

1 The effect of peat and wood fly ash on the porosity of mortar

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11

12 Abstract

13 Fluidized bed combustion fly ash (FBCFA), notably different from regular (coal) fly ash, is a promising industrial side
14 stream to be used as a supplementary cementitious material (SCM). Peat and wood are important sources of biomass
15 for energy production in Nordic countries and generate formidable amounts of un-used ash yearly. Two FBCFAs from
16 the co-combustion of peat and wood, fly ash from coal combustion, and limestone filler were used to replace 10 wt %,
17 20 wt %, and 40 wt % of cement in mortar specimens. The compressive strength, porosity, water absorption, water
18 vapor permeability, and drying shrinkage of the mortars were measured and compared. It was found that in almost all
19 properties FBCFAs outperformed un-reactive limestone filler. Compared to coal fly ash, FBCFAs produced mortars with
20 comparable compressive strength although with higher porosity, water absorption, and water vapor permeability.

21

22 Keywords: biomass fly ash, fluidized bed combustion, porosity, capillary water absorption, water vapor permeability,
23 drying shrinkage

24 1. Introduction

25 Fly ash originating from fluidized bed combustion (FBC) is a significant side stream of energy production that does not
26 have well established reutilization practices yet. The FBC method is suitable for different fuel types with a variable
27 quality, such as biomass, peat, municipal waste and coal. Fluidized bed combustion fly ashes (FBCFA) resulting from
28 renewable sources can be expected to be an available and relevant material in the future. This may not be the case for

29 pulverized coal combustion fly ashes, as societies are striving to reduce CO₂ emissions and their dependency on fossil
30 fuels. It has been already reported that in some regions demand for supplementary cementitious materials is already
31 higher than their production [1]. From the viewpoint of a circular economy, it is important to find a variety of
32 applications to utilize side streams instead of landfilling them as waste. One promising application for FBCFA is to use it
33 as a partial cement replacement material, as several studies have already shown [2–7]. The utilization of fly ashes can
34 also reduce the CO₂ emissions of concrete [1,8,9]. Ideally, fly ash should be utilized near its production site, because it
35 has been studied [1] that long transportation distance of this material can produce enough CO₂ emissions to outweigh
36 the benefits of the utilization.

37 Porosity is an important property of cement-based materials due to its effect on hardened-state properties, including
38 strength, water absorption, and durability. It is well known that partial cement replacement by unreactive fillers can
39 increase the porosity of mortars/concretes as the water to cement ratio (W/C) of the paste increases. However, FBCFAs
40 differ from most traditional fillers and fly ashes due to their characteristics: they usually possess highly irregular particle
41 shape [2,3,5,7,10–16], which can affect the workability and microstructure of the hardened cement paste matrix.
42 Additionally FBCFAs can possess hydraulic properties [3], meaning that material can react with water and produce
43 cementitious reaction products. Fly ashes from the pulverized coal combustion have usually pozzolanic properties,
44 meaning that material contains high amount of reactive silica that can react together with calcium hydroxide to form
45 calcium silicate hydrate, the main reaction product of cement. On the other hand, in addition to pozzolanic properties,
46 fly ashes from pulverized coal combustion can contain also hydraulic properties if material contains enough reactive
47 calcium. These type of fly ashes may demonstrate similar self-cementitious properties as FBCFAs.

48 Only a few studies have investigated how cement replacement by FBCFAs effects the porosity [17–19] of
49 mortars/concretes and, consequently, the other related properties, such as water absorption [20] , water vapor
50 permeability, and drying shrinkage [20–23] . In addition, there are also few studies investigating how cement
51 replacement by FBCFAs affects the properties of air-entrained concrete [20,24,25].

52 It should be noted that almost all previous studies on the aforementioned topics were conducted on fly ashes originated
53 from the combustion of coal, and only one [20] dealt with non-coal derived fly ash. It is still uncertain how well the
54 results obtained by FBCFAs from coal combustion can be applied to FBCFAs from non-coal based fuels. Since it is known
55 that coal-derived fuels often have high sulfur content, which requires limestone injection to eliminate SO_x emissions,
56 the amount of injected limestone can be as much as 30%–50% of burned fuel [26]. It is reasonable to assume that a
57 different fuel source together with a higher amount of injected limestone can have a significant effect on the physical

58 and chemical characteristics of formed fly ashes. Additionally, the effect of fluidized bed combustion fly ash on water
59 vapor permeability has not been studied before. Therefore, there is a need to further study the effect of fly ash from
60 fluidized bed combustion of peat and wood on porosity and properties related to it.

61 This research aims to investigate the effect of cement replacement by FBCFAs from the co-combustion of wood and
62 peat on the porosity, water absorption, water vapor permeability, and drying shrinkage of mortars. Additionally,
63 specimens in which cement was replaced by coal fly ash or limestone filler are prepared and tested as references.

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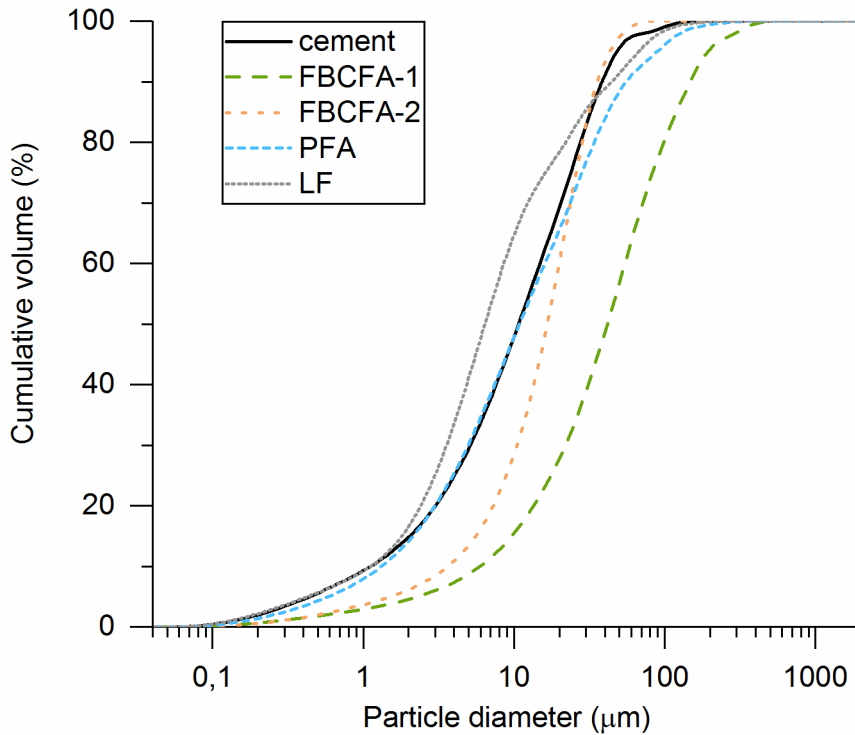
65 2. Materials and Methods

66 2.1 Materials

67 Fly ash samples from the circulating fluidized bed combustion of wood and peat were obtained from two different
68 plants. Both fly ashes had promising chemical characteristics for utilization as cement replacement material, but they
69 differed in particle morphology and bulk density. The fly ashes, referred to as FBCFA-1 and FBCFA-2, were not exposed
70 to humid conditions prior to experiments. Fly ash originating from pulverized coal combustion (PFA) and limestone filler
71 (LF) (CA 100, Cava Gola della Rossa) were used as reference materials for comparison.

72 The type of cement used was CEM II/A-LL 42,5 R (Colacem), according to standard SFS-EN 197-1 [27]. The aggregate was
73 a commercial sand ranging in size from 0 to 3 mm (SABBIA0/3, Esincalce). The used super plasticizer (SP) was an acrylic
74 polymer based (Dynamon SP1, Mapei).

75 Both FBCFAs had slightly higher particles size than cement (Fig. 1). The median particle sizes of FBCFA-1 and FBCFA-2
76 were 39.7 μm and 15.9 μm , respectively, whereas the median particle size of cement was 10.2 μm . The particle size
77 distributions of PFA and LF were really close to that of cement; their median particle sizes were 10.4 μm and 6.1 μm ,
78 respectively.



80

81 Fig. 1. Particle size distribution of cement and replacement materials.

82

83 Both FBCFAs used in this study were mainly composed of Ca, Si, Al, and Fe, but containing also P, S, Mg, and K (Table 1).

84 The content of Cl was 0.1% for both FBCFAs, which satisfies the requirement of fly ash standard EN 450-1 [28]. SO₃85 content of FBCFA-1 was 2.1%, which was below the limit of fly ash standard EN-450-1 (3.0%) while the SO₃ content of86 FBCFA-2 (3.5%) slightly exceeded that limit. P₂O₅ content as well as content of alkalis (calculated as Na₂O equivalent)

87 of both FBCFAs was below the limits of fly ash standard EN-450-1. FBCFA-1 contained 2.5% of free CaO, whereas FBCFA-

88 2 only the 0.1%. Loss on ignition was low for both materials, indicating a low content of carbon unburned. The PFA was

89 composed mainly of Ca, Si, and Al, but it also contained some Fe and small quantities of other elements. Due to high

90 calcium content of PFA, it is likely that this ash contains also hydraulic properties in addition to pozzolanic properties.

91 Loss on ignition was 2.9%. Unsurprisingly, LF seems to almost pure calcium carbonate, since it has CaO content of 97.5%.

92 This conclusion is also supported by the fact that LF has very high LOI value (44.0%) as the calcium carbonate decompose

93 roughly at the temperature of 825 °C. The chemical composition of the cement is typical for cement type CEM II/A-LL

94 42,5 R, consisting mainly of Ca, Si, and Al. The loss on ignition value for cement was 5.2%, which was probably due to

95 the added limestone.

96

97 Table 1. Chemical composition (wt %) of materials.

	FBCFA-1	FBCFA-2	PFA	LF	cement	sand
CaO	16.3	12.0	42.6	97.5	65.5	79.8
SiO ₂	41.8	30.8	33.4	0.2	19.1	17.8
Al ₂ O ₃	13.1	15.1	10.1	0.1	5.0	0.8
Fe ₂ O ₃	13.6	26.7	5.5	0.1	2.5	0.4
Na ₂ O	2.1	1.1	0.8	0.0	0.4	0.0
K ₂ O	2.3	1.6	1.2	0.0	0.9	0.3
MgO	2.5	2.5	2.0	1.0	1.3	0.6
P ₂ O ₅	3.5	4.9	0.3	0.0	0.3	0.1
TiO ₂	0.5	0.4	0.5	0.0	0.2	0.0
SO ₃	2.1	3.5	1.9	0.2	3.5	0.1
Cl	0.1	0.1	0.0	0.0	0.1	0.0
Free CaO	2.5	0.1	NA	NA	NA	NA
Loss on ignition 950 °C	0.3	1.5	2.9	44.0	5.2	6.2

98

99 2.2 Methods

100 2.2.1 Characterization of materials

101 The chemical composition of the materials was analyzed by X-ray fluorescence method (XRF). The analysis was done
102 from melt-fused tablets using a wavelength dispersive XRF spectrometer (AxiosmAX, PANanalytical). The tablets were
103 prepared by melting 1.5 g of the examined material with 7.5 g of X-ray flux Type 66:34 (66% Li₂B₄O₇ and 34% LiBO₂) at
104 1150° C. Loss on ignition at 950° C was determined using thermogravimetric analysis (prepASH, Precisa Gravimetrics
105 AG). The free CaO content of the FBCFAs was analyzed according to standard EN 451-1 [29]. The particle size distribution
106 of materials was investigated using a laser diffraction particle size analyzer (LS 13 320, Beckman Coulter). Measurements
107 were done in wet mode in isopropanol, and the data were analyzed using the Fraunhofer optical model.

108

109 2.2.2 Preparation of mortar specimens

110 The mix design of the reference specimen was based on European cement testing standard EN 196-1 [30] with a few
111 modifications as a bigger batch size, the use of a SP. Instead of CEN-standard sand, different sand product having
112 different chemical composition, particle size distribution, and moisture content, was used. Aggregate sand was
113 composed mainly from calcium containing phases, whereas CEN-standard sand is composed almost completely from
114 silica. Particle size of aggregate sand ranged between 0 to 3 mm, whereas CEN-standard sand ranges between 0.08 and

115 2 mm. Moisture content of aggregate sand varied from 1.8 to 4.4%, whereas maximum moisture content of CEN
 116 standard is 0.2%.

117 In the specimens, FBCFA-1, FBCFA-2, PFA, or LF replaced the 10, 20, or 40% by weight of cement. In the mortar specimen
 118 labelled as FBCFA-1 10%, 10% of the cement was replaced by FBCFA-1; the same logic applies to label the other mortar
 119 specimens. The mix design of the different mortars is presented in Table 2. In order to ensure the same amount of sand
 120 and effective water hydrating cement, the amounts of sand and water were accordingly adjusted in every mix as
 121 reported in Table 2.

122

123 Table 2. Mix design of mortars.

Sample	sand (kg/m ³)	cement (kg/m ³)	water (kg/m ³)	super plasticizer (kg/m ³)	FBCFA-1 (kg/m ³)	FBCFA-2 (kg/m ³)	PFA (kg/m ³)	LF (kg/m ³)	Water/ Cement	Water/ Powder ^a	Sand/Powder ^a
Reference	1516	505	252	1.36	0	0	0	0	0.50	0.50	3.00
FBCFA-1 10%	1512	454	252	1.97	50	0	0	0	0.56	0.50	3.00
FBCFA-1 20%	1509	403	252	2.72	101	0	0	0	0.63	0.50	3.00
FBCFA-1 40%	1501	300	250	4.1	200	0	0	0	0.83	0.50	3.00
FBCFA-2 10%	1514	454	253	3.66	0	50	0	0	0.56	0.50	3.00
FBCFA-2 20%	1515	404	253	7.69	0	101	0	0	0.63	0.50	3.00
FBCFA-2 40%	1516	303	253	9.22	0	202	0	0	0.83	0.50	3.00
PFA 10%	1514	454	252	1.05	0	0	50	0	0.56	0.50	3.00
PFA 20%	1510	403	252	1.29	0	0	101	0	0.63	0.50	3.00
PFA 40%	1505	301	251	1.42	0	0	201	0	0.83	0.50	3.00
LF 10%	1513	454	252	1.38	0	0	0	50	0.56	0.50	3.00
LF 20%	1507	402	251	1.42	0	0	0	101	0.63	0.50	3.00
LF 40%	1504	301	251	1.46	0	0	0	200	0.83	0.50	3.00

^a Powder includes cement and possible cement replacement material

124

125 To prepare mortars, the ingredients were mixed in buckets using a drill. First cement, fillers, water and SP were mixed
 126 for 2 minutes (min). Then sand was slowly added and the mortar mixed for further 4-9 min. To manufacture FBCFA-2
 127 20% and FBCFA-4 40% specimens, an extra amount of SP had to be added to ensure proper workability.

128 After mixing, the workability of the mixtures was evaluated by measuring the diameter of the spread mortar according
 129 to the flow table method described in EN 1015-3 standard [31] with slight modifications. In this method, a truncated,
 130 cone-shaped mold (100 base diameter, 70 mm top diameter, and 60 mm height) is first placed on a jolting table and

131 filled with mortar. Next, the mold is removed and the table is jolted 15 times in 15 seconds. After that, the diameter of
132 the mortar is measured from two directions at right angles, and the average of these measurements is calculated. This
133 procedure was repeated, and, finally, the average diameter was reported. The workability of the mortars was adjusted
134 to give a 12 ± 1 cm spread on the flow table test.

135 Finally, the mortars were cast in molds according to EN 1015-11 standard [32]. Next, the specimens were sealed with a
136 plastic film to ensure a relative humidity (RH) of $95 \pm 5\%$. After 2 days, the specimens were demolded and kept sealed
137 (RH $95 \pm 5\%$) for the following 5 days. After 21 days, the mortars were exposed to an RH of $65 \pm 5\%$. The curing of the
138 specimens was conducted at a temperature (T) of 20 ± 1 °C.

139

140 2.2.3 Properties of hardened mortars

141 The compressive strength of the mortars was measured after 28 ± 1 days of curing, according to EN 1015-11 standard
142 [32] with a hydraulic press (Galdabini).

143 The porosity was analyzed using a mercury intrusion porosimeter (MIP) (Pascal 240, Thermo Fischer). Analysis were
144 performed on 3 fragments of the specimens, originating from 28 days compressive strength measurements.

145 The capillary water absorption and capillary water absorption coefficient (AC) ($\text{kg}/\text{m}^2 \cdot \text{s}^{0.5}$) of the mortar specimens were
146 analyzed according to standard EN 15801 [33]. In this method, a cubic mortar specimen (40 x 40 x 40 mm), sawn from
147 a mortar prism, is placed on a wet bedding layer. The water amount absorbed through the area of the cube's (Q_i) (kg/m^2)
148 face is determined by weighting the specimen after certain time intervals. The AC is determined from the linear part of
149 the plot in which Q_i is presented as a function of the square root of time ($\text{s}^{0.5}$). The AC is a slope of line, which is fitted
150 to the first five data points, which are measured during the first 60 minutes of experiment.

151 A water vapor permeability test was performed according to standard EN 1015-19 [34]. Cylindrical specimens were
152 prepared ($d = 12.5$ cm, $h = 3.0$ cm). After 28 days of curing, the curved sides of the specimens were sealed to a specimen
153 holder so that water vapor, released under controlled conditions by using a saturated solution of potassium nitrate
154 (KNO_3), was able to diffuse only through the flat ends of the mortar specimens. Inside the specimen holder, the RH was
155 $93 \pm 3\%$, and the temperature was 20 ± 2 °C, while outside, the RH and T were $50 \pm 5\%$ and 20 ± 2 °C, respectively.
156 Before the analysis of porosity, capillary water absorption and water vapor permeability, the specimens were dried in
157 an oven at 60 °C until they reached constant mass. The reported results are the average values from three

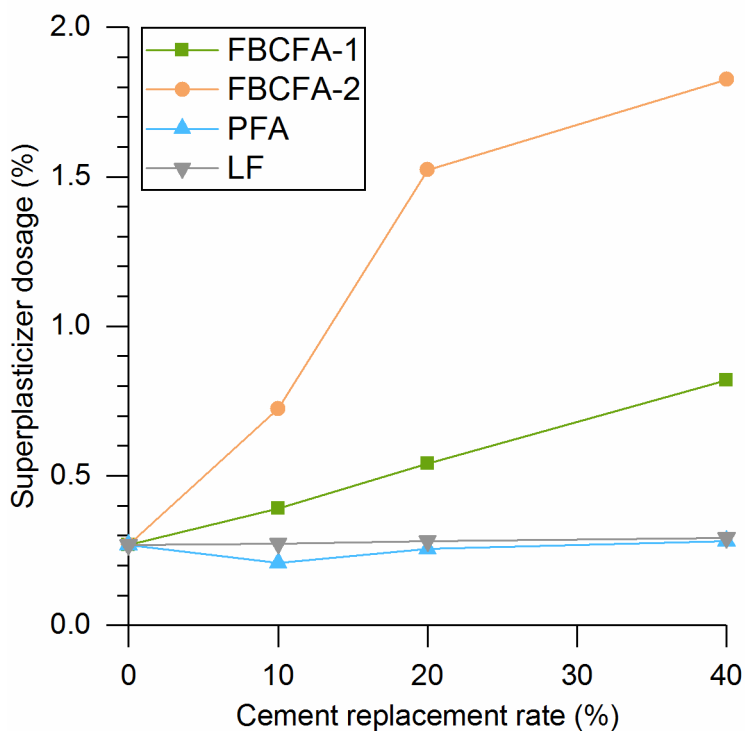
158 measurements. The drying shrinkage and mass loss of the mortars were studied using methods described in standard
159 EN 12617-4 [35].

160

161 3. Results and Discussion

162 3.1 Workability of mortars

163 Fig. 2 shows that both FBCFAs required a much higher dosage of super plasticizer than reference mortar to obtain similar
164 workability and the amount of SP increased with the replacement level. These results are well in line with other studies,
165 which have reported increased water requirement [5,6,10,36–38] or increased SP dosage [7] caused by FBCFAs due to
166 the higher water absorption typical for biomass ash [39,40]. In particular, mortars with FBCFA-2 required more SP than
167 mortars with FBCFA-1. In our previous study [10], ash samples obtained from the same power plants were compared
168 concluding that the share of highly irregularly shaped ash particles varies among ashes affecting workability in different
169 way. Therefore, it is reasonable to assume that FBCFA-2 contains more irregularly shaped ash particles, although this is
170 hard to quantify, than FBCFA-1, which increases SP dosage. The amount of added SP in the mortars containing PFA and
171 LF was quite close to that in the reference mortar indicating that the effect of these materials on workability was low
172 (Fig. 2).



173

174 Fig. 2. Super plasticizer dosage of mortar specimens, as a percentage of powder's (cement and possible
175 replacement material) mass.

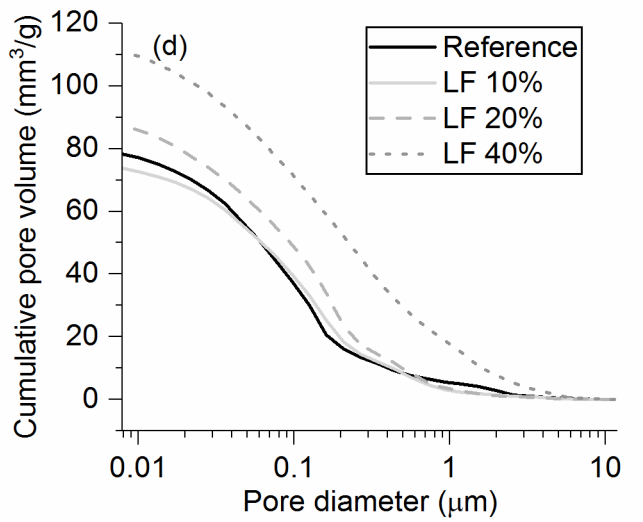
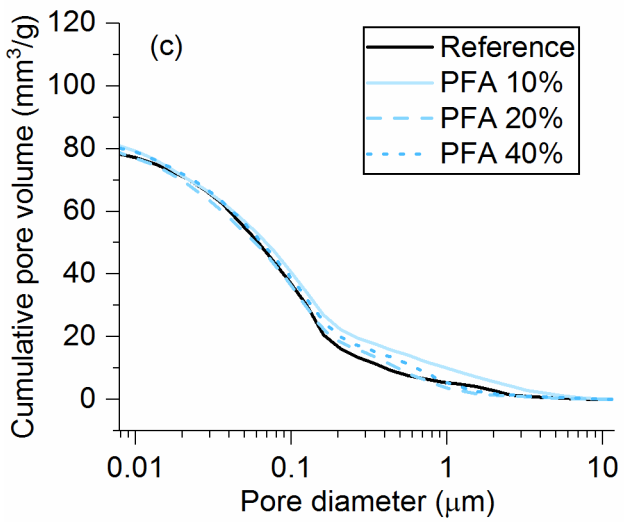
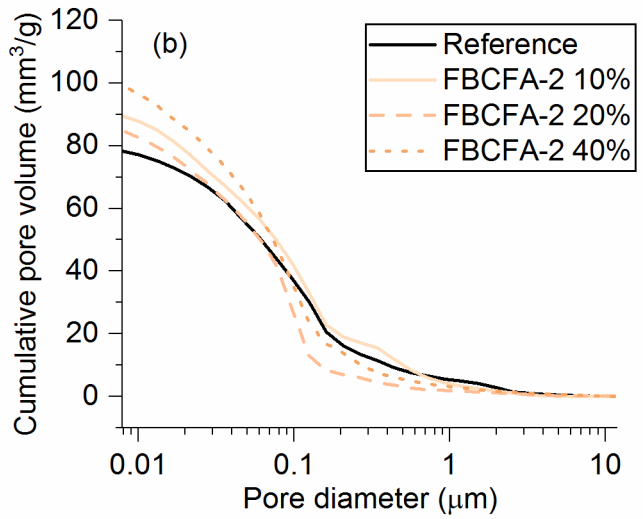
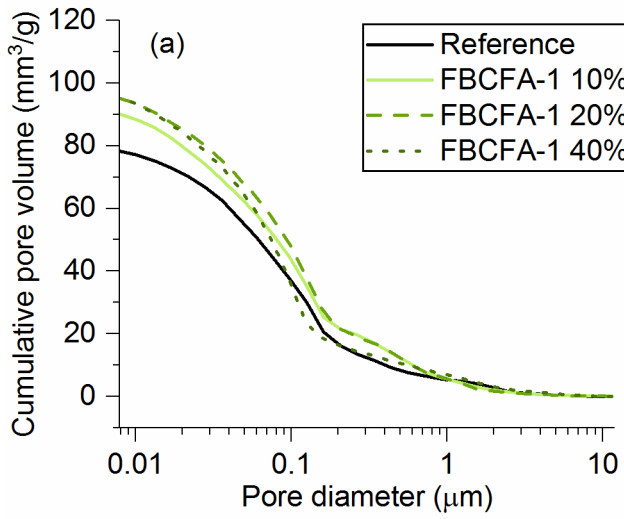
176 3.2 Porosity of mortars

177 The cumulative porosity of the mortar increased compared to the reference specimen when cement was replaced with
178 FBCFAs, but without a clear trend (Fig. 3a and b). The number of pores with $d > 0.1 \mu\text{m}$ (Fig. 4a and b) especially increased
179 in all specimens with FBCFA. Furthermore, in FBCFA-2 20% and FBCFA-2 40% specimens, the number of pores with d
180 $< 0.1 \mu\text{m}$, slightly decreased (Fig. 4b). These results are in line with those of Zhao et al., which also reported increased
181 number of pores in the size range of $0.01\text{--}0.1 \mu\text{m}$ [17] when FBCFA from coal gangue combustion was used as a cement
182 replacement material. Similarly, Lin et al. reported that cement replacement using FBCFA from coal combustion
183 increased total porosity; however, in their case, porosity was especially increased in the size range of $1\text{--}10 \mu\text{m}$ [18]. Also
184 Janowska-Renkas et al. reported significant increase in total porosity when 30 and 45% of cement was replaced with
185 FBCFA from coal [19]. The increased number of small pores in specimens containing FBCFAs could be caused by highly
186 irregularly shaped ash particles. It is possible that these ash particles contain pores, which are not filled with hydration
187 products during the 28 day curing time, as reported for paper ash by [41].

188 The PFA did not have much effect on the porosity of the specimens compared to the reference specimen regardless of
189 the replacement rate (Figs. 3c and 4c). It is possible that because PFA has a similar particle size distribution to cement
190 and smooth particle morphology, it has good filler properties, which mitigates the increase in porosity due to increased
191 W/C. Apparently, as the replacement rate increases, the hydraulic and pozzolanic properties of PFA can still compensate
192 the increase in porosity caused by an increased W/C.

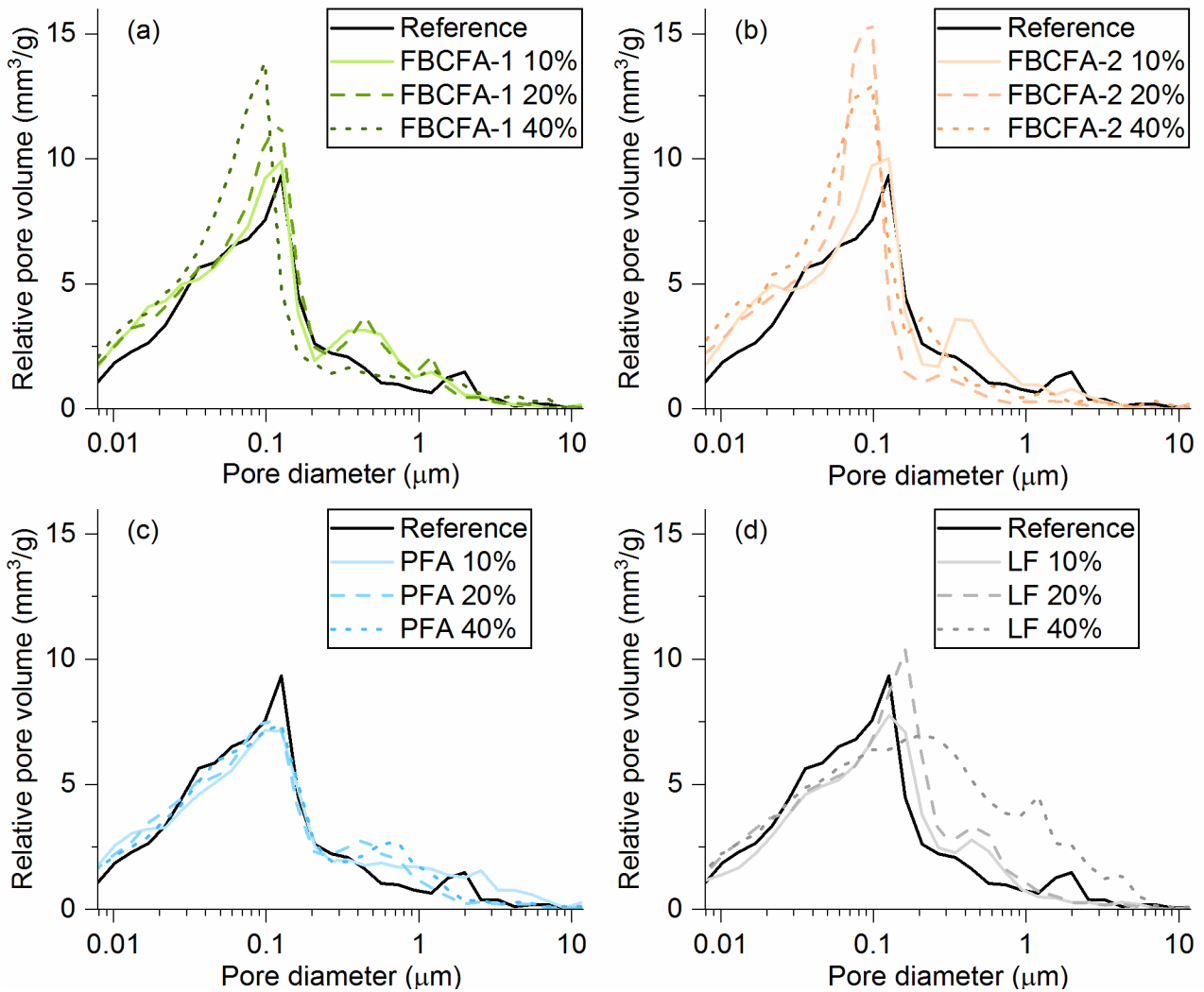
193 When LF replaced 10% of the cement, there was only a small decrease in total porosity. However, when higher
194 replacement rates of 20% or 40% were used, the porosity of the mortars clearly increased (Figs. 3d and 4d) due to the
195 increased W/C. Based on the MIP results, in LF 40% specimen cement replacement especially increased the number of
196 large pores (diameter $> 0.3 \mu\text{m}$), which may have a more detrimental effect on strength properties.

197 It should be noted that the MIP analysis was performed on a pore size range of 8 nm to $11 \mu\text{m}$. This means that small
198 gel pores ($< 8 \text{ nm}$), larger capillary pores ($> 11 \mu\text{m}$), and pores from entrained air were not detected by this method,
199 although there may be differences among the specimens in this pre-mentioned pore sizes, which may affect mortar
200 properties.



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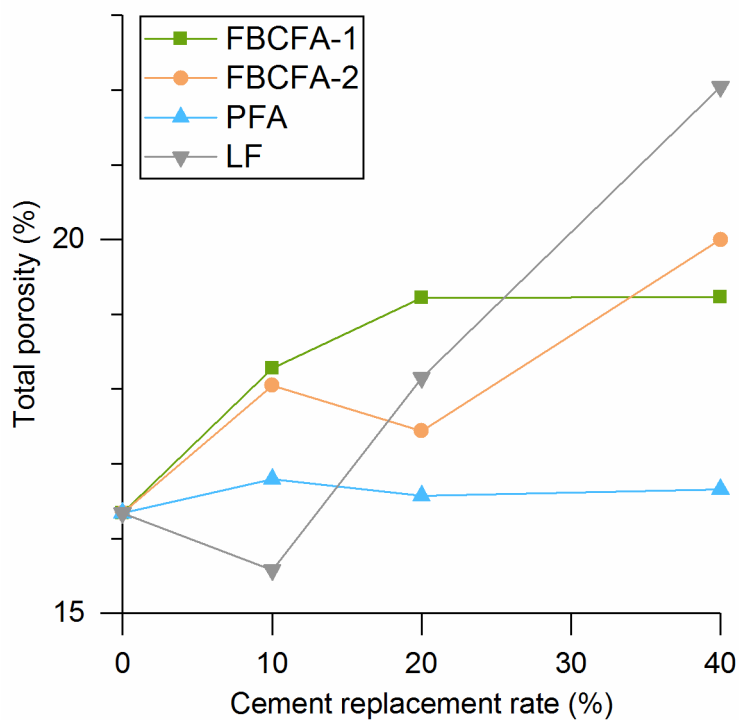
202 Fig. 3. Cumulative pore volume of mortars.



203

204 Fig. 4. Relative pore volume of mortars.

205 Total porosity of mortar specimen (Fig. 5) was in line with cumulative pore volume (Fig. 3). Both FBCFs in generally
 206 increased total porosity as replacement rate increased. There were however few exceptions to this trend. In FBCFA-1
 207 total porosity did not increased when replacement rate was increased from 20% to 40%. In the case of FBCFA-2 total
 208 porosity decreased from 18.1% to 17.4% as replacement increased from 10% to 20%. Increased total porosity has
 209 been also when cement has been replaced using FBCFA originating from combustion of coal gangue [17]. Cement
 210 replacement using PFA did not had significant effect on total porosity (Fig. 5). When LF was to used as cement
 211 replacement material at 10% replacement rate it decreased porosity, but when replacement ratios increased, also the
 212 total porosity increased significantly (Fig. 5).



213

214 Fig. 5. Total porosity of mortars

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216 3.3 Compressive strength of mortars

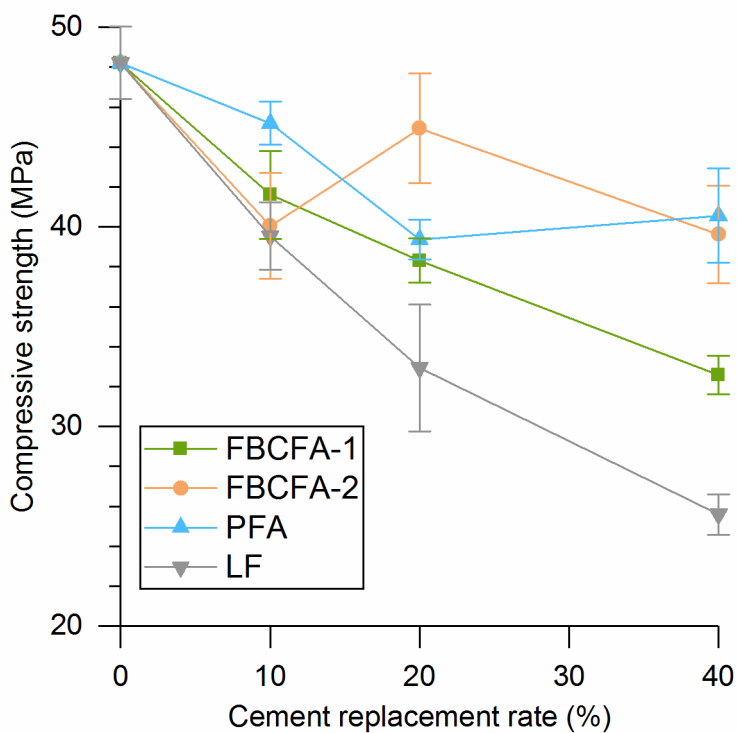
217 Based on the compressive strength measurements, it seems that both FBCFAs are reactive materials and they contribute
 218 to strength development in a way similar to PFA. When more than 10% of the cement was replaced, they clearly
 219 outperformed unreactive LF.

220 FBCFA-1 clearly decreased the compressive strengths as replacement rate increased (Fig. 6). The behavior of FBCFA-2
 221 was more ambiguous; at a 10% replacement rate, strength of the corresponding mortar was at the same level of that
 222 with FBCFA-1 and LF, but when the replacement rate was increased to 20%, strength increased to near the strength of
 223 the reference sample (48.2 MPa). It is possible that good performance of FBCFA-2 at 20% replacement rate was due to
 224 reduced porosity on small pore size. Finally, at the replacement rate of 40%, FBCFA-2 mortar still reached a strength of
 225 39.6 MPa, similar to that of PFA 40% mortar and only 18% lower than that of the reference specimen.

226 Cement replacement by PFA produced lower compressive strength than the reference specimen at every replacement
 227 rate (Fig. 6). From all the PFA-containing specimens, PFA 10% had the best performance, whereas the compressive
 228 strengths of PFA 20% and PFA 40% were slightly lower. Although PFA did not reach compressive strengths as high as the
 229 reference specimen, it clearly had hydraulic properties, since the compressive strengths were much higher than with

230 unreactive LF. In generally total porosity correlated quite well with the compressive strength within different
231 replacement materials, not with PFA. It is possible that hydration products of PFA develops weaker binder matrix they
232 have only limited effect on porosity.

233 The compressive strengths of the mortars containing LF decreased as the share of LF increased (Fig. 6). This was
234 expected because LF is an unreactive filler material, unlike FBCFAs and PFA. The compressive strengths of 10% LF
235 specimens were quite close to other specimens in which the same replacement ratio was used, but when the
236 replacement ratio was higher, strengths were clearly lower than with other materials.



237

238

239 Fig. 6. Compressive strength of mortars after 28 days of hardening.

240

241 3.4 Capillary water absorption of mortars

242 The capillary water absorption curves presented in Fig. 7 can be divided into two parts; the first linear part is related to
243 the filling of bigger capillary pores, whereas the second non-linear part is related to the filling of smaller pores [42–45].

244 The capillary water absorption coefficient (CA) presents the rate of water absorption and is determined from the linear
245 part of the curve. The capillary water absorption curve also shows the total amount of absorbed water, which is related
246 to the total porosity, pore size, and connectivity of the pores [45].

247 Regarding the reference specimen, the capillary water absorption curve had not yet stabilized at the end of the
248 experiment, which means that the specimen had the potential to absorb even more water if the test had continued.

249 With a few exceptions, FBCFAs generally increased the capillary water absorption of the mortars (Figs. 7a and b)
250 compared to the reference specimen, whereas the effect on the water absorption coefficient was more limited (Fig. 8).

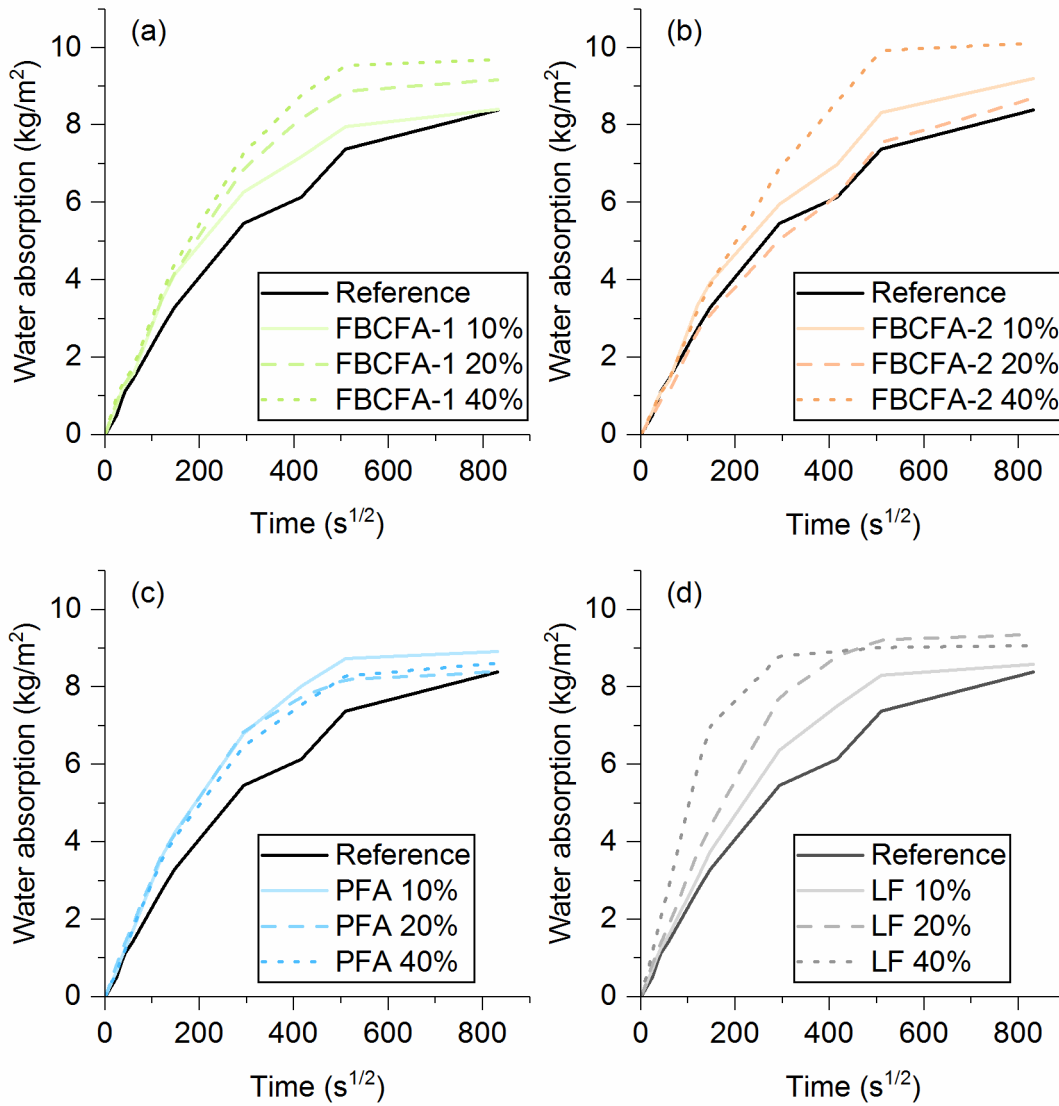
251 FBCFA-1 10% absorbed almost the same amount of water during the experiment as the reference specimen, whereas
252 the total water absorptions of FBCFA-1 20% and FBCFA-1 40% were clearly higher. The mortar specimens containing
253 FBCFA-1 had slightly higher water absorption coefficients than the reference specimen (Fig. 8). The water absorption of
254 the specimens containing FBCFA-2 was similar to the specimens with FBCFA-1 (Fig. 7). The total amounts of absorbed
255 water in FBCFA-2 10% and FBCFA-2 20% were slightly higher than in the reference specimen, whereas FBCFA-2 40% had
256 the highest amount of absorbed water in this study. However, the capillary water absorption coefficients of FBCFA-2
257 10% and FBCFA-2 40% specimens were really close to the reference specimen, whereas in FBCFA-2 20% specimen, the
258 water absorption coefficient was clearly lower (Fig. 8). It is possible that, although FBCFA increases the porosity of the
259 specimens, it had only a limited effect on the water absorption rate because the ashes mainly increased the number of
260 small pores (0.01–0.2 μm diameter). Apparently, these small pores do not have a significant impact on the rate of water
261 absorption. However, with enough time, these small pores were also filled with water, which led to a higher amount of
262 absorbed water at the end of experiment. Results of this study are in line with those of Nagrockienė and Daugėla who
263 studied the effect of cement replacement using 0-30% wood fly ash (combustion method was not mentioned) on water
264 absorption of concrete. They found that up to 15% replacement rate water absorption was slightly decreased. At higher
265 replacement rates ($\geq 20\%$) water wood fly ash slightly increased water absorption [46]. On the other hand results of this
266 study differ from those of Ipatti who studied the effect of cement replacement with peat fly ash and coal fly ash on
267 water tightness of concrete and came to conclusion that peat fly ash slightly improved the water tightness while coal
268 fly ash made concrete slightly less water tight, however differences between specimen were only marginal [20].

269 The effect of PFA on the amount of absorbed water was quite limited, and there was no clear trend between the amount
270 of absorbed water and the replacement rate (Fig. 7c). This result is well in line with the fact that these specimens had
271 quite similar microstructures according to the MIP results (Figs. 3c and 4c). However, the results of the MIP experiments
272 indicated that there could be a small increase in the amount of large capillary pores in these specimens, which could
273 explain the slightly increased water absorption coefficients (Fig. 8c) caused by cement replacement using PFA. Results
274 differ significantly from the study of Hussain et al. in which cement replacement using 0-60% coal fly ash significantly

275 increased water absorption while wood fly ash caused much smaller increase in water absorption with similar
276 replacement rates [47].

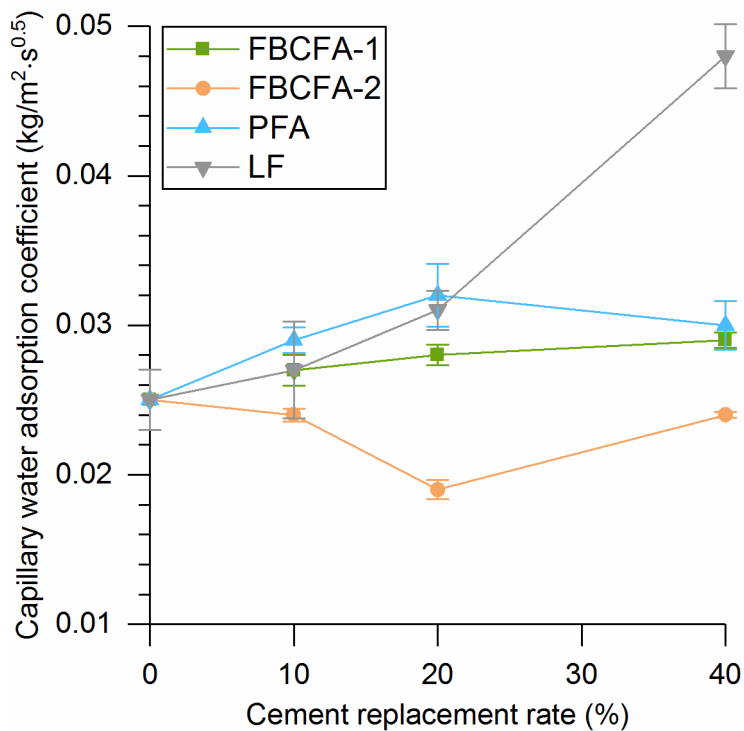
277 Cement replacement using more than 10% LF seemed to increase the water absorption of the specimens; this result is
278 generally in line with the MIP results (Figs. 3d and 4d). However, in the MIP experiment, the difference between the
279 total porosities of LF 20% and 40% was quite high, although the difference in the amount of total absorbed water was
280 almost insignificant. The effect of LF on the capillary water absorption coefficient was clearer; the increase in the
281 coefficient was proportional to the cement replacement rate (Fig. 8d). This is in line with the porosity results (Figs. 3d
282 and 4d), which indicated that cement replacement using LF increased the amount of large pores when the replacement
283 rate was higher than 10%. Apparently, cement replacement using unreactive filler material increases the amount of
284 large capillary pores more than when reactive replacement materials such as FBCFAs or PFA are used. Consequently,
285 this also leads to an increased rate of water absorption.

286



287

288 Fig. 7. Capillary water absorption of mortars.



290

291 Fig. 8. Capillary water absorption coefficients of mortars.

292

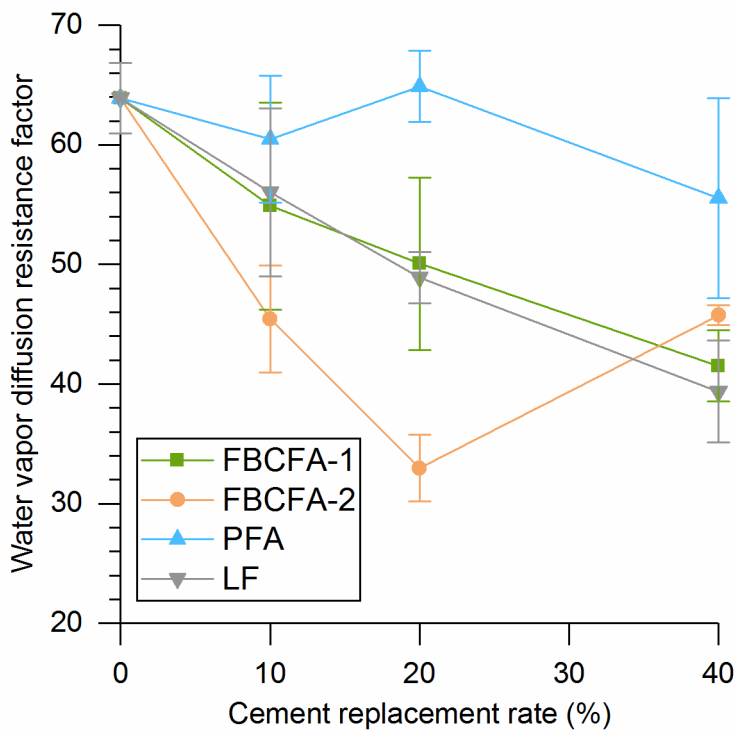
293 3.5 Water vapor permeability of mortars

294 In most of the specimens, the cement replacement materials lowered the water vapor resistance factors compared to
 295 the reference specimen, which indicates an increased permeability to water vapor (Fig. 9).

296 Cement replacement by FBCFA-1 clearly lowered the water vapor resistance factors as the replacement rate increased,
 297 and the results were almost identical with unreactive LF (Fig. 9). In mortars containing FBCFA-1 decreased water vapor
 298 resistance factor correlated quite well with increase in total porosity (Fig. 5). In a similar way, FBCFA-2 decreased the
 299 water vapor resistance factor of the mortar, but there was no clear trend related to the replacement rate; for some
 300 reason, the lowest resistance factor was measured in specimen FBCFA-2 20% (Fig. 9). Additionally, in the case of FBCFA-
 301 2 there was no clear correlation with total porosity. The results could indicate that an increased number of small pores,
 302 observed in mortar specimens in which FBCFAs were used, also contributed to water vapor permeability, meaning that
 303 water vapor can diffuse through material more easily.

304 It seems that the effect of PFA on water vapor permeability was negligible when the replacement rate was 10 or 20%
 305 (Fig. 9). However, when the replacement rate was increased to 40%, the water vapor resistance factor slightly

306 decreased, but it is not clear if this decrease is significant, since deviation between the specimens was quite high.
 307 Apparently, the overall effect of PFA on the pore structure of the mortars is relatively low, since results from the MIP,
 308 capillary water absorption, and water vapor permeability experiments show only a low correlation with the cement
 309 replacement rate. It is possible that reaction products from hydraulic and pozzolanic reactions can fill the pores in the
 310 cement paste matrix and decrease the permeability of the water vapor to the level of the reference specimen.
 311 When cement was replaced using LF, the water vapor resistance factor clearly decreased incrementally as the
 312 replacement rate increased (Fig. 9). Obviously, this was due to increased total (Fig. 5) porosity and pore size, which was
 313 caused by increased W/C.
 314



315
 316 Fig. 9. Water vapor diffusion resistance factors of mortars.

317
 318 3.6 Shrinkage and mass loss of mortar

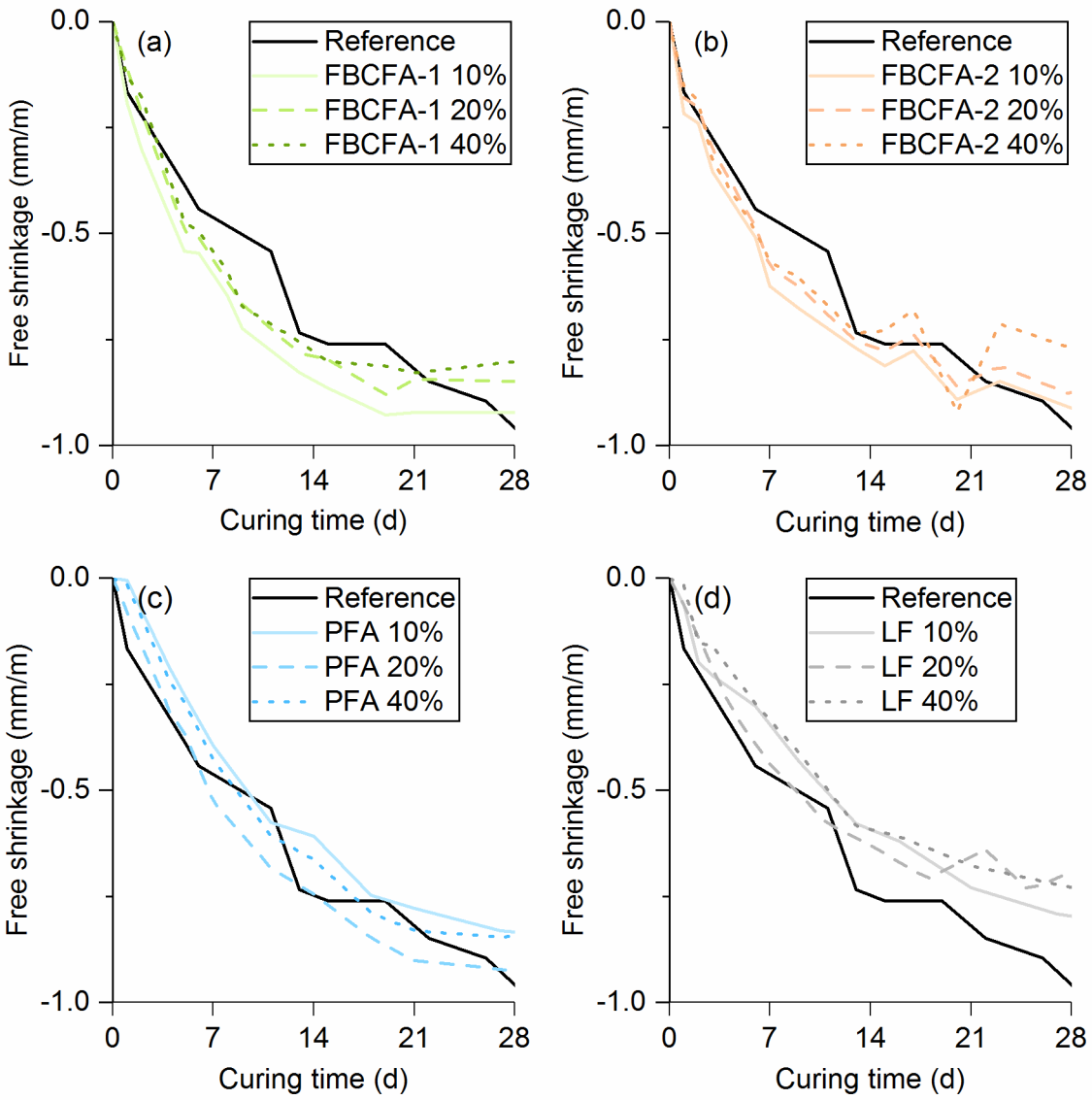
319 FBCFAs and PFA did not had significant effect on shrinkage of mortars whereas LF seemed slightly decrease the shrinkage
 320 compared to reference specimen (Fig. 10). The shrinkage measurements were performed by several operators that may
 321 have induced some variation to measurement data. However, measurement data gives general trend and magnitude of
 322 mortars drying shrinkage.

323 It is well known that drying shrinkage is mainly caused by the loss of physically absorbed water from calcium silicate
324 hydrate. However, the loss of free water from larger pores does not cause significant shrinkage. Only when water is lost
325 from smaller capillary pores, it causes negative internal pressure, resulting in shrinkage. This could explain why the
326 mortars with FBCFAs and PFA exhibited similar drying shrinkage than the reference specimen, although their mass loss
327 clearly increased with an increasing replacement rate (Fig. 11).

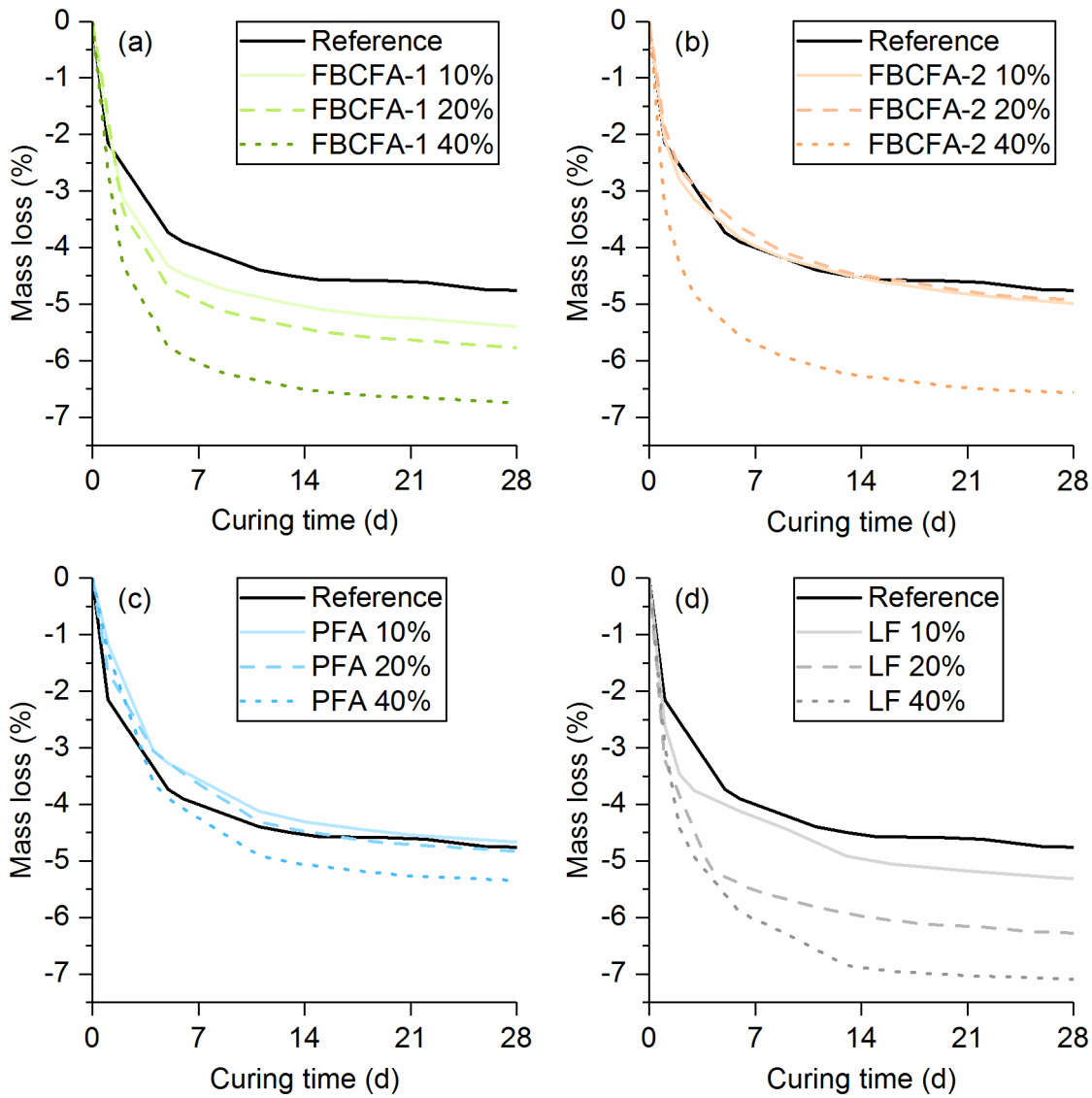
328 In the case of FBCFAs, it seems that the cement replacement using these ashes does not affect significantly drying
329 shrinkage (Figs. 9a and b). Similarly, at the study of Ipatti there was no significant difference between drying shrinkage
330 of reference concrete and concrete in which 37% of cement was replaced by peat fly ash [20]. On the contrary, mass
331 loss seemed to increase as the replacement rate increased. Apparently, FBCFAs react at a much slower rate than cement,
332 which leaves more free water in the material. Free water can probably exit easily from these mortars due to their
333 increased porosity.

334 Similarly, cement replacement using PFA has no significant effect on drying shrinkage. Apparently, pozzolanic and
335 hydraulic reactions caused by PFA lowered the amount of free water, which resulted in mass loss values close to those
336 of the reference specimen, especially at replacement rates of 10% and 20%. These results conflict with those found
337 from other studies in which non-conventional fly ashes are compared to coal fly ash. Ipatti reported that increased drying
338 shrinkage of concrete when 59% of cement was replaced using coal fly ash [20]. On the other hand Wu et al. reported
339 that conventional coal fly ash decreased the length change of mortars significantly. When coal fly ash from circulating
340 fluidized bed combustion (CFBC) was used together with conventional coal fly ash length change increased as the share
341 of CFBC increased [23].

342 When cement was replaced by unreactive LF, the specimens exhibited the lowest shrinkage (Fig. 9d) although their mass
343 loss was the highest (Fig. 10d). These specimens contain more free water than other specimens do because LF does not
344 react with water. The specimens also have high porosity, which may have increased the rate of mass loss. Apparently,
345 cement replacement using LF increases the size of capillary pores, which decreases the drying shrinkage. Additionally
346 cement replacement with LF reduced the cement paste content of specimens, which also reduced shrinkage.



349 Fig. 10. Free shrinkage (mm/m) of mortars.



351

352 Fig. 11. Mass loss (%) of mortars.

353

354 4. Conclusions

355 Untreated fluidized bed combustion fly ashes(FBCFAs) from the combustion of peat and wood could be utilized as
 356 supplementary cementitious material if a sufficient amount of super plasticizer (SP) is used to ensure proper workability,
 357 keeping the water/powder ratio constant. In this study, specimen FBCFA-2 40% had the highest requirement for (SP).
 358 At the water/powder ratio of 0.5, for this specimen SP dosage of 1.8% (from the mass of powders) was enough to
 359 produce mortar that could be easily cast to molds.

360 For replacement up to 10% FBCFAs, the porosity is clearly increased. However, pores induced by FBCFAs had smaller
361 diameters than pores induced by unreactive limestone filler (LF). These small pores increased the amount of capillary
362 absorbed water as well as water vapor permeability. However, these small pores had a limited effect on the rate of
363 capillary water absorption.

364 Despite the increased porosity, the studied FBCFAs increase the mechanical properties of mortars as fly ash from coal
365 combustion (PFA), and both FBCFAs clearly outperformed unreactive LF in terms of compressive strength.

366 Cement replacement using FBCFAs and PFA did not had significant effect on drying shrinkage whereas LF slightly
367 decreased drying shrinkage.

368

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375

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