

1 **Properties of an Innovative Multi-functional Finish for the Improvement of Indoor Air** 2 **Quality**

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16 17 **Abstract**

18 Due to lifestyle changes, people spend most of their time indoors at present; thus, Indoor Air Quality
19 (IAQ) is a matter of utmost importance. Multi-functional and innovative finishes can help to passively
20 improve the IAQ, benefitting the health and comfort of occupants. For this study, reference and pre-
21 mixed commercial mortars are compared to a new multi-functional hydraulic lime mortar for indoor
22 finishes, in which conventional aggregates are substituted by a highly porous adsorbent material and
23 biomass waste ashes. The up to 20% higher accessible porosity of the multi-functional finish led to
24 lower density (30%), higher thermal insulation properties (30%), higher water vapor permeability
25 (more than 40%), and improved moisture buffering capacity (three times higher), when compared to
26 the reference mortar. Different types of photocatalytic agents (TiO₂) were also added into the new
27 multi-functional hydraulic lime mortar, in order to investigate their effect on the de-polluting
28 properties of the finish. Even if the photocatalytic efficiency remained unexpressed under indoor
29 conditions, due to the predominance of the adsorption process, the de-polluting properties of the new
30 mix were more than 30% higher than that of the reference mortar. The obtained results confirm that
31 the developed innovative multi-functional finish—besides fulfilling the ordinary requirements—is

32 better than commercial mortars, as it can improve the IAQ passively, thus benefitting the health and
33 comfort of occupants.

34

35 **Keywords**

36 *Indoor Air Quality (IAQ), thermo-hygrometric behaviour, mould susceptibility, photocatalytic*
37 *oxidation (PCO), adsorption, de-polluting properties.*

38

39 **1. Introduction**

40 The sustainability of the building sector has recently received increased attention, as around 40% of
41 global carbon dioxide (CO₂) emissions are directly related to activities associated with the
42 construction industry [1]. Consequently, several strategies have already been formulated to tackle
43 their impact on the environment [2]. European Directives and laws have become stricter, in terms of
44 energy efficiency; in particular, with more sealed buildings, if an appropriate air change rate is not
45 guaranteed, the Indoor Air Quality (IAQ) may worsen as a result.

46 IAQ impacts the health, comfort, well-being, and cognitive performance of a building's occupants.

47 As up to 90% of the population's time is spent indoors, on average [3], the risks related to being
48 exposed to the different pollutants, such as Volatile Organic Compounds (VOCs), moulds, airborne
49 particles, and inadequate levels of Relative Humidity (RH) are growing. Indoor humidity affects
50 warm respiratory comfort, skin humidity, perceived IAQ, and well-being of occupants [4][5].

51 Moreover, high RH affects the durability of building materials and promotes biological attack of the
52 surfaces, thus increasing maintenance costs [6]. Fungi and micro-organisms are also unhealthy for
53 occupants, as they produce spores, allergens, toxins, and other metabolites that contribute to the
54 degradation of IAQ [7] with consequent allergies, irritations, and respiratory and skin diseases. These
55 negative effects can be short- or long-term, such as the well-known Sick Building Syndrome (SBS).
56 SBS has been recognized by the U.S. National Institute for Occupational Safety and Health (NIOSH),
57 the major cause of which is poor ventilation quality.

58 To settle this problem, the main techniques used at present to improve IAQ are ventilation, source
59 control, and air cleaning [8] by means of active systems. However, thanks to their wide surface of
60 exposure, building materials such as finishes can positively interact in a passive way with the
61 surrounding environment [9], so these materials can help active systems to be more effective, using
62 a lower amount of energy [10]. It has been demonstrated that highly porous materials applied indoors
63 can act as buffers for RH which affects the indoor concentrations of humidity and VOC such toluene,
64 as reported in XX, both numerically and by means of laboratory studies [11] [12].



65 Adsorption is one of the most effective de-polluting processes, in which a gas or liquid is passively
66 removed from a fluid and transferred on the solid surface of the adsorbent, where the adsorbate covers
67 the surface of the adsorbent with a molecular layer. The capturing methods differ in terms of the
68 polarity (electrostatic forces), molecular weight (the higher the molecular weight, the higher the
69 boiling temperature and the easier the deposition on the adsorbent surface), and size (molecular
70 sieves) of the adsorbate and adsorbent. As it is focused on the surface of the solid, the adsorption
71 process requires a high specific surface of the solid adsorbent and a high solid–fluid contact area.
72 Therefore, in order to obtain high removable efficiency of pollutants, very porous materials with a
73 wide specific surface area (of about 300–3000 m²/g) are required as adsorbents.

74 Heterogeneous Photocatalytic Oxidation (PCO) also represents a promising option to ensure healthier
75 environments, exploring the possibilities of the use of nanotechnology in building materials [13][14].
76 PCO is a superficial phenomenon [15], induced when a photocatalytic agent, such as titanium dioxide
77 (TiO₂), is added to a finish—either in bulk or as coating—for the mineralization of adsorbed
78 pollutants into less harmful compounds. This process involves different steps [16][17]: PCO first
79 requires UVA light wavelengths (320–400 nm) to activate the agent, generating a hole/pair. Then,
80 the pollutants are adsorbed onto the TiO₂. PCO capacity is significantly affected by the presence of
81 macro/micro pores [18][19] and hydraulic lime mortars generally have higher de-polluting properties
82 than cement-based ones [16][20][19][21][22], due to the higher presence of micro-porosity; on the
83 other hand, the higher quantity of gel pores (nanopores) than in cementitious products may hide the
84 catalytic sites [18][19][23].

85 To improve the sustainability of finishes [24] [25], biomass by-products (e.g., ashes from the thermal
86 valorisation of biomasses) have been used as unconventional aggregates [26]. Biomass ashes are
87 considered carbon neutral as, in binder-based materials, they bind the same amount of CO₂ released
88 in the combustion process [27]. Ashes typically contain silicon, calcium, potassium, phosphorus,
89 manganese, iron, zinc, sodium, and boron, in the form of oxides, and studies have demonstrated that
90 their use does not negatively influence the ecotoxicity of mortars [28]. Rather, they generally improve
91 the mechanical and durability properties of the resulting concrete/mortar products, while also
92 lowering the release of hazardous elements [29]. “Green” building materials containing biomass ashes
93 able to decrease level of ozone [30] and indoor Volatile Organic Compound (VOC) de-pollution
94 properties due to their adsorption ability [31], have already been successfully developed [31].
95 Biomass ashes also enhance the thermo-hygrometric performance of finishes/renders [32] [33][34].
96 As long as an RH level from 40–70% can be guaranteed, the primary requirements for indoor
97 materials are high transpirability to water vapour, avoiding the storage of humidity, and the ability to
98 act as an hygroscopic buffer by absorbing and desorbing moisture [35]. The capacity of a finish to



99 absorb water vapor during exposure at high levels of RH and to release water vapour when the level
100 of RH decreases is expressed as the Moisture-Buffering Capacity (MBC) [36] [37].

101 This research is aimed at the development and testing of an innovative multi-functional mortar (Italian
102 Patent 102017000033750) for indoor finishes, which is capable of passively improving IAQ, thus
103 benefitting the health and comfort of occupants, in terms of permeability, MBC, de-polluting activity
104 and inhibition of mould growth, besides fulfilling the ordinary requirements. In the developed mix,
105 conventional sand is replaced by volume with biomass ashes and an unconventional aggregate,
106 characterized by high adsorption properties and currently not used in the building sector but, instead,
107 in water/air depuration processes [38]. The interaction with different TiO_2 agents activable under UV
108 and VIS radiation [39], as declared in the product's datasheet, is also considered. If TiO_2 is present,
109 the unconventional adsorbent aggregates give large contacting areas to the catalytic agent which,
110 thanks to their high specific surface, can potentially improve the de-pollution activity of the material.
111 In this way, saturation of the adsorbent material could also be avoided, providing the benefit of
112 maintaining the same efficiency over time [40]. As references, a traditional mortar and two different
113 commercial pre-mixed products for the same indoor application were manufactured and tested.

114

115 2. Experimental program

116

117 2.1. Materials

118 A hydraulic lime, classified as LIC 3.0 according to UNI EN 15368:2010, with a density of 2650
119 kg/m^3 (commercial product Plastocem, by Italcementi) was used as binder.

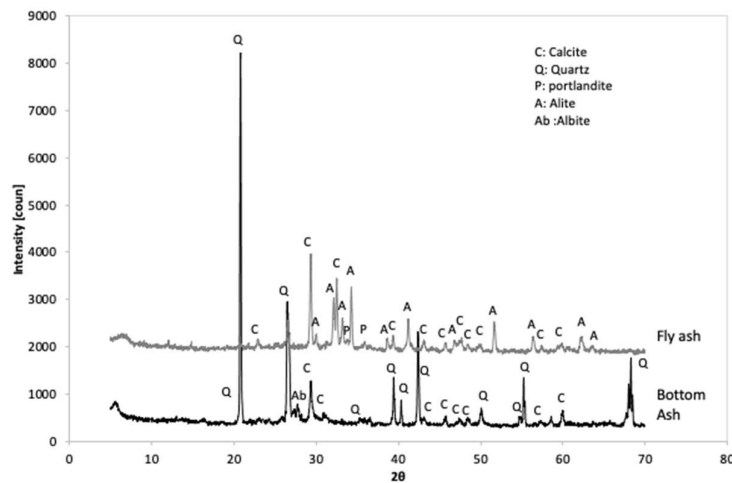
120 The commercial sand, CA 400 (provided by Cava Gola della Rossa, Italy), was chosen as
121 conventional aggregate for the reference mortar, which is a calcareous sand with 98% purity, and
122 water absorption to reach the saturated surface dry (ssd) condition of 5% in weight and ssd density of
123 2650 kg/m^3 .

124 The aggregates used in the patented mix are a porous adsorbent material and fly/bottom ash obtained
125 from a biomass (corn cobs; USA) thermal plant. The porous adsorbent material is currently not used
126 in the building sector, but in water/air depuration processes [38]. It was obtained in the form of
127 granules about 10 mm in diameter, and its water absorption to reach the ssd condition is 86% with
128 corresponding density of 1310 kg/m^3 . Before being added into the mix, it was ground and sieved at
129 $300 \mu\text{m}$, in order to reach a grain size suitable for indoor finishes.

130 Fly and bottom biomass ashes are a mixture of inorganic and organic compounds, due to the presence
131 of unburnt components. The water absorption to reach the ssd condition is 49% and 20%, and the ssd
132 density is 1410 and 1940 kg/m^3 for fly and bottom ashes, respectively. The ashes were chemically

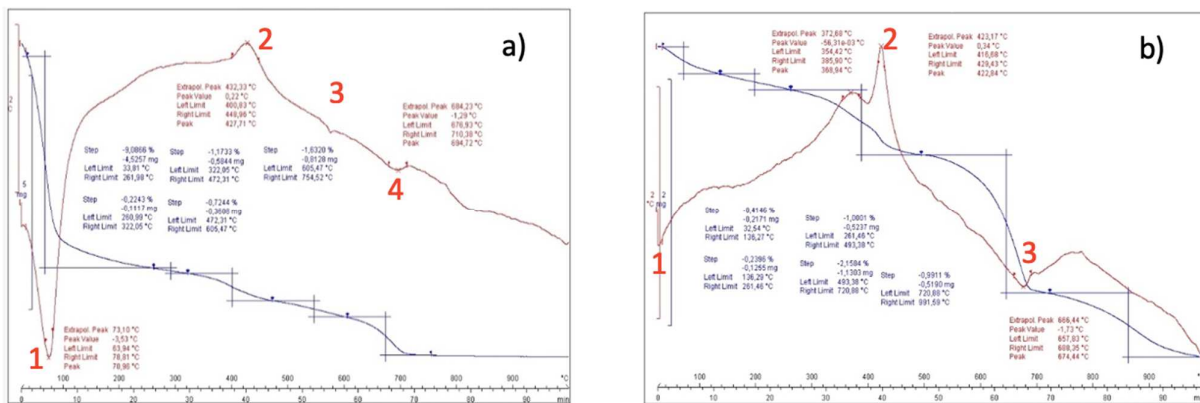


133 characterized by X-ray analysis, using an RX Philips PW 1730 X-ray diffractometer (operating
 134 voltage/current 40 kV/30 mA; scan mode: continuous speed, 3°/min), and by thermogravimetric
 135 differential (TG-DTA) analysis (STA 409 EP) up to a temperature of 1000 °C with a heating rate of
 136 10 °C/min in an oxidizing environment. Ashes were crushed and sieved at 90 µm before the analysis,
 137 in order to provide a homogeneous sample.
 138



139
 140 **Figure 1. X-ray diffraction patterns of biomass fly and bottom ashes.**
 141

142 For both ashes, the most present crystalline phase was quartz, followed by calcium carbonate (CaCO₃)
 143 (Figure 1). In the case of fly ash, the presence of alite was also detected, as well as traces of calcium
 144 hydroxide due to hydration of CaO under environmental humidity. In case of bottom ashes, trace
 145 albite was observed. As the presence of amorphous phases gives a curve baseline in the range of 30°
 146 [41], no amorphous phases were detected in the current ashes.
 147



148
 149 **Figure 2 TG/DTA of fly (a) and bottom ashes (b)**
 150

151 Calcium carbonate was detected also in TG, as an endothermic peak at $T \sim 700$ °C (step 4 in Figure
152 2a; step 3 in Figure 2b), and was quantified as 3.71% and 4.91% of the total weight in fly and bottom
153 ashes, respectively. Moreover, the endothermic peak at $T = 80\text{--}130$ °C (step 1 in Figure 2a, b) was
154 related to the loss of the unbound water. Step 2 in Figure 2a, b, related to an exothermic reaction at T
155 ~ 400 °C, was due to the loss of unburnt organic carbon, quantified as 0.32% and 0.27% of the total
156 weight of fly and bottom ashes, respectively. In fly ash, the presence of $\text{Ca}(\text{OH})_2$ was confirmed by
157 the endothermic reaction (labelled step 3 in Figure 2a) from 470 to 650 °C, and quantified as 2.98%
158 of the total weight. Before addition into the proposed mix, the bottom ash was milled and sieved at
159 $d_{\text{max}} = 500$ μm , providing a grain size suitable for indoor finishes.

160 The effects of three different types of nano- TiO_2 on the de-polluting properties of the new finishes
161 was also considered. The selected products were P-25 Aeroxide® by Evonik (TiO_2A), KRONOClean
162 7000 (TiO_2K), and KRONOClean 7404 (TiO_2J) by KRONOS International Inc., which are activable
163 by VIS radiation. According to the technical data sheet, P-25 is a mixture of anatase–rutile–
164 amorphous phases (78, 14, and 8% in weight, respectively) [42]. The particles had a nano-size of
165 about 20–50 nm. The specific surface, measured by BET, was 35–65 m^2/g . The pH of a 4% water
166 dispersion was 3.5–4.5, and the density was reported as 3.1 g/cm^3 . KRONOClean® is a TiO_2
167 photocatalyst that degrades pollutants both under VIS and UV radiation. As reported in the data sheet,
168 the content of TiO_2 is higher than 97.5%, with a prevalence of anatase phase. The particle size is
169 approximately 15 nm. The specific surface area, measured by BET, was higher than 225 m^2/g , the
170 density was 2.9 g/cm^3 , and the pH range of 4–9 in water dispersion, as declared in the technical
171 datasheet. The main difference between the last two photocatalytic products is the carbon content
172 which, for TiO_2J , was up to 6.1% when measured by EDAX analysis, as shown in [43] and in TiO_2K
173 was 0.7%, measured by means of TGA, evaluated as organic carbon.

174 Boron salts (from Durga) were added as a biocide admixture, to prevent possible biological attack in
175 some specimens of the new mixes.

176 Two different commercial inorganic finish pre-mixes (Commercial 1 and Commercial 2), sold for the
177 same indoor application, were selected and tested for further comparison.

178

179 2.1.1 Mix design

180 According to a previous work [44], the optimum water to binder ratio to reach the best consistency
181 was evaluated as 0.58. Table 1 provides the mix design parameters of the tested mortars. Aggregate
182 was used in ssd condition; in this state, the aggregate does not absorb or release water in the mix,
183 without any changes to the water to binder ratio (w/b).

184



Table 1. Mix design (g/l) of the tested mortars. * KRONOClean 7000 (TiO₂K), and **KRONOClean 7404 (TiO₂J)

Mix	Water	Hy-lime	Sand	Adsorbent	Bottom ash	Fly ash	TiO ₂		Premixed mortar
							P-25	KronoClean	
	g/l	g/l	g/l	g/l	g/l	g/l	g/l	g/l	g/l
Reference (REF)	256	437	1535						
Reference TiO ₂ (REF TiO ₂)	256	437	1535				26		
Commercial 1 (COM1)	286								1360
Commercial 2 (COM2)	408								1360
MIX1	256	437	-	379	281	204			
MIX1 TiO ₂ A	256	437	-	379	281	204	26		
MIX1 TiO ₂ K	256	437	-	379	281	204		26*	
MIX1 TiO ₂ J	256	437	-	379	281	204		26**	

186

187 The two commercial pre-mixes (Commercial 1 and Commercial 2) were prepared by adding the
188 amount of water suggested in the technical data sheets.

189 In presence of biomass ashes, boron salt was added in MIX1, MIX1 TiO₂A, and MIX1 TiO₂J
190 specimens as a water-based solution (active principle of 16%), at the dosage indicated in the technical
191 data sheet (0.06 g for each litre of cast).

192

193 2.1 Methods

194 2.1.1 Fresh, mechanical, and micro-structural properties

195 The fresh properties of mortars were evaluated in terms of workability class. The slump was measured
196 with a truncated conical mould and jolting table, according to the standard UNI EN 1015-3:2007.

197 After 28 days of curing at Temperature (T) of 20 ± 2 °C and Relative Humidity (RH) at 95 ± 5% for
198 the first 7 days and 65 ± 5% for the following 21 days, the hardened properties of mortars were
199 investigated. The compressive strength was evaluated, according to the standard UNI EN 1015-



200 11:2007, in $4 \times 4 \times 16$ cm specimens, by means of a Galdabini hydraulic press with a precision of
201 1%.

202 The density of hardened mortars (ρ , in kg/m^3) was evaluated by weighing and measuring (with a
203 calliper) the dimensions of the different dried specimens. Specimens were considered dry when, after
204 exposure in an oven at $T = 50$ °C, two different weights in a time interval of at least 24 hours did not
205 exceed 0.1%.

206 In hydraulic lime-based materials (as well as cementitious ones), the porosity can be divided as
207 follows: i) gel pores—nanopores from about 0.5 to 10 nm, inside the hydration products; ii) capillary
208 pores—micropores from 10 nm to $5 \mu\text{m}$, mainly affected by the degree of hydration and the w/b ratio;
209 iii) macropores—higher than $5 \mu\text{m}$, due to the entrainment of microbubbles; and iv) the porosity
210 within the aggregate, which is related to the type of aggregate [34]. The pore size distribution of
211 mortars was studied in the pressure range of 0.01–200 MPa by means of Mercury Intrusion
212 Porosimetry (MIP) using a PASCAL 240 (Thermo Fisher Scientific, Waltham, MA, USA). For each
213 type of mortar, three fragments with dimensions of about 1 cm^3 were sampled and tested, and the
214 average results are reported.

215

216 2.1.2 Thermo-hygrometric properties

217 As reported in a previous study [18], small amounts of TiO_2 do not affect the thermo-hygrometric
218 behaviour of mortars. In this case, TiO_2 was added at low percentages (2% by solid weight); thus, the
219 thermo-hygrometric properties, in terms of water vapor permeability, Moisture Buffering Capacity
220 (MBC), and thermal conductivity, were tested only for mixes without TiO_2 .

221 The water vapor permeability was tested according to UNI EN 1015-19:2007, and data were
222 processed according to UNI EN ISO 12572:2007, which evaluates the water vapor resistance factor
223 (μ) in cylindrical specimens ($h = 3$ cm, $d = 13$ cm).

224 The MBC of finishes was assessed on cylindrical specimens ($h = 3$ cm, $d = 10$ cm), by means of a
225 simplified version of the NORDTEST methods [44].

226 The thermal properties were measured in terms of thermal conductivity, according to UNI EN
227 12664:2002, at $T = 20 \pm 2$ °C and $\text{RH} = 50 \pm 5\%$. Equation (1) was used to evaluate the data:

$$228 \lambda = \frac{Jd}{(T_2 - T_1)^2} \quad (1)$$

229 where J is the heat flux (W/m^2), d is the distance between the thermocouples (m), and T_1 and T_2 are
230 the temperatures at the two different sides of the sample (K). For each mix, three specimens were
231 tested and the average values are reported.

232

233 2.1.3 Inhibition of mould growth test



234 Evaluation of the mould growth inhibition effect was performed according to UNI EN 15457:2014
235 using *Aspergillus niger* (DMS 126) which generally grows in humid indoor environments such as
236 bathrooms, and which can cause health problems including allergies and asthma, especially during
237 prolonged exposure. The methodology for quantification of the inoculum was performed as described
238 in previous works [45][46]. The specimens ($5 \times 5 \times 0.5$ cm prisms) were casted and cured for 28
239 days, as described in Section 2.2.1. After the curing period, the pH of the mortars was tested, in order
240 to ensure that the initial basicity of the mortar was lost, as alkalinity generally inhibits mould growth
241 [46]. To guarantee sterile conditions for the inoculum, the specimens were sterilized in an oven at T
242 $= 150$ °C for 2 hours. Then, the inoculum was provided on an agar substrate under a laminar flow
243 hood and the specimens were inserted into petri boxes, in order to maintain sterility outside the hood,
244 and incubated at 24°C.

245 Mould growth was monitored for 28 days under two different conditions: without any irradiance (dark
246 condition) and under visible irradiation for at least 8 hours of exposure per day. As blank tests, three
247 samples of *Aspergillus* grown on sterile filter paper were provided for each condition of inoculum, to
248 verify the vitality of *Aspergillus*. Quantification of the inoculum was conducted once a week for 28
249 days, and the average results of three inocula are reported. To quantify the percentage of colonized
250 area on the surface of the specimens, photos of the specimens were taken and elaborated by means of
251 the ImageJ and GIMP2 software. The results obtained from the inoculum after 28 days are reported
252 and discussed below.

253

254 2.1.4 De-polluting properties

255 The photocatalytic activity of mortars was assessed according to UNI 11247:2009, which measures
256 the activity of inorganic photocatalytic materials in terms of the degradation rate of nitrogen oxides
257 (NO_x) in plug flow condition. The photocatalytic activity (PC) was tested without irradiation (dark)
258 and under both UVA and VIS irradiation and was expressed in terms of the ratio of mineralized NO
259 over total NO_x flux directed in a plug flow reactor. The NO_x flux was provided by a tank at 500 ppb
260 ± 5 NO (SAPIO S.r.l., Monza, Italy), kept constant by mixing with air at $T = 25$ °C and $\text{RH } 50 \pm 10\%$
261 using a dilution system (Calibrator 8188, Rancon Instruments S.p.A., Milan, Italy). In the outlet flux,
262 the concentrations of NO and NO_2 were continuously monitored using a chemiluminescence NO_x
263 analyser (nitrogen oxide analyzer model 8841; Monitor Labs, Englewood, CO, United States).
264 According to the standard, the borosilicate glass photoreactor had a volume of 3 litres and the
265 specimen (cylinder with $d = 8$ cm and $h = 0.8$ cm) was placed in the centre. Then, 20 W/m^2 UVA
266 irradiance, measured by a photo-radiometer (Delta Ohm, HD2102.2, Padua, Italy, equipped with
267 LP471 probe), was provided using a UV 300 W metal halogen lamp, placed 25 cm from the surface



268 of the specimen outside the reactor. VIS light was provided by placing an UV screen protector
269 between the lamp and the reactor. Irradiation of the sample was guaranteed until stable conditions
270 were reached (usually 30 min).

271 The de-polluting property regarding VOCs adsorption was evaluated by means of batch tests, where
272 the VOCs concentration decay was measured over time using a Gas Chromatographer (GC-Carlo
273 Erba Gas 8000 Top). Tests were conducted in dark conditions, and under UV and VIS radiation, in
274 order to evaluate the possible photocatalytic activity. Under UVA radiation, an irradiance of 10
275 W/m²—as measured by a photo-radiometer (Delta Ohm, HD2102.2, Padua, Italy, equipped with
276 LP471 probe)—was provided by a 9 W UV-A lamp (Philips); whereas, for VIS radiation, a 9 W lamp
277 was used (Osram). The de-pollution test was performed on cylindrical specimens (h = 0.8 cm, d = 8
278 cm) following the procedure described in [46], for a total exposed surface of 50 cm². Methyl ethyl
279 ketone (MEK) was used as a model pollutant, injected into a borosilicate glass box where the
280 specimen was inserted before. MEK concentration was assessed each 10 minutes for 120 minutes. A
281 pseudo-second-order (PSO) [47] adsorption kinetic model was used to describe the adsorption
282 kinetics of MEK into the specimen, as described by the following equation:

$$283 \quad q_t = q_e(1 - e^{-k_1 t}) \quad (2)$$

284 where q_t and q_e (mg/cm²) are the adsorptive removal capacity at equilibrium and a specific time (t),
285 respectively, normalized on the sample surface; and k_1 is the adsorption rate constant. The difference
286 between the initial MEK concentration without specimen (C_0 , equal to 2402 mg/m³) and the
287 concentration monitored over the time (C_i) was used to determine q_t . The test was repeated at least
288 three times, and was directly fitted to Equation 2.

289

290 **3 Results and Discussion**

291

292 **3.1 Fresh, mechanical, and micro-structural properties**

293 All the prepared mortars were classified as stiff, according to UNI EN 1015-6:2007.

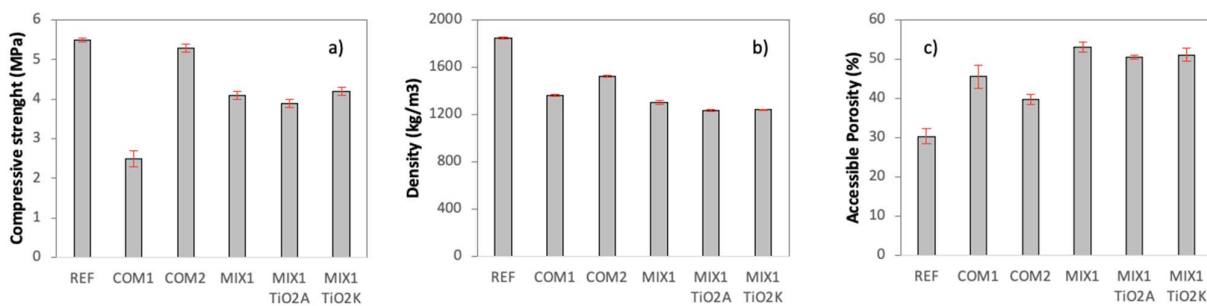
294 The compressive strength, density, and accessible porosity results, evaluated after 28 days of curing,
295 are reported in Figure 3. Both commercial and new mortars had lower compressive strength than the
296 reference mortar. In the case of COM1 and COM2, the compressive strength values were 5% and
297 55% lower than reference mortars, respectively (Figure 3a). MIX1 (with and without TiO₂) showed
298 a decrease in compressive strength of about 25% (when compared to reference mortar) or 20% (when
299 compared to COM2). The reduction of compressive strength in the proposed mix was due to the
300 addition of the very porous adsorbent aggregate [44], even if the addition of biomass ashes facilitated
301 partial recovery of the mechanical resistance loss, thanks to their pozzolanic activity [48][49][50,51].



302 The presence of alite in the ashes, as detected by XRD (Figure1), would favour the formation of
303 calcium silicate hydrates in the binding phase, thus enhancing the mechanical performance. As
304 expected, the mechanical behaviour was not significantly changed with the addition of TiO_2 .

305 According to UNI EN 998-1:2010, the maximum value of density for indoor mortar to be classified
306 as lightweight is 1300 kg/m^3 . As the highly porous adsorbent aggregate and biomass ashes have lower
307 density than conventional sand, the obtained mortars were lighter than the reference and both
308 commercial mortars. As reported in Figure 3b, MIX1 and COM1, classified as lightweight mortars,
309 and the new mortar had lower density than the commercial products and the reference mortars, by
310 about 15% and 35%, respectively, regardless of TiO_2 presence.

311



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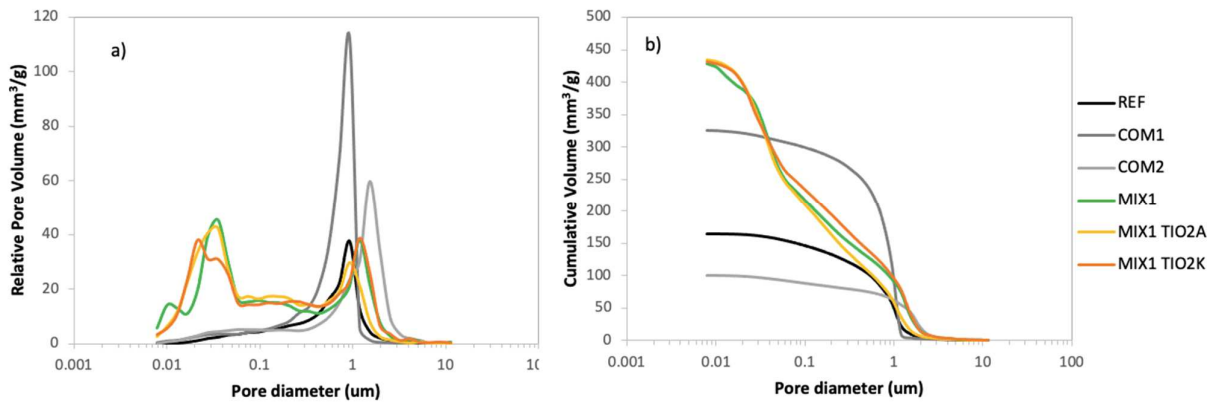
313 **Figure 3. Compressive strength (a), density (b), and accessible open porosity (c).**

314

315 Accessible open porosity was evaluated by MIP (Figure 3c), and both the relative and cumulative
316 pore volume distribution curves are shown in Figure 4. The REF mortar had the lowest total porosity.
317 COM1 and COM2 had 10% and 15% higher total porosity than REF, respectively, whereas the new
318 mortar showed the highest value of total porosity (again, regardless of TiO_2 addition). Furthermore,
319 the pore size distribution was not significantly affected by the type or the presence of TiO_2 , as shown
320 in Figure 4, in which the pore distribution curves of reference, commercial, and multi-functional
321 mortars are compared. REF and commercial mortars present unimodal distributions, whereas the new
322 mortar shows a bi-modal distribution. The main factor usually affecting the porosity of the matrix is
323 the type of binder. In hydraulic lime mortar, the pore diameter threshold—which is the first inflection
324 point of the curve—generally ranges from 1 to 3 μm ; that is, hydraulic lime introduces a higher
325 medium-pore size porosity into the matrix than cement [20]. In this case, the REF mortar had a modal
326 pore diameter of 0.93 μm and a threshold pore diameter of 1.54 μm ; while COM1 and COM2 had
327 modal diameters of 1.60 μm and 0.90 μm and threshold pore diameters of 3.27 μm and 1.20 μm ,
328 respectively. Regardless of the type of TiO_2 , the unconventional mortars all had the first peak at 1.2
329 μm , except for M1 TiO_2A , which was at 0.93 μm . The second peak, at a lower pore diameter, was at
330 0.04 μm for M1 and M1 TiO_2A , and 0.04 μm and 0.021 μm for the mortars with titanium dioxides.



331



332

333 **Figure 4** Relative (a) and cumulative (b) pore volume distribution of different mortars

334

335 **3.2 Thermo-hygrometric properties**

336 As the influence of TiO₂ on hygro-thermal behaviour of mortars at low percentages has been reported
 337 to be negligible [18], only mortars without TiO₂ were tested for thermo-hygrometric properties. The
 338 obtained results, with relative standard deviations, are reported in Table 2.

339

340 **Table 2. Thermo-hygrometric properties of mortars: water vapor resistance factor, moisture buffering value, and**
 341 **thermal conductivity.**

Mix	μ (-)	s.d.* (-)	MBV (g/m ² *RH%)	s.d.* (-)	λ (W/mK)	s.d.* (-)
REF	19.8	1.2	0.15	0.01	0.124	0.016
COM1	14.0	0.2	0.30	0.05	0.096	0.001
COM2	15.8	0.9	0.25	0.01	0.101	0.004
MIX1	11.4	0.2	0.61	0.18	0.083	0.001

s.d. standard deviation

342

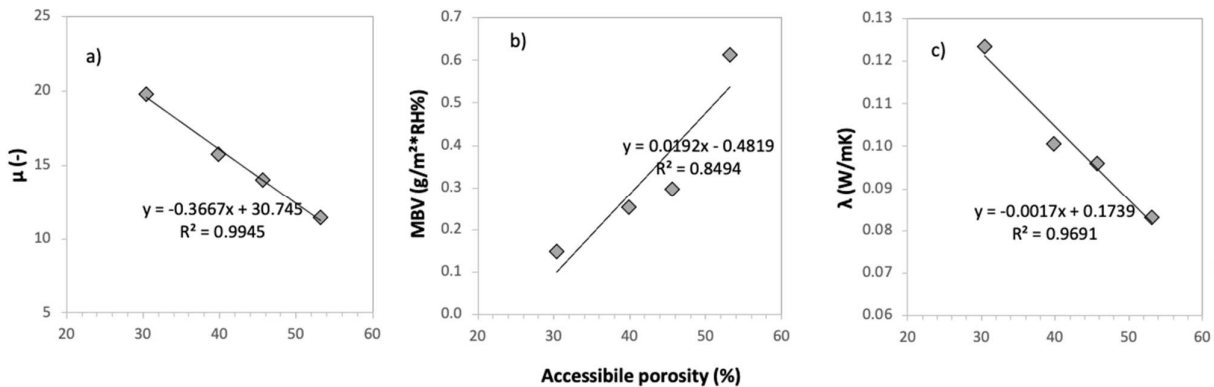
343 Low μ factor values indicate high permeability. REF mortar showed the highest μ values and,
 344 therefore, lower water vapor permeability. Commercial mortars had a μ value lower than that of the
 345 REF mortar: of about 20% for COM1 and 30% for COM2. M1 had the lowest value of μ —40% lower
 346 than REF and about 20 and 25% lower than COM1 and COM2 mortar—and, therefore, the highest
 347 water vapor permeability. Taking into account that the same hydraulic binder was used for all the
 348 tested mortars, the differences were mainly due to the different porosities of aggregates. In particular,

349 the results follow the well-known principle in which a higher pore threshold radius (Figure 4) leads
350 to higher transpirability [34] [52] . The total amount of porosity is also very influential on
351 permeability: in the case of M1, the higher volume of pores, in terms of total porosity (Figure 3c), led
352 to a lower hygroscopic resistance factor, as reported in Figure 5a.

353 The interaction between mortars and the indoor RH was also studied, by measuring the change in
354 moisture content of specimens (Δm) exposed at different RHs. Table 2 shows Δm normalized on the
355 surface of specimens, representing the MBV values. The REF mortar had the lowest ability to
356 exchange water vapor and, consequently, the lowest MBV. Both commercial mortars had a higher
357 capacity to exchange water vapor than the REF mortar, about 100% and 75% for COM1 and COM2,
358 respectively. The unconventional mortar had up to three times higher MBV, compared to the
359 reference mortar. Furthermore, in this case, the result was strictly related to the porosity of the mortar,
360 as a higher total volume of pores implies a higher available mortar surface with large pore volume,
361 providing sufficient space for the adsorbate (in this case, water) to be trapped in [53]. The obtained
362 results confirmed that the higher the accessible porosity, the higher the MBV value (Figure 5b);
363 unconventional mortars had the highest total porosity (Figure 3c) and, therefore, the highest MBV
364 values (Table 2). Moreover, higher permeability usually indicates higher water vapor-exchanging
365 ability [54] [55].

366 Concerning the ability of the materials to contribute to wall system insulation [53], the REF mortar
367 had the highest thermal conductivity, while commercial mortars both had about 20% lower thermal
368 conductivity values than the reference mortar. MIX1 had the lowest value: 35% lower than the REF
369 and about 20% lower than COM 1 and COM 2. Furthermore, in the case of thermal conductivity, the
370 higher the porosity, the lower the thermal conductivity; with a good linear correlation, as shown in
371 Figure 5c.

372



373

374 **Figure 5. Relationships between accessible porosity and water vapor permeability factor (a), moisture buffering**
375 **value (b), and thermal conductivity (c).**

376

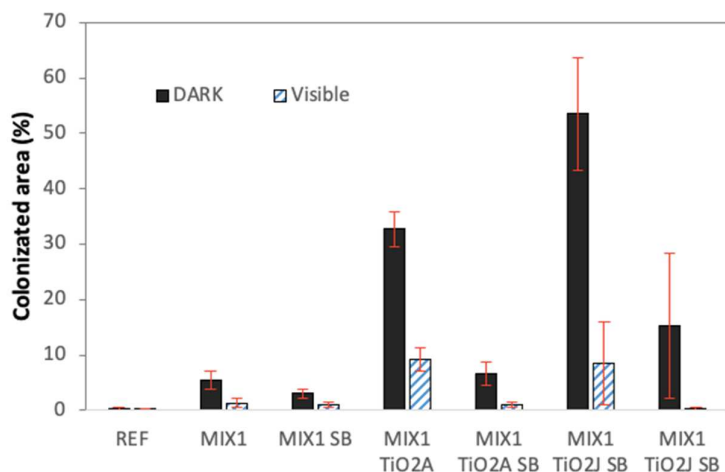
377 3.3 Inhibition of mould growth test

378 REF mortar had the highest resistance to the biological attack under both test conditions [46], as
379 shown in Figure 6, probably due to the absence of any nutrient on the mortar surface (e.g., organic
380 carbon or potassium). P-25 was chosen as benchmark TiO₂ (TiO₂A) and KRONOClean 7404 (TiO₂J)
381 was tested, due to the high presence of organic carbon [43], which creates favourable environmental
382 conditions, in terms of mould growth. Commercial mortars were not tested, as their datasheets have
383 already reported their mould growth inhibition ability.

384 All M1 mortars (without and with TiO₂) were unable to completely inhibit the growth of micro-
385 organisms on their surfaces. The main reasons for this could be the presence of biomass ash, which
386 favours mould growth, as well as the roughness of the surface [56]. The high accessible porosity of
387 the M1, as reported in Figure 3c, also assists in the capillary uptake of the agar nutrient in the mortar,
388 providing a better substrate for mould growth, when compared to REF.

389 The presence of inactivated TiO₂ (under dark condition) increased the susceptibility of mortars to
390 biological attack, with an enhancement in the colonized area up to 6 and 10 times for TiO₂A and
391 TiO₂J, respectively, when compared to M1 without TiO₂. This was probably due to the hydrophilicity
392 of TiO₂: the test was conducted in dark, but the inoculum was provided under VIS radiation, which
393 could be enough to activate the photo-induced hydrophilicity [57][58][44]. In the case of TiO₂J, the
394 higher biological attack could be due to the higher content of the organic carbon [43], which provides
395 additional nutrients for the growth of mould.

396 Therefore, surface treatment of the materials should be considered necessary to prevent biological
397 attack [59] when the control of environmental factors (e.g., irradiation, composition of the material,
398 and humidity) cannot be guaranteed [60]. In this case, the addition of boron salt (SB) relevantly
399 decreased mould growth under dark conditions. In the case of M1 TiO₂A, the colonized area
400 decreased from 33 to 7% and, in the case of M1 TiO₂J, it decreased from 54 to 15%. When specimens
401 were prepared with both TiO₂ and boron salt and tested under VIS conditions, the colonized area
402 became negligible, showing that the well-known antibacterial ability of activated TiO₂ under UV
403 assists the biocidal action of the boron salts [61][62] [63].



404

405 **Figure 6. Percentage of colonized area.**

406

407 3.4 De-polluting properties

408 As expected, NO decomposed only in the presence of TiO₂, as shown by the NO_x abatement factor
 409 and the NO₂/NO removal results reported in Figure 7. The values lower than 5% detected in REF,
 410 COM1, and MIX 1 under UV radiation were considered negligible.

411 The removal of up to 6% of NO in the reference sample was ascribed to photolysis of the pollutant,
 412 and to the sorption and conversion of NO into nitrous acid [64][65].

413 In the case of commercial mortars, the presence of TiO₂ was declared for COM2, and 18% NO_x
 414 removal was assessed under UV radiation. The abatement was detected also under VIS radiation, but
 415 only at a negligible percentage (8%).

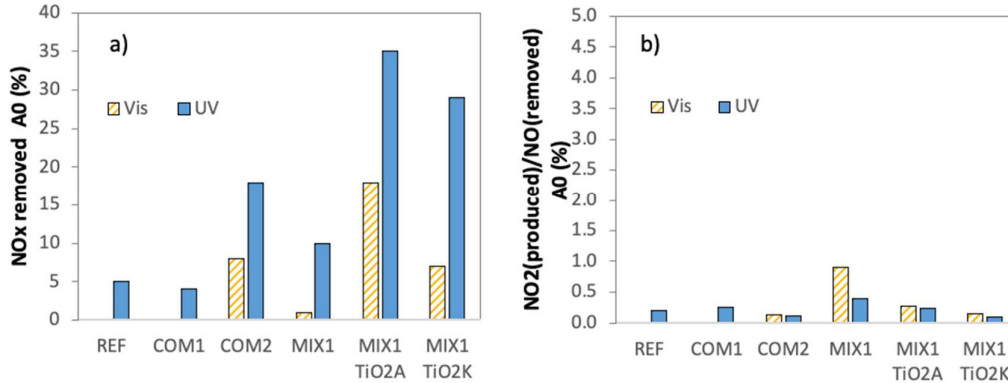
416 Both types of TiO₂ enhanced the photocatalytic degradation of M1 mortar: up to 30% under UV
 417 irradiation. The photocatalytic efficiency was also demonstrated under VIS radiation, but with lower
 418 values (of about 18% and 7% for TiO₂A and TiO₂K, respectively).

419 The ratio between NO₂ and NO was also considered: the higher the ratio, the higher the production
 420 of unwanted NO₂ during the reaction. Figure 7b shows that the production of unwanted NO₂ was
 421 negligible for all the mortars. For M1 mortars, with adsorbent materials as unconventional aggregate,
 422 this can be ascribed to the synergistic effect between adsorption and photocatalysis, as has already
 423 been detailed in previous studies [66][67]. The enhanced reactivity with Ti-OH by NO₂ disproportion
 424 is due to the higher amount of water adsorbed in the proximity of TiO₂ [68] and the consequent
 425 reaction of NO_x with alkaline products [53]. Additionally, the pore structure of MIX 1 with the
 426 addition of TiO₂ enhanced the photocatalytic effect [69]. These pores act as a booster for
 427 photocatalytic activity, as has been stated in [70], in cement mortars with TiO₂. These authors
 428 concluded that the formation of capillary pores in the range of 0.01–0.05 μm was critical for the



429 enhancement of the photocatalytic activity (NO abatement), which has also been demonstrated in
 430 [71], where higher NO degradation was detected in specimens with outstanding increase in capillary
 431 pores.

432



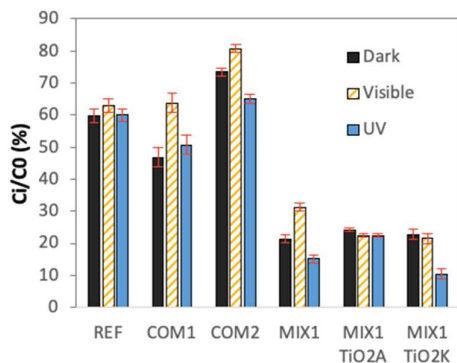
433

434 **Figure 7. Percentage of NO_x removed (a), and ratio between NO₂ produced and NO removed (b) during plug-flow**
 435 **test.**

436

437 During the VOCs de-polluting capacity test, the specimen was sealed inside a box, into which a
 438 known quantity of the model pollutant MEK was injected. Then, the MEK residual concentration was
 439 monitored for 120 mins. The MEK concentrations at the end of the tests are shown in Figure 8.

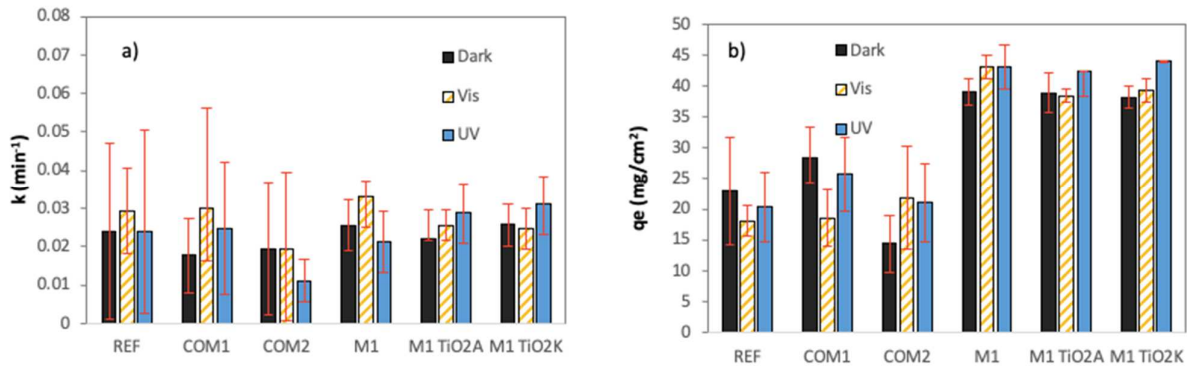
440 The commercial products COM1 and COM2 showed 20% higher and 20% lower de-polluting
 441 properties than REF mortar, respectively. M1 mortar showed a de-polluting capacity about 40%
 442 higher than the REF mortar and both COM mortars. After 2 hours of the test, the residual MEK in
 443 the box was only 20% when a M1 finish was used [38]. The addition, both types of TiO₂ did not
 444 significantly influence the de-polluting capacity under dark conditions. Under VIS radiation, the
 445 effect of TiO₂K was not observed; whereas, under UV radiation, the percentage of residual MEK
 446 inside the box ranged from 11–22%. TiO₂A provided negligible (about 4%) enhancements in de-
 447 polluting capacity under both visible and UV radiation.



448



449 **Figure 8. Residual concentration inside the box after 120 min of monitoring.**



450

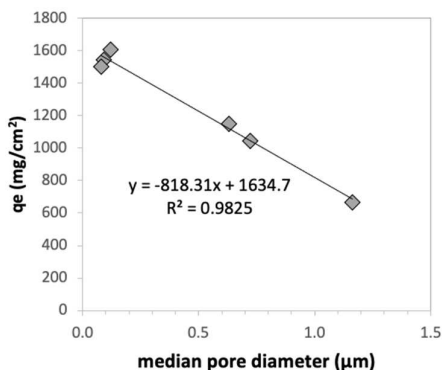
451 **Figure 9. Adsorption rate constant k_1 a) and the adsorption rate constant and specific MEK removal capacity at**
 452 **equilibrium q_e b).**

453

454 The unconventional aggregates added in M1 cause the adsorption process to be predominant over the
 455 PCO, even under visible and UV radiation, as has been shown in [72]. In fact, when the monitored
 456 residual concentration data were represented in terms of adsorption kinetics, in terms of constant
 457 adsorption rate, the results could be classified in two groups with similar values (Figure 9a): one
 458 related to the M1 mortars with adsorbent unconventional aggregate, and the other to the REF and
 459 COM mortars. High errors bars occurred during the elaboration of the data for REF, COM1, and
 460 COM2, as well as when the specific MEK removal capacity at equilibrium q_e was calculated (Figure
 461 9b), highlighting that no significant adsorption mechanism was present in those specimens [73]. On
 462 the other hand, MIX1, MIX1 TiO₂A, and MIX1 TiO₂K had higher MEK removal capacities (approx.
 463 30%) than REF and COM mortars, and the test conditions (UV, VIS, or dark) did not affect the
 464 adsorbent properties.

465 The adsorption efficiency of the finish was extremely enhanced when the adsorbent aggregate was
 466 used and, consequently, a high specific surface interacted with the environment. Moreover, with a
 467 higher pore size reduction, a higher de-polluting capacity was observed [71], with good linear
 468 correlation, as reported in Figure 10.

469



470

471 *Figure 10. De-polluting properties, in terms of specific MEK removal capacity at equilibrium q_e under dark*
472 *conditions.*

473

474 The de-polluting property was higher in M1 mortars than REF and COM mortars, due to the
475 combination of the adsorbent process and PCO. The photocatalyst activity can be enhanced with a
476 high presence of micro–nano-pores [74]: the larger the volume of pores with diameter higher than 80
477 nm, the higher the PCO efficiency [19]. Therefore, the high porosity of M1 mortars allowed for the
478 absorption of pollutants into their internal structure [22]. Moreover, during the test, STD and COM
479 mortars were close to reaching saturation conditions, while the new mortars continued to adsorb and
480 decompose the loaded MEK, as has been detailed in [46].

481

482 **4 Conclusions**

483 An unconventional mortar for use as an indoor finish was designed, characterized, and compared to
484 a traditional reference mortar and two commercial pre-mixes for the same application. In the new
485 finish mortar, an unconventional aggregate—based on an adsorbent material which is generally not
486 used in the building sector, but in air/water purification processes—was adopted, instead of traditional
487 sand. The patented new finish has potentially high sustainability, thanks to the use of hydraulic lime
488 and biomass wastes, instead of cement and conventional aggregates.

489 The innovative mix was able to fulfil all traditional requirements for an indoor finish. The new mortar
490 can be classified as a lightweight mortar ($\rho < 1300 \text{ kg/m}^3$), with density 30% lower than the REF
491 mortar and around 10% lower than COM mortars. This low density was due to the use of
492 unconventional lightweight adsorbent aggregates, which also led to 20%, 10%, and 5% higher
493 porosity, when compared to the REF, COM1, and COM2 mortars, respectively.

494 Regardless of the high porosity and low density, only a 5% decrease in compressive strength, with
495 respect to the REF mortar, was detected; as such, the proposed mortar can be considered acceptable
496 for the proposed application. Compared to commercial products, the compressive strength was 5%
497 lower or around double, according to the type of mortar. The new formulation had thermal
498 conductivity 35% lower than that of the REF mortar and about 20% lower than those of commercial
499 mortars; as well as the lowest water vapor resistance factor (about 40% lower than REF and about 20
500 and 25% lower than the COM mortars). This ensures the highest transpirability, preventing the
501 stagnation of water vapour.

502 In terms of moisture buffering capacity, the best performance was also detected in the new mortar,
503 with three times higher MBV than the reference mortar and the double and 75% higher, when
504 compared to the respective COM mortars.



505 Concerning the de-polluting ability, the patented finish reached the NO_x abatement of 35%, 75%
506 higher than COM mortar with the addition of TiO₂ activated under UV radiation, whereas the
507 adsorbent properties were enhanced by 50% with respect to REF and both commercial mortars.
508 Finally, the behaviour of the innovative mortar, in terms of biological attack, was improved when a
509 biocide was added into the mix.

510

511 **Patent**

512 *Italian Patent 102017000033750*

513

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518

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