

RESISTANCE TO SEAWATER OF A NEW CEMENT MORTAR BASED ON ADDITIONS OF RECYCLED PRODUCT

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A b s t r a c t

This paper aims to explore some sustainability indicators including mainly the resistance to seawater of mortars containing 5% kaolin filler in the sand and different types of eco-products as cement additions. To this end, an experimental study was carried out with the aim of evaluating the effects of three types of cement additions (glass powder (GP), metakaolin (MK) and brick waste (BW)) with contents of 5, 15 and 25% as well as binary and ternary combinations of these same additions. The results obtained made it possible to show the effectiveness of filler and cement additions (glass powder (GP) or / and metakaolin (MK) compared to brick waste (BW)) in improving the durability of mortars against aggressive agents such as seawater. In the end, the cement mortars made from 25% MK, 25% GP and this compound of 5% GP and 25% MK showed good resistance to attack by seawater.

Keywords: durability, eco-products, brick, glass powder, kaolin filler, metakaolin, new mortar

1. INTRODUCTION

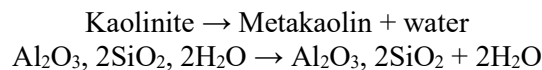
Cement is the most used material in the world of construction thanks to its technical qualities. However this massive use harms our environment because of

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the pollution that accompanies its manufacturing process. To remedy this, and failing to make the process less polluting, engineers and researchers have devised other ways to reduce the production of cement and thus reduce this pollution. Among these means, the partial substitution of cement by products from the recycling of eco-products or waste during the manufacture of concrete and mortars. These findings have confirmed the results of several other recent studies [9, 11, 8, 23, 4] that have shown that glass powder (GP) has very interesting pozzolanic properties and can be used as an alternative cementitious additive. The results of these studies also revealed that GP gives better results in concretes such as resistance to penetration of chlorine ions and give compressive strengths comparable to those obtained with concretes and mortars containing silica fumes (SF). Other studies [14, 3, 18] have shown that the use of brick waste (BW) and granulated blast furnace slag as an addition in concrete and mortar give completely satisfactory results, also the feasibility of producing mixtures of slag cements activated by alkalis from a crushed mixture of slag and red clay brick waste. In general, brick-based mortar or concrete (used as powder or aggregates), gives lower mechanical strengths than the control concrete because the brick has a high porosity and a high power of water absorption.

In addition to the recycling of eco-products: Metakaolin (MK) is a synthetic pozzolan, resulting from the treatment of kaolinite, a clay quite common on the surface of the globe. Metakaolin belongs to the family of ultrafine, mineral particles smaller than 10 μm , used in concrete to improve mechanical properties and durability.

Metakaolin (MK) is obtained by calcining kaolinite at a temperature between 500°C and 800°C. Industrially there are essentially two methods of calcination, one slow calcination (around five hours in production units) and flash calcination (very fast in comparison with slow calcination). At a high temperature, the kaolinite loses its water by dehydroxylation according to the following equation:



Metakaolin is considered to be a pozzolanic material, that is, it reacts in the presence of lime to form hydrated calcium silicates and hydrated calcium silico-aluminates.

We can also promote local materials such as Kaolin (K) which is clay which found in abundance in most parts of the world. The use of kaolin (K) and metakaolin (MK) as additions in the manufacture of mortars and concretes has been the subject of several studies [19, 10, 6, 2, 1]. Most researchers show that metakaolin (MK) is a good addition that reduces the pores and makes the structure impermeable. The results revealed that the inclusion of metakaolin

significantly reduced shrinkage, but increased the mechanical strength of concretes and mortars. The researchers showed that concrete with metakaolin (MK) gave an improvement in sulphate resistance compared to simple concrete. In a previous article [5], we proved the feasibility of substituting a percentage of cement by combinations of glass powder (GP), brick powder (BW) and metakaolin (MK) dust in the manufacture of mortar. The additions are used as substitution of 5, 15 and 25% in the cement. On the other hand, we used kaolin (K) as filler in the sand with a content of 5% and also used after conversion into metakaolin as a binding activity in the cement to improve the compactness of the cement matrix. This clay was calcined in a laboratory oven at a temperature of 750 ° C for a period of 4 hours, after cooling, the calcined clay was milled. The calcination of the clay at 750 ° C allowed the departure of the water of constitution (the dehydroxylation) and the formation of the metakaolinite with an amorphous structure which makes it more reactive than the starting clay kaolin (K). We have been able to conclude that the incorporation of filler into the sand and the additions into the cement made it possible to obtain mortars with physico-mechanical and durability properties (porosity and absorption by immersion as well as sulphate resistance). More or less superior to that of the reference mortar.

In terms of durability, in seawater the dissolved salts are mainly chlorides and sulphates. The attack of concrete and mortar is a result of separate but more or less simultaneous reactions between sulphates and chlorides and the constituents of the cement. Chlorine can be inserted into the crystal lattice of C-S-H. It makes the fibers disappear and creates a honeycomb structure: a lattice is formed whose mesh becomes more and more loose when the amount of dissolved lime increases. Besides, when exposing to chloride environment, the chloride binding capacity of hardened matrix can be significantly increased by metakaolin addition. The literature indicates that MK may be a promising material for limiting the diffusion of chloride [22, 15, 13, 20, 16, 21]. The mechanism of these degradations involves many parameters related to the characteristics of cement and concrete, the nature of the aggressive agent and the conditions of exposure. We will consider here some factors that condition the chemical resistance of concrete and mortar vis-à-vis the seawater. Most of the additions play a very important role in aggressive environments such as seawater because these additions give a compact structure because of its small porosity which leads to the small absorption of seawater. As the metakaolin (MK) participates in the closing of the pores and consolidates the structure which is more resistant to external aggression and avoids the deterioration of the specimen [12, 7].

In this work, which completes the previous one, we study the influence of the substitution rate of mineral additions on the behavior of unary, binary and

ternary mixtures of mortars in a marine environment. It is about the absorption of the water by capillarity and the aggressions of the sea water while making a comparison with mortars kept in the fresh water.

2. MATERIALS AND METHODS

2.1. Raw materials

This work being the continuation of the previous work [5], we used the same raw materials as in the previous study, we simply give here a summary and we invite the reader to read the article in question for more details. The physicochemical characteristics of raw materials used are summarized in Tables 1 and 2.

Cement

The cement used is an artificial portland cement CPA-CEM1-42.5 (CRS 400) coming from the Ain Touta cement plant (East of Algeria).

Sand

We used a coarse sand from the Tebessa sand pit (eastern Algeria), to which was added 5% of kaolin fillers

Mineral additions

The kaolin (K) used is a very fine dust from the ETER ceramics factory in Guelma (eastern Algeria). Its fineness determined using the Blaine digital device is: $SSB = 12603 \text{ cm}^2 / \text{g}$

The Glass powder (GP) was obtained by grinding bottle waste using a ball mill. Its measured fineness is: $SSB = 5067 \text{ cm}^2 / \text{g}$

The brick waste (BW) was micronised into powder by grinding. The powder obtained has a fineness of $SSB = 7718 \text{ cm}^2 / \text{g}$.

The metakaolin (MK) was obtained by calcination of the clay at 750°C . for 4 hours and then after ball milling a fineness of $SSB = 8781 \text{ cm}^2 / \text{g}$ was obtained.

Adjuvant

The adjuvant used is the 'MEDAPLAST40' produced by the company GRANITEX (Algeria). It is a super plasticizer high water reducer.

Table 1. Chemical compositions of cement and mineral additions

	CaO	SiO ₂	Fe ₂ O ₃	Al ₂ O ₃	MgO	K ₂ O	Na ₂ O	SO ₃	Cl ⁻	CaO libre	PAF 1000°C
C	60.16	23.03	4.25	4.18	1.32	0.82	0.44	2.76	0.018	0.81	2.98
K	0.20	41.97	0.12	38.00	0.07	None	None	0.75	None	None	16.8
GP	8.05	69.10	1.80	0.89	1.65	0.28	18.18	0.05	0.004	None	None
BW	2.31	69.05	1.52	23.02	1.05	2.59	1.28	0.04	0.089	None	None
MK	1.29	57.98	0.75	38.31	0.11	0.21	0.89	0.36	0.006	None	None

Table 2. Physico-mechanical characteristics of cement

Poids spécifiques (g/cm ³)	Surface spécifiques (cm ² /g)	Consistance Normale (%)	Temps de prise (mn)		Retrait 28 j (um/m)	Résistance traction 28 j (MPa)	Résistance compression 28 j (MPa)
			Début	Fin			
3.06	3901	25.92	135	200	660	7.40	47.28

2.2. Formulation of mortars

In order to study the influence of the various mineral additions used, 22 mixtures of mortars were composed.

Unary combination mortars with a single addition, others were formulated from binary and ternary combinations. For each composition, 3 test pieces were tested.

Adopted combinations include (5 and 25%) assays for six binary combinations and (5, 5 and 15%) for three ternary combinations.

The different types of mortar mixtures have been prepared according to the standards in force (EN 196-1). The slump test was carried out on fresh mixtures. For tests in the hardened state, prismatic specimens (4x4x16) cm³ were made. Samples are taken 24 hours after demolding.

After 28 days of storage in the freshwater (curing), the specimens for chemical treatment are submitted in seawater to monitor their behavior for 28, 90, 180 and 360 days.

In what follows we use the following notations:

C1: mortar without K and without SP. 5GP, 15GP and 25GP: mortar with 5%, 15% and 25% GP

C1-SP: mortar without K and with SP. 5MK, 15MK and 25MK: mortar with 5%, 15% and 25% MK

C2: mortar with K and without SP. 5BW, 15BW and 25 BW: mortar with 5%, 15% and 25% BW

C2-SP: mortar with K with SP.

5GP + 25MK: mortar with 5% GP and 25% MK.

5GP + 25BW: mortar with 5% GP and 25% BW.

5MK + 25BW: mortar with 5% MK and 25% BW.

5GP + 5BW + 15MK: mortar with 5% GP, 5% BW and 15% MK.

15GP + 5BW + 5MK: mortar with 15% GP, 5% BW and 5% MK.

5GP + 15BW + 5MK: mortar with 5% GP, 15% BW and 5% MK.

3. RESULTS OF THE TESTS AND INTERPRETATION

3.1. Workability

The measurement of the workability of the mortars was evaluated just after the end of mixing using the manual shaking table.

The incorporation of a superplasticizer into the mortar causes a change in fluidity. The latter is directly related to the chemical nature and dosage of superplasticizer.

The results obtained are summarized in (Figure 1). It is clear that the addition of a superplasticizer increases the workability of the mortar. This is well shown that it is between the mortars C1 and C1-SP or the mortars C2 and C2-SP

Regarding the role of mineral additions, the substitution of cement by glasse powder (GP) gives the mortar a high plasticity which increases with the substitution rate because it is a non-absorbent material. On the other hand, metakaolin (MK) and brick waste (BW) substituted for cement reduce the workability in proportion to the addition rate. This is probably due to the greater fineness of the binder because the MK used in this study is thinner than cement and the BW is an absorbent material. This finding was given by the researchers [17] where they showed that the use of MK as an addition in the mortar gives an influence on the demand for superplasticizer that is to say that the superplasticizer dosage increases with the increase in MK content.

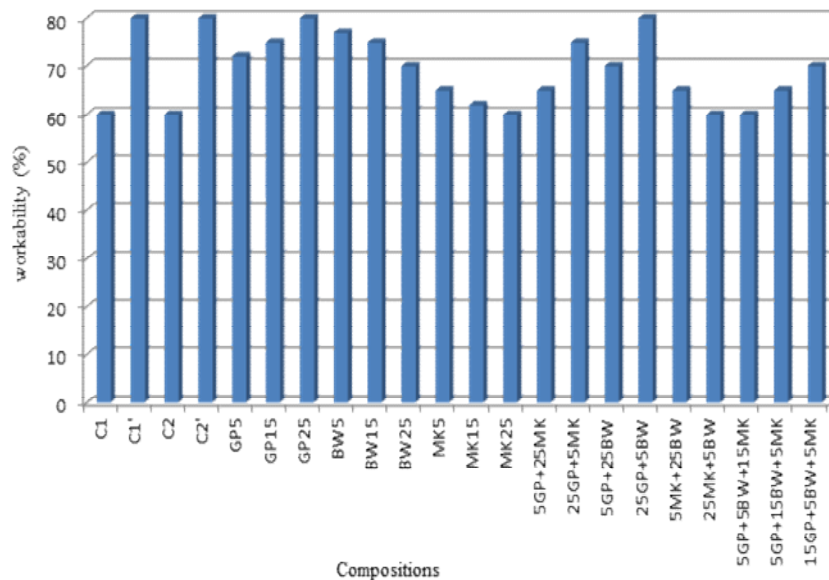


Fig. 1. Effect of superplasticizer addition on workability of mortar compositions

3.2. Absorption by capillarity

Unary combination

Figure 2 shows the evolution over time of the absorption of fresh water by capillarity for the thirteen different unary combinations. The specimens were immersed in water for up to 28 days. Measurements were taken for 24 hours at time intervals varying gradually from 5 minutes to 12 hours.

These results show a continuous increase in capillary water absorption in the compositions.

The mortar (C2-SP) is less absorbent than the mortars (C1), (C1-SP) and (C2), this is probably the gain in compactness due to the use of kaolin fillers and the super-plasticizer.

MK-based and GP-based mortars showed a decrease in capillary water absorption with increasing substitution rates.

This can be explained on the one hand by the fact that glasse powder (GP) is non-absorbent and on the other hand by the fact that metakaolin (MK), having a greater fineness that allows to fill the existing pores between the grains of the mixture and thus increases the compactness.

As for mortars made with BW, they showed an increase in water absorption with the increase of the substitution rate. This can be explained by the fact that brick is a very porous material that promotes absorption. The influence of the rate of the addition on the capillary absorption of the mortar is clear in FIG. 3 and Fig.4

It can also be seen that the kinetics of the absorption is characterized by two zones, first a low speed up to 2 hours. , then the speed increases after 2h.

This pace is observed for all mortars regardless of the type of addition or substitution rate.

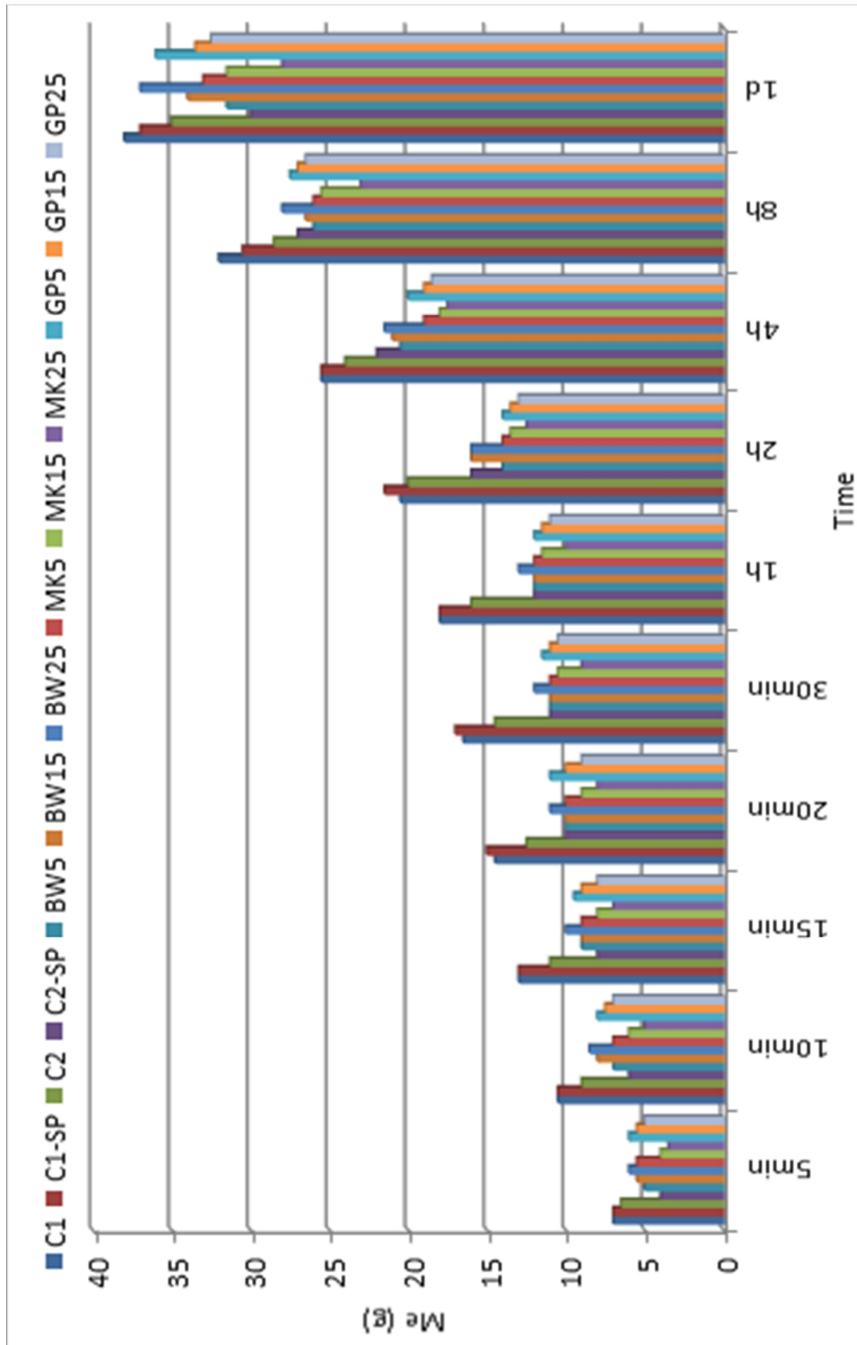


Fig. 2. Kinetics of water absorption by capillarity: Effect of the nature of the addition

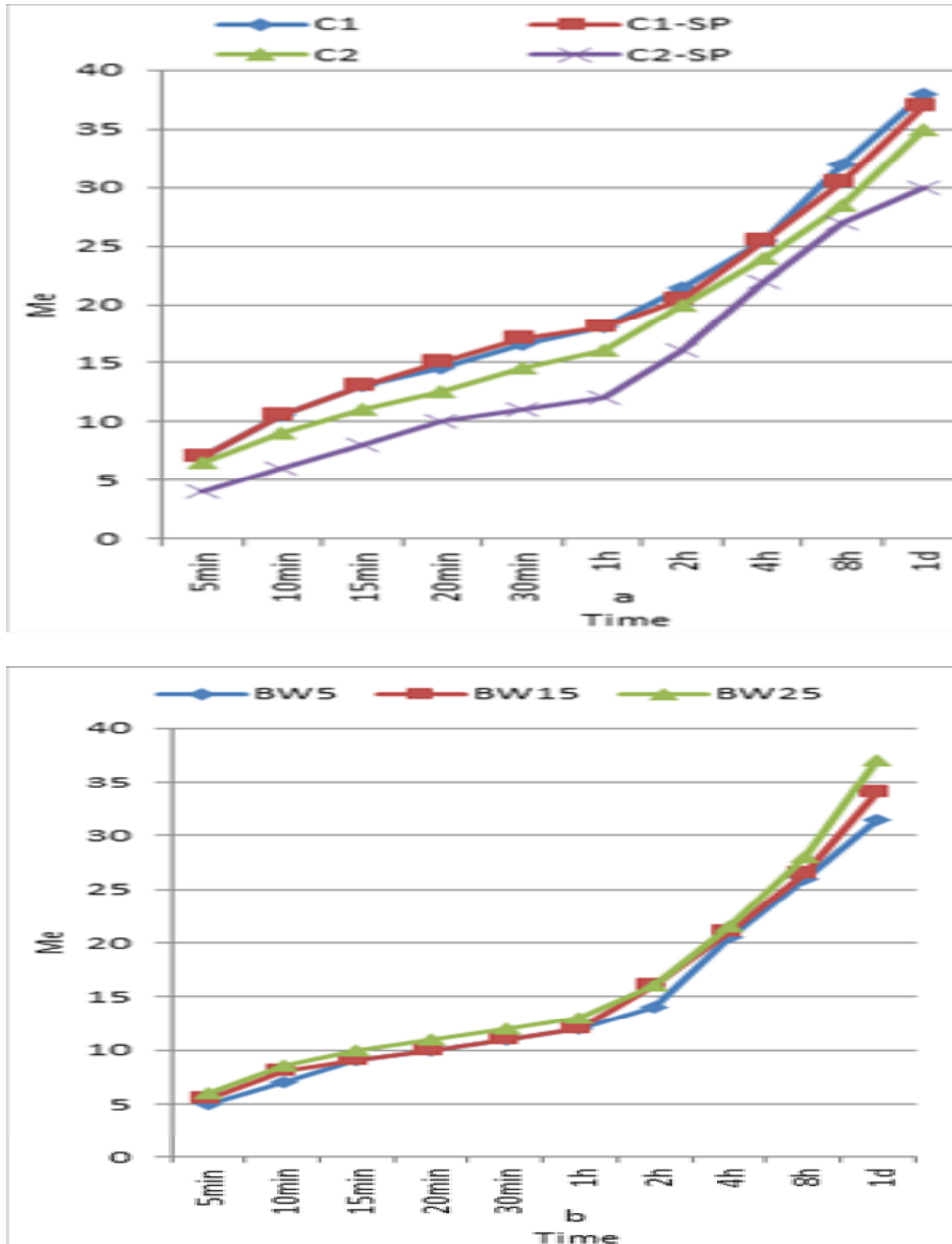


Fig. 3. Kinetics of water absorption by capillarity of mortars:
 (a). Effect of SP ; (b). Effect of BW

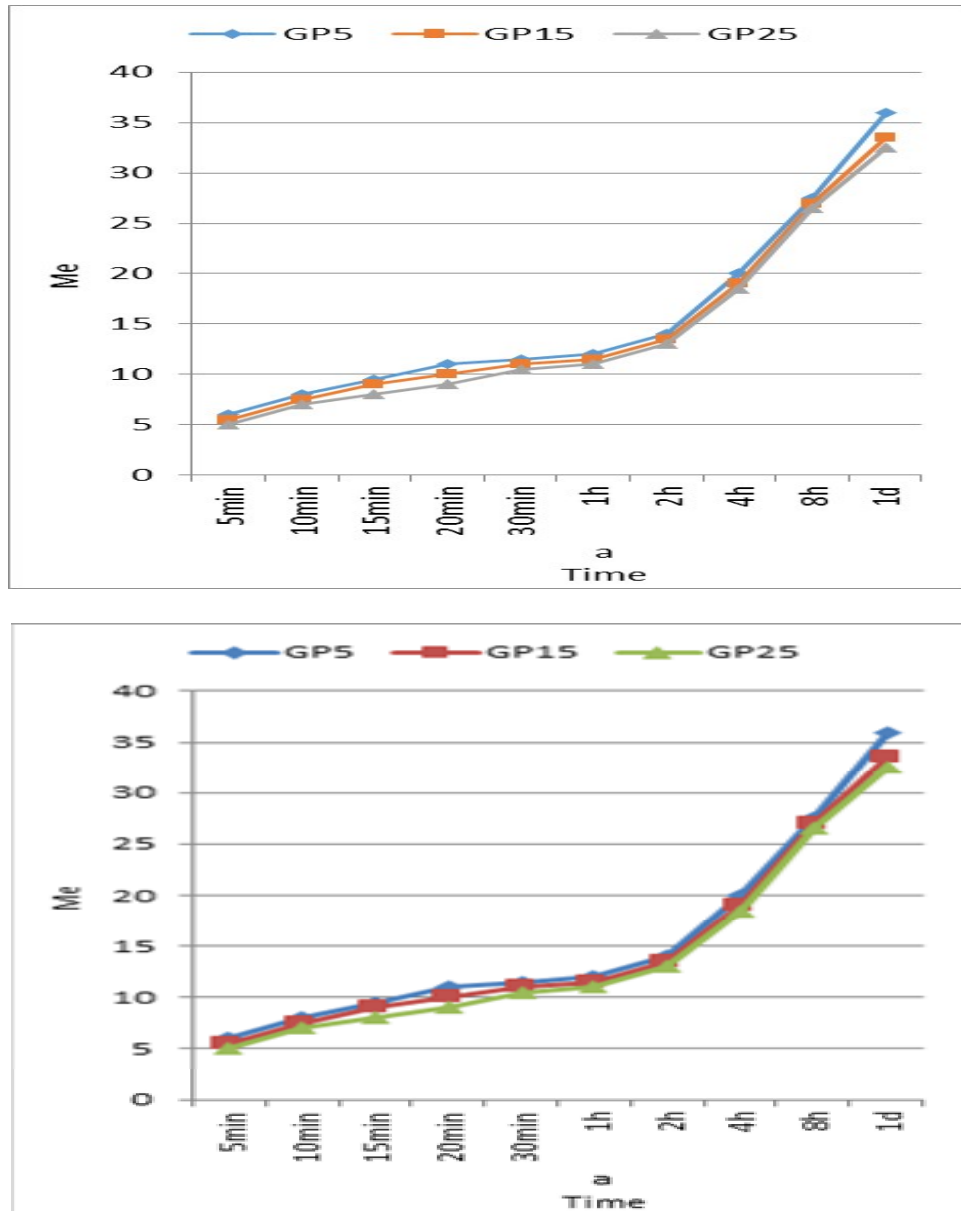


Fig. 4. Kinetics of water absorption by capillarity of mortars:
(a). Effect of GP; (b). Effect of MK

Binary and ternary combinations:

Figure 5 show that the two compositions (5GP + 25MK), (25GP + 5MK) gave the lowest values of capillary water absorption. This confirms the results of the unary compositions.

These results can be explained by the fact that the MK used has a great fineness and thus fulfills a maximum of pores and thus makes the structure of the mortar more compact which gives less water absorption by capillarity. On the other hand, glasse powder (GP) being a non-water absorbing material, when mixed with MK, the two additions give a more compact mortar.

The results of Figure 6 and Figure 7 show that it is the combinations (5GP + 25BW, 5MK + 25BW and 5GP + 15BW + 5MK) give the highest values of water absorption by capillarity especially when increasing the rate of brick waste (BW). The BW is an absorbent material with a high porosity, so it is logical that the mortars containing this mineral addition have a high water absorption by capillarity and increases accordingly with its substitution rate.

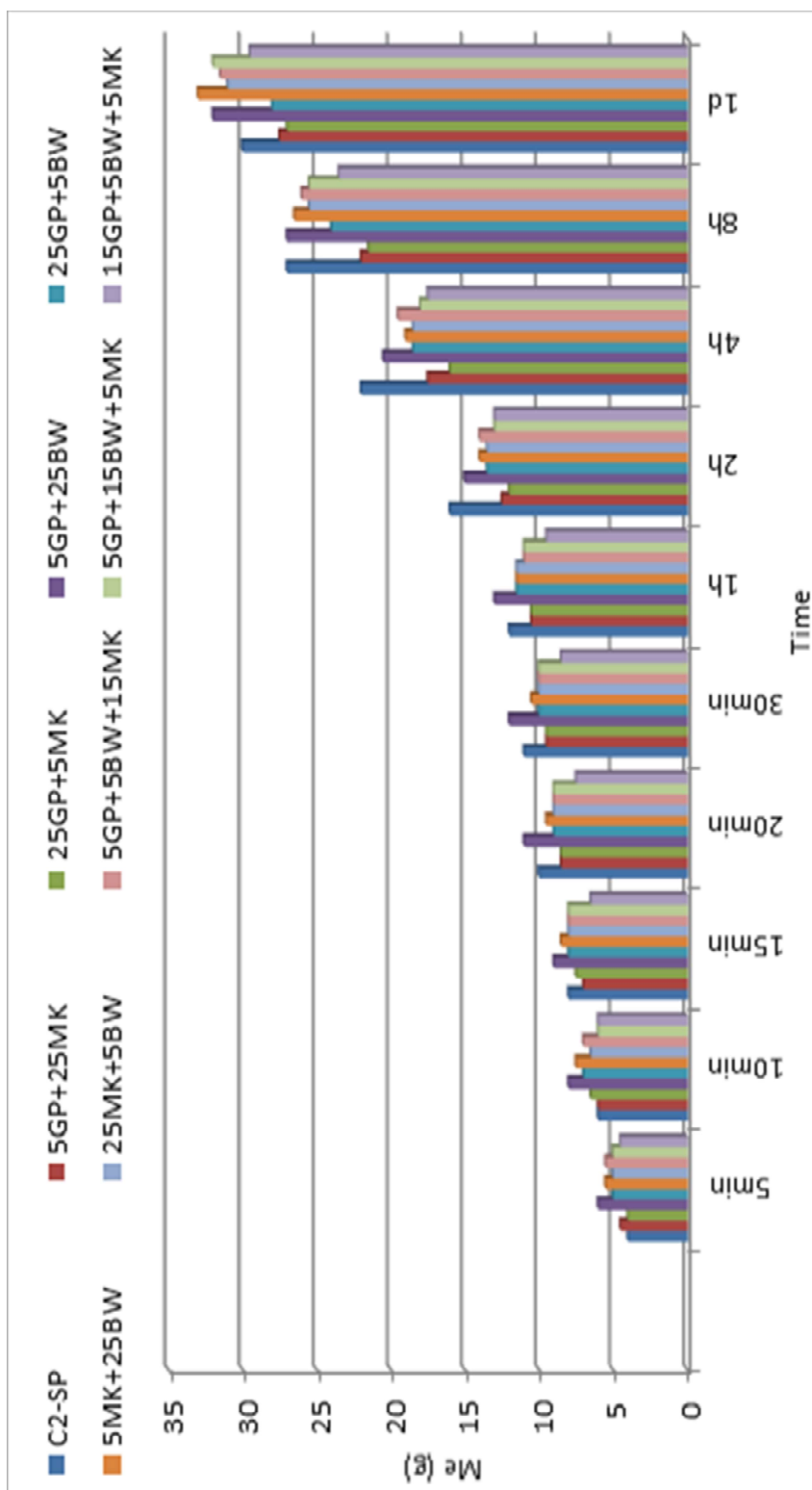


Fig. 5. Kinetics of water absorption of binary and ternary combinations

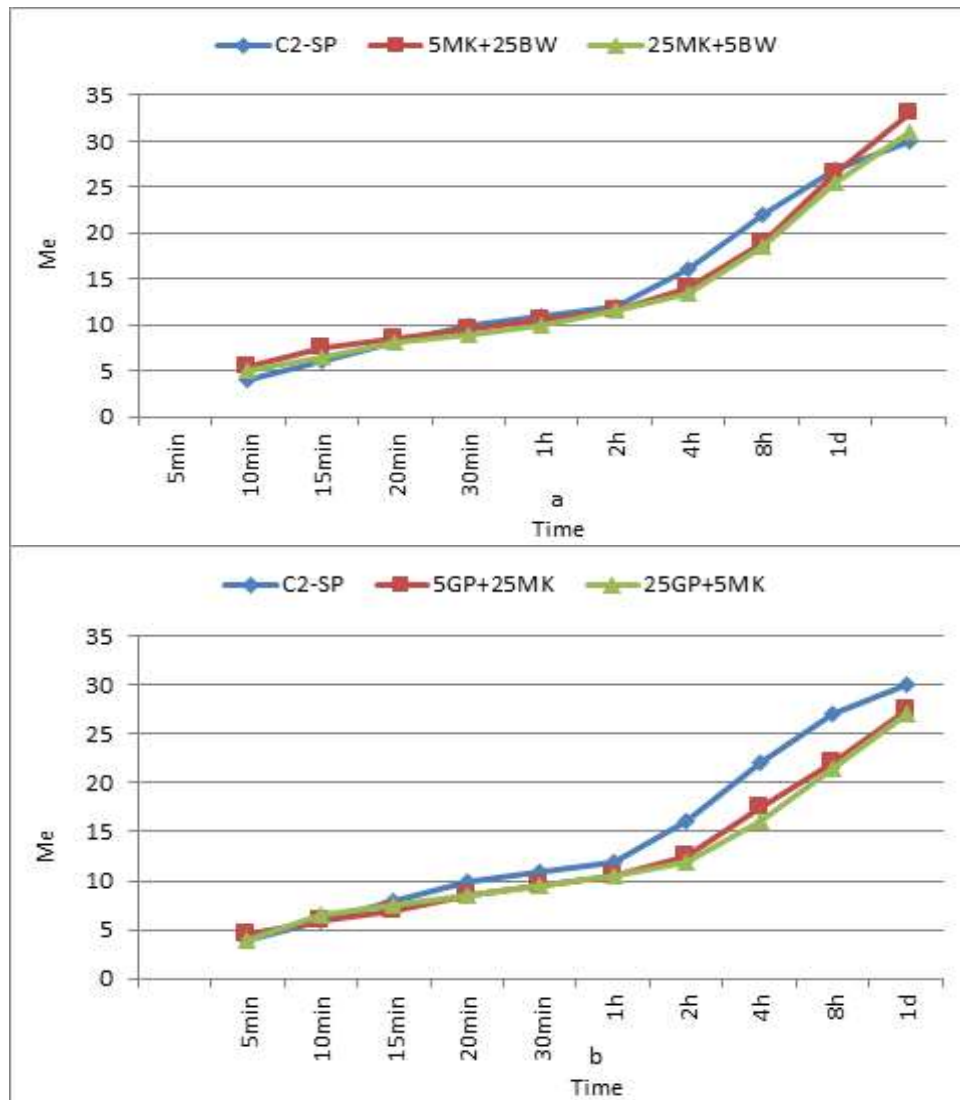


Fig. 6. Kinetics of water absorption by capillarity of binary and ternary combinations: (a). Effect of (MK and BW); (b). Effect of (GP and MK)

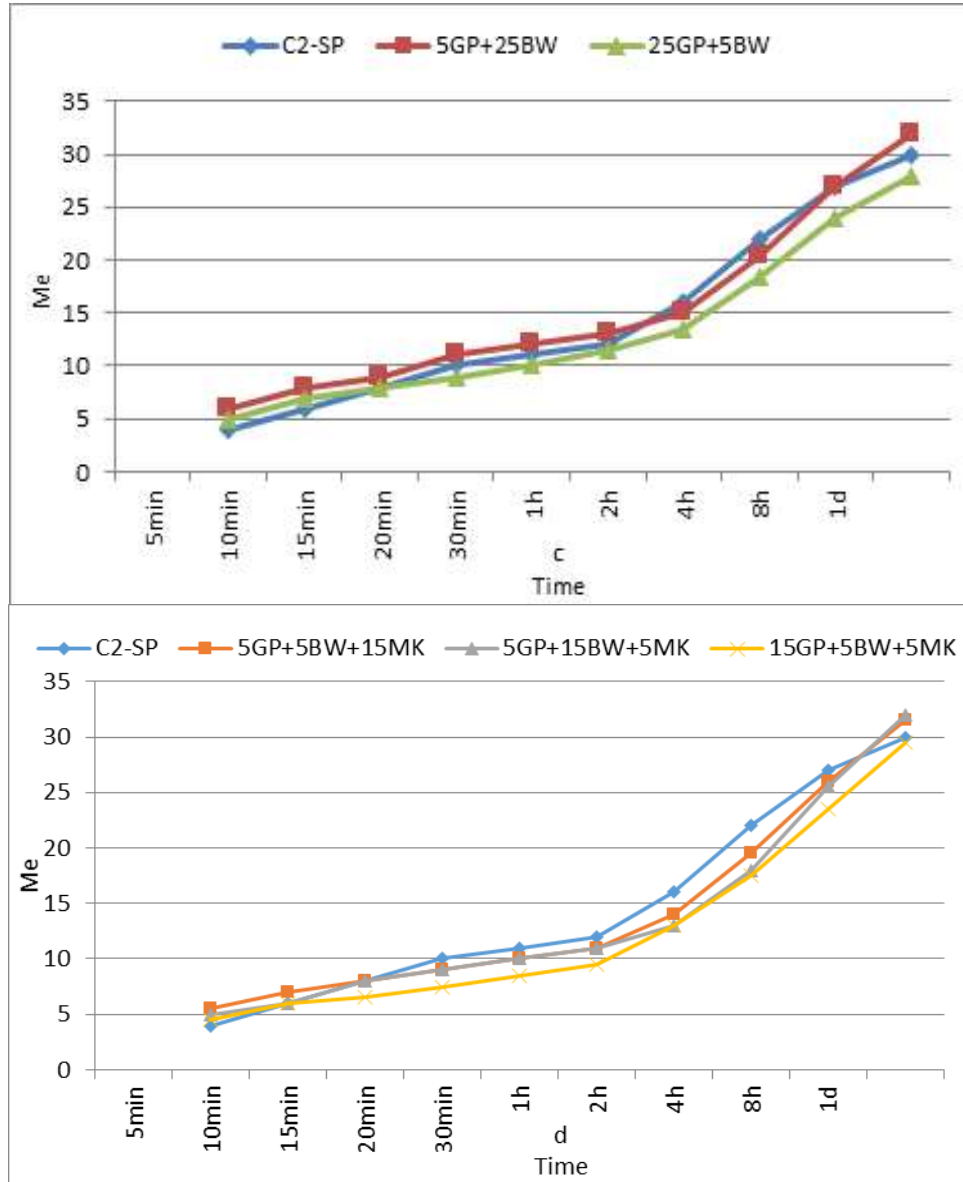


Fig. 7. Kinetics of water absorption by capillarity of binary and ternary combinations: (c). Effect of (GP and BW); (d). Effect of (GP, BW and MK)

3.3. Mechanical resistances

Healthy environment (freshwater)

The specimens are stored in fresh water for the same durations as in seawater. The results of compressive strength are shown in Figure 8.

We observe that the evolution of age-related compressive strength of all mortars used in freshwater increases steadily with age and shows no drop until the age of 360 days. Thus, we notice that maximum strength is obtained for the MK25 cement mortar at the age of 360 days.

The strength of the mortars containing (15%, 25%) GP and (5%, 15%, 25%) of metakaolin (MK) is closer to that of the control mortar (C2-SP).

At 90 days, the strength of the mortar [MK + GP] exceeds other cements. This result is explained by the slow activity of the pozzolanic reaction which is triggered between the reactive silica located in the glassy part of pozzolan or metakaolin, and the portlandite $\text{Ca}(\text{OH})_2$ released by the hydration of CPA.

The pozzolanic reaction is not predominant at young age, this leads to a less intense hydration at young ages by inducing weak resistance. This effect is due to the fact that in the long term, the pozzolanic reaction continues its effect by forming an additional C-S-H which improves the paste-granulate interface, as well as the reduction of the capillary porosity of the mortar.

The tensile strength results are shown in Figure 9. Observations made for the compression tests are the same for the tensile tests.

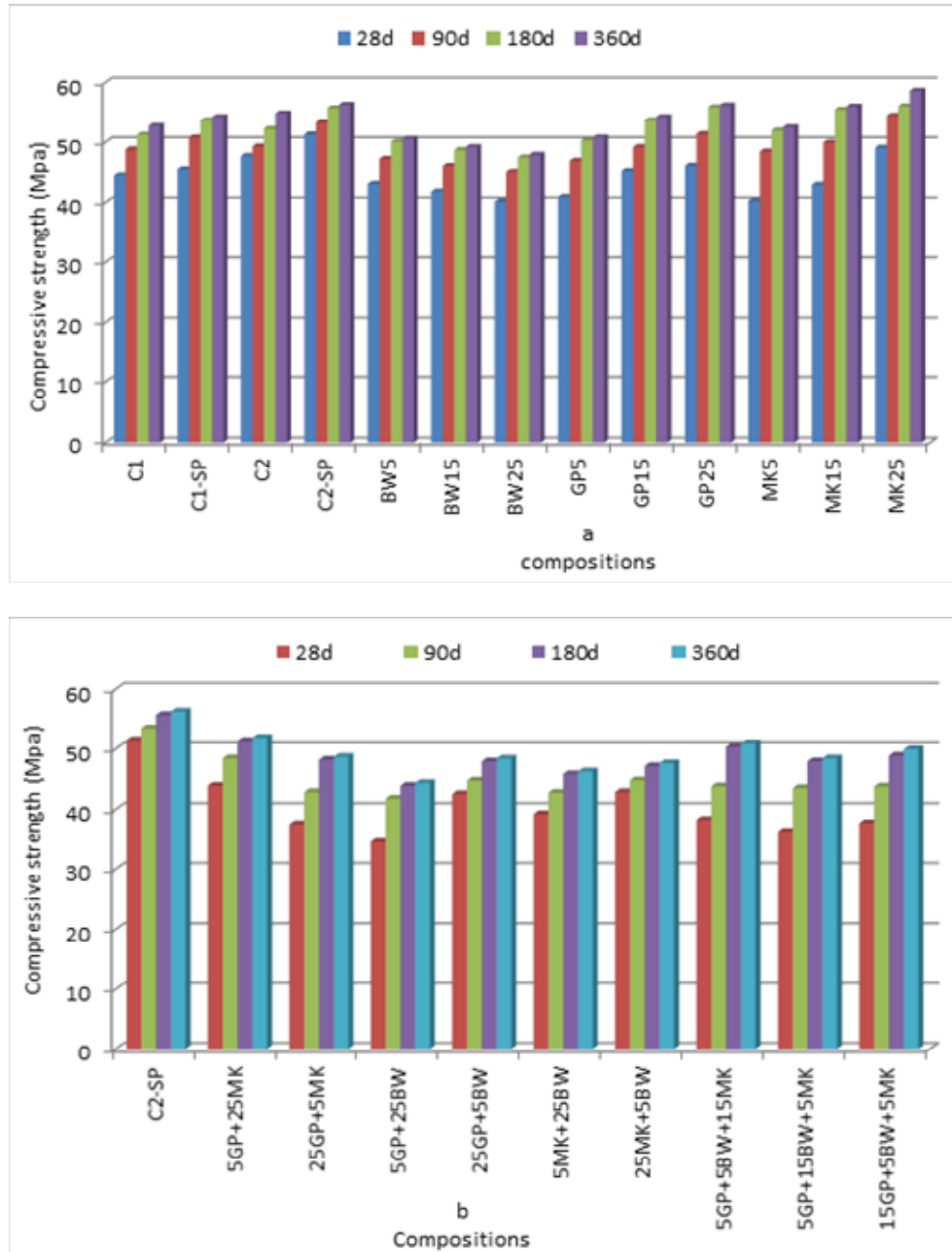


Fig. 8. Resistance to compression of the specimen immersed in water (H2O)
 (a): separate additions; (b): combined additions

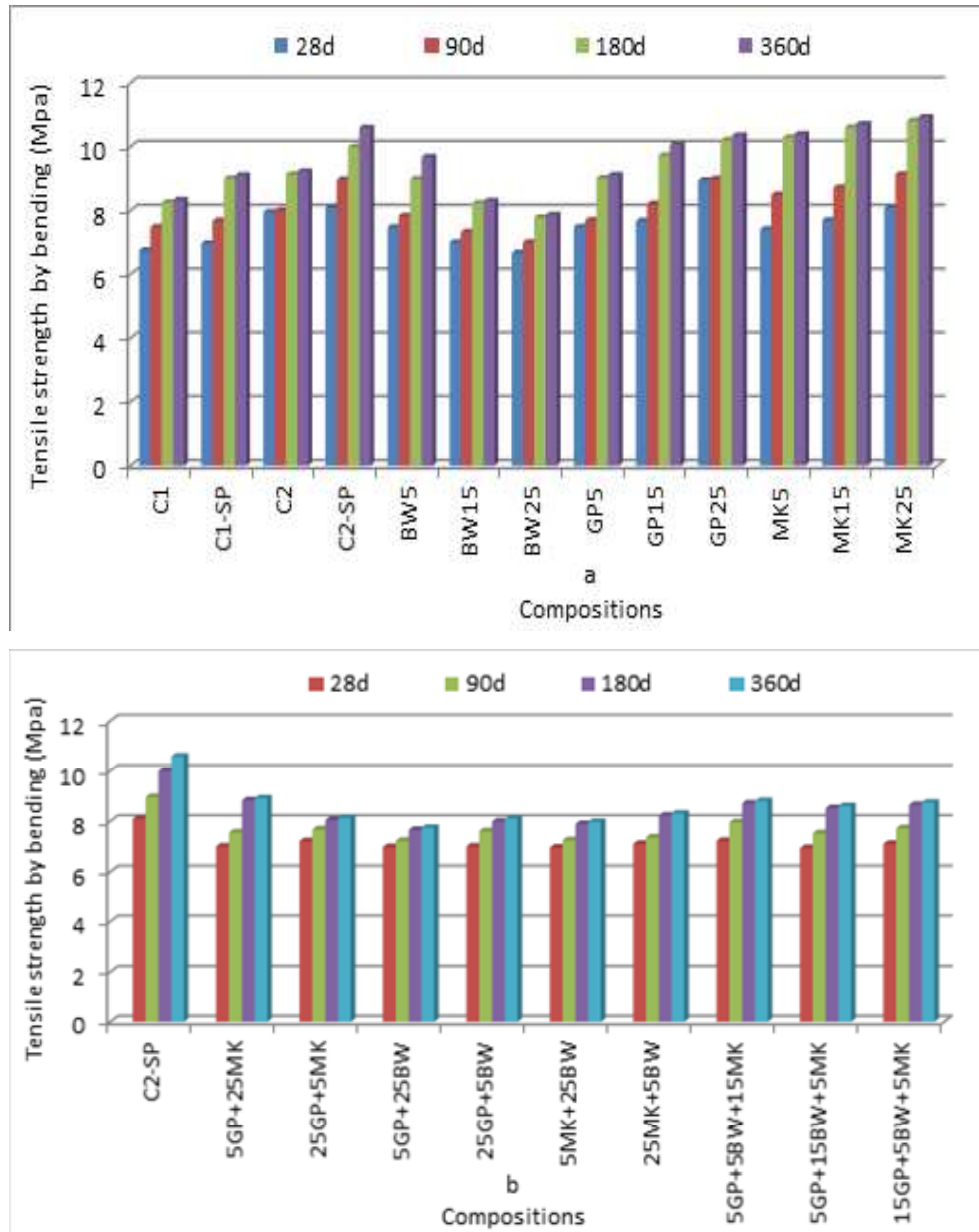


Fig. 9. Tensile strength of specimens immersed in water (H₂O)
 (a): separate additions; (b): combined additions

Aggressive environment (seawater)

The results of the compressive and flexural tensile strength of specimens held in seawater at different ages after a 28-day water cure are shown in Figures (10 and 11).

The results show for all mortars had a progressive increase in compressive strength up to 90 days and flexural tensile up to 180 days immersion in seawater. Beyond this duration a generalized fall in resistance for all mortars was recorded.

Beyond 90 days of cure, seawater reacts chemically within the cement matrix. These reactions are initiated by the numerous ions dissolved in seawater (chlorides, sulphates and carbonates for the principal ones) by a mechanism of dissolution and lixiviation of the calcium compounds of the mortars Ca (OH) 2 and C-S-H. These reactions cause an increase in the porosity of the mortar.

The decrease in resistance observed from the age of 180 days is probably due to the expansive effect (éttringite) insoluble compounds more or less protective, the formation of an inflating hydrate (expansive) leads to the degradation of the material.

With regard to the additions used separately, it is clear that the increase in the percentage of substitution positively influences seawater resistance in the case of metakaolin (MK) and to a lesser degree for glass powder (GP). On the other hand, it is the opposite effect that is observed in the case of BW, so for 25% of substitution there is even a high drop in the resistance of the specimen at the age of 180 days of immersion (FIG. 9).

In the case of combined additions, it is observed that the mortars obtained with a combination containing a high content of brick waste (BW) such that (5MK + 25BW and 5GP + 25BW) have the lowest resistance to seawater attack. In contrast, the mortars obtained with a combination containing a high metakaolin (MK) content such that (25MK + BW and 25MK + 5GP and 5GP + 5BW + 15MK) have the strongest resistances.

This is explained by the fact that the addition of BW gives a non-compact structure because of its high porosity which leads to the great absorption of sea water which has degraded the performance of the mortars especially for the high content of water. BW. Note that this drop in performance is not associated with a deterioration of the external appearance of the test pieces.

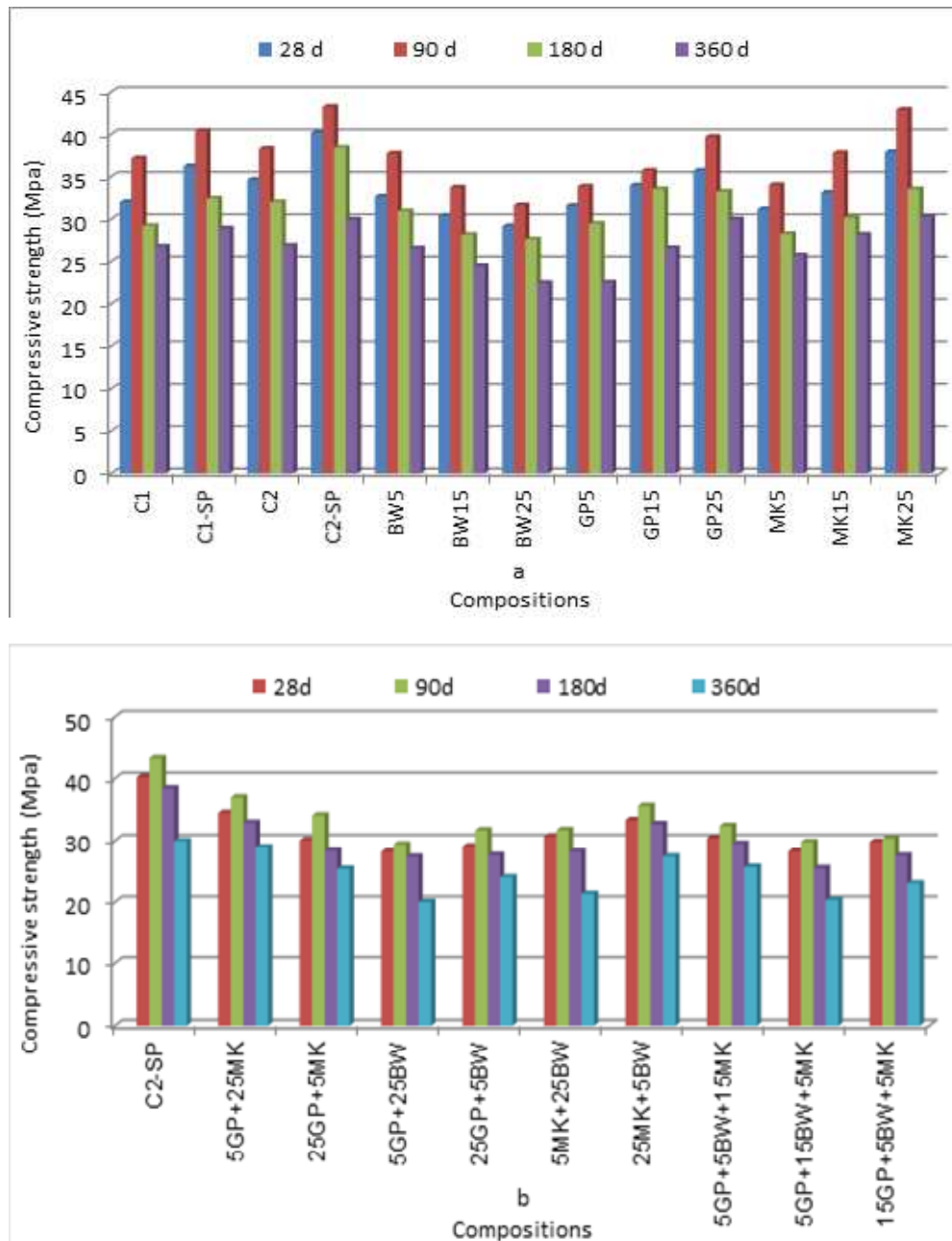


Fig. 10. Resistance to compression of the specimen immersed in seawater
 (a): separate additions; (b): combined additions

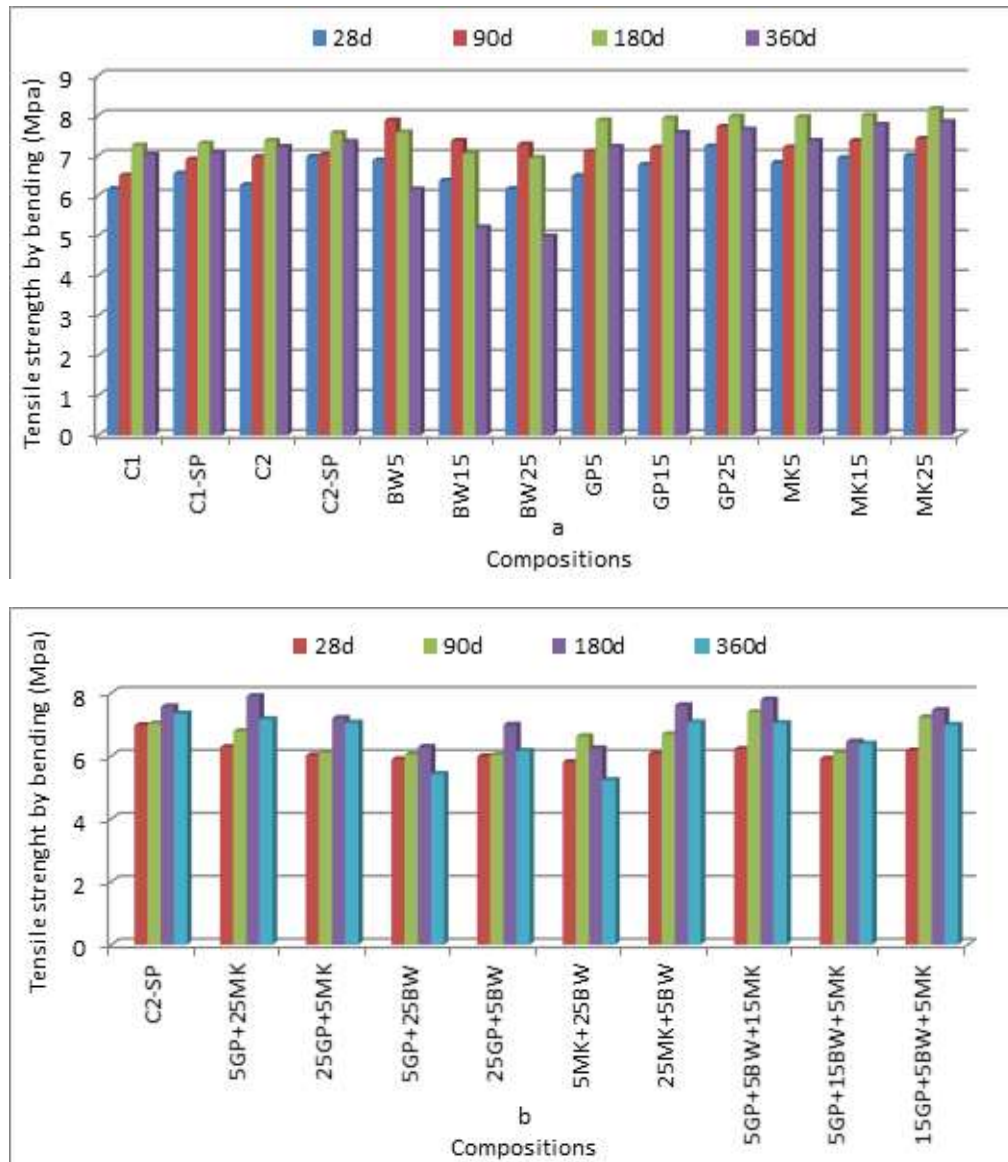


Fig. 11. Tensile strength of specimens immersed in seawater (a): separate additions; (b): combined additions

Comparing the results obtained in the two environments, seawater and freshwater, we found the following:

- A drop in resistance beyond 90 days of mortars immersed in seawater for compressive strengths (R_c) and 180 days of cure for tensile strengths (R_t).
- A degradation of the performances of the mortars especially for the high contents of brick waste (BW) with no degradation of the external aspect of the specimens preserved in the sea water.
- Specimens kept in fresh water show increases in resistance up to 360 days, however, there is an increase in the resistance of specimens kept in seawater up to 90 days only and drops in resistance in the water. 180 days old.
- The resistance of the specimens immersed in fresh water are higher than those immersed in the aggressive marine environment.

4. CONCLUSION

The main purpose of this study is to study the seawater durability of a cement mortar obtained by introducing fillers into the sand and substituting part of the cement with mineral additions combined with each other.

The fillers used are those of kaolin, the mineral additions used are glass powder, brick powder and Metakaolin.

The use of calcined clay (metakaolin) and high-dose glass powder (25%) and low-dose brick powder (5%) help improve durability because of their pozzolanic reactivities that decrease porosity and also the absorption of water which leads to limit the expansion of the aggressions and the degradation of the material.

On the other hand, the high dosage of the briquetting powder increases the porosity and increases the water absorption by capillarity of the test tube, which favors the infiltration of external aggressions and consequently the degradation of the material. For this purpose, the BW must be used at very low levels (at 5% in our case).

The capillary absorption coefficient of metakaolin (MK) and glass powder (GP) decreases with increasing percentage of metakaolin and glass powder. This decrease was explained by a thinner pore structure of metakaolin and the non-absorbency of glass powder, this means that the incorporation of metakaolin and glass powder has a very positive effect on the resistance of mortars to water infiltration by capillarity.

REFERENCES

1. Antoni, M, Rossen, J, Martirena, F and Scrivener, K 2012. Cement substitution by a combination of metakaolin and limestone. *Cem.concr. Res.* **42 (12)**, 1579–1589.
2. Cheng, SK, Shui, ZH, Sun, T, Yu, R, Zhang, GZ and Ding, S 2017. Effects of fly ash, blast furnace slag and metakaolin on mechanical properties and durability of coral sand concrete. *Appl. Clay Sci.* **141**, 111–117.
3. Gonzalez-Corominas, A and Eetxeberria, M 2014. Properties of high performance concrete made with recycled fine ceramic and coarse mixed aggregates. *Const. Build. Mater.* **68**, 618–626.
4. Harbec, D, Tagnit-Hamou, A and Gitzhofer, F 2016. Waste-glass fume synthesized using plasma spheroidization technology: reactivity in cement pastes and mortars. *Constr. Build. Mater.* **107**, 272–286.
5. Harbi, R, Derabla, R and Nafa, Z 2017. Improvement of the properties of a mortar with 5% of kaolin fillers in sand combined with metakaolin, brick waste and glass powder in cement. *Construction and Building Materials*. Vol. **152**, 632-641.
6. Janotka, I, Puertas, F, Palacios, M, Kuliffayova, M and Varga, C 2010. Metakaolin sand-blended-cement pastes: Rheology, hydration process and mechanical properties. *Construction and Building Materials*. Vol. 24.791-802p.
7. Kim, MJ, Kim, KB and Ann, KY 2016. The influence of C3A content in cement on the chloride transport. *Adv. Mater. Sci. Eng.* 1–8.
8. Omran, A, Harbec, D, Tagnit-Hamou, A and Gagne, R 2017. Production of Roller- Compacted Concrete Impact of alternative cementitious material on mechanical and transfer properties of concrete Using Glass Powder: Field Study. *Constr. Build. Mater.* **133**, 450–458.
9. Omran, A, Dumas-Morin, E, Harbec, D and Tagnit-Hamou, A 2017. Long-Term Performance of Glass-Powder Concrete in Large-Scale Field Applications. *Constr. Build. Mater.* **135**, 43–58.
10. Pavlíková, M, Brtník, T, Keppert, M and Černý, R 2009. Effect of metakaolin as partial Portland-cement replacement on properties of high performance mortars. *Cement Wapno Beton*. Vol. **3**,115-122.
11. Pavoine, A, Harbec, D, Chaussadent, T, Tagnit-Hamou, T and Divet, L 2014. *ACI Mat J* **111**, 1–6.
12. Ping, D, Zhonghe, S, Wei Chen, A and Chunhua, S 2012. Influence of metakaolin on pore structure-related properties and thermo dynamic stability of hydrate phases of concrete in seawater environment. *Construction and Building Materials*. **36**, 947–953.

13. Poon, CS, Kou, SC and Lam, L 2006. Compressive strength, chloride diffusivity and pore structure of high performance metakaolin and silica fume concrete. *Constr. Build. Mater.* **20 (10)**, 858–865.
14. Reig, MM, Tashima, L, Borrachero, MV, Monzó, J, Cheeseman, CR and Paya, J 2013. Properties and microstructure of alkali-activated red clay brick waste. *Constr. Build. Mater.* **43**, 98–106.
15. Saikia, N, Kato, S and Kojima, T 2006. Thermo gravimetric investigation on the chloride binding behaviour of MK-lime paste. *Thermochim. Acta* **444 (1)**, 16–25.
16. Shi, ZG and al. 2017. Role of calcium on chloride binding in hydrated Portland cement metakaolin - limestone blends. *Cem. Concr. Res.* **95**, 205–216.
17. Si-Ahmed, M, Kenai, S and Ghorbel, E 2013. *Influence du métakaolin sur la durabilité des mortiers et bétons*. 31èmes Rencontres de l’AUGC, E.N.S. Cachan, 29 au 31 mai.
18. Sun, Z and al. 2013. Synthesis and thermal behavior of geopolymer-type material from waste ceramic. *Constr. Build. Mater.* **49**, 281–287.
19. Thomas, MDA, Hooton, RD, Scott, A and Zibara, H 2012. The effect of supplementary cementitious materials on chloride binding in hardened cement paste. *Cem. Concr. Res.* **42 (1)**, 1–7.
20. Vance, K, Aguayo, M, Oey, T, Sant, G and Neithalath, N 2013. Hydration and strength development in ternary portland cement blends containing limestone and fly ash or metakaolin. *Cem. Concr. Compos.* **39**, 93–103.
21. Wang, GM, Kong, Y, Shui, Z, Li, Q and Han, JL 2014. Experimental investigation on chloride diffusion and binding in concrete containing metakaolin. *Corros. Eng. Sci. Technol.* **49 (4)**, 282–286.
22. Wang, Y, Shui, Z, Huang, Y, Sun, T and Duan, P 2018. Properties of coral waste-based mortar incorporating metakaolin: Part II. Chloride migration and binding behaviors. *Constr. Build. Mater.* **174**, 433–442.
23. Zidol, A 2014. *Durabilité en milieux agressifs des bétons incorporant la poudre de verre*. Université de Sherbrooke.

Editor received the manuscript: 14.12.2020