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1 **Compensation of the negative effects of micro-encapsulated phase change**
2 **materials by incorporating metakaolin in geopolymers based on blast**
3 **furnace slag**

4
5 Bouha EL MOUSTAPHA^{1,2,3,*}, Stéphanie BONNET², Abdelhafid KHELIDJ², Nordine LEKLOU²,
6 Daniel FROELICH¹ , Isselmou AHMEDOU BABAH³ , Carole CHARBUILLET¹, Abderahmane
7 KHALIFA⁴

8 * Corresponding Author

9 ¹Arts & Métiers sciences and technologies —Innovation and Product Design Laboratory, 151 Boulevard
10 de l'Hôpital, F-75013 Paris, France.

11 ² University of Nantes - IUT Saint-Nazaire, GeM, CNRS UMR 6183, Research Institute in Civil
12 Engineering and Mechanics, 58 rue Michel Ange (BP 420), 44606 Saint Nazaire Cedex, France.

13 ³ University of Nouakchott AL-Aasriya , Faculty of Sciences and Techniques/UCME, BP 880,
14 Nouakchott, Mauritanie.

15 ⁴MAGMA group laboratory (www.magma-groupe.com), villa 051 bis-BP 2942- Nouakchott –
16 Mauritania.

17 Corresponding author e-mail: Bouha.el_moustapha@ensam.eu

18 **HIGHLIGHTS**

- 19 • Geopolymer-MPCM mortars have shown better mechanical performance compared to Portland
20 cement-MPCM mortars.
- 21 • The CASH and NASH gels formed after the inclusion of metakaolin also improved the
22 mechanical and thermal performance of the geopolymer-MPCM mortars.

- 23 • Good linear correlations were obtained between compressive strengths on the one hand and
24 Young's modulus and thermal conductivity on the other.

25

26 **Abstract**

27 This study aims to use NASH (sodium alumina silicate hydrate) gel and CASH (calcium alumina
28 silicate hydrate) gel to overcome the negative effects of incorporating microencapsulated phase
29 change materials (MPCM) on the mechanical strength and thermal conductivity of blast furnace
30 slag-based geopolymer mortars. The coexistence of CASH and NASH gels was obtained by using
31 an inclusion of a small amount of metakaolin in a geopolymer matrix based on blast furnace slag;
32 this results in a geopolymer with high mechanical strength. Several tests were performed to
33 characterize different mortars (with and without MK) such as workability, microstructural
34 properties, water porosity, mechanical properties (compressive strength, dynamic Young modulus)
35 and thermal properties (thermal conductivity, the specific heat capacity). The results obtained
36 showed that the coexistence of NASH and CASH gel brought improvements in terms of mechanical
37 properties and thermal conductivity compared to GBFS-based geopolymer-MPCM only. Indeed,
38 the addition of 10 and 20% of metakaolin was sufficient to obtain this coexistence. With a
39 concentration of MPCM up to 10% in the geopolymer mortars, the compressive strength was
40 increased by about 10 MPA and the thermal conductivity was increased by about 31%, which led
41 to an improvement in the specific heat capacity of up to 1280 J/Kg.K. These improvements were
42 due to the high reactivity of MK under the activation conditions used. This favored the good
43 dissolution of silica and aluminum in the MK, which participated well with the calcium in the GBFS
44 to create the NASH and CASH gels. In fact, it was felt that these two gels filled the small pores
45 caused by the MPCM and that this compensated well for their negative effects on the geopolymer
46 matrix.

47 Geopolymer-MPCM based mortars after their optimizations with NASH and CASH gel coexistence
48 showed good workability, compressive strength, and thermal performance. On the other hand, a
49 reduced water porosity compared to Portland cement-MPCM based mortars.

50 **Keywords: Geopolymer; Microcapsule Phase Change Materials; Granulated Blast Furnace**
51 **Slag; Metakaolin**

52 **1. Introduction**

53 The building sector is the world's largest energy consumer, consuming 36% of the world's total final
54 energy and emitting more than 40% of total direct and indirect carbon dioxide (CO₂) emissions [1].
55 Most of the energy consumption in the building is reserved for thermal comfort improvement in the
56 hot and cold seasons (building use phase) [2]. Furthermore, 80% of the greenhouse gas emissions
57 are related to this phase [3].

58 Several researchers in recent decades have studied thermal storage through latent heat in order to
59 reuse it when the building needs the heat (e.g., store heat during the day and use it at night) [4–6].
60 Among the techniques to improve thermal comfort is the use of Microencapsulated Phase Change
61 Materials (MPCM). These materials help to regulate heat exchange in building compartments and
62 reduce the requirement for heating and cooling systems [4–8]. MPCM store at a constant
63 temperature (ambient temperature around human comfort) a large amount of energy in the form of
64 latent heat during their phase change (solid liquid), which prevents the heat flow from entering the
65 building during peak hours. This amount of heat is stored during the day and released at night [9].
66 This technology ensures the desired thermal comfort and reduces the need for air conditioning and
67 heating, as well as reducing CO₂ emissions related to energy consumption in the building. A recent
68 literature review showed that the use of these materials in buildings reduces annual energy
69 consumption by about 50% for cooling and heating [2].

70 After mentioning the enormous energy consumption of the building sector, there is also another
71 problem in this sector, namely is the massive use of Portland cement in concrete. The cement

72 industry is responsible for 7% of the world's CO₂ emissions [10] and causes other pollution in the
73 air, water, etc.

74 The reflection of a strategy to reduce the environmental impacts related to CO₂ emissions and the
75 improvement of the durability of concrete encouraged the researcher Davidovits [11] to invent a
76 new ecological cement (with less environmental impacts) called the Geopolymer. This material is
77 the result of the activation of aluminosilicate materials by alkaline solutions [11]. Aluminosilicate
78 materials are industrial wastes, by-products, or types of clays such as Granulated Blast Furnace Slag
79 (GBFS), metakaolin (MK), fly ash, and red mud, etc. The use of these materials to replace Portland
80 cement will reduce CO₂ emissions and waste caused by the industries, thus helping to reduce the
81 impact on the environment. Wang et al. [12] showed that the CO₂ emissions caused by the
82 production of geopolymer are reduced by about 70 to 80% compared to the production of clinker.
83 Furthermore, geopolymer has several advantages over traditional cement-based materials, such as
84 higher initial mechanical strength, low drying shrinkage, high fire resistance, shorter curing time,
85 higher resistance to acid attack [13–16]. Moreover geopolymer concrete is more resistant to chloride
86 penetration because of its reduced overall permeability compared to Portland concrete [17].

87 However, the incorporation of MPCM in the geopolymer can be a solution to decrease the
88 environmental impacts on two major phases of the building life cycle, which are the building
89 operation phase (energy use to improve thermal comfort) and the cement production phase.

90 Shadnia et al. [18] constructed three small cells of activated fly ash geopolymer mortars. Among
91 these three cells, two contain MPCM. Measurements of their internal temperature showed a
92 reduction of 4.5 and 5.5°C for the two MPCM geopolymer cells compared to the reference cell. Cao
93 et al. [5] numerically investigated the influence of different conditions on the energy efficiency of a
94 wall constructed from geopolymer-MPCM concrete using the finite difference method. With the
95 addition of 5.2% MPCM, there was a reduction in the interior wall temperature of approximately
96 3°C and a reduction in energy consumption of 25% to maintain the interior temperature at 23°C.

97 In contrast, to date, and based on all current research, most studies show that MPCM exhibits a
98 negative effect on the mechanical, physical and thermal conductivity performance of the cement
99 and geopolymer matrix [7,18–20].

100 Cao et al. [7] studied the effect of MPCM addition on the compressive strengths of Portland cement-
101 based concrete and geopolymer-based concrete. Their results show that the presence of MPCM
102 increased the heat capacity of both types of concrete studied up to the value of 1500 (J/kg ° C), but
103 the compressive strengths were reduced by about 42 and 51%. The same observations were reported
104 by Shadnia et al. [18] that cited a decrease in compressive strength of up to 25%. The study by Cao
105 et al. [21] showed that the decrease in mechanical performance is related to the increase in open
106 porosity when the concentrations of MPCM increase in the fly ash geopolymer concrete matrix.

107 Disadvantages of incorporating MPCM into the cement matrix include the rate of decrease in the
108 thermal conductivity of the total cement matrix [22]. Recall that in the case of a material not
109 containing MPCM, its thermal insulation performance is related to its low conductivity, which will
110 delay the penetration of heat into the building. Nevertheless, in the case of a material containing
111 MPCM, the decrease in its thermal conductivity will disrupt the latent heat of MPCM to complete
112 their charge-discharge cycle due to the low heat flux transmitted. This is a problem in this field and
113 has been highlighted by several researchers [23–25]. The effect of reducing thermal conductivity
114 after the addition of MPCM is similar to its effect on increasing porosity (porosity caused by the
115 presence of MPCM in the cement matrix) [22,23]. Several researchers have attempted to improve
116 the thermal conductivity of pure PCM (without capsules) after their incorporation into a cement
117 matrix by using some carbon-based or metal-based materials that have high thermal conductivity
118 [26–29]. Nevertheless, unfortunately, few studies have been conducted to improve the thermal
119 conductivity of a cement or geopolymer matrix after the incorporation of MPCM.

120 The scientific question of this study is how to overcome the negative effects of incorporating MPCM
121 into the geopolymer matrix to improve the mechanical and thermal performances of this new
122 material.

123 Concerning the use of geopolymer, we note that the main gel produced in the geopolymerization
124 process is in the form of a gel called sodium alumina silicate hydrate (NASH), resulting from the
125 activation of alumina and silica found in metakaolin or fly ash [30]. This gel is the result of the
126 chemical reaction between $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$.

127 In recent years, several researchers have shown that it is possible to have in a single geopolymer
128 matrix another hydration gel in addition to the geopolymer gel itself. This gel can be obtained in a
129 matrix that is rich in calcium, silica and alumina to produce calcium alumina silicate hydrate
130 (CASH), and it is obtained from the chemical reaction between $\text{Na}_2\text{O}-\text{CaO}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{H}_2\text{O}$ [31–
131 34]. The CASH gel results from the activation of blast furnace slag [35]. The use of calcium-rich
132 blast furnace slag with the inclusion of a small amount of silica (SiO_2) and alumina (Al_2O_3) rich
133 metakaolin (MK) contributes to a better workability and a structure mainly composed of the
134 coexisting gels mentioned above. This coexistence of gels, once obtained, improves the mechanical
135 performance and durability of the geopolymer [34].

136 However, most of the research that has been carried out on the geopolymer-MPCM is focused on a
137 geopolymer based on a unique base material such as GBFS, fly ash (FA), MK, etc. In addition, there
138 are few studies on combining two primary materials rich in calcium, alumina, and silica at the same
139 time, and no studies have investigated the inclusion of MK into a GBFS-based matrix containing
140 MPCM.

141 The aim of this study is to investigate the feasibility of developing the coexistence of CASH and
142 NASH gels in geopolymer-MPCM mortars to overcome the negative effects of MPCM
143 incorporation on the mechanical and thermal performance of these mortars. This coexistence of gels
144 will be achieved by adding three percentages of MK inclusion (0, 10 and 20%) in a GBFS-based
145 mortar. The geopolymer-MPCM mortars were also compared to Portland-MPCM cement-based
146 reference mortars.

147 Several characterizations of two types of mortar are carried out in this study, such as workability,
148 microstructure properties (SEM, EDS and XRD), water porosity, mechanical properties

149 (compressive strength, dynamic Young's modulus resistance) and thermal properties (thermal
150 conductivity, the specific heat capacity).

151 **2. Materials and experiments design**

152 **2.1. Materials**

153 The MPCM studied here is considered in the form of white spherical microcapsules marketed by the
154 laboratory Microteck-United States. Its technical name is Nextek 28 D. The microcapsule consists
155 of two parts, a polymer wall and the core material (paraffin). Its composition is $\geq 97\%$ paraffin
156 powder. The polymer is a urea polymer, cross-linked with a modified low molecular weight
157 polyethylene. MPCM possesses a melting temperature between 26 and 28°C and a heat capacity
158 between 180 and 190 (J/g).

159 The density was measured by the pycnometer method according to NF T 20-053 and is equal to 0.84
160 g/cm³, almost equal to the density of MPCM in the literature [7,18]. Its thermal conductivity was
161 measured with the Hot Disk 1500 device (the measurement procedure will be explained in the
162 following sections). It is equal to 0.2 W/mK. This value is the same as that found in the literature
163 [25]. MPCM grains range in size from 6.19 to 38.22 μm .

164 The GBFS was delivered by the company ECOCEM in France while the MK was provided by the
165 company KENZAI (ecological materials) in France. The cement used is CEM II (32.5). The
166 chemical compositions and physical properties of cement II, GGBS and MK are presented in Table1.

167 Table 1. Chemical composition and physical properties of CEM II, GBFS and MK.

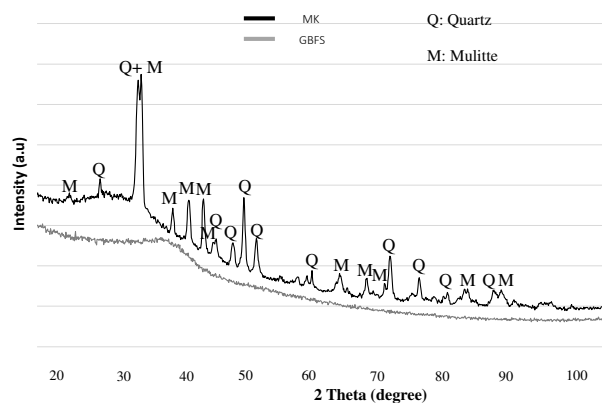
Chemical composition	CEM II	GBFS	MK
(%)			
SiO ₂	7,47	37,3	55
Al ₂ O ₃	2,18	10,7	41
Fe ₂ O ₃	2,84	0,2	1,2
CaO	69,02	43,0	0,1

MgO	-	6,5	0,2
TiO ₂	-	0,7	0,4
(Na ₂ O + K ₂ O)eq	-	0,8	1,8
Specific gravity	3.03	2.90	2.40
Blaine specific surface area (m ² /g)	0.37	0.445	17
average grain size (μm)	8.47	13.25	7.13

168

169 According to Table 1, GBFS is composed of a sum of 48% SiO₂ and Al₂O₃ and a high content of
 170 42.3% CaO while metakaolin MK is rich in SiO₂ and Al₂O₃ with a sum of 96%.

171 The mineralogical structure is shown by X-ray diffraction (XRD) in Figure 1. MK shows a sharp
 172 crystalline peak at 32°, which is attributed to the presence of amorphous quartz structure (SiO₂) and
 173 mullite crystalline phases (Al₆Si₂O₁₃). GBFS is composed mainly of an amorphous phase.

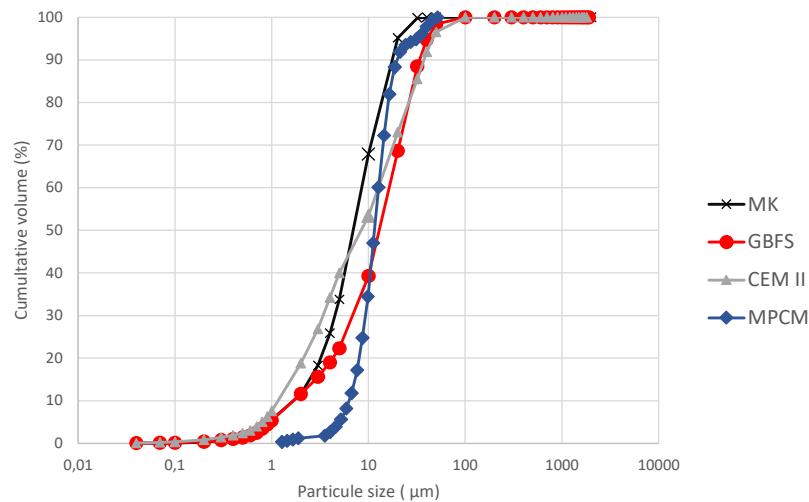


174

175 Figure 1. XRD patterns of MK and GBFS.

176 Figure 2 shows the particle size distribution of GBFS, MK, CEM II and MPCM, which was determined
 177 using a laser granulometer type Cilas 1190. The measurements were performed using the wet method.
 178 Indeed, we had doubts about the fact that water would not disperse MK particles, but the results of the
 179 measurements are close to the results quoted in the study of Kamath et al. [35].

180 It is also noted that MK has finer particles than GBFS with an average particle size that is equal to 7.13
181 μm while the average particle size of GBFS and cement is equal to 13.25 μm and 8.47 μm . On the other
182 hand, their specific surface areas are equal to 17; 0.44; and 0.37 m^2/g .



183

184 Figure 2. Particle size distribution of MK, GBFS, CEM II and MPCM.

185

186 The activation solution is a mixture of sodium silicate and sodium hydroxide. According to the supplier,
187 the composition by mass of the sodium silicate is 27.53% SiO_2 , 11.47% Na_2O and 61% H_2O . Sodium
188 hydroxide NaOH is a caustic soda of 98% purity. The two solutions were provided by the company
189 E2EM in France.

190 A standardized sand (CEN NF 196-1). This type of sand is generally used for laboratory tests. The
191 supplier is the same as for two alkaline solutions. The purpose of using standardized sand is to eliminate
192 the secondary effects on the binder of unfavorable impurities that natural sand may contain.

193 2.2. Mixing method and curing condition

194 Twelve formulations were investigated in this study; three were based on standardized mortar, and nine
195 were based on geopolymer mortar. The water/binder ratio of 0.5 and the sand/binder ratio of 3 have been
196 fixed for the two types of mortars, knowing that the equivalent binder for geopolymer (equivalent to
197 cement) is considered as the total of GBFS, MK, and alkaline solution (solid part). For this, the mass

198 ratio (GBFS+MK)/SA was set to 3, SA corresponding to the solid part of the alkaline solution. These
199 choices are recommended by the study of Hasnaoui et al [14].

200 MK will replace part of the GBFS in the geopolymer matrix with three substitution rates (0, 10 and
201 20%). Bernal et al. [34] cited that increased MK content resulted in significant reductions in compressive
202 strength, due to the incomplete reaction of MK in the activation solution used. In fact, the optimal
203 parameters were determined in this study to provide a high alkalinity of the activation solution to get
204 good reactivity of the MK. Regarding this observation, a mass ratio of 2.5 between sodium silicate (SS)
205 and sodium hydroxide (SS/NaOH) was set according to the study of Bernal et al. [34].

206 The sodium hydroxide solution was prepared with a concentration of 12M and mixed with the sodium
207 silicate and allowed to cool for 24 hours before the mixing procedure.

208 A high concentration of NaOH (above 14M) can disrupt the dissolution of calcium in the matrix [36],
209 resulting in an absence of CASH gel, while a low concentration of NaOH does not allow the dissolution
210 of silica and aluminum in GBFS and MK, while the latter require sufficient Na^+ to be dissolved. Huseien
211 et al. [37] carried out a study on the effect of NaOH concentration in a geopolymer matrix. They
212 indicated that the mechanical properties and microstructures of the geopolymer were improved with a
213 concentration of 12M as chosen in this study.

214 Regarding MPCM, three concentrations were chosen of MPCM in the two types of mortar (cement-
215 based and geopolymer) such as 0; 5 and 10. The method chosen for the incorporation of MPCM in the
216 geopolymer is the method of replacement of sand by MPCM. This method is the most used in this field
217 and, according to the study by Pilehvar et al. [38], its use allows the least reduction of the mechanical
218 performance.

219 Table 2 illustrates the mixing proportions in kg/m^3 . The first three formulations are based on
220 standardized cement mortar while the last nine are based on geopolymer.

221 The quantity of water in the two alkaline solutions is considered to have a water/binder ratio equal to
222 0.5.

223 Table 2. Formulations of cement II mortars and geopolymer mortars (kg/m^3).

224

225

226

Sample	Mixtures proportions (kg/m ³)								
	Cement	GBFS	MK	Sand	MPCM	Na ₂ SiO ₃	NaOH	MGP water	MCII water
MCII _{0/0/0}	585,9	-	-	1757,8	-	-	-	-	293,0
MCII _{0/0/5}	585,9	-	-	1673,8	28,4	-	-	-	293,0
MCII _{0/0/10}	585,9	-	-	1582,0	56,8	-	-	-	293,0
MGP _{100/0/0}	-	439,5	-	1757,8	-	267,9	127,7	42,6	-
MGP _{100/0/5}	-	439,5	-	1673,8	28,4	267,9	127,7	42,6	-
MGP _{100/0/10}	-	439,5	-	1582,0	56,8	267,9	127,7	42,6	-
MGP _{90/10/0}	-	395,5	43,9	1757,8	-	267,9	127,7	42,6	-
MGP _{90/10/5}	-	395,5	43,9	1673,8	28,4	267,9	127,7	42,6	-
MGP _{90/10/10}	-	395,5	43,9	1582,0	56,8	267,9	127,7	42,6	-
MGP _{80/20/0}	-	351,6	87,9	1757,8	-	267,9	127,7	42,6	-
MGP _{80/20/5}	-	351,6	87,9	1673,8	28,4	267,9	127,7	42,6	-
MGP _{80/20/10}	-	351,6	87,9	1582,0	56,8	267,9	127,7	42,6	-

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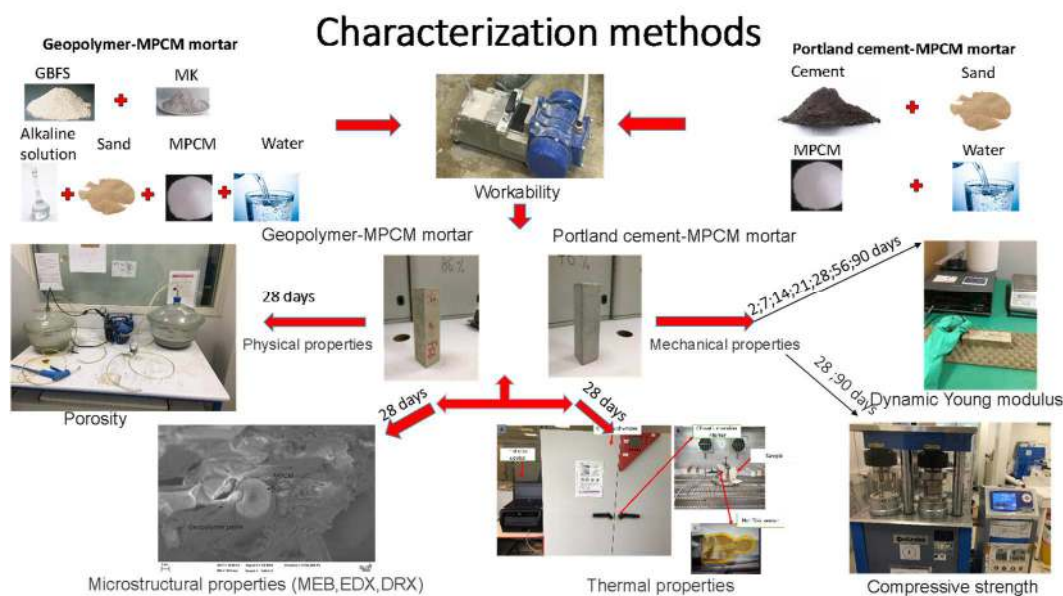
228 The standard NF 196-1 [39] was followed for the fabrication of standardized cement mortars, which
 229 were chosen as a reference for comparison with geopolymer-based mortars. However, regarding the
 230 mixing of geopolymers, GBFS and MK were mixed with alkaline solution and water for 90 seconds to
 231 produce a homogeneous paste [19,20]. The mixing was followed by the addition of sand and mixed for
 232 5 minutes and finally MPCM was added and mixed for 2 minutes.

233 The idea of increasing the mixing time is recommended by Ahmari et al. [40] and is intended to allow
 234 the efficient dissolution of silica, alumina, and calcium (Ca) in the alkaline solution. After the mixing

235 procedures, standardized cement mortars and geopolymers were used for the measurement of
 236 workability. After this step, the casting is done in specimens of $40 \times 40 \times 160 \text{ mm}^3$ and vibrated using
 237 an impact table and are stored in an air-conditioned room (temperature $20 \pm 1^\circ \text{C}$ and relative humidity
 238 $50 \pm 5\%$) for 48 hours before demolding. All samples except the standardized cement mortars (water
 239 conservation) were covered with plastic films.

240 2.3. Experimental methods

241 In this research, all the experiments tests were carried out at room temperature, i.e., between 19°C and
 242 21°C , excepted the thermal properties (see dedicated item). All the tests, including age of mortars, are
 243 listed in Figure 3. The samples used have the same dimensions ($40 \times 40 \times 160 \text{ mm}^3$) for mechanical
 244 tests, water porosity and thermal properties determination.



245

246

Figure 3. Characterization methods

247 The workability test was carried out directly after mixing according to standard NF P18-452 [41] using
 248 a workability meter. The test consists in measuring the flow time of mortars under the effect of the
 249 vibration caused by the vibrator. The water porosity was determined according to the NF P 18-459
 250 standard [42].

251 Morphological analyses were performed using a ZEISS CVO®4 scanning electron microscope (SEM)
 252 combined with a BRUKER marker energy dispersive X-ray spectroscopy (EDS) analyzer. The analyses

253 were performed on fractured samples after 28 days of curing. Indeed, the geopolymer samples were
254 placed in the oven at 60°C to limit hydration before the SEM analyses. XRD analysis was performed on
255 samples ground to powder using a mortar pestle, then sieved using an 80 µm sieve after 28 days of
256 curing. Hydration was then stopped using an acetone quench. Measurements were performed using a
257 SEIFERT MZ VI E X-ray diffractometer with Co K α radiation and a constant scanning speed in the
258 range $2\theta = 15^\circ$ - 115° .

259 Compressive strengths were performed according to the NF EN 196-1 [40] standard. These tests were
260 carried out on six samples to ensure good repeatability. Dynamic Young's modulus measurements were
261 performed following the ASTM E 1876-01 standard [43] and using the Grindosonic apparatus. These
262 measurements were performed on three samples of each formulation at 2, 7, 14, 21, 28, 56 and 90 days.
263 The technique is based on pulse excitation of the vibrations. This modulus is calculated with the use of
264 the equation 1 and by using the flexural frequency (f_f) obtained during the measurement with the
265 dimensions and weight of the samples:

$$E = 0.9465 \times \left(\frac{m \times f_f^2}{b} \right) \times \left(\frac{L^3}{t^3} \right) \times T_1 \quad (1)$$

266

267 With E, f_f , m, L, b, t represent the Young's modulus (Pa), flexural frequency (Hz), mass in (g), length
268 in (mm), width in (mm) and thickness in (mm) of the sample. T_1 represents correction factor that
269 accounts for the finite thickness of the bar, Poisson's ratio, and so forth.

270 One of the main objectives of this work is to evaluate the thermal properties of the materials studied
271 here to determine their thermal performance so that they can be used in the future in a real application.

272 In the research field, there are two techniques for measuring the thermal properties of a material; namely,
273 the steady-state measurement technique and the non-steady-state (transient) measurement technique
274 [44]. Transient measurements have attracted a great deal of research because of their speed and accuracy.

275 Among these is the hot disk technique which uses the transient planar source technique (Hot Disk TPS).

276 The advantage of this method is that it can measure the thermal properties of a wide range of materials
277 and measure the thermal conductivity in the range of 0.005 to 500 W/m.K.

278 The Hot Disk method can measure three thermal properties at the same time, such as thermal
279 conductivity, thermal diffusivity, and specific heat capacity, using the TPS sensor, which has two roles
280 (heat transmitter and temperature measurement receiver) [45].

281 Nevertheless, thermal property measurements with the TPS Hot Disk are often performed in an
282 environment where the temperature is ambient (between 20 and 22°C). This is not possible in our case
283 due to the melting temperature of the MPCM used in this study (between 26 and 28°C). Given this
284 constraint, the measurements were carried out in a climate controlled chamber with a temperature range
285 between 15 and 40°C and a constant humidity of 50%. The climatic chamber used is the KBF 720 type.
286 Its temperature ranges are between 0 and 70 ° C while the relative humidity ranges are between 10 and
287 80 %.

288 Figure 4 shows a picture of the climatic chamber and the hot disk device with the measurement samples.



289

290

291 Figure 4. Thermal measuring device, A: climatic chamber and hot disk; B: interior view of the
292 climatic chamber; C: the measuring sensor

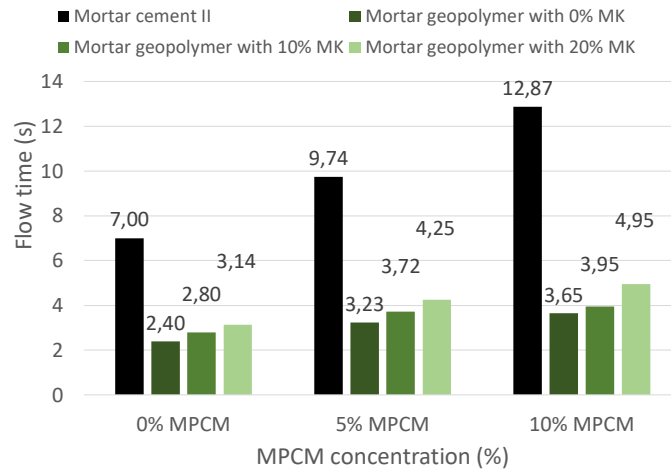
293 The thermal conductivity is influenced by the moisture content of the measurement samples. Due to this
294 influence, the measurements were carried out on samples dried at 60 °C until they had a constant mass.
295 This drying method was recommended by Zhang et al. [46]. Thermal measurements are performed by
296 placing the sensor on the upper surface of the sample, which is connected to a data acquisition unit as
297 shown in Figure 4.

298 3. Results and discussion

299 3.1. Workability

300 Figure 5 shows the flow time of the different formulations studied. It should be noted that the main ratios
301 [Water/Binder, (GBFS+ MK)/SA, SS/NaOH and NaOH concentration] that affect the workability of the
302 geopolymer were fixed. The increase in the ratio (GBFS+ MK)/SA increases progressively the flow
303 time, which means that the workability of geopolymer mortars decreases with the incorporation of raw
304 materials (GBFS+ MK). This decrease is due to the high water demand of raw materials, especially
305 metakaolin, which has a high specific surface [14]. On the other hand, increasing the SS/NaOH ratio
306 and the concentration of NaOH increases the viscosity of the activation solution and thus reduces the
307 flow of the mortar. This is due to the very high viscosity of sodium silicate and sodium hydroxide [47].
308 Then the only variables in this study are the MPCM in both types of mortar (cement-based and
309 geopolymer) and the MK in the geopolymer mortar. Cement mortar without MPCM showed a flow time
310 of about 7 seconds that is confirmed from the literature [14].

311 Furthermore, as the MPCM concentration increases, the flow time increases for both types of mortars,
312 meaning that workability is reduced. This effect is attributed to the agglomeration of MPCM during
313 mixing due to its small size. This causes a larger water adsorption surface compared to the sand surface.
314 Indeed, MPCM traps water and prevents it from penetrating the matrix, which results in a decrease in
315 workability. In fact, MPCM strongly affected cement mortars while it did not show significant effects
316 on geopolymer mortars. This difference is explained by the very high workability of geopolymers
317 compared to cement-based mortars. This is related to the difference in activation of aluminosilicate
318 materials compared to cement-based materials, as confirmed by Muhammad et al. [48] and Deb et al.
319 [49].



320

321

Figure 5. Workability of different formulations

322

323 It is further observed that there is a slight increase in the flow time in all the mortars of geopolymers
 324 with 10 and 20% MK. This increase is related to the water demand of MK due to its large specific
 325 surface area.

326 Based on the comparison with the flow time of cement-based mortars, geopolymer-MPCM mortars
 327 showed good workability using up to 10% MPCM and 20% MK where the maximum flow time is equal
 328 to 4.9 seconds that is lower than the flow time of Portland cement-based mortar (7 seconds).

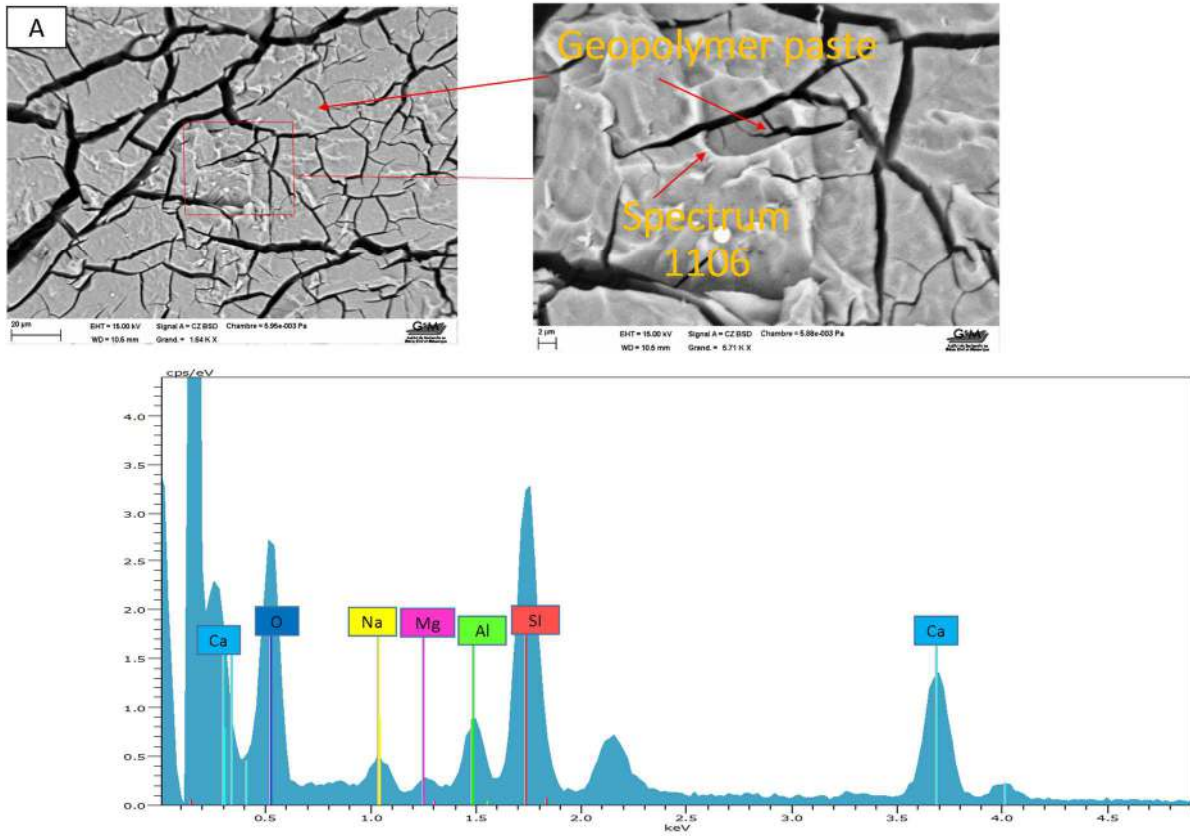
329 3.2. Microstructural analysis

330 Morphological analyses were performed with SEM for the observation of the microstructure of the
 331 geopolymer samples and EDS for the analysis of the elemental composition. Figure 6 compares the
 332 fractured geopolymer samples MGP_{100/0/0} (Figure 6.A) and MGP_{80/20/0} (Figure 6.B).

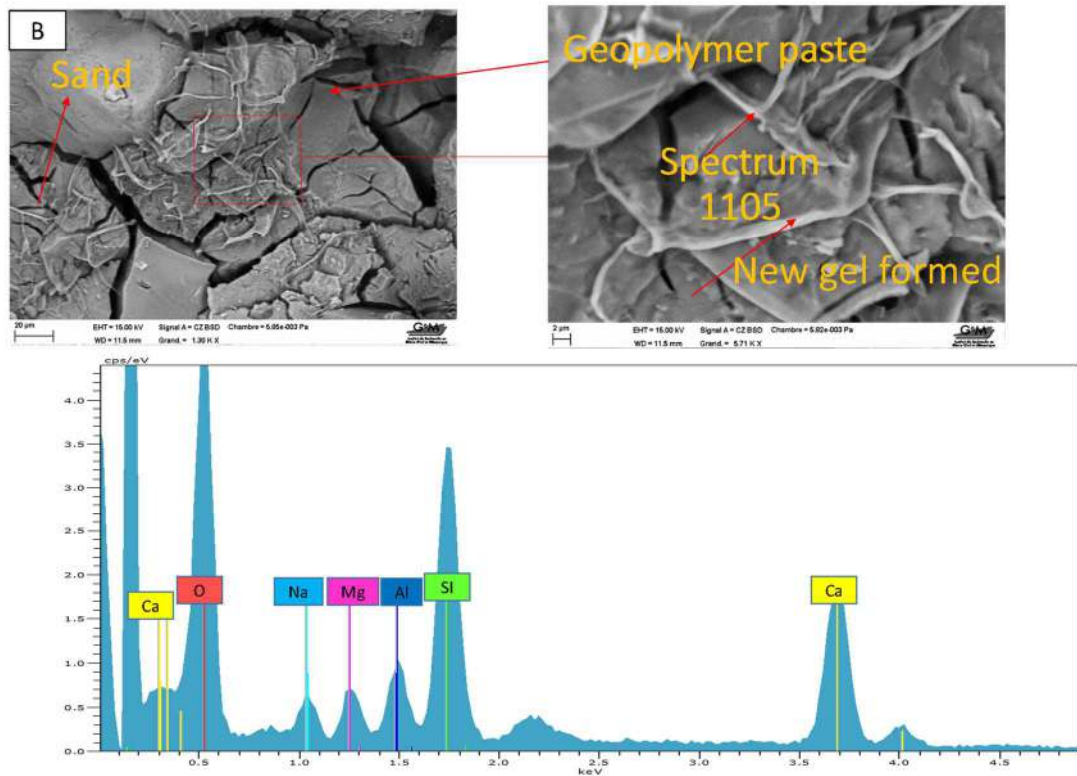
333 The SEM observations clearly show the appearance of additional gels in the matrix with 20% MK
 334 (Figure 6.B), while this appearance of gels was not observed in the matrix with 0% MK. The EDS
 335 analyses showed the presence of the main elements Si, Al, Ca, Na, which shows the formation of CASH
 336 gel, the main component of the GBFS activation [35,50]. Indeed, the analyses also show a higher content
 337 of the main elements mentioned previously for the MGP_{80/20/0} formulation compared to the MGP_{100/0/0}
 338 formulation. The increase in Si, Al, Ca, Na confirms the presence of NASH in addition to CASH gel in

339 the MGP_{80/20/0} formulation. According to Mijarsh et al. [30], the coexistence of these two gels results a
340 single stable gel called (C, N)ASH.

341



342



343

344

Figure 6. Microstructures of fractured samples at 28 days, A: MGP_{100/0/0}; B: MGP_{80/20/0}.

345

Table 3 compares the elemental compositions obtained by EDS of the geopolymer mortars MGP_{100/0/0},

346

MGP_{100/0/10}, MGP_{80/20/0}, and MGP_{80/20/10}.

347

Table 3: Elemental compositions of MGP_{100/0/0}, MGP_{100/0/10}, MGP_{80/20/0}, MGP_{80/20/10}.

348

Specimen

Elemental compositions (%)	MGP _{100/0/0}	MGP _{80/20/0}	MGP _{100/0/10}	MGP _{80/20/10}
O	60,69	54,71	65,91	58,17
Si	14,68	18,16	14,32	20,97
Al	3,64	3,94	2,79	3,74
Ca	15,05	19,35	12,29	14,06
Na	3,31	3,20	3,66	2,25

Si/Al

4,03

4,60

5,13

5,60

349

350 Table 3 shows that the concentrations of silica, calcium and aluminum increased after the addition of
351 20% MK. The mass content of silica is equal to 14.68% in the case of MGP_{100/0/0} while it increased to
352 18.16% with 20% MK (MCP_{80/20/0}). This content also increased from 14.32 to 20.97% in the geopolymer
353 mortar which contains 20% MPCM after the inclusion of 20% MK. Indeed, the main precursors of MK
354 activation are Si and Al, which may indicate that the high silica and alumina content of MK was well
355 dissolved by alkaline activation to form Si-O-Si and Si-O-Al bonds. On the other hand, the increase in
356 silica content increased the Si /Al ratio, confirming that the Si-O-Si bond was dominant over the Si-O-
357 Al bond. The Si/Al ratio increased from 4.03 in the MGP_{100/0/0} matrix to 4.60 in the MGP_{80/20/0} matrix,
358 and from 5.13 in MGP_{100/0/10} to 5.6 in MGP_{80/20/10}. The increase of the Si/Al ratio improves the
359 microstructure of the geopolymer, which results in an increase in its mechanical performance [30,51,52].
360 Moreover, it is also noted that the calcium content increased after the addition of MK, showing that the
361 calcium reacted well to the addition of MK and that its dissolution is not disturbed by the presence of
362 the 12M NaOH concentration. The increase in the calcium content increases the mechanical
363 performance of the geopolymer, as it will be shown in section 3.5 [34,53,54].

364 In addition, the presence of CASH and NASH gel can be confirmed by the mass ratio values in
365 percentages of Na/Si, Al/Si, Ca/Si. The Na/Si ratios are between 0.10 and 0.22 and Al/Si ratios are
366 between 0.17 and 0.24. They are in the range of the study by Ibrahim et al. [51] from which they
367 confirmed the appearance of these gels. On the other hand, the Ca/Si ratios are between 1.02 and 1.06,
368 which are higher than their ratios. This difference is due to the use of up to 80% GBFS in this study with
369 high calcium content.

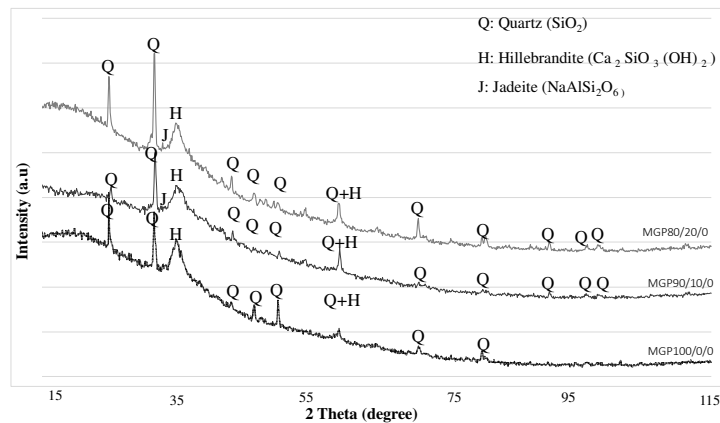
370 3.3. X-ray diffraction analysis

371 Figure 7 shows the XRD analyses performed on the powders of samples MGP_{100/0/0}, MGP_{90/10/0} and
372 MGP_{80/20/0} at 28 days. From a global point of view, it appears that all the samples show crystalline phases.
373 Around the 35° peak, the presence of hillebrandite of chemical formula Ca₂ SiO₃ (OH)₂ is observed in

374 all samples. Indeed, hillebrandite belongs to the CSH family of gels; Mijarsh et al. [30] explained that
375 with the presence of aluminum, the main composition of CSH turns into CASH gel. These results were
376 observed in EDS in the previous section (3.2) and are due to the use of up to 80% GBFS which leads to
377 the creation of CASH gel in general.

378 Several researchers have confirmed the presence of hillebrandite around this peak at 35° [30,55,56]. On
379 the other hand, Mijarsh et al. [30] have noted that hillebrandite serves the improvement of the
380 mechanical performance of the geopolymer. These results are in good agreement with the mechanical
381 performance results in Section 3.5.1.

382



383

384 Figure 7. X-ray diffraction of samples MGP_{100/0/0}; MGP_{90/10/0}; MGP_{80/20/0}) at 28 days.

385 After MK inclusion, the peak of jadeite (NaAlSi₂O₆) increased around 33°. Jadeite is a member of the
386 NASH family (the main gel for MK activation) [30]. It has a crystal structure with tetrahedral Si
387 coordinated in single chains and Al and Na in octahedral coordination [30,57]. The presence of
388 hillebrandite and jadeite in a single matrix confirms the coexistence of NASH and CASH gel and results
389 also in a high mechanical strength [51,55]. According to Ibrahim et al. [51], the CASH and NASH gel
390 renders the microstructure very compact and dense and this serves to improve the mechanical strength.
391 Indeed, this can be explained by the dissolution of silica, alumina, and calcium, which serve to fill the
392 small pores in the geopolymer matrix [50]. EDS analysis also showed that the silica, alumina, and
393 calcium contents were increased after the addition of MK.

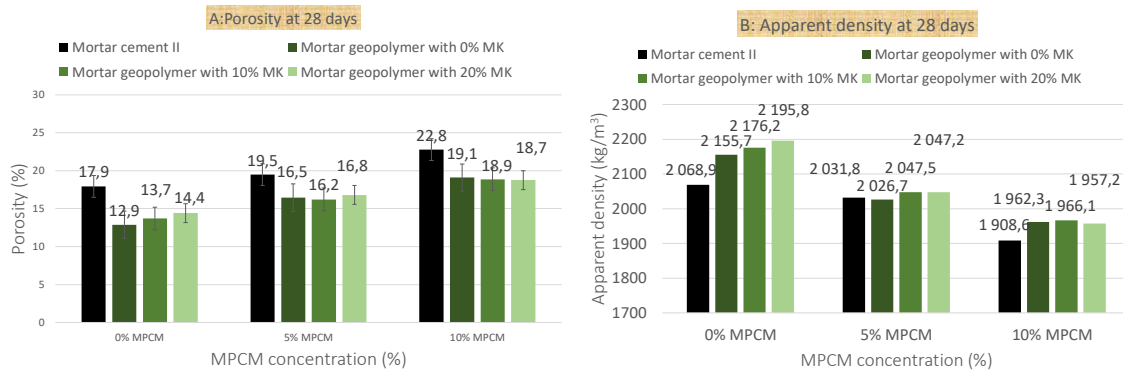
394 On the other hand, around the 60° peak, the intensity of the hillebrandite plus quartz (SiO₂) peak was
395 increased. This clustering may be related to the participation of calcium after the inclusion of MK in the
396 formation of the CASH gel with the presence of the NASH gel. The EDS confirmed the same
397 observations where the calcium content was increased after the inclusion of MK. Regarding this
398 observation, it is not possible to confirm these results because the samples used are in the form of mortars
399 (presence of sand). Quartz is therefore the main element of the sand.

400 **3.4. Water porosity and apparent density**

401 Figure 8 shows the water porosity results as well as the density of the studied samples. It indicates that
402 the cement-based mortar without MPCM presents a porosity of 17.90%. This value is approximately
403 similar to that reported in several studies that have investigated porosity for standardized cement mortars
404 [14,58].

405 It is noted that the porosity values of geopolymer mortars without MPCM are lower than those of
406 cement-based mortars. This difference may be related to the difference in workability between the two
407 types of mortars in section 3.1. Yang et al. [59] mentioned similar observations and cited that the
408 improvement of the workability of geopolymer reduces its porosity.

409 In addition, the increase in MPCM concentration caused an increase in porosity and a decrease in density
410 of all the samples studied. The decrease in density may be caused by the difference in density between
411 the sand (2.6 g/cm³) and the MPCM (0.84 g/cm³). Furthermore, the increase in porosity may be due to
412 the MPCM not filling the matrix cavities due to their agglomeration surface area being larger than that
413 of the sand. This agglomeration surface serves to adsorb a small amount of the binder paste, while this
414 can produce voids during mixing and increase porosity.



415

416

Figure 8. Water porosity and apparent density at 28 days.

417 However, the addition of MK shows no effect on porosity, showing that MPCM exerts a major effect
 418 on controlling the porosity of the geopolymer samples studied here. In contrast to the porosity results,
 419 the density was improved after the addition of MK in the samples without MPCM. This improvement
 420 was due to the high reactivity of MK compared to GBFS, which promoted the good dissolution of silica
 421 and aluminum to form new NASH and CASH gels in the matrix, as shown in Sections 3.2 and 3.3.

422 3.5. Mechanical properties

423 3.5.1. Compressive strength

424 Figure 9 shows the results of the compressive strength at 28 and 90 days. Globally, it should be noted
 425 that there is no significant difference between the compressive strength between 28 and 90 days for
 426 geopolymer mortars, contrary to cement-based mortars, which show an increase in their compressive
 427 strength between 28 and 90 days. Geopolymer is considered as a material that can gain most of its
 428 mechanical strength in the first few days of curing due to its strong chemical bonding, unlike cement-
 429 based materials.

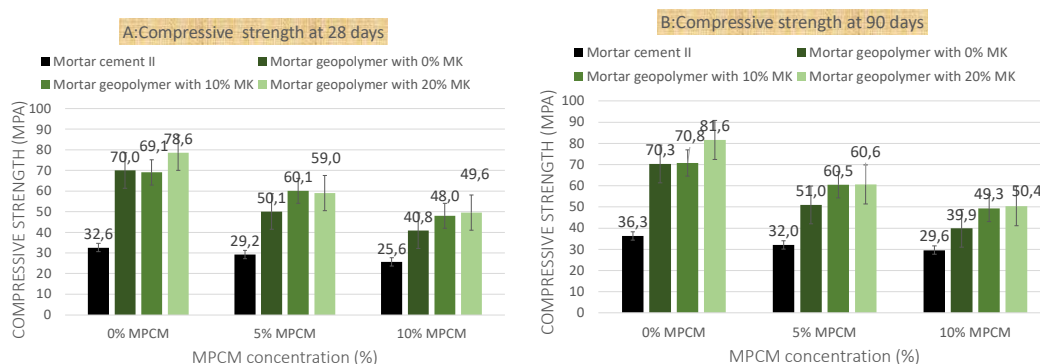
430 Bernal et al. [34] found that there is no difference in the compressive strength between 28 and 90 days
 431 for geopolymer without the addition of metakaolin (100% GBFS), while Bai et al. [60] pointed out that
 432 the increase in the GBFS rate serves to accelerate the development of the mechanical strength of the
 433 geopolymer. This may explain why there is no great difference between the 28- and 90-day time frames
 434 in this study.

435 The compressive strength of geopolymer mortars (with and without MPCM) is higher than the
 436 compressive strength of cement-based mortars. This may be related to the optimal formulation ratios
 437 that have been set according to the literature [14,34,37] and that have contributed to the improvement
 438 of geopolymerization.

439 The compressive strength of cement-based mortars decreases as the rate of MPCM incorporation
 440 increases. After 28 days, the compressive strength decreased from 32.6 to 29.2 MPA with 5% MPCM
 441 and then to 25.6 MPA with 10% MPCM. The effect is remarkably similar after 90 days. In the case of
 442 geopolymer-based mortars containing 0% MK, the same results are obtained. The compressive strength
 443 value in this case decreased from 69.9 to 50 MPA with 5% MPCM and reached 40.8 MPA with 10%
 444 MPCM after 28 days. Therefore, the rate of reduction of mechanical performance of geopolymer mortars
 445 with 10 and 20% MK is less compared to geopolymer mortars without MK.

446 The compressive strength value for the 10% MK geopolymer mortar is reduced from 69.08 to 60.12
 447 with 5% MPCM, and then it is reduced to 47.9 MPA with 10% MPCM, while for the 20% MK
 448 geopolymer mortar, the value is reduced from 78.5 to 58.9 until it reaches a value of 49.5 MPA.

449 This reduction in compressive strength is caused by the effect of replacing MPCM with sand. Recall
 450 that MPCM has replaced a certain volume percentage of sand in the matrix. Therefore, MPCM has low
 451 stiffness and mechanical strength compared to sand and can easily break under compressive force
 452 [7,18,23]. Furthermore, the increase in matrix porosity after MPCM incorporation is one of the causes
 453 of the reduction in mechanical strength as observed in the results detailed in the section. **3.4.**



454

455

Figure 9. Compressive strength; A: 28 days; B: 90 days.

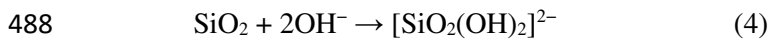
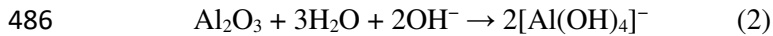
456 In addition, it can be observed that the geopolymer mortars with 0% MK showed a high mechanical
457 strength. This increase is caused by the high activator content and is due to the high Si/Al ratio [61]. The
458 presence of this high ratio in this study can be explained by the good dissolution of silica ions and
459 alumina of the GBFS due to the high PH of the solution, which results in a gel with high mechanical
460 performance [34].

461 Moreover, the addition of 10% MK in geopolymer mortars with MPCM concentrations (5 and 10%) led
462 to an increase in compressive strength at 28 days. The value increased from 50.06 to 60.12 MPA
463 (geopolymer mortar with 5% MPCM) and from 40.8 to 47.9 MPA (geopolymer mortar with 10%
464 MPCM) compared to the mortar without MK. The same effect of the addition of MK on the increase of
465 compressive strengths is observed also at 90 days. On the contrary, the addition of 10% MK did not
466 show any improvement in the compressive strength for geopolymer mortars without MPCM. About this
467 observation, the CASH and NASH gels created after the addition of MK showed a significant effect on
468 the mortars that have both concentrations of MPCM. Indeed, these mortars showed a high porosity,
469 while CASH and NASH gels are generally used to fill small pores [51]. This observation may be the
470 reason for the improved compressive strength.

471 In the case of the addition of 20% MK, the compressive strength is almost the same in the case of 10%
472 MK compared to the geopolymers with 5 and 10% MPCM, but for the geopolymers without MPCM,
473 the strength was increased from 69.9 to 78.5 MPA at 28 days and it reaches 81.6 MPA at 90 days. This
474 effect of increased compressive strength can be explained by the small particle size and large specific
475 surface area of MK compared to GBFS, which allow for accelerated reactivity [31,62] on the one hand,
476 and the large amount of Al_2O_3 and SiO_2 in MK. These two effects led to an improvement in
477 geopolymerization by producing sodium aluminosilicate hydrate (NASH) gel and calcium silicate
478 hydrate (CASH) gel in addition to the CASH produced by the activation of GBFS, which are rich in
479 calcium [34].

480 In a matrix that contains both NASH and CASH, the geopolymerization process proceeds through the
481 following steps:

482 1. The NASH gel is formed in a system containing Na₂O-Al₂O₃-SiO₂-H₂O and starts with the
483 dissolution and hydrolysis of the silica and alumina of the aluminosilicate material to form Si-
484 OH and Al-OH bonds and continues with a condensation. The hydrolysis of Si and Al is
485 presented in the following equations (2), (3), (4) [30,63]:



489 However, the activator content (Na and Si concentrations) has a very important role in the
490 condensation process, which strongly influences the properties of the geopolymer [30]. The use of
491 a high activator content as in this study favored the condensation to form oligomeric silicates, which
492 generate rigid polymeric 3D structures [poly (sialate-siloxo) and poly (sialate-disiloxo)] with Si/Al
493 ratios greater than 2. Otherwise, the use of a low concentration of activator results in poly (sialate)
494 polymer structures with Si/Al ratio equal to 1 and accompanied by low mechanical strength [30].

495 The EDS analyses in section 3.2 confirm that the Si/Al ratio is higher than 2, which explains the
496 better mechanical strengths found in this study.

497 2. The CASH gel is formed in a system containing CaO-Na₂O-Al₂O₃-SiO₂ -H₂O. Calcium
498 influences in a very important way the acceleration of the hardening and the improvement of
499 the mechanical performances of the geopolymer because of its additional nucleation sites
500 [30,34]. This may be one of the reasons for the improvement in mechanical performance, and
501 the EDS analysis shows that the calcium content was increased after the addition of MK. The
502 increase in this content confirms that it was indeed involved in the formation of both gels. These
503 are two mechanisms cited by other researchers [30,40,64] to explain how calcium is involved
504 in the geopolymer system. The first states that Ca²⁺ acts as a charge balancer and this allows it
505 to be integrated into the geopolymer network. The second mechanism indicates that Ca is a
506 contributing element to the formation of a CASH gel which has the possibility to coexist with
507 the NASH geopolymer gel as explained above.

508 The reason for the increase in mechanical performance may be that the CASH and NASH gels
509 caused by MK activation filled the pores caused by MPCM, which was not observed in the
510 geopolymer with 0% MK.

511 The other reason for the improved mechanical performance may be related to the strong chemical
512 structure of the geopolymer after the addition of MK. This effect is confirmed by EDS, which shows
513 that the silica content increased after the addition of MK, which increased the Si/Al ratio. With the
514 increase in Si content, more silicate species are available for condensation and reaction between
515 them, resulting in more oligomeric silicates and they become dominant [62]. Researchers have
516 shown that oligomeric silicates of Si-O-Si bond are stronger than those of Si-O-Al and Al-O-Al.
517 This can explain the increase in compressive strength of geopolymer mortars after the addition of
518 MK, on the other hand [14,65].

519 **3.5.2. Dynamic Young's modulus**

520 The results of the Young's modulus at different times (2, 7, 14, 21, 28, 56 and 90 days) are shown
521 in figure 10. First, it can be noticed that this modulus develops with the curing time for both types
522 of mortar. This observation is an indication that the hydration of the cement and the
523 geopolymerization remain continuous during this time. Comparing the hydration time for cement
524 mortar and geopolymer, it is observed that the values of this modulus continue to increase for cement
525 mortars up to 90 days, while for geopolymer their values have been stabilized from the 14th day.
526 This is due to the very rapid acceleration of the mechanical strength of these materials compared to
527 cement-based materials [60].

528 Contrary to the compressive strength values, the Young's modulus values of the cement-based
529 mortar are higher than those of the geopolymer mortar. Although this effect has been granted
530 according to the literature [14,58,66], which cites that geopolymer possesses low rigidity, in contrast
531 the values of this modulus in this study are very close to those of standardized mortars with a
532 maximum value equal to 35.1 GPA at 90 days (geopolymer mortar with 20% MK without MPCM).

533 Figure 10 shows that the inclusion of MPCM decreased the Young's modulus for the two mortars
534 (cement and geopolymer). These results are like the compressive strength results in the previous
535 section (3.5.1).

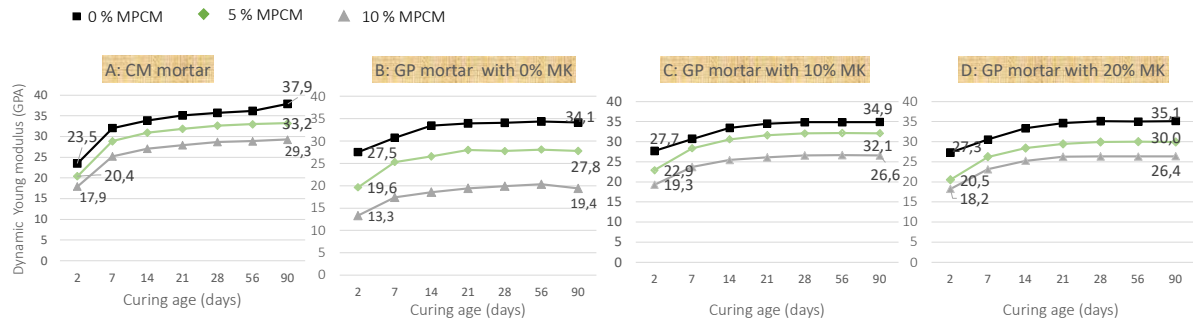
536 The values of Young's modulus of cement-based mortar decreased from 35.7 GPA to 32.6 and 28.6
537 GPA with 5 and 10% MPCM at 28 days. This decrease after the incorporation of MPCM was
538 accorded with the study of Šavija et al. [67]. Moreover, the same effects are observed in geopolymer
539 mortars with 0% MK from which the values decreased from 34.0 to 27.7 and 19.8 GPA with 5 and
540 10% MPCM.

541 The reason for the decrease in Young's modulus is explained in section 3.5.1 of compressive
542 strength. The Young's modulus depends on the strength of the material [68], so the inclusion of
543 MPCM decreases the rigidity of the matrix due to its low rigidity compared to sand. The second
544 reason may be due to the porosity caused by these materials, which decreases the flexural resonance
545 frequency (f_r) while the latter is the main element that controls this modulus.

546 Figures 10.C and 10.D show that the MK inclusion rates improved the Young's modulus for all
547 geopolymer samples. The addition of 10% MK increased the Young's modulus value of geopolymer
548 mortars from 34.1 to 34.9 GPA (0% MPCM) and from 27.7 to 32.1 GPA (5% MPCM) and from
549 19.9 to 26.6 GPA (10% MPCM).

550 Similar remarks are reported in Figure 10.D (20% MK), which shows that the addition of 20% MK
551 increased the Young's modulus values to 35.1, 29.9 and 26.4 GPA.

552 The optimum MK content is 10% for geopolymer-MPCM mortars, but for geopolymer mortars
553 without MPCM, the optimum content is 20% MK. These results are in good agreement with the
554 compressive strengths and can be explained by the pore filling effect caused by the activation of
555 silica and alumina in the MK.

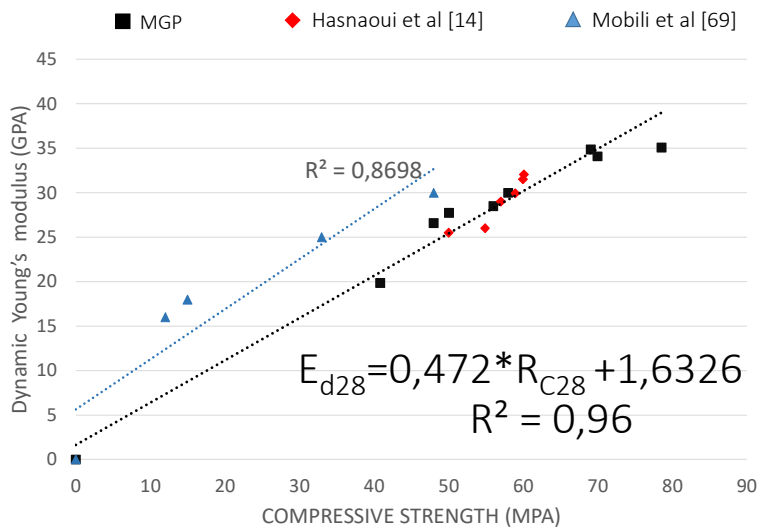


556

557 Figure 10. Young's modulus as a function of curing time, A: (cement-based mortar), B:
 558 (Geopolymer mortar with 0% MK); C: (Geopolymer mortar with 10% MK); D: (Geopolymer mortar
 559 with 20% MK).

560 Figure 11 represents a comparison of dynamic Young's modulus results on geopolymer mortars with
 561 other researchers who have used dynamic methods to evaluate the values of this modulus [14,69].

562



563

564 Figure 11. Correlation between dynamic Young's modulus and compressive strength at 28 days.

565 According to figure 11, there is a good correlation between this modulus and the compressive
 566 strength expressed by a linear variation with a correlation coefficient $R^2 = 0.96$ for our geopolymer
 567 mortars and those of Hasnaoui et al. [14] and a linear correlation for the mortars of Mobili et al.

568 [69]. Indeed, Duxson et al. [65] indicated that the Young's modulus of the geopolymer is correlated
569 with its compressive strength.

570 The results presented in this study are higher than the reference results mentioned. This is probably
571 the consequence of the use of 80/20 GBFS and MK, in contrast to Hasnasoui et al. [14] who used
572 50/50 GBFS and MK, and the use of Mobili et al. [69] up to 100% fly ash. The CASH gel formed
573 by GBFS activation is denser compared to the NASH gel and has a higher pore filling capacity
574 [70,71], which may explain the better mechanical performance obtained in this study compared to
575 the two references above.

576 **3.6. Thermal properties**

577 Figure 12 shows the thermal properties of two types of mortars at different measuring temperatures.
578 These measurements were performed in a temperature range that is between 15 °C (MPCM in solid
579 state) and 40°C (MPCM in liquid state). It can be clearly shown that the rate of increase of MPCM
580 concentration caused a decrease in the thermal conductivity of two mortars (cement-based mortar
581 and geopolymer-based mortar) independently of the measurement temperature. The reason for this
582 decrease in the two cases can be explained by the two effects below:

- 583 • The first effect is attributed to the low thermal conductivity of MPCM (about 0.2 W/m.K),
584 which replaced sand while the latter has a higher thermal conductivity. This effect was
585 confirmed by the measurements that were carried out in the materials section (2.1).
- 586 • The second effect is the appearance of voids in the matrix after the incorporation of MPCM
587 (see figure 8). These voids will transfer the heat flow from the state of conduction to the
588 state of convection, resulting in a decrease in thermal conductivity.

589 However, the thermal conductivity in the solid state (measured at 15°C) of the two mortars with the
590 two MPCM concentrations is higher compared to the thermal conductivity in the liquid state. This
591 is confirmed by the study of Cui et al. [25] and Cao et al. [7] and due to the thermal conductivity of
592 the solid state paraffin, which is generally higher than its thermal conductivity in the liquid state.
593 Additionally, Sasaguchi et al. [72] have shown that the thermal conductivity of paraffin-type phase

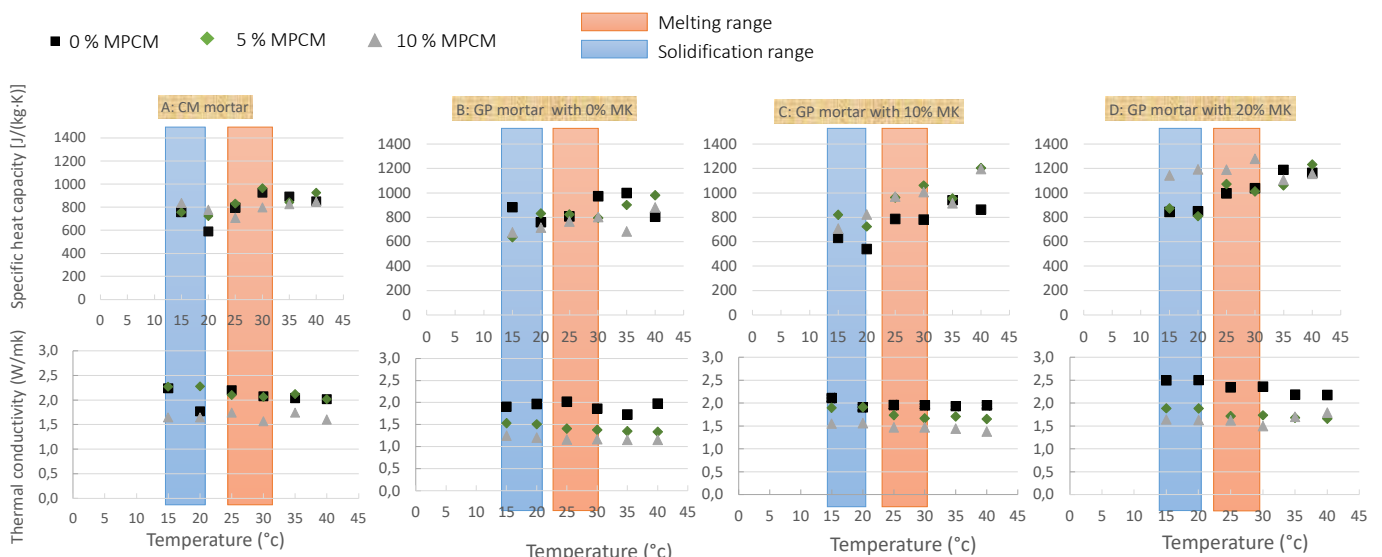
594 change material (n-octadecane) in the solid state is equal to 0.358 W/mK while in the liquid state is
595 equal to 0.148 W/mK.

596 Nevertheless, the MPCM did not show an improvement in the specific heat capacity of the cement
597 mortars and that of the geopolymer without MK (figures 12 A and B).

598 This consequence can be related to the low thermal conductivity of the samples presented in figure
599 12 and due to the presence of more voids which appeared (figure 8 A). Indeed, these voids did not
600 allow the ambient heat to transmit to the microcapsules to be stored on the one hand and can be
601 related to the hot disk sensor which did not capture the latent heat stored by the MPCM on the other
602 hand.

603 These results are not in agreement with the following references [9,18,73] which found an
604 improvement in heat capacity with the use of the DSC method. When using the DSC method, the
605 sample tested is in the form of a homogeneous powder with a mass of about 1-10 mg. Therefore, this
606 method does not consider thermal conductivity, unlike the hot disk method used in this study.

607



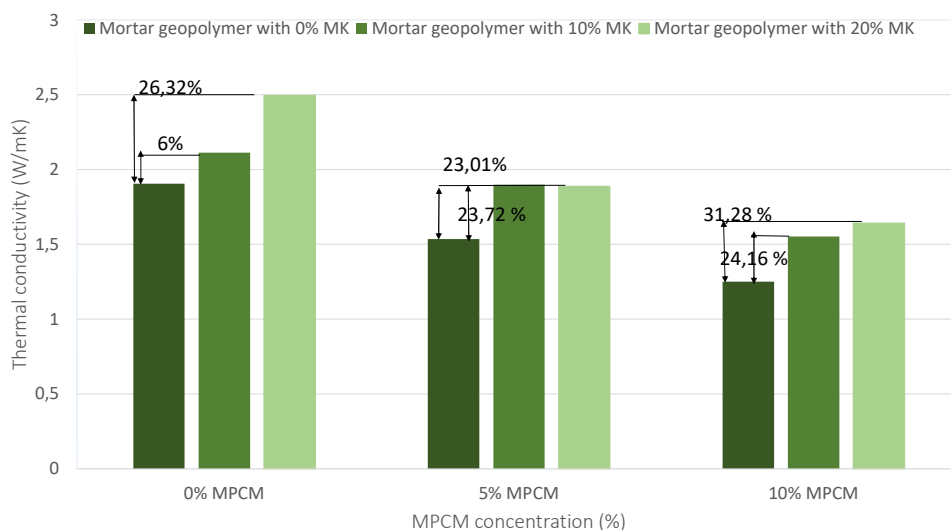
608

609

610 Figure 12. Thermal properties at 28 days; A: (cement-based mortar), B: (Geopolymer mortar with
 611 0% MK); C: (Geopolymer mortar with 10% MK); D: (Geopolymer mortar with 20% MK).

612 In contrast to geopolymer mortars without MK, all samples with 10 and 20% MK showed the highest
 613 specific heat capacity at the melting point of MPCM (between 25 and 30°C) and at the solidification
 614 point (between 15 and 20°C). The highest value of specific heat capacity reached 1280.76 J/Kg.K at
 615 the melting range for MGP_{80/20/10} (20% MK and 10% MPCM). This value is close to the one that is in
 616 the study of Cao et al. [7] who found a value of 1500 J/kg.K at the melting point of MPCM while they
 617 used up to 72 kg/m³ of MPCM while in this study the concentration of MPCM is equal to 56.8 kg/m³.
 618 The same improvements were accorded by Shadnia et al. [18], who found an improvement in specific
 619 heat capacity up to 1200, 33 J/kg.K. With this value of specific heat capacity, the conclusion of the
 620 study by Shadnia et al. [18] indicates that the internal temperature measurements of two cells
 621 constructed with geopolymer-MPCM mortar showed a reduction of 4.5 and 5.5 ° C compared to a
 622 reference cell that does not contain MPCM. In this case, the improvement of the specific heat capacity
 623 is related to the increase of the thermal conductivity.

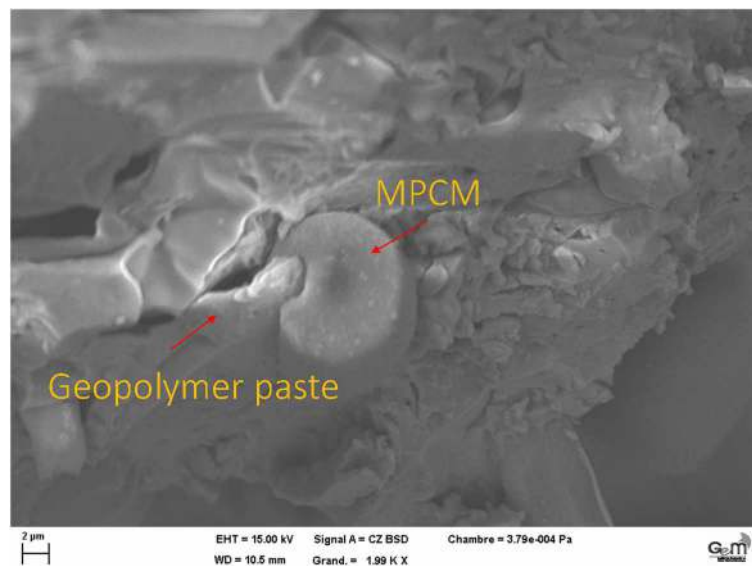
624 Figure 13 is extracted from Figure 12. It shows the effect of the addition of MK on the thermal
 625 conductivity of geopolymer mortars in the solid state where the measurements are taken at 15°C. This
 626 point was chosen because the effect of adding MK on the thermal conductivity is roughly similar
 627 compared to the other measurement points.



629 Figure 13. Thermal conductivity of geopolymer mortars at 15°C as a function of MPCM
630 concentration before and after MK addition.

631 In Figure 13, it is clearly shown that the inclusion of two MK rates (10 and 20%) increased the
632 thermal conductivity by 6 and 26.32% for the geopolymer mortar with 0% MPCM. This increase
633 translates into an improvement in thermal conductivity of about 0.137 W/mK and 0.52 W/mK. On
634 the other hand, an improvement in thermal conductivity of 23.01; 23.72; 24.16 and 24.16% is
635 observed for geopolymer mortars with 5 and 10% MPCM. The increase of thermal conductivity
636 after the inclusion of MK can be explained by the decrease of voids in the matrix due to the creation
637 of gels in the geopolymer matrix, as several researchers have mentioned the advantage of the good
638 reactivity of MK [14,34,53]. In addition, Ibrahim et al. [51] pointed out that the high Si content
639 observed in the EDS leads to the filling of the pores of the studied matrix.

640 Figure 14 was obtained from the SEM for MGP_{80/20/20} (20% MK and 20% MPCM). It shows a good
641 cohesion of the geopolymer paste with the MPCM, which can explain the improvement of the
642 thermal conductivity of the total matrix after the addition of MK and might be due to the increase
643 of the thermal flux that was transmitted during the measurement.

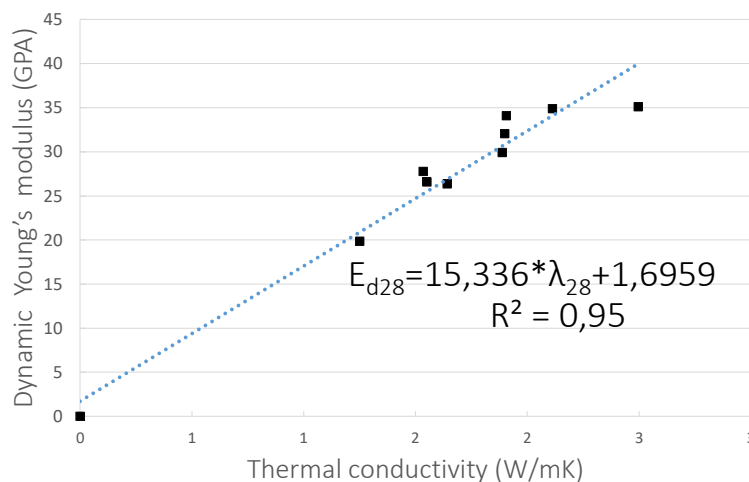


644
645 Figure 14. Cohesion of the geopolymer paste with MPCM.

646

647 XU et al. [74] added carbon nanotubes to a cement matrix to increase its thermal conductivity after
648 incorporation of PCM (without capsule). They found an increase of about 45% in thermal conductivity.
649 Their study reveals that the increase in thermal conductivity improved the heat storage performance of
650 PCM. On the other hand, they monitored the time taken for the temperature to increase from 27 to 31°C.
651 Their results showed that the heat storage and release time decreased compared to the reference sample
652 by about 181 seconds for the heat storage process and 266 seconds for the heat release process.
653 Increasing the thermal conductivity of geopolymer mortars will help the MPCM to charge and discharge
654 its latent heat, thus improving its thermal performance in actual construction.

655 Figure 15 shows a correlation between the Young's modulus and thermal conductivity of geopolymer
656 mortars at 28 days. A linear correlation expressed by a correlation coefficient $R^2 = 0.95$ is noted. These
657 two properties are dependent on the density of the material, which may explain this linear correlation.
658 A similar correlation between dynamic Young's modulus and thermal conductivity is presented in the
659 study of Barnaure et al. [75] for other material constructions.



660

661 Figure 15. Correlation between dynamic Young's modulus and thermal conductivity at 28 days.

662 4. Conclusion

663 The objective of this study was to use the coexistence of two NASH and CASH gels by the inclusion
664 of metakaolin to overcome the negative effects of MPCM incorporation in blast furnace slag-based

665 geopolymer mortars. The conclusions of the different tests after the inclusion of metakaolin are as
666 follows:

- 667 • The geopolymer-MPCM mortars showed good workability with up to 10% MPCM and 20%
668 MK. The maximum flow time was equal to 4.9 seconds, which is lower than the standard mortar
669 flow time of 7 seconds.
- 670 • SEM, EDS and XRD analysis confirmed that incorporation of metakaolin into the blast furnace
671 slag matrix produced CASH and NASH gel as stable reaction products.
- 672 • EDS analysis revealed a higher Si/Al ratio, which means it has the potential to produce a dense
673 and compact bonding geopolymer.
- 674 • The compressive strength of geopolymer-MPCM mortars is between 49 and 61 MPa and is
675 sufficient for structural applications.
- 676 • The CASH and NASH gels formed after the addition of MK increased the thermal conductivity,
677 and the latter improved the specific thermal capacity of geopolymer-MPCM mortars. The
678 maximum value obtained is equal to 1280 J/Kg.K This value is higher than the thermal capacity
679 values of Portland cement mortars.

680 Finally, the increase in MPCM rate showed a reduction in mechanical strength and an increase in
681 porosity, which limit the amount of MPCM that can be added to the geopolymer mortar. Future research
682 could focus on developing microcapsules with low agglomeration, excellent compatibility with the
683 concrete matrix and high mechanical strength (robust microcapsule) to overcome these constraints.
684 These could be a viable option to reduce the loss of compressive strength and the increase of porosity.
685 On the other hand, this solution will increase the amount of MPCM in the geopolymer, thus increasing
686 the total thermal energy storage capacity of the geopolymer.

687 **Declaration of Conflict of Interests**

688 The authors declare that they have no known competing financial interests or personal relationships that
689 could have appeared to influence the work reported in this paper.

690 **Credit authorship contribution statement**

691 **Bouha EL MOUSTAPHA:** Conceptualization, Methodology, Investigation, Writing - original draft.
692 **Stéphanie BONNET:** Validation, Resources, Visualization, Supervision, review & editing. **Abdelhafid**
693 **KHELIDJ:** Validation, Resources , Visualization, Supervision, review & editing. **Nordine LEKLOU :**
694 Validation, review & editing. **Daniel FROELICH:** Validation, review & editing , Supervision.
695 **Isselmou AHMEDOU BABAH:** review & editing, Supervision. **Carole CHARBUILLET:** review &
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697

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703

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