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Effect of metakaolin addition on the mechanical performance of granulated blast furnace slag based geopolymer mortar with micro-encapsulated phase change materials

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Abstract

The incorporation of microcapsule phase change materials (MPCM) in geopolymer is one of the effective technologies that contribute to improve the thermal comfort of buildings on the one hand and replace the use of Portland cement-based materials on the other hand. Although MPCM increases the thermal capacity of the cementitious matrix, whether it is based on cement or geopolymer, their incorporation has unfortunately several negative effects on the mechanical performances. This scientific problem is still unsolved and several researchers have pointed it out.

This study aim to investigate the effect of 10 and 20% metakaolin(MK) inclusion on the mechanical performance of geopolymer-MPCM mortars based on granulated blast furnace slag (GBFS) and to compares them with Portland cement-MPCM based mortars. Different tests were performed for to types of mortar in the aim to characterize their properties such as workability, porosity, compressive strength, dynamic Young's modulus and dynamic shear modulus.

The results show that the inclusion of two proportions of metakaolin compensated well for the loss of mechanical strength related to the incorporation of MPCM. Therefore, with up to 20% of MPCM the inclusion of metkaolin increased the compressive strength by about 10 MPA. In addition, all geopolymer-MPCM mortars showed high compressive strength, improved workability and reduced porosity compared to Portland cement-MPCM based mortars.

1. Introduction

The building sector is the largest energy consumer in the world, accounting for 40% of final energy consumption and 30% of greenhouse gas emissions[1].

The environmental impacts of this sector are produced throughout its life cycle during the phase of extraction of raw materials, the phase of manufacture of building materials, the phase of use and finally the phase of its end of life.

However, most of the energy consumption in the building is reserved for the use phase of the building to improve thermal comfort in the hot and cold seasons [2]. Furthermore, 80% of the greenhouse gas emissions throughout the life cycle of the building are related to this phase when electricity is used for the building's requirements [3].
Moreover, the fabrication phase of construction materials such as Portland cement is the second most impacting phase on the environment in this sector [4]. The fabrication of Portland cement to produce concrete is responsible for 7% of CO_2 emissions [5] in the world and causes other pollution in the air, water, etc.
Add to all these negatives environmental impacts, ordinary concrete has durability issues against external aggressive attacks.
Several researchers have shown in recent years the effectiveness of using MPCM to improve thermal comfort and reduce the requirement for heating and cooling systems in buildings [6,7,8,9].

Indeed MPCM store at a constant temperature (ambient temperature around human comfort) a large amount of energy in the form of latent heat during their phase change (solid-liquid) which prevents the heat flow to enter the building during peak hours, this amount of heat is stored during the day and released at night [10].

Regarding the fabrication phase of construction materials such as Portland cement, we note that the use of geopolymers has attracted a lot of interest in the research field because of their low environmental impact and their superior technical advantages compared to Portland cement-based materials. Geopolymer is the result of activation of aluminosilicate materials by alkaline solutions, these aluminosilicate materials are industrial by-products or types of clay such as granulated blast furnace slag (GBFS), metakaolin (MK), fly ash, and red mud, etc [11].

The use of these materials to replace Portland cement will reduce CO_2 emissions and waste caused by the industries, which will help reduce the negative environmental Impacts.
A study shows that CO_2 emissions caused by the production of geopolymer are reduced by about 70 to 80% compared to the manufacture of Portland cement [12]. In addition geopolymer has several advantages over Portland cement-based materials, such as higher initial mechanical strength, low drying shrinkage, high fire resistance, shorter curing time, superior acid resistance and improved durability [13,14,15,16].

In recent years several researchers have shown that the incorporation of microcapsule phase change materials (MPCM) into geopolymer can be a promising solution to overcome CO_2 emissions related to energy consumption in buildings and Portland cement production.

Shadina et al (2015) [17] constructed three small cells out of fly ash based geopolymer mortars, among these three cells two containing MPCM.Their internal temperature measurements showed a reduction of 4.5 and 5.5 °C for the two MPCM geopolymer cells compared to the reference cell. Secondly, Cao et al (2019) [7] numerically investigated the influence of different climatic conditions on the energy efficiency of a wall constructed of geopolymer concrete-MPCM. The reduction of the interior wall temperature was about 3°C and the reduction of energy consumption was 25%, while maintaining the interior temperature at 23°C.

Unfortunately, previous research studies show that MPCM have negative effects on the mechanicals performances of the cementitious matrix [7,9,10,17].

Cao et al [9] studied the effect of the addition of MPCM on the compressive strengths of Portland cement-based concrete and geopolymer-based concrete. Their results show that although MPCM generated an increase in the heat capacity of the two types of concrete studied up to the value of 1500 (J/kg ° C), the compressive strengths were reduced by about 42 and 51%. The same observations were reported by Shadina et al (2015) [17] who reported a decrease in compressive strength of up to 25%.

The scientific question in this study is how to overcome the negative effects of incorporating MPCM into a geopolymer matrix, for this reason, no studies have been conducted to date in the research field to address this issue.

However, several researchers have shown that the inclusion of a small amount of metakaolin in a geopolymeric matrix based on granulated blast furnace slag improves mechanical performance and durability properties [18,19,20]. Bernal et al (2012) [18] studied the effect of adding 10 and 20% metakaolin in a geopolymer matrix based on granulated blast furnace slag, their results show that the mechanical performance and durability were improved due to the very high reactivity of the metakaolin which was accompanied by a high activator content [18].The same observations were presented by Huseien et al [19], their results showed that compressive strength increased by 33% compared to the geopolymer matrix based on granulated blast furnace slag with 15% metakaolin inclusion. Kumar et al (2020) [20] also reported that the optimum inclusion rate of metakaolin was 20% and the compressive strength was improved by about 24% with this rate. However, Bernal et al (2012) [18] pointed out that the reaction of metakaolin is conditioned by high alkalinity of the alkaline solution and if it is on the contrary metakaolin had negative effects on the mechanical strength and durability properties [21,22].

If we recall most of the research studies have been carried out on the geopolymer-MPCM, we find that they concentrate on the geopolymer based on a single base material such as granulated blast furnace slag, fly ash, metakaolin, while very few studies can be found on combination of two materials at the same time and no research work has been done on the inclusion of metkalaolin in granulated blast furnace slag and varying the alkalinity of the activator.

The aim of our study is to investigate for the first time the effect of including 10 and 20% of metakaolin in in geopolymer-MPCM mortars based on blast furnace slag and to compare them with reference Portland cement-MPCM based mortars. Several characterization studies are performed in this study, such as workability, total water porosity, compressive strength, dynamic Young's modulus and dynamic shear modulus.

2. Materials and experiments design

2.1 Materials

The MPCM considered in this study are in the form of a white spherical micro-encapsule marketed by the laboratory Microteck-United States.Its technical name is Nextek 28 D, its melting temperature is equal to 28 ° C while density equal to 0.84 g/cm^3.They possess sizes between 6.190 and 38.22 µm.

The granulated blast furnace slag is provided free of charge by the company ECOCEM in France and the metakaolin was provided by the company KENZAI (ecological materials) in France. The cement used is a CEM II (32.5).

The chemical compositions and physical properties of cement II,GGBS and MK are presented in Table1.

Table 1: Chemical composition and physical properties of CEM II, GGBS and MK.

Chemical composition (%)	**CEM II**	**GBFS**	**MK**
SiO_2	7,47	37,3	55
Al_2O_3	2,18	10,7	41
Fe_2O_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
$(Na_2O+K_2O)eq$	-	0,8	1,8
Specific gravity	3.03	2.9	2.4
Blaine specific surface area (m^2/g)	0.37	0.445	17
average grain size (µm)	8.47	13.25	7.13

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

A sand standardized CEN NF 196-1 with a density of 2.6 g/cm^3 , this type of sand is generally used for laboratory tests. The provider is the same as that of two alkaline solutions. The purpose of using standardized sand is to eliminate the secondary effects on the binder of unfavorable impurities that natural sand might contain.

2.2 Mixing method and curing condition

Twelve formulations were investigated in this study, three based on standardized mortar and nine based on geopolymer mortar.
The water/binder ratio of 0.5 and the sand/binder ratio of 3 have been fixed for both types of mortars. The binder (equivalent to cement) of geopolymer is considered as the total of GBFS, MK and the alkaline solution (solid part).We fixed a mass ratio of 3 of (GBFS+MK)/SA, with SA corresponds to the solid part of the alkaline solution. These choices are recommended by the study of Hasnaoui et al (2019)[23].

The inclusion percentages of MK are 0, 10 and 20 %.The ratio of sodium silicate (SS) to sodium hydroxide (SS/NaOH) is 2.5 in all geopolymers formulations.This high sodium silicate content will maintain an optimal amount of silica after the addition of our higher percentage of metakaolin (20% MK) [18].

The MPCM will replace the same percentage by volume of sand with three concentrations such as 0, 5 and 10% in the two types of mortar.

Table 2 shows the mixing proportions in kg/m^3 where the first three formulations are based on Portland cement mortar while the last nine are based on geopolymer mortar.
The amount of water in the two alkaline solutions is considered to have a water/binder ratio equal to 0.5.

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

Sample	Cement	GGBS	MK	Sand	MPCM	Na_2SiO_3	NaOH	MGP water	MCII water
$MCII_{0/0/0}$	585,9	-	-	1757,8	0,0	-	-	-	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	0,0	1757,8	0,0	267,9	127,7	42,6	-
$MGP_{100/0/5}$	-	439,5	0,0	1673,8	28,4	267,9	127,7	42,6	-
$MGP_{100/0/10}$	-	439,5	0,0	1582,0	56,8	267,9	127,7	42,6	-
$MGP_{90/10/0}$	-	395,5	43,9	1757,8	0,0	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	0,0	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	-

We followed the NF 196-1 standard [24] for the manufacture of the standardized mortar based on Portland cement while it was chosen as reference in order to compare it with the geopolymer-based mortar.

However for the geopolymer, the GBFS and MK were mixed with the alkaline solution and water for 90 seconds to have a homogeneous paste. The mixture was followed by the addition of sand and mixed for 5 minutes and finally the MPCM were added and mixed for 2 minutes. After the mixing procedures, the standardized mortars and geopolymers were used for the measurement of workability, after this step the casting is carried out in specimens of 40 × 40 × 160 mm^3 and vibrated using a shock table and are stored in an air-conditioned room (temperature 20 ° C and relative humidity 50%) for 48 hours before demolding.

2.3 Characterization methods

The workability test was carried out directly after mixing according to standard NF P18-452 [25]. The test consists in measuring the flow time of mortars under the effect of the vibration caused by the vibrator.

The water porosity was determined according to the NF P 18-459 standard [26].
The relation to calculate the porosity is as follows:

$$n=\frac{Mair-Mdry}{Mair-Mw}\times 100 \qquad (1)$$

M_{air} is the mass of the sample saturated in free air, M_{dry} is the mass of the sample in the dry state after its drying while M_w is its mass in water (hydrostatic weighing).

The compressive strengths were realized following the NF EN 196 standard, these tests were carried out on six samples in order to ensure a good repeatability.
For the measurements of dynamic Young's modulus and dynamic shear modulus, we followed the ASTM E 1876-01 standard [27] using an apparatus called Grindosonic.
These measurements were performed on three samples of each formulation at 2,7,14,21,28,56 and 90 days. The technique is based on a pulse excitation of the vibrations.
Dynamic Young's modulus and dynamic shear modulus are calculated using the equations below using the flexure (f_f) and torsion (f_t) frequencies obtained during measurements with the dimensions and weight of the samples:

$$E_{dyn}=0{,}9465\left(\frac{m.f_f^2}{b}\right)\left(\frac{L^3}{t^3}\right)T \qquad (2)$$

$$G_{dyn}=\frac{4.L.m.f_t^2}{b.t}R \qquad (3)$$

With m, L, b, t represent the mass in (g), length in (mm), width in (mm) and thickness in (mm) of the sample.
T and R represent the coefficients of correction, which can be defined by following certain steps in ASTM E 1876-01[27].

3. Results and discussion

3.1 Workability

The figure 1 shows the flow time of the different formulations studied.We recall that we have set the main ratios that affect the workability of the geopolymer and that of the cement, Indeed the only variations in the two types of mortar (cement-based and geopolymer) are the concentrations of MPCM and MK.

Cement mortar without MPCM has a flow time of about 7 seconds which is confirmed by Hasnaoui et al (2019) [23].

The difference between the flow time of cement mortars and geopolymer mortars is related to the difference between the rheology of geopolymer and cement as Muhammad et al (2019) [28] pointed out.
We observe from figure 1 that with increasing concentration of MPCM,the flow time increases for both types of mortars which means that the workability is reduced.

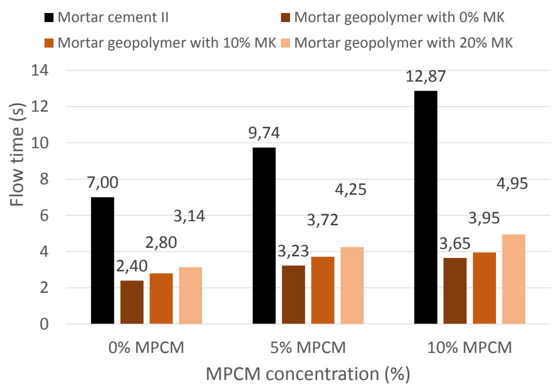


Figure 1: Workability of different formulations

The effect of decreased workability is attributed by the agglomeration of MPCM during mixing with its small sizes which are between 2.190 and 38.22 µm and this causes a larger water adsorption surface compared to the sand surface.

Actually, MPCM traps water and prevents it from penetrating the matrix, which results in a decrease in workability. These effects are in agreement with the PILEHVAR et al study (2018) [29].

We remark that there is a slight increase of the flow time in all the geopolymer mortars with 10 and 20% of MK.This increase is related to the water demand of MK which makes the mortar a little viscous and caused by its large specific surface which is equal to $17m^2/g$ compared to the specific surface of GGBS which is equal to 0.445 m^2/g [19,23].

Based on the comparison with the flow time of cement-based mortars, our geopolymer-MPCM mortars have good workability using up to 10% MPCM and 20% MK where the maximum flow time is equal to 4.95 seconds which is lower than the flow time of Portland cement-based mortar.

3.2 Porosity

The figure 2 presents the results of the porosity as well as the density of the studied samples, it shows us that the cement-based mortar without MPCM has a porosity of 17.90%.This value is approximately similar to the one mentioned in several studies that investigated the porosity for standardized mortars [23,30].

We note that the porosity values for geopolymer mortars without MPCM are lower compared to cement-based mortars. The reason for this difference might be the good workability obtained by the geopolymer mortar compared to the cement mortar, this effect is observed in the study of Yang et al (2020) [31] while they mention that the improvement of the workability of the geopolymer reduces its porosity.

On the other hand, the increase in the concentration of MPCM caused an increase in the porosity and a decrease in the density of all the samples studied (geopolymer mortar and cement).

The decrease in density is due to the difference in density between the sand (2.6 g/cm^3) and the MPCM (0.84 g/cm^3).The increase in porosity might be caused by the fact that the MPCM could not fill the cavities in the matrix due to their agglomeration during mixing and this is due to their agglomeration surface which is larger than the surface of the sand. This agglomeration surface serves to adsorb a quantity of the binder paste whereas this can produce voids during mixing and increases the porosity [32].

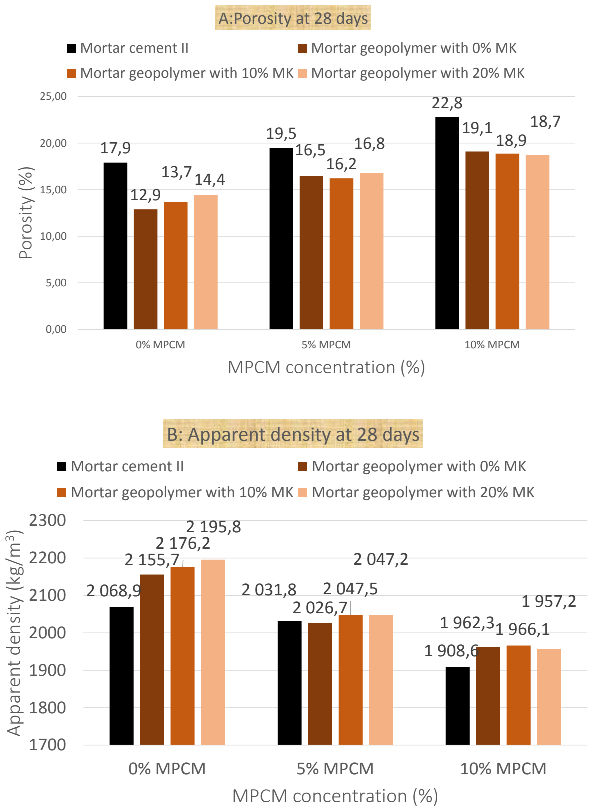


Figure 2:A: Porosity; B: bulk density at 28 days.

However, we find that the addition of MK has no effect on the porosity, this shows that MPCM tend to control the porosity of the geopolymer samples studied here.

In contrast to the porosity results, we note that the density was improved after the addition of MK in the samples without MPCM. This improvement is caused by the high reactivity of MK compared to GBFS which promoted the good dissolution of silica and aluminum to form new gels in the matrix called NASH and CASH gels [18].

These results are in good agreement with the study of HUSEIEN et al (2018) [19] who studied the effect of different replacement of GBFS by MK (0, 5.10 and 15%) and found that with the increase of MK content the density increases due to the creation of NASH and CASH gel.

3.3 Mechanical properties

3.3.1 Compressive strength

The figure 3 shows the results for compressive at 28 and 90 days.

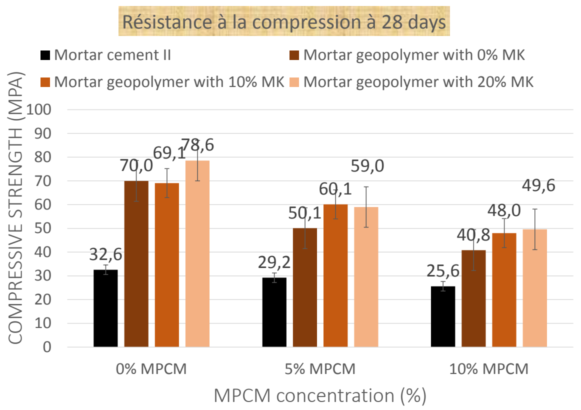
From a global point of view, we observe that there is not a large difference between the compressive strengths between 28 and 90 days for the geopolymer mortar on the contrary of the cement-based mortar which shows an increase in its compressive strength between 28 and 90 days.

Actually, the geopolymer can gain most of its mechanical strength in the first days of its curing due to its strong chemical bonding [18].

The two figures 3.A and 3.B show that the rate of increase of MPCM of 5 and 10% decreased the mechanical strengths of cement-based and geopolymer-based mortars until reaching the value of 25.6 MPA and 40.8 MPA at 28 days. A similar effect is observed for the 90-day period.

The reduction in compressive strength is due to the effect of replacing MPCM with sand, as MPCM has low stiffness and mechanical strength compared to sand and can easily fracture under compressive force [10,17].

On the other hand, the increase of porosity in the matrix after the incorporation of MPCM is one of the causes of the reduction of the mechanical strength as we observed in our results detailed in the previous section [32].



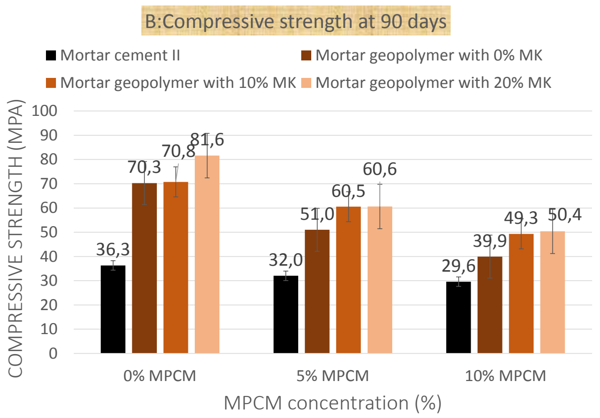


Figure 3: Compressive strength A: 28 days; B: 90 days.

We notice that the geopolymer mortar with 0% MK has a fairly high mechanical strength. NAZARI et al (2015) [33] reported this increase is caused by the high content of activator.This is explained by the high Si/Al ratio and the good dissolution of silica ions and alumina from the GBFS due to the high PH of the solution, resulting in a very efficient gel mechanically [33].Bernal et al [18] (2012) noted that this increase in compressive strength is caused by the calcium that was released during the reaction between Na ions and Si and Al ions, which allowed the production of the CASH gel, while this gel is rich in Al unlike the CSH gel observed in traditional concrete.A similar effect was observed by JIMÉNEZ et al (2003) [34].

Furthermore, we remark that the addition of 10% and 20% MK in the geopolymer mortars that have both concentrations of MPCM (5 and 10%) lead to the increase of the compressive strength at 28 days of all the samples. When using up to a concentration of 10 MPCM the compressive strength increased from 40.8 to 49.6 MPA. The same effect of the addition of MK on the increase of compressive strengths is observed also at 90 days.

On another hand, the addition of 10% MK did not showed any improvement in the compressive strength for the geopolymer mortar without MPCM on the contrary to the addition of 20% MK. Concerning this observation we can note that the addition of 10% MK was beneficial on the filling of small voids caused by MPCM, which improved the mechanical performance [35].

This effect of increasing the mechanical strength can be explained by the small particle size and large specific surface area of MK compared to GBFS, allowing it to have accelerated reactivity [36]. The second reason is due to the high amount of Al_2O_3 and SiO_2 in MK[37] which was accompanied by the high activator content[19]. This high activator content promoted the good dissolution of silica and aluminum in the MK, resulting in improved geopolymerization by producing hydrated sodium aluminosilicate gel (NASH) and hydrated calcium silicate gel (CASH), in addition to the CASH produced by the activation of GGBS which is rich in calcium [18] [19].

3.3.2 Dynamic Young's modulus and dynamic shear modulus

The results of the Young's modulus and dynamic shear at different times (48 h, 7,14,21,28,56 and 90 days) are shown in the figure 4 and 5. We notice first of all that these moduli increase with the increase of the curing time for the cement-based mortars whereas for the geopolymer mortars the time of geopolymerization is stabilized at 14th days.This observation is an index which shows us that the hydration of cement remains continuous during this time contrary to the geopolymerization.This was observed in the results of the compressive strength.

Contrary to the compressive strength values, the Young's modulus values of the cement-based mortar are higher than those of the geopolymer mortar. The geopolymer in general case has a low stiffness [23,38], but we note rather that our values are very close to those of the standardized mortars with a maximum value that is equal to 35. 14 GPA at 90 (GP mortar with 20%MK without MPCM).This may be related to the good parameters of the formulations set according to the literature.

Figure 4 and 5 shows that the inclusion of MPCM decreased both moduli for both mortars (cement and geopolymer).These results are similar to the compressive strength results in the previous section.

This incidence is explained in the section of compressive strength, these two moduli depend on the strength of the material [39] so the inclusion of MPCM decreases the stiffness of the matrix due to their low stiffnesses compared to sands.The second reason may be due to the porosity caused by these materials that decreases the resonance frequencies of flexure (f_f) and torsion (f_t) while these are the main elements that control these two moduli.

Comparing the geopolymer mortars with and without the addition of MK we see from figure C and D in both figures 4 and 5 that the addition rates of MK improved the Young's modulus and the shear modulus for all geopolymer samples. With up to a concentration of 10 MPCM the Young's modulus increased from 19.4 to 26.6 GPA.

The optimal rate of MK on Young's modulus is 10% for geopolymer mortars with MPCM, but for geopolymer mortar without MPCM the optimal rate is 20% of MK. These results are in good agreement with the compressive strengths and can be explained by the pore filling effect caused by the activation of silica and alumina in the MK.The study of NGERNKHAM et al (2014) [37] quoted that the geopolymer matrix after the increase of SiO_2 and Al_2O_3 contents as in our case becomes very dense because of CASH and NASH gels and this makes them resistant.

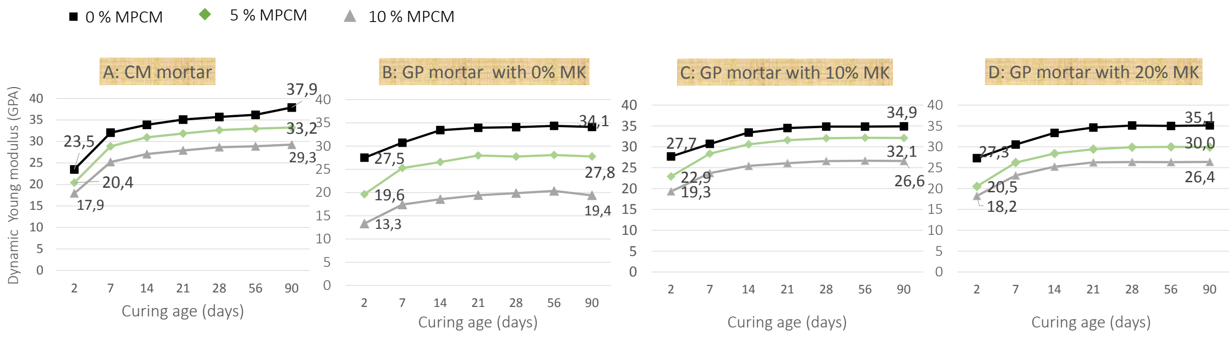


Figure 4: Dynamic Young's modulus as a function of curing time, A:(cement-based mortar), B:(GP mortar with 0% MK); C:(GP mortar with 10% MK); D:(GP mortar with 20% MK).

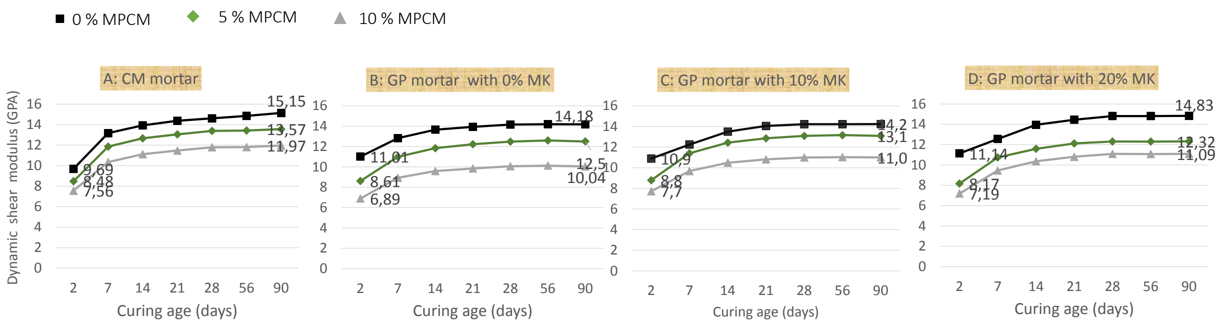


Figure 5: Dynamic shear modulus as a function of curing time, A:(cement-based mortar), B:(GP mortar with 0% MK); C:(GP mortar with 10% MK); D:(GP mortar with 20% MK).

The figure 6 represents a comparison of our results of the dynamic Young's modulus of geopolymer mortars with other researchers who have used dynamic methods on geopolymer mortars [23,40].

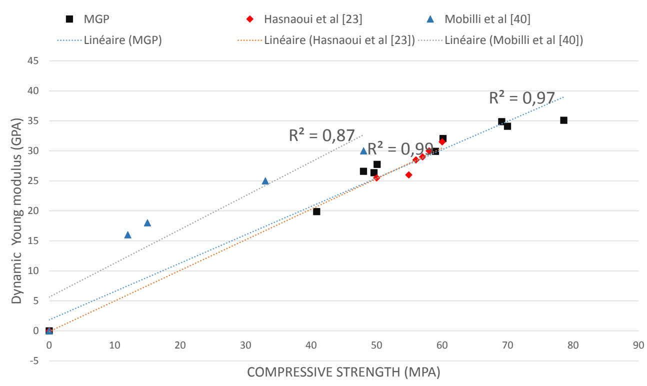


Figure 6 : correlation between dynamic Young's modulus and compressive strengths at 28 days.

We notice from figure 6 a good correlation between this modulus and the compressive strength expressed by a linear variation with a correlation coefficient R2 = 0.97 in our case, and linear correlations in the two references mentioned. Our results are higher than the mentioned reference results.This is probably the consequence of the use in our case of 80/20 of GBFS and MK, unlike Hasnaoui et al (2019) [23] who used 50/50 of GBFS and MK and the use of Mobili et al (2016) [40] up to 100% of fly ash.The main gel observed in the GBFS activation process is the CASH gel while this gel is denser compared to the NASH gel (gel obtained by the activation of metakaolin or fly ash) and has a stronger capacity to fill the pores, which can explain the better mechanical performances obtained in our case compared to the two references above.

4.Conclusion

The objective of this study was to investigate the effect of the inclusion of 10 and 20% of metakaolin in a matrix of geopolymer-MPCM mortar based on granulated blast furnace slag and to compare it with a mortar based on Portland cement-MPCM.

The conclusions of the different tests are the following:

- The geopolymer-MPCM mortars showed good workability with up to 10% MPCM and 20% MK. The maximum flow time is equal to 4.95 seconds which is less than the standard mortar flow time (7 seconds).
- Despite the good results obtained for the total water porosity of geopolymer mortars (without MK) compared to cement-based mortars, we did not obtain accurate results on the porosity of geopolymers after the addition of MK, this can be explained by the fact that the porosity was controlled only by the incorporation of MPCM.
- Geopolymer-MPCM mortars have shown good compressive strengths compared to cement-based mortars.This is related to the strong mechanical structure of these materials highlighted by other researchers.
- The inclusion of 10 and 20% metakaolin increased the compressive strength, Young's modulus and dynamic shear modulus of all geopolymer-MPCM mortar samples. The minimum compressive strength of the geopolymer-MPCM mortar after the inclusion of MK is equal to 49.3 MPA, which is higher than the strengths required by the construction standards.

Finally, we conclude that our geopolymer-MPCM mortars developed in this study are apparently capable of meeting a wide range of applications in the field of construction. Due to the good mechanical, physical (reduced porosity) and workability performances compared to Portland cement-MPCM based mortars.

Nomenclature

MPCM: Microencapsulated phase change materials
GP: Geopolymer
GGBS : blast furnace slag
MK : Metakaolin
SS: Sodium silicate

Declaration of Conflict of Interests

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

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