry

Pores size distribution strongly influences mortars durability and hygro-thermal properties. Pores size distribution was investigated by Mercury Intrusion Porosimetry (MIP) and results are reported in Fig. 11 where the total open porosity was divided into micropores (<0.1 μm), mesopores (0.1–1 μm) and macropores (>1 μm). The results clearly confirmed what discussed in the previous

paragraphs: at increasing natural sand replacement, a decrease of micropores and a contextual increase of macropores were observed. The lower capillary water absorption coefficient (Table 2) measured for LWMs is correlated to the lower amount of micropores. On the contrary, the increase of macropores in LWMs determines the higher water vapour permeability (Table 2).

4. Conclusions

In this study, hygro-thermal and durability properties of a lightweight mortar containing foamed end-of-waste plastic aggregates were investigated. Artificial plastic aggregates were manufactured according to the procedure described in a previous article in a three step process: foamed strand production, grinding and sieving [24]. Such procedure determined higher adhesion with the cementitious matrix thanks to the presence of interlocking positions onto aggregates surface thanks to the high porosity and rough surface of foamed aggregates. Three lightweight mortars (LWMs) were produced replacing natural quartz sand, in volume, by 10%, 25% and 50% of foamed end-of-waste plastic aggregates. Experimental investigations showed that the pores morphology of LWMs differs from the reference samples. In particular, due to the pores microstructure variation at increasing LWAs, there is an increase

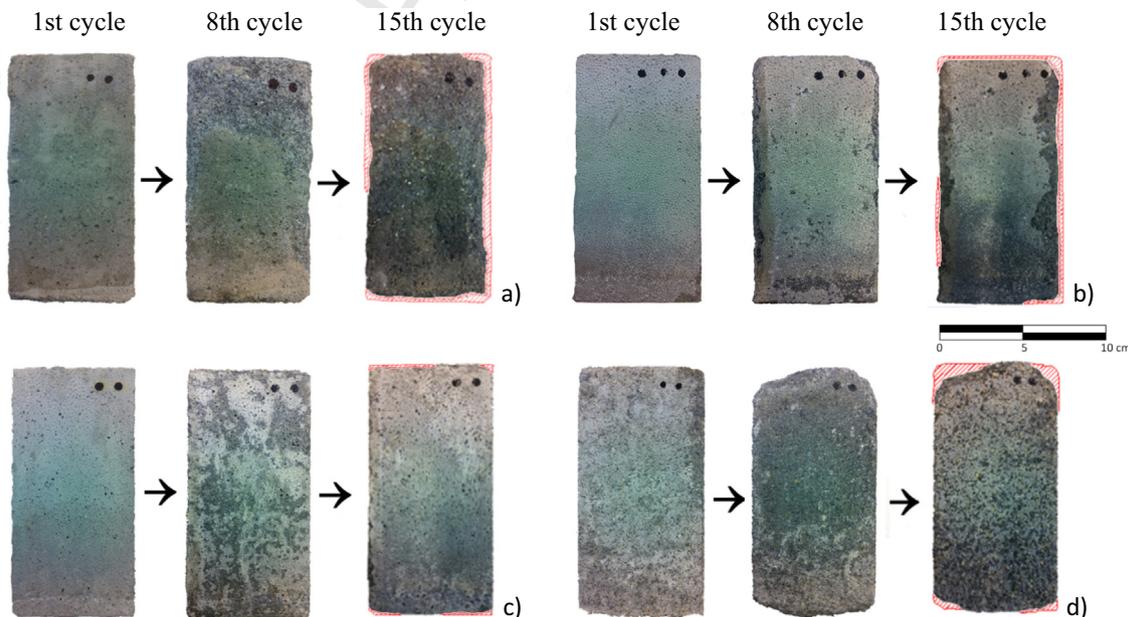


Fig. 10. Samples at the beginning, after 8 and 15 cycles of sulfate attack. a) Reference, b) LWM10, c) LWM25 and d) LWM50.

of macropores, which positively influence thermal conductivity and water vapour permeability, reducing at the same time the capillary water absorption. Thus, the presence of plastic foamed aggregates decreased mortar density (up to 36%, compared to the reference sample, for the 50% of natural sand volume replacement) as well as thermal conductivity (10% for the 50% volume replacement). Moreover, water vapour resistance significantly decreased at increasing natural sand replacement whereas capillary water absorption decreased. Also resistance to sulfate attack achieved good results, thanks to the different pores morphology. In order to confirm the hypothesis about pores size distribution, mercury intrusion porosimetry (MIP) investigations were performed on LWMs. MIP results confirmed the increase of macropores and the contextual decrease of micropores, at increasing LWAs content. In the end, this study demonstrated that polymeric end-of-waste materials can be used to produce artificial aggregates for the replacement of natural sand in LWMs manufacturing. Such replacement leads to a more sustainable lightweight mortar with better hygro-thermal properties.

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