

PRESSED NON-FIRED BRICKS FROM PHOSPHOGYPSUM WASTE FOR NON-LOAD BEARING WALL

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ABSTRACT

In several countries of the world, phosphogypsum represents a large quantity of waste that poses serious problems of environmental and groundwater pollution. This study aims at recovering phosphogypsum, in its raw state without treatment, in the manufacture of non-load-bearing non-fired bricks. The study starts with the analysis of the radionuclide activity of the materials constituting the bricks, in particular phosphogypsum, in order to avoid any human health problems after the manufacture and use of the bricks. Then, several compositions are tested with several preservation methods in order to optimize the composition. The physical, chemical and mechanical resistance is determined. The results show the possibility to produce non-load-bearing bricks based on untreated phosphogypsum which comply with the standards requirements, using low energy. Indeed, among the considered mixtures, two compositions (60% of PG and of 75% of PG) perfectly verify the physical and mechanical tests. Also, storage of the mixtures for two days in the laboratory and then three days in an oven at 70°C, allows to obtain the best resistance to compression. Thus, the obtained resistance is much higher than the minimum value required for non-load-bearing bricks.

KEYWORDS

Non-fired bricks, Phosphogypsum, Mechanical properties, Radionuclide activity

INTRODUCTION

The industrial production of phosphoric acid generates, following the treatment of phosphate rock, a large quantity of a waste called phosphogypsum (PG). In fact, to produce one tonne of phosphoric acid, around 5 tonnes of PG are generated [1]. Thus, world production of PG, mainly composed of calcium sulphate dehydrate ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), is enormous. It exceeds 280 million tonnes per year [2] of which 22 million tonnes in China [3], 10 million tonnes per year in Tunisia [4] and more than 6 million tonnes in India [5].

The majority of PG is nowadays deposited, without treatment, in large stocks near factories in coastal regions [6]. The problems caused by the enormous amount of PG stored is not limited by the large surface areas occupied, but also extends to environmental problems. Indeed, the presence of heavy metals and radioactive nuclides in PG increases the risk of pollution of the soil, water and atmosphere around the storage areas [1, 3].

In order to reduce stored quantities, several attempts have been made to valorise the PG, mainly in agriculture [6] and in construction sectors. Thus, the use of PG in road structures has been studied. But the results were not encouraging, and the attempts were quickly abandoned in

France as in the USA (Florida) because of the heavy rains [7]. On the other hand, its use in regions with low rainfall, such as southern Tunisia, remains possible [8]. A recent work [9] has examined the use of phosphogypsum for the production of bituminous materials and has shown that PG can improve the mechanical properties of the asphalt binder as well as its performance against rutting.

The use of PG in soil stabilization [10], [11] and in embankment [12] has also been studied. Also, since PG has qualities and properties like natural gypsum, several researchers have studied its use in the manufacture of gypsum [13-15]. Other attempts to valorise PG have considered its use in cement manufacturing [16-18]. Kuryatnyk et al [19] used it as a hydraulic binder but the formation of ettringite led to a loss of strength. In Tunisia, PG has been studied for the manufacture of cement under the name of ultimax cement [20].

One of the most studied uses for PG is the manufacture of bricks, whether non-fired or fired bricks [1, 21-23]. Ajam et al. [7,24] used PG for the manufacture of fired bricks and studied its radioactivity. Their work has shown that the radioactivity measured is acceptable and below the limits prohibited by standards. Zhou et al. [23] mention that, for non-fired bricks, some studies use the autoclaving curing process where green bricks are formed at pressures between 20 and 40 MPa then autoclaved at 100-180°C for 4-8 hours at pressures of 0.8-1.2 MPa. In other studies, the green bricks are formed under high pressure of around 80 MPa. Another process used by Zhou et al. [23] consists of the pre-treatment of Chinese phosphogypsum using two-step hydration process, one before the formation of bricks under a pressure of 20 bars, and the other after. Although these approaches provide excellent mechanical performance, they lead to a considerable consumption of energy in the manufacturing process, which causes a significant amount of greenhouse gases to be released into the environment.

Within this framework, the present work envisages the introduction of untreated PG as a raw material in the manufacture of unfired bricks made of pressed sand for use in non-load-bearing walls. The advantage of the manufacture of unfired bricks is the limitation of the energy consumption during the manufacturing process, which preserves the environment by limiting the amount of the released CO₂. In a first part, and to avoid human health problems after the use of the manufactured bricks, a study of the radioactivity of the bricks materials is presented, the results encourage the use of the PG in bricks. In a second part, several formulations with PG contents that vary between 33 and 85 % are considered with several preservation methods. Characteristics such as appearance, water absorption and compressive strength are determined to find the formulations that meet the requirements of the standards.

MATERIALS

Sand

In this study, the used sand is a 0/5 silica sand from the Khelidia quarry (northern Tunisia). The particle size analysis is carried out by wet sieving. Figure 1 shows the granulometric distribution of the sand.

The uniformity coefficient (C_u) and the curvature coefficient (C_c) are evaluated at 1.54 and 3.62 respectively. The obtained value by the sand equivalent test is 51.81, that of the methylene blue test is 0.76. Thus, it is deduced that the sand contains non-clay fines.

The chemical composition of the sand (Table 1) is determined using the X-ray fluorescence spectrometer. According to table 1, it is noticed that the used sand is relatively low in alumina which acts directly on the plasticity of the mixture, with a high silica content of more than 86% which serves to constitute the skeleton of the mixture, and with some elements which play the role of fluxing agents (K₂O, Na₂O,...).

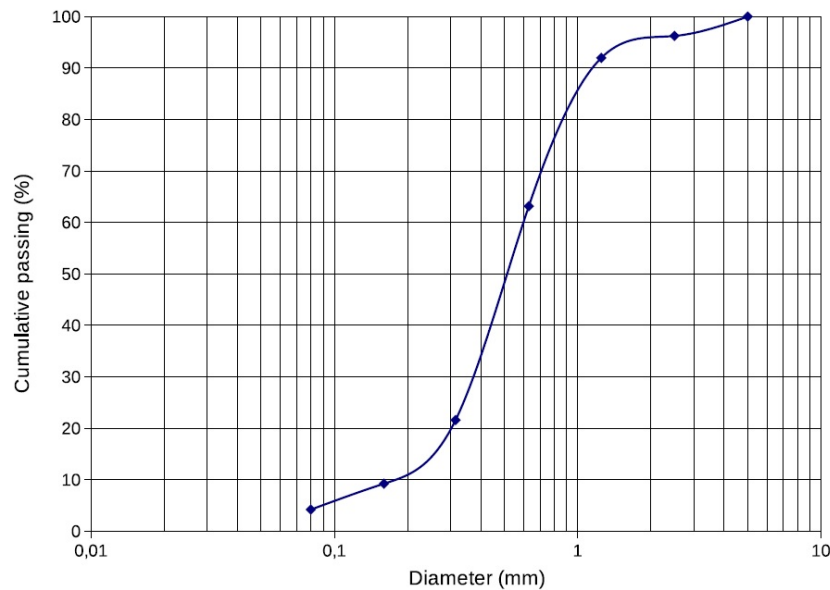


Fig. 1 – Granulometric curve of sand

The real grain density is determined by the pycnometer method. The studied sand has a density of about 2420 kg/m³.

Tab. 1: Chemical composition of the used sand

Designation	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO	Na ₂ O	K ₂ O	Loss Ignition
Content(%)	86.15	3.22	1.27	0.86	1.32	0.76	1.13	5.42

Phosphogypsum

The used phosphogypsum comes from the Sfax region in Tunisia. Two tons of phosphogypsum are taken from one of the two slag heaps (Figure 2), which is 12 m high.



Fig. 2 – Phosphogypsum waste heap

In order to have a sample that represents as much as possible the characteristics of the heap, three locations are chosen for the sampling of the PG: the left and right sides of the bottom and at

an altitude of about 10 m. The PG is then homogenized by means of shovels, before being placed in 50 kg bags for laboratory tests.

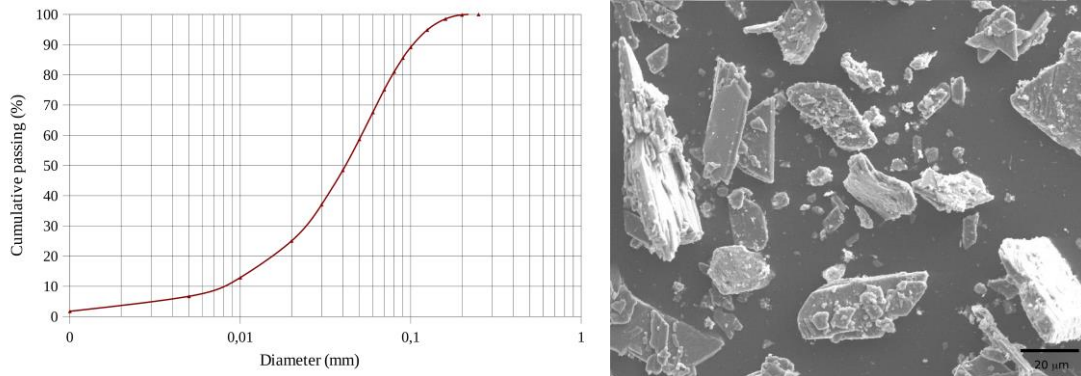


Fig. 3 – Granulometric curve and SEM photo of the studied Tunisian PG

Phosphogypsum is characterised by a grey colour. The granular distribution of PG (Figure 3) is obtained by laser diffraction (laser granulometry) and shows that PG looks like fine sand (<250 µm and about 80 % fine), with a uniform granulometry and a permeability equal to $2.6 \cdot 10^{-6}$ m/s. Figure 3 shows also a SEM photograph of phosphogypsum. This figure shows a tabular form of crystals and that shows their length is between 20 and 200 µm, with a median length of 50 µm.

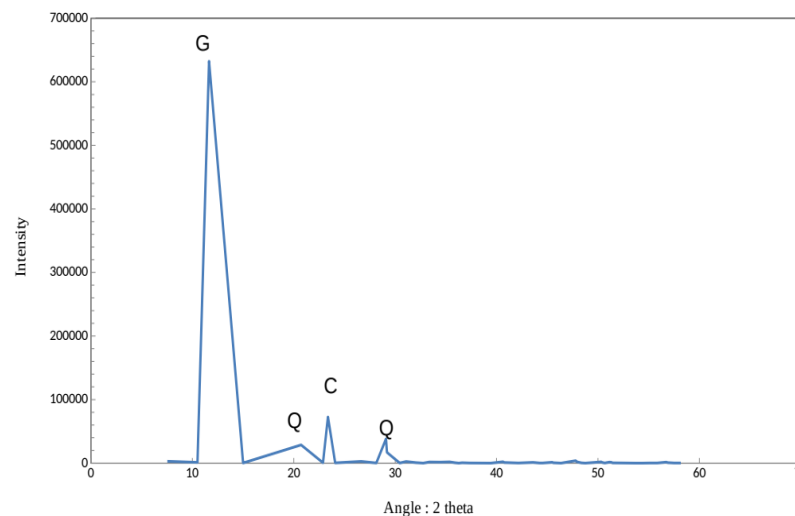


Fig. 4 –XRD patterns for the studied PG

Figure 4 shows the mineralogical phases determined using X-ray diffraction (XRD). It shows the presence of gypsum (86.15 %), calcite (10.76 %) and quartz (3.07 %). Table 2 presents the chemical analyses of the PG, which consists mainly of calcium sulphate (77 % of CaSO_4). Table 2 also reveals that the amount of silica present in PG is very low (1.37 % of SiO_2).

Tab 2: Chemical composition of PG

CaO	SO ₃	P ₂ O ₅	F	SiO ₂ _x005F_x005F_x005F_x005F_x0001_	Fe ₂ O ₃	Al ₂ O ₃	MgO	Ignition loss at 1000°C
32.8	44.4	1.69	0.55	1.37	0.33	0.11	0.007	22.3

Pycnometer method is used to determine the real density of PG. The density measured for the PG studied is 2.31 g/cm^3 . This value is very close to the 2.32 g/cm^3 of natural gypsum.

METHODS

Elaboration of the composite

In their work, Felfoul et al. [25] show that the acidity of PG is an unfavourable parameter for the mechanical properties and water resistance. The study of Yang and al. [22] shows that hydrated lime, by neutralising the acidity of PG, eliminates the negative effect of acids and organic impurities on mechanical resistance of bricks.

In the present work, a percentage of hydrated lime with a Ca(OH)_2 content of 97.1 % is used to neutralise the PG. Thus, the optimization of the mixtures is based on the results of the evolution of the pH of the PG with the addition of the lime presented in Table 3.

Tab 3: Effect of lime on phosphogypsum pH

% of added lime	0	5	10
pH measurement	3.8	6.7	7.5

It is noticed that the variation of the lime content between 5 and 10 % has little influence on the neutralization. So, 6 % of lime may be sufficient to neutralize the PG. In addition to PG, and lime, a percentage of sand is added as aggregate and a binder is used to help the hardening of the bricks. The binder is a Portland limestone cement II/A.L32.5 used in small percentages varying between 5 and 10%. Table 4 shows the composition (percentage by weight) of the different mixtures of the studied blocks.

Tab 4: Composition of the different mixes (% weight)

Mixture	PG	Sand	Cement	Lime
M ₀	33	67	0	0
M ₁	45	45	10	0
M ₂	60	25	9	6
M ₃	75	10	5	10
M ₄	65	20	0	15
M ₅	85	0	0	15

Manufacturing process

In this study, miniature phosphogypsum-based pressed sand bricks (prismatic samples $10 \text{ cm} \times 5 \text{ cm} \times 1.7 \text{ cm}$ in size) are made. Before mixing the materials, the optimum moisture content for the maximum dry density for each type of mixture must be determined. For this purpose, the Proctor test is carried out (Table 5).

The dry phosphogypsum is sieved through a 0.25 mm sieve. The weighed quantity of phosphogypsum, sand, lime and cement are first carefully mixed for a period of 10 minutes in order to obtain a uniform mixture. Then the quantity of water is added, and the mixing continues for 1 min at slow speed and 2 min at fast speed. All pressed phosphogypsum bricks are manufactured with a

semi-automatic hydraulic press under static pressure of 25 MPa (Figures 5 and 6) under laboratory conditions (humidity $\approx 70\%$ and temperature $t \approx 22^\circ\text{C}$).

Tab 5: Optimal water content of mixtures

	M_0	M_1	M_2	M_3	M_4	M_5
γ_d (g/cm^3)_x005F_x005F_x0001	2.18	1.9	1.85	1.78	1.6	1.55
$W_{op}(\%)$	11	17	17.7	18.2	24.1	29



Fig. 5 – Hydraulic press



Fig. 6 – Manufacture of pressed bricks

Finally, cylindrical specimens with a diameter of 5 cm and a height of 10 cm are made for the compressive strength measurement. These specimens are statically pressed at 25 MPa and then stored under different conditions in order to seek the best characteristics. It is noted that for each composition, three samples are developed for each test. This allows the determination of the average measurement and the standard deviation.

Experimental techniques

Radio-element contents

The radio-element content measurements are carried out by gamma spectrometry using a high-purity germanium detector. This method allows to estimate the activities of the studied samples by identifying the different gamma emitting radioelements and calculating their activities. Thus, the phosphogypsum sample is crushed and placed in a Marinelli Beaker type container (Figure 7). The container is then hermetically sealed with paraffin to prevent the escape of radon gas.

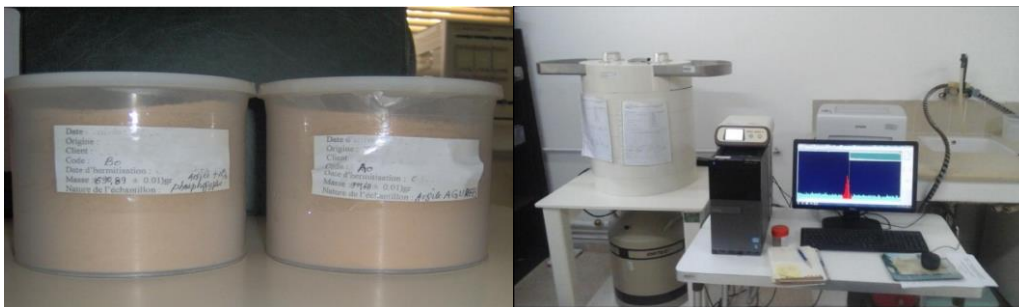


Fig. 7 – Samples to be analysed and the Germanium detector

Physical characteristics

The determination of the optimum water content required for each type of mixture is carried out with the Proctor test. The water absorption, appearance and spalling tests are carried out according to NF EN 772 [26] and to Tunisian standards NT 21-287 [27].

Compression tests

Compression tests are carried out, in accordance with standard NF EN 772 [26], on cylindrical specimens using a 3000 kN "C70-Matest" mechanical press.

RESULTS

Radioactivity

Any extract from the earth, including PG, contains some radioactivity. The determination of this radioactivity is essential for any use of construction materials. Indeed, this radioactivity must not exceed the tolerated limit set at $Ra_{eq}=370$ Bq/kg ([28]) in order to avoid posing radiological risks to human health.

The radionuclide content of materials used in the manufacture of non-load-bearing bricks is measured by gamma spectrometry; the results are an important factor in the human health risk analysis for the use of this type of brick after manufacture. The results of the radionuclide activity analysis are given in Table 6.

Tab 6: Radionuclide activities (Bq/kg) for different materials

	^{238}U	^{214}Pb	^{214}Bi	^{226}Ra	^{40}K	^{232}Th	^{228}Ac	^{212}Pb
Sand Khelidia	19.75	7.01	6.18	6.6	65.95	6.82	7.85	5.78
PG	39	191	209	200	15	18	17	18
Cement	25.57	11.54	10.84	11.19	265.09	11.75	13.42	10.09

The gamma-radionuclides present in soil are mainly ^{40}K , ^{238}U and ^{232}Th [29]. The obtained results for these radionuclides are close to the results obtained by Reguigui et al. [30] in Tunisian soil. Indeed Reguigui et al. show that radioactivity values for the natural radionuclides vary from 10 to 25 Bq/kg for ^{238}U , from 11 to 30 Bq/kg for ^{232}Th and from 30 to 520 Bq/kg for ^{40}K .

Several parameters can be calculated to estimate whether the use of construction materials is safe for human health, among these parameters radium-equivalent activity, absorbed dose rate, external and internal hazard indices, indoor and outdoor annual effective dose and radioactivity level index.

Radium-Equivalent Activity (R_{eq})

Let consider A_{Ra} , A_{Th} and A_K , respectively the specific activities of ^{226}Ra , ^{232}Th , and ^{40}K . To be able to estimate the total gamma activity of a building material, the Radium-Equivalent Activity (Bq/kg) can be estimated by the Equation 1 [28].

$$R_{eq}(\text{Bq/Kg})=A_{Ra}+A_{Th} \times 1.43+A_K \times 0.077 \quad (1)$$

Absorbed Dose Rate (D_γ)

Building materials at a height of 1 m above the earth's surface provides a gamma dose rate that can be estimated using the same conversion factors as in [28] (Equation 2).

$$D_\gamma(\text{nGy/h}) = 0.462 \times A_{Ra} + 0.604 \times A_{Th} + 0.0417 \times A_K \quad (2)$$

D_γ must be less than the maximum limit of 55 nGy/h.

External and Internal Hazard Indices (H_{ex} and H_{in})

The external and internal hazard indices of building materials [28], defined in Equations 3 and 4.

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

A permissible risk of irradiation is characterised with an index value less than one (≤ 1).

Indoor and Outdoor Annual Effective Dose (E_{in} and E_{out})

The annual effective dose rates to the general public is estimated using the following expression ([28]):

$$E_{in}(\text{mSv/year})=D_\gamma(\text{nGy/h}) \times 10^{-6} \times 8760 \times 0.8 \times 0.7 \leq 1 \quad (5)$$

$$E_{out}(\text{mSv/year})=D_\gamma(\text{nGy/h}) \times 10^{-6} \times 8760 \times 0.2 \times 0.7 \leq 1 \quad (6)$$

Radioactivity Level Index (I_γ)

The radioactivity level index (I_γ) ([28]) of a building material is expressed by :

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \leq 1 \quad (7)$$

Table 7 gives the different parameter values. For all used materials, the calculated R_{eq} = 226.895 Bq/kg is less than the maximum value 370 Bq/kg. The gamma dose rate D_γ obtained for the PG are higher than the suggested limit value. However, Tunisian Phosphogypsum registers a much lower level of absorbed gamma dose rate than Indian PG characterised by 198.5 nGy/h ([28]).

Tab 7: Radionuclide activities (Bq/kg) for different materials

	Ra_{eq} (Bq/kg)	D_{γ} (nGy/h)	H_{ex}	H_{in}	E_{in}	E_{out}	I_{γ}
Sand	21.43	9.92	0.06	0.08	0.05	0.01	0.16
PG	226.9	103.9	0.61	1.15	0.51	0.13	1.52
Cement	48.4	23.32	0.13	0.16	0.11	0.03	0.37
Limit	≤ 370	≤ 55	≤ 1	≤ 1	≤ 1	≤ 1	≤ 1

The external hazard index, the indoor effective dose and outdoor effective dose of all materials were well within the safety limit (<1).

The internal hazard index and the radioactivity level index calculated for the PG were superior to the permissible level. Therefore, the building materials used have a tolerable radiation level for external use in buildings. For internal use, the percentage of PG must be limited to limit the level of radioactivity.

Physical Characterization of Bricks

The bricks should be free of visible defects such as cracks, fractures, deformations. Some cracks may be tolerated at a percentage of the product and if their number does not exceed the limits defined by the standards. All the formulations, except M_0 and M_1 , have a good appearance. The poor appearance of the M_0 formulation is due to the presence of a large quantity of sand which is a pulverizing material in addition to the quantity of PG (50 % of the sand) which has a character of a fine sand.

Spalling

The specimens of the bricks obtained with the different mixtures (except M_0 as it presents a bad aspect), are carefully examined in order to detect any spalling. Next, these specimens are immersed in water at 80°C , then they are kept for 3 hours at boiling temperature. In order to be considered as not scaled, the external surface of the specimen must satisfy two conditions: no pop outs with an average diameter greater than 10 mm/dm^2 , and no more than 3 pop outs with an average diameter between 5 and 10 mm. The test has shown that the different mixtures M_1 , M_2 and M_3 are stable chemically and mechanically on bursting (Table 8).

Tab 8: Appearance test results

M_0	M_1	M_2	M_3	M_4	M_5
-	Stable	Stable	Stable	Instable	Instable

Water absorption

Water absorption is a main factor for the durability of the product and its behaviour in the natural environment. High water absorption contributes to the rapid deterioration of this type of material.

Table 9 gives the value of the water absorption of the different mixtures except that of the M_0 mixture which deteriorates rapidly in the presence of water. This deterioration is due to the large amount of sand in the formulation (67 %) and to the absence of the binder.

For M_2 and M_3 mixtures, the absorption coefficient is less than the tolerated limit value. Although the value of mixture M_3 is close to the required limit by the standard (15 %) [27, 31]. The increase in the absorption coefficient for mixtures M_4 and M_5 is due to the fineness of the PG and associated sand (around 85 %). For the mixture M_1 , it shows a lower absorption coefficient than that of M_4 and M_5 although the percentage of sand and PG is higher (90 %). This is due to the presence of cement (10 %).

Tab 9: Water absorption for different mixtures (%)

Mixtures	M_1	M_2	M_3	M_4	M_5
Water absorption	25.53	6.36	14.49	33.82	32.77

Storage methods and compressive strength

In order to optimize the mechanical resistance, several storage methods are considered. The modes are chosen in order to improve the mechanical resistance on the one hand and to seek a method of economic conservation on the other. Table 10 represents the considered five modes of conservation.

Tab 10: Storage method

Mode	Storage method
Mode 0	water conservation
Mode 1	5 days in the laboratory
Mode 2	3 days in the laboratory + 2 days in an oven at 70°C _x005F_x0001_
Mode 3	2 days in the laboratory + 3 days in an oven at 70°C _x005F_x0001_
Mode 4	2 days in the laboratory + 5 days in an oven at 70°C

Tables 11, 12, 13 give the compressive strength for different mixtures and storage method on the 7th, 14th and 28th days.

Tab. 11: Compressive strength (MPa) at 7 days

	Mode 0		Mode 1		Mode 2		Mode 3		Mode 4	
	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
M_0	-	-	0.350	0.009	0.398	0.019	0.480	0.035	0.276	0.007
M_1	0.703	0.034	0.703	0.034	0.735	0.048	0.763	0.019	0.764	0.024
M_2	0.479	0.047	1.113	0.228	0.534	0.113	1.266	0.217	0.969	0.066
M_3	0.754	0.016	0.905	0.064	1.493	0.225	0.621	0.059	0.504	0.244
M_4	0.205	0.006	1.013	0.171	2.135	0.150	1.691	0.299	1.597	0.173
M_5	0.243	0.005	0.745	0.132	3.217	0.296	1.727	0.644	2.174	0.200

Tab. 12: Compressive strength (MPa) at 14 days

	Mode 0		Mode 1		Mode 2		Mode 3		Mode 4	
	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
M_0	-	-	0.522	0.104	0.593	0.016	0.652	0.026	0.427	0.055
M_1	0.860	0.075	0.860	0.075	0.906	0.032	0.930	0.038	0.892	0.019
M_2	0.851	0.013	1.292	0.239	1.709	0.207	1.913	0.441	1.608	0.249
M_3	0.669	0.011	1.025	0.092	2.588	0.073	1.080	0.134	1.608	0.249
M_4	0.232	0.011	1.402	0.062	2.013	0.116	1.638	0.380	1.129	0.306
M_5	0.315	0.004	1.311	0.038	2.967	0.575	1.715	0.589	1.593	0.434

Tab. 13: Compressive strength (MPa) at 28 days

	Mode 0		Mode 1		Mode 2		Mode 3		Mode 4	
	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.	Average	S.D.
M_0	-	-	0.715	0.067	0.663	0.050	0.710	0.026	0.516	0.086
M_1	0.800	0.030	0.800	0.030	0.932	0.012	0.974	0.017	0.932	0.010
M_2	2.389	0.210	1.550	0.209	2.400	0.157	3.490	0.341	2.671	0.097
M_3	0.643	0.07	1.281	0.117	3.860	0.141	4.120	0.278	2.510	0.098
M_4	*	*	1.452	0.115	1.953	0.142	1.427	0.145	1.060	0.078
M_5	*	*	1.227	0.105	2.787	0.172	1.600	0.144	1.573	0.180

The mixture M_0 , consisting only of sand and PG and considered as a reference mixture, cannot be stored in water. Indeed, the sample loses its shape during the conservation. Moreover, the obtained strength for the M_0 mixture (without binder) is almost insensitive to the storage mode. In addition, when using the mode 0 and beyond the 14th day, the M_4 and M_5 mixes burst and only the M_2 mix retains a good appearance and good mechanical resistance.

At 14th day, the conservation mode 2 provides the best mechanical resistance, with more than 2 MPa. At 28th day, only the M_2 and M_3 mixtures have a mechanical strength greater than 2 MPa for the 2, 3 and 4 storage methods. This is expected since the physical, appearance, bursting and water absorption tests are fully verified only for M_2 and M_3 mixtures.

By comparing M_2 and M_3 mixtures at 28th day, it is clear that the mechanical strength of M_2 is higher than that of M_3 for storage modes 1 and 4, while the strength of M_3 is higher for storage modes 2 and 3. Moreover, it is noticed that Mode 3 gives a better resistance compared to the other modes and that in Mode 4 the resistance of all mixtures decreases. This clearly shows that the storage time in the oven at 70°C has a significant effect on the mechanical resistance.

Mode 1 is a laboratory storage mode, while in mode 2 and 3 the sample is placed in the oven at 70°C for 2 and 3 days respectively. Thus, the improvement in mechanical strength can only be attributed to the longer storage time in the oven. Exposure to 70°C for up to 3 days, although it positively affects the mechanical strength of all samples, the extent of the improvement differs from one sample to another since the composition of the samples in phosphogypsum, lime and sand varies.

The experimental values obtained for the M_2 and M_3 mixtures in modes 2, 3 and 4 are mechanically comparable to most non-load-bearing bricks on the market. Indeed, the Tunisian standard imposes a minimum value of 2.3 MPa for non-load-bearing bricks [27, 31], and consequently, solid pressed phosphogypsum-based bricks can replace fired clay-based bricks.

CONCLUSION

As an economic and environmental solution for the PG deposited in heaps near the factories, this study considers the valorisation of PG in the manufacture of unfired bricks as a substitute for clay. Thus, a study of the radioactivity of the used materials was carried out and the results show that all the materials have radionuclide activities below the standard for outdoor use. For indoor use, the amount of phosphogypsum must be limited.

Several mixtures have been tested in several preservation methods. Among these mixtures, the compositions M_2 (composed of 60 % PG) and M_3 (composed of 75 % PG) fully verify the physical and appearance tests in addition to a good mechanical resistance. Therefore, the third mode of conservation (2 days in the laboratory + 3 days in an oven at 70°C) has allowed to have the best resistance to compression, which is much higher than the minimum value required for non-load-bearing bricks.

In conclusion, this study resulted in a formulation of phosphogypsum-based unfired bricks. The use of this type of non-load-bearing brick requires a low amount of energy and consumes a large amount of waste, which largely reduces environmental pollution, in addition to the high economic and social benefits. In addition, this study shows that the radioactive emission of the components of this brick is below the limit values recommended by the standards, and therefore its use is safe.

REFERENCES

- [1] Yang, L., Yan, Y., & Hu, Z. H. (2012). Utilization of Phosphogypsum as raw Material for manufacturing of non-fired load-bearing wall bricks. In *Advanced Materials Research* (Vol. 374, pp. 787-791). Trans Tech Publications Ltd.
- [2] Zhou, J., Sheng, Z., Li, T., Shu, Z., Chen, Y., & Wang, Y. (2016). Preparation of hardened tiles from waste phosphogypsum by a new intermittent pressing hydration. *Ceramics International*, 42(6), 7237-7245.
- [3] Yang, L., Zhang, Y., & Yan, Y. (2016). Utilization of original phosphogypsum as raw material for the preparation of self-leveling mortar. *Journal of Cleaner Production*, 127, 204-213.
- [4] Mechi, N., Ammar, M., Loungou, M., & Elaloui, E. (2016). Thermal study of Tunisian phosphogypsum for use in reinforced plaster. *Current Journal of Applied Science and Technology*, 1-10.
- [5] Naresha, R., Laxminarayana, P., & Sailaja, K. S. D. V. (2016). Yield and Moisture Studies of Rabi Groundnut as Influenced by Moisture Regimes and Phosphogypsum Levels. *Research Journal of Agricultural Sciences*, 7(3), 487-491.
- [6] Saadaoui, E., Ghazel, N., Ben Romdhane, C., & Massoudi, N. (2017). Phosphogypsum: potential uses and problems—a review. *International Journal of Environmental Studies*, 74(4), 558-567.
- [7] Ajam, L., Hassen, A. B. E. H., & Reguigui, N. (2019). Phosphogypsum utilization in fired bricks: Radioactivity assessment and durability. *Journal of Building Engineering*, 26, 100928.
- [8] Felfoul, H. S., Clastres, P., Carles, G. A., & Ouezdou, M. B. (2002). Amélioration des caractéristiques du phosphogypse en vue de son utilisation en technique routière. *Waste Sci Tech*, 28, 21.
- [9] Amrani, M., El Haloui, Y., Hajikarimi, P., Sehaqui, H., Hakkou, R., Barbachi, M., & Taha, Y. (2020). Feasibility of using phosphate wastes for enhancing high-temperature rheological characteristics of asphalt binder. *Journal of Material Cycles and Waste Management*, 1-11.
- [10] Degirmenci, N., Okucu, A., & Turabi, A. (2007). Application of phosphogypsum in soil stabilization. *Building and environment*, 42(9), 3393-3398.
- [11] Krishnan, K. D., Deepika, M., Ravichandran, P. T., Sudha, C., & Kottupillil, A. K. (2016). Study on Behaviour of Soil with Phosphogypsum as Stabiliser. *Indian Journal of Science and Technology*, 9(23).

- [12] Amrani, M., Taha, Y., Kchikach, A., Benzaazoua, M., & Hakkou, R. (2020). Phosphogypsum recycling: New horizons for a more sustainable road material application. *Journal of Building Engineering*, 30, 101267.
- [13] Ennaciri, Y., Zdah, I., El Alaoui-Belghiti, H., & Bettach, M. (2020). Characterization and purification of waste phosphogypsum to make it suitable for use in the plaster and the cement industry. *Chemical Engineering Communications*, 207(3), 382-392.
- [14] Singh, M. (2002). Treating waste phosphogypsum for cement and plaster manufacture. *Cement and Concrete Research*, 32(7), 1033-1038.
- [15] Singh, M. (2005). Role of phosphogypsum impurities on strength and microstructure of selenite plaster. *Construction and building materials*, 19(6), 480-486.
- [16] Altun, İ. A., & Sert, Y. (2004). Utilization of weathered phosphogypsum as set retarder in Portland cement. *Cement and Concrete Research*, 34(4), 677-680.
- [17] Islam, G. S., Chowdhury, F. H., Raihan, M. T., Amit, S. K. S., & Islam, M. R. (2017). Effect of phosphogypsum on the properties of Portland cement. *Procedia engineering*, 171, 744-751.
- [18] Kacimi, L., Simon-Masseron, A., Ghomari, A., & Derriche, Z. (2006). Reduction of clinkerization temperature by using phosphogypsum. *Journal of hazardous materials*, 137(1), 129-137.
- [19] Kuryatnyk, T., da Luz, C. A., Ambroise, J., & Pera, J. (2008). Valorization of phosphogypsum as hydraulic binder. *Journal of Hazardous Materials*, 160(2-3), 681-687.
- [20] Karray, M. A., & Mensi, R. (2000, April). Etude de la deformabilite des poutrelles en beton arme a base de ciment Ultimax. In *ANNALES DU BATIMENT ET DES TRAVAUX PUBLICS* (No. 2).
- [21] Kumar, S. (2002). A perspective study on fly ash–lime–gypsum bricks and hollow blocks for low cost housing development. *Construction and Building Materials*, 16(8), 519-525.
- [22] Yang, J., Liu, W., Zhang, L., & Xiao, B. (2009). Preparation of load-bearing building materials from autoclaved phosphogypsum. *Construction and Building Materials*, 23(2), 687-693.
- [23] Zhou, J., Yu, D., Shu, Z., Li, T., Chen, Y., & Wang, Y. (2014). A novel two-step hydration process of preparing cement-free non-fired bricks from waste phosphogypsum. *Construction and Building Materials*, 73, 222-228.
- [24] Ajam, L., Ouezdou, M. B., Felfoul, H. S., & El Mensi, R. (2009). Characterization of the Tunisian phosphogypsum and its valorization in clay bricks. *Construction and Building Materials*, 23(10), 3240-3247.
- [25] Felfoul, H. S., Clastres, P., & Benouezdou, M. (2005). Gestion des sous-produits industriels et developpement durable: cas du phosphogypse de sfax (tunisie). *Sciences & Technologie. B, Sciences de l'ingénieur*, 66-81.
- [26] EN, B.: 772. Methods of test for masonry units (2011)
- [27] Inorpi: Nt 21-287. Specification des elements en maconnerie (2004)
- [28] Sankaran Pillai, G., Shahul Hameed, P., & Mazhar Nazeeb Khan, S. M. (2016). Radioactivity in building materials and assessment of risk of human exposure in the Tiruchirappalli District of Tamil Nadu, India. *Journal of Hazardous, Toxic, and Radioactive Waste*, 20(3), 04016004.
- [29] United Nations Scientific Committee on the Effects of Atomic Radiation. (1994). Sources and effects of ionizing radiation. UNSCEAR 1994 report to the General Assembly, with scientific annexes.
- [30] Reguigui, N., Ben Kraiem, H., & Latrous, H. (1999). Monitoring and measurements of radioactivity around the valley of Madjerdah river in Tunisia (No. IAEA-TECDOC--1094).
- [31] AFNOR: Nf en 771-1. Specification des elements en maconnerie (2003)