Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872

http://www.jmaterenvironsci.com/

Mathematical approach for research of new formulation for immobilization of radioactive waste in cementitious matrices

T. El Ghailassi[1*], M. Aoulad Belayachi[2], A. Bouih[1], S. Labied[3], T. Guedira[2], O. Benali[3]

[1] Unit of Management of radioactive waste, National Center of Nuclear Energy, Sciences and Nuclear Techniques (CNESTN), Center for Nuclear Studies of Maamoura (CENM).PB 1382, 10001 Kenitra, Morocco

[2]Laboratory of Materials Electrochemistry and Environment, Faculty of Science, University Ibn Tofail, BP 133, 14000 Kenitra, Morocco

[3]Laboratory of separation Processes, Faculty of Science, University Ibn Tofail, BP 133, 14000 Kenitra, Morocco

Received 12 April 2016,
Revised 10 March 2017,
Accepted 10 March 2017

Keywords

- ✓ Confinement;
- ✓ radioactive waste;
- ✓ cementation;
- ✓ immobilization;
- ✓ compressive strength
- ✓ porosity

T. El ghailassi
elghailassi@cnesten.org.ma
elghailassi@gmail.com
(+212) 6 66 04 50 74

Abstract

The immobilization of low and intermediate level radioactive wastes in cementitious matrices is the most commonly used technique to produce inexpensive waste matrix that complies with regulatory requirements. The interest of this work is to ensure confinement, durability and an integrability of the immobilization of radioactive waste inside the packages stored in the CNESTEN, and also to highlight the influence of the water-cement (W/C) report on the behavior of the studied matrix. For this purpose, an experimental program allowed the necessary mathematical approach for the determination of an optimal formulation of the mortar. Mechanical testing of different matrices shows different behaviors. The best performance is observed in a report W/C = 0, 4 and S/Mt = 0, 3. This study finds the matrix with the optimal behavior to confine the radioactive waste.

1. Introduction

Radioactive waste management appeared more and more as a major concern of Governments, scientists and users of nuclear techniques. This management must be performed within a rigorous framework to ensure the safe management solutions for national radioactive waste products across all without losing sight of the standing requirement to protect present and future generations and the environment from the risk posed by such wastes [1, 2, 3]. Cementation is one of many methods used for radioactive waste solidification. The immobilization of low and intermediate level radioactive wastes is a mode of treatment among the adopted process; it requires a cementitious matrix that responds to certain essential conditions as good resistance to compression [4, 5, 6].

This present study focuses on the research of the optimal conditions of the confinement of radioactive waste. This work fits into the context of research [7, 8], within the unit of management of radioactive waste, on the behavior of materials in order to find a good matrix of confinement of radioactive waste treated by the CNESTEN. This matrix must ensure storage in accordance with the requirements of safety and security [9]. The choice of materials must be judicious and optimized on the basis of scientific, technical and economic criteria. In this light, an experimental program allowed the necessary mathematical approach for the determination of an optimal formulation of the mortar (Portland cement, sand and water) by varying the (water/cement) (W/C) ratio and the quantity of sand. Characterization of different matrices has been accomplished by measuring the compressive strength and the porosity of different matrices destined for confinement of radioactive waste.

2. Experimental

Our work was firstly to make tests of resistance to compression on mortars. For this purpose, we have followed an experimental program to test several formulations consisting of cement, water and sand mortar.

In this work, the (water/cement) (W/C) ratio and the quantity of sand are varied until obtainment of formulation that has good compressive strength.

2.1. *Materials*
2.1.1. *Equipments*
The press used to measure the mechanical resistance to compression, is a manual type hydraulic press Carver Model (4350.L) S/N (4350-362), capacity up to 24000 Pounds and is enslaved by weight.
Mixing the components of the formulation is carried out with an automatic mixer, type 3R, this device has a capacity of 5 L, it ensures a uniform and homogeneous mixture.
The Column of Sieve of type 3R used to determinate the distribution of the mass of sand particles in the dry state.

2.1.2. *Portland Cement*
The cement used in the manufacture of test specimens is a (CPJ 35) from the factory Lafarge cements Meknes composite Portland cement and meets 10.1.004 and NM 10.1.005
Physical, mechanical characteristics (on standardized mortars) and chemical are presented respectively in tables 1 and 2.

Table 1: Physical characteristics (on standard mortar) used cement

Cement	**CEM I**
28 days compressive strength	32 Mpa
Beginning of outlet at 20 ° C	1 h 30 min
Expansion hot	< 10mm
28 d withdrawal	800 µm/m
SO_3 content	4%

Table 2 : The CPJ 35 cement composition

Clinker	67%
C_3S	69%
Limestone	22.5%
C_2S	13%
C_3	11%
C_4AF	7%
CPL	6%
Gypsum	4.5%

2.1.3. *Sand*
The sand used in this study is characterized by its siliceous morphology. This sand is complies with NM 10.1.020 (EN 196-1) [10]. The physical characteristics of the sand of study are represented below and the particle size distribution is shown in table 3.

- Particle size analysis [11]

Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872

http://www.jmaterenvironsci.com/

Table 3 : The result of the particle size analysis of sand.

Sieve (mm)	Cumulative refusal (%)	Cumulative percentage (%)
1.6	0.5	99.5
0.800	2	98
0.400	6.3	93.7
0.200	88	12
0.080	99.2	0.8
Background	100	0

Previous results have shown that the use of sand the size of 0.2 mm reinforces the strength of cement matrix. According to the result of particle size analysis [12], the majority of the grains constituting studied sand has an average size of 0.2 mm and a pretty fine high 0.08 mm which shows that our sand is well class and well sorted.

- Fineness (MF) Module [13]

The finesse is equal to the sum of the cumulative percentage retained on the sieve series listed in the table above is divided by 100.
The result found is: MF = 1.9555
From this result, the MF < 10, this shows that our sand is fine.

- Sand Equivalent [14]

Said sand equivalent test to determine the degree of cleanliness of sand (table 4):

$$ES = \frac{h1}{h2} \times 100 \qquad Eq(1)$$

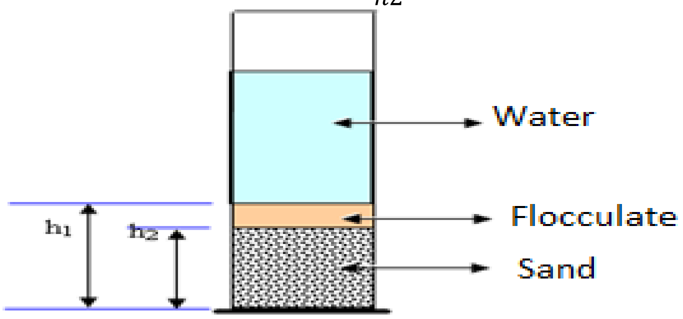

Figure 1: Method for the determination of the equivalent of sand

-Height h_1: clean sand + fine elements.
-Height h_2: sand clean only.

Table 4 : Presentation of the value average of the equivalent of sand.

	Height	Height	Average value
H_1	7.3 cm	6.7 cm	**96.76%**
H_2	7.1 cm	6.45 cm	
ES	97.26%	96.26%	

The average value is greater than 85%, which means that our sand is very clean.

2.2. Formulations of the test

The formulation that we have adopted in our study is based on an experimental program that allowed a mathematical approach to determine an optimal formulation of the mortar.
Either M_t the total mass of the mixture to be prepared: $M_t = S + C + W$ Eq(2)
With:

- S = mass of dry sand.

Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872
http://www.jmaterenvironsci.com/

- C = the mass of cement
- W = the mass of water required for the hydration of the cement in the mix
- M_t = the total mass

We pose $Y = S/M_t$, and $Z = W / C$

Therefore the mass of cement becomes: with Y < 1

$$C = \frac{(1 - Y)}{1 + Z} \times MtEq(3)$$

In this formula of the mortar, it adopted the following protocol; since we have two variables y and z (because M_t = 3200 g), we assigned the values of W/C ranging from 0.4; 0.45; 0.5 to 0.55. While the variable y = S/M_t, we have ranged it from 0.1 to 0.6 (y < 1).

2.3. Preparations of cement matrix

In our study all tests will be carried out on 5 height 10cm and 5.7 cm diameter plastic molds. These molds are distributed in the following way:

- Mold 1: test of resistance to compression during 7 days.
- Mold 2: test of resistance to compression 14 days.
- Mold 3: test of resistance to compression during 21 days.
- Mold 4: test of compressive strength for 28 days.
- Mold 5: for porosity testing.

Tables 5, 6, 7 and 8 combine all tests or formulations studied in this work:

Table 5: Tests contain W/C = 0.4 for different ratio S/Mt

Test	Weight %/(g)	Cement	Sand	Water	S/M_t	W/C
Test 1	%	64.28	10	25.71	0.1	0.4
	g	2057	320	822.8		
Test 2	%	57.14	20	22.85	0.2	
	g	1828.57	640	731.43		
Test 3	%	50	30	20	0.3	
	g	1600	960	640		
Test 4	%	42.8	40	17.2	0.4	
	g	1371.4	1280	548.6		
Test 5	%	35.7	50	14.3	0.5	
	g	1142.8	1600	457.2		
Test 6	%	28.57	60	11.42	0.6	
	g	914.28	1920	365.71		

Table 6: Tests contain W/C = 0.45 for different ratio S/M_t

Test	Weight %/(g)	Cement	Sand	Water	S/M_t	W/C
Test 7	%	62.06	10	27.93	0.1	0.45
	g	1986.21	320	893.79		
Test 8	%	55.2	20	24.8	0.2	
	g	1765.52	640	794.48		
Test 9	%	48.3	30	21.7	0.3	
	g	1544.8	960	695.2		
Test 10	%	41.38	40	18.62	0.4	
	g	1324.14	1280	595.86		
Test 11	%	34. 48	50	15.52	0.5	
	g	1103.45	1600	496.55		

Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872
http://www.jmaterenvironsci.com/

Table 7: **Tests contain W/C = 0.5 for different ratio S/M_t**

Test	Weight %/(g)	Cement	Sand	Water	S/M_t	W/C
Test 12	%	60	10	30	0.1	0.5
	g	1920	320	960		
Test 13	%	53.33	20	26.66	0.2	
	g	1706.66	640	853.33		
Test 14	%	46.66	30	23.33	0.3	
	g	1493.33	960	746.67		
Test 15	%	40	40	20	0.4	
	g	1280	1280	640		
Test 16	%	33.33	50	16.66	0.5	
	g	1066.66	1600	533.33		

Table 8: **Tests contain W/C = 0.55 for different ratio S/M_t**

Test	Weight %/(g)	Cement	Sand	Water	S/M_t	W/C
Test 17	%	58.06	10	31.93	0.1	0.55
	g	1858.06	320	1021.93		
Test 18	%	51.61	20	28.38	0.2	
	g	1651.61	640	908.38		
Test 19	%	45.16	30	24.83	0.3	
	g	1445.16	960	794.84		
Test 20	%	38.71	40	21.29	0.4	
	g	1238.71	1280	681.28		
Test 21	%	32.26	50	17.74	0.5	
	g	1032.26	1600	567.74		

2.4 Compressive strength and porosity

Uniaxial compression tests are carried out on rights cylinders with circular bases (Figure 2) at specific intervals that correspond to 7, 14, 21, 28 days (Figures 3, 4, 5 and 6). Resistance to compression for each formulation test is carried out at different ages of the matrix: 7, 14, 21, 28 days. The porosity is measured according to the following formula [15]:

$$P = \frac{mf - mi}{Vap} \times 100 \; Eq(4)$$

3. Results and discussion

Variations of the compressive strength depending on the time of solidification for all formulations W/C = 0.4; 0.45; 0.5; 0.55 and S/M_t = 0.1; 0.2; 0.3; 0.4; 0.5; 0.6 are in shown in Figures 2, 3, 4, 5, 6, 7 and 8.

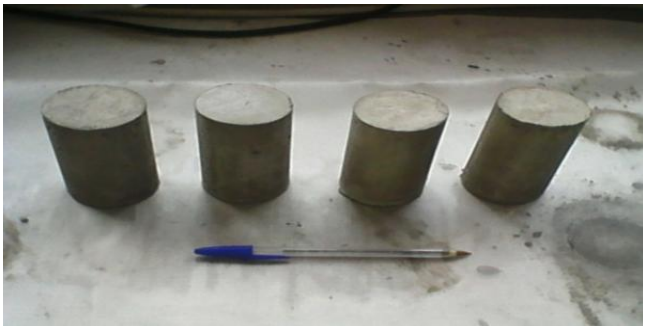

Figure 2: **Photography test matrix**

Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872
http://www.jmaterenvironsci.com/

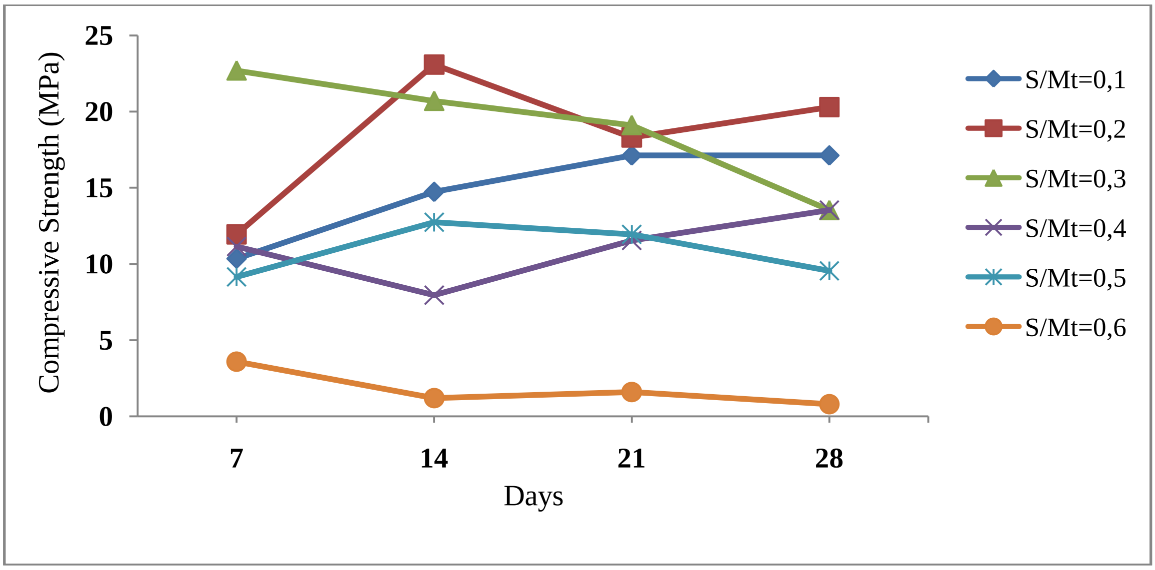

Figure 3: Variation of the compressive strength of the mortar with age. W/C = 0.4

The results of trial campaigns show that the matrix contain $S/M_t = 0.3$ and W/C = 0.4 (Figure 3) develops a better mechanical resistance to compression at 7 days which is explained by the presence of a large amount of C3S and C2S which are responsible for the mechanical performances (formation of C-S-H gel) [16], but beyond 7 days there is a decay age compressive strength with values ranging between 22.69 MPa and 13.54 MPa. This decrease may be due to the change in the kinetics of transformation of the cement [17] anhydrous responsible for mechanical performance (C3S and C2S) and also for degradation products of hydration of C3S and C2S (C-S-H). We notice that beyond the point $S/M_t = 0.5$ and W/C = 0.4 the compressive strength decreases abruptly.

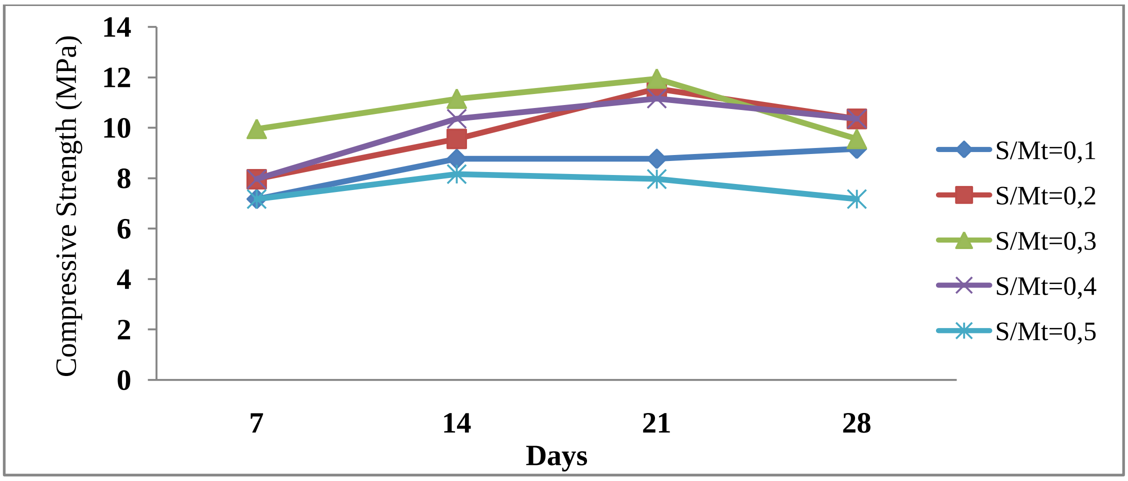

Figure 4 : Variation of the compressive strength of the mortar with age. W/C = 0.45

We note in figure 4 that the best resistance is observed at the point $S/M_t = 0.3$ and W/C = 0.45, but the strength of this matrix remains always lower than the matrix that contain $S/M_t = 0.3$ W/C = 0.4

Journal of Materials and Environmental Sciences
ISSN : 2028-2508

JMES, 2017 Volume 8, Issue 11, Page 3864-3872

http://www.jmaterenvironsci.com/

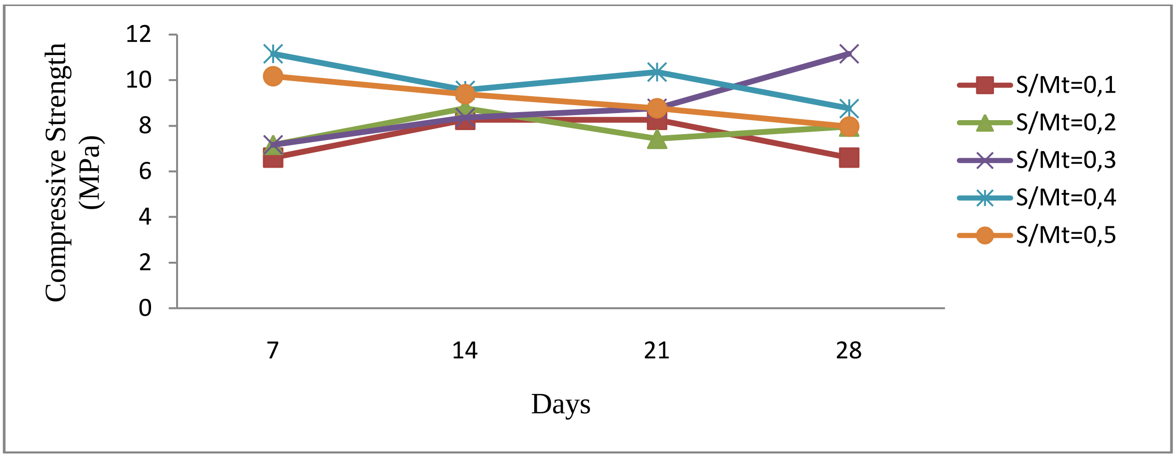

Figure 5 : Variation of the compressive strength of the mortar with age. W/C = 0.5

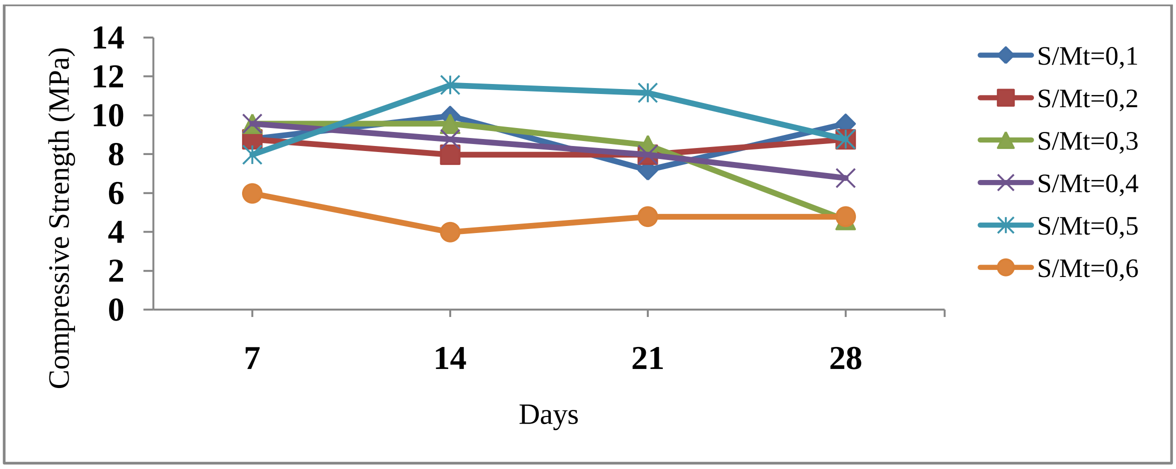

Figure 6: Variation of the compressive strength of the mortar with age. W/C = 0.55

The values of the compressive strength (Figures 5 and 6) of all point remain lower than the matrix that contain $S/M_t = 0.3$ and $W/C = 0.4$

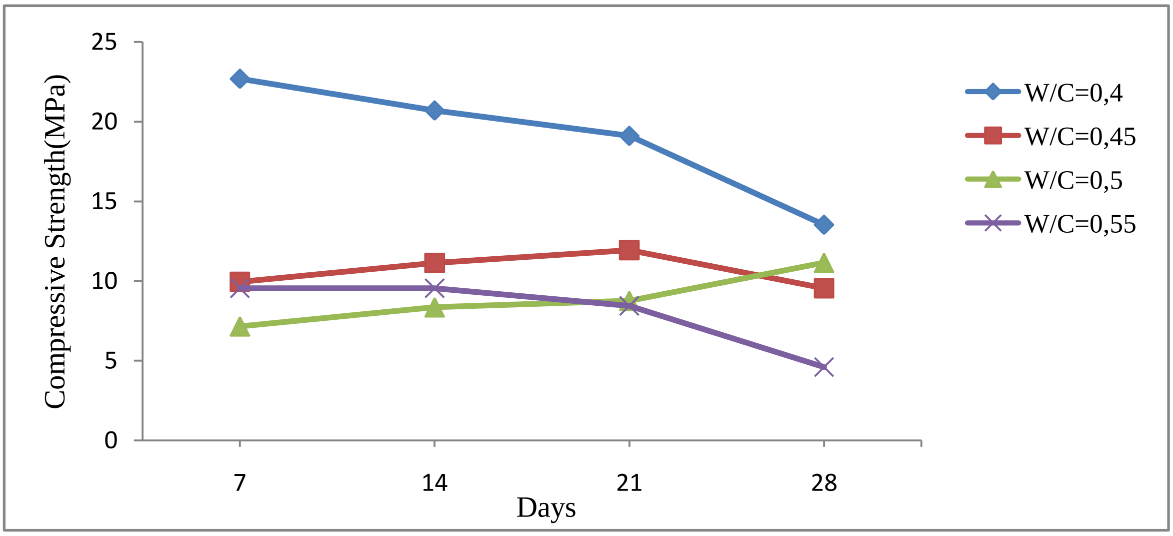

Figure 7 : Variation of the compressive strength of various reports W/C for S/M$_t$ = 0, 3.

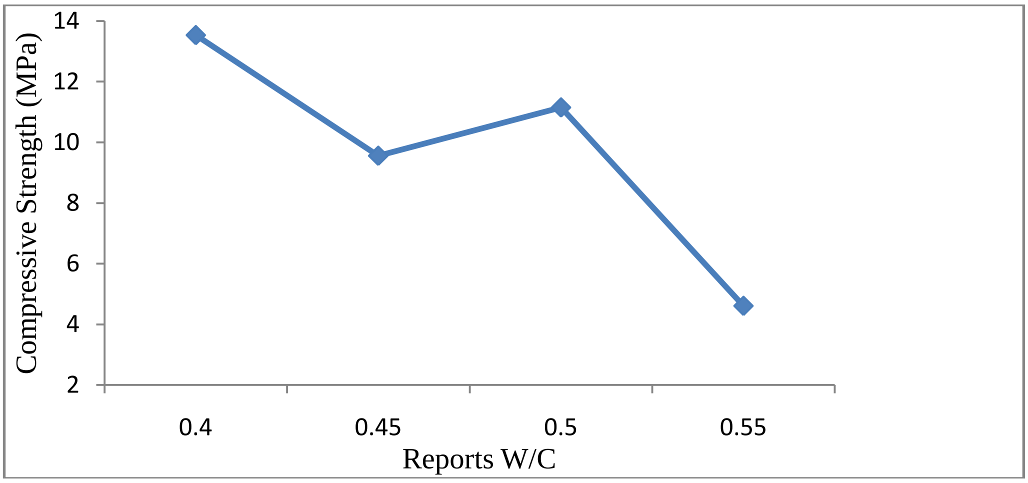

Figure 8: Variation of the compressive strength of various reports W/C for S/M$_t$ = 0, 3 at 28 days

In Figures 7 and 8 we clearly notice that matrix contain W/C = 0.4, S/M$_t$ = 0.3 is characterized by the best value of the compressive strength in comparison with other matrices that contains ratio W/C = 0.45, W/C = 0.5 and W/C = 0.55. This increase is mainly due to the low amount of water compared to other issues [18, 19, and 20].

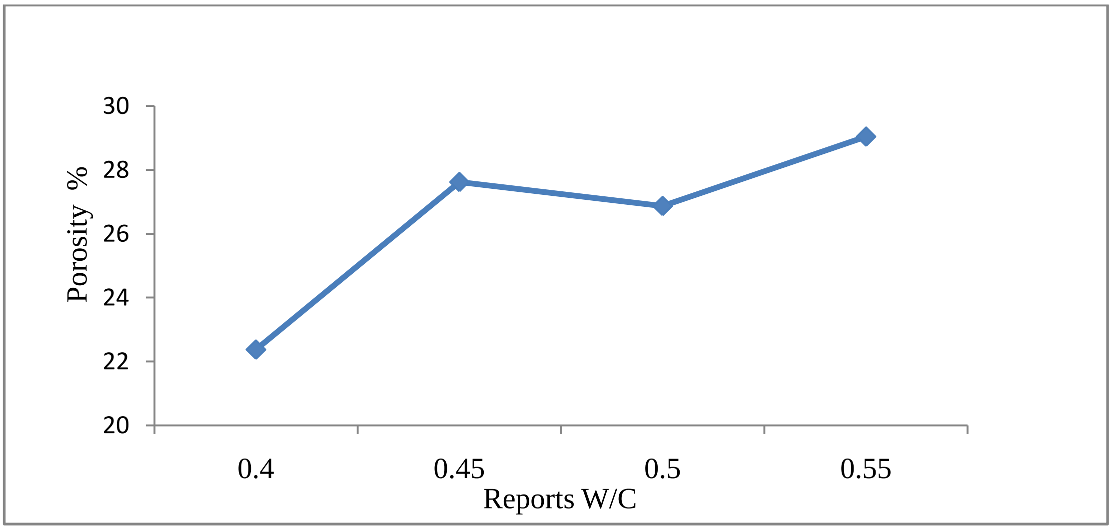

Figure 9: Porosity of the various reports W/C for S/M$_t$ = 0, 3

The porous nature of the mortar is very important since the resistance is linked to its porosity as shown in the figure 9, we find that more the report W/C is great over a porous volume important in the microstructure of the paste of cement and consequently the decrease of the mechanical performances [21, 22].

Conclusion

Our contribution within the unit of management of radioactive waste, for development of materials in order to find a good matrix of confinement of radioactive waste and for storage in accordance with the requirements of safety and security, is as follows:

- ✓ Determination of an optimum formulation of mortar based on a mathematical approach which is characterized by a better mechanical strength.
- ✓ Influence of the water-to-cement ratio on the porosity and more specifically on the mechanical resistance to compression of the mortar.
- ✓ Best performance was observed in a report W/C = 0, 4 and S/M_t = 0, 3 (50% of cement 30% of sand and 20% of Water), which translates into a better mechanical resistance to compression and low porosity.

References

1. AIEA, Collection normes de sureté, *Gestion des déchets radioactifs avant stockage définitifs y compris le déclassement* n° WS-R-2 (2004) VIENNE.
2. El Ghailassi T., *Système de gestion des déchets radioactifs entreposage des déchets radioactifs et combustible Usé Triga Mark II* » Rapport de titularisation (2008) 8-9.
3. El Hilal B., Lambarki El Allioui T., Bouih A., El Harfi A., *J. Mater. Environ. Sci*, 6 N°4, (2015) 969-976.
4. PERLOT C., Thèse de doctorat, *Influence de la décalcification de matériaux cimentaires sur les propriétés de transfert : application au stockage profond de déchets radioactifs*, Engineering Sciences. Université Paul Sabatier de Toulouse III (2005) 6-7.
5. Junfeng Li, Jianlong Wang, *J. Hazard Mater*, B135 (2006) 443–448.
6. Erdal Osmanlioglu A. *Waste Management*, 22 (2002) 481-483
7. Faiz Z.,BouihA., FakhiS., LaissaouiA., HannacheH., IdrissiA., *J. Mater. Environ. Sci*. 6 (1) (2015) 289-296
8. EL Hilal B., Lambarki el Allioui T., Bouih A., Bekhta A., EL Harfi A., *IJIAS*. 7 (2014)
9. AIEA, Collection Normes de Sûreté, N°GSR Part, *Gestion Des Déchets radioactifs avant stockage définitif* (2009) 20-21.
10. Office national de l'eau potable, Tome 4 Génie Civil 2009-11-16, Morocco, (2009) 2-3.
11. Fournier J., Bonnot-Courtois C., Paris R.,Voldoire O., Le Vot M., *Analyses granulométriques, principes et méthodes*. CNRS, Dinard, (2012) 99
12. Faiz Z., Fakhi S., Bouih A., Idrissi A., Mouldouira M., *J. Mater. Environ. Sci*. 3 (6) (2012) 1129-1136.
13. Luke M. Snell and Bryan C, *Fineness Modulus For Sand*, Proceedings of the 10th Annual Mongolian Concrete Conference (2011)
14. Sand Equivalent test laboratory - pavement materials Nanyang Technological University School of Civil and Structural Engineering.
15. Makani A., Thèse de Doctorat, Influence de la nature minéralogique des granulats sur le comportement mécanique différé des bétons, Université de Toulouse, France (2011) 156-157
16. Fabbri A., Thèse de Doctorat, *Physico-mécanique des matériaux cimentaires soumis au gel-dégel*, Université de Marne-La-Valle, France (2006)
17. Rafik Affes, thèse de Doctorat, *Relations microstructure-fissuration-perméabilité dans les milieux granulaires cimentés* Université Montpellier II (2012) 16-18
18. Abdel Rahman R.O., *Chemical Engineering Journal*. 155 (2009) 698-708.
19. Lombardi F., Mangialardi T., PigaL., Sirini P., *Waste Management*, 18 (1998) 99-106.
20. Roy A., Harvill E.C., Carteledge F.K., *J. Hazard. Mater*. 30 (1992) 297-316.
21. Fantozzi-Merle C., Thèse, Villeurbanne: *Institut National des Sciences Appliquées de Lyon*, (2003) 80-89.
22. Todorvic, Ecke H., Lagerkvist A., *Waste Management*. 23 (2003) 621-629.

(2017) ; http://www.jmaterenvironsci.com