ASSESSMENT OF MOROCCAN NATURAL ADDITIONS IMPACT ON THE CEMENTATION PROCESS QUALITY OF SPENT ION EXCHANGE RESINS: STRENGTH, ^{134}CS LEACHING RESISTANCE AND MORPHOLOGICAL STRUCTURE

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1. Background and Goal of the present work

Cementation is the most widely worldwide used stabilization/solidification process for long-lived low and medium level radioactive waste such as spent Ion Exchange Resins (IER). This process uses cement as a binding material. It transforms the waste into a form easier to control and transport. The most commonly used cement are the Ordinary Portland Cements (OPC). IER cementation process is characterized by the production of final waste with a higher volume and lower mechanical resistance.

The present work is undertaken within the general framework of this problematic. It aims to improve the conditioning quality by cementation of the spent IER used in the fluids purification of TRIGA Mark II research reactor used by the nuclear study center of Maamoura (CENM), Morocco. In order to ensure a sustainable and safe conditioning of spent IER, this study focuses on improving the physical, chemical and mechanical properties, as well as the radionuclides retention capacity of the cemented IER form. To meet these objectives, the impact of the introduction of three natural additions to the IER cementitious formulation was studied.

2. Materials et methods

2.1. *Additions*

The used additions characterized by their diversity and their mineralogical richness are: the limestone (CA) formed essentially by calcite, the red clay (MA) rich in silica and kaolinite and the marly limestone (CM)which contains smectite and kaolinite.

CHEMICAL COMPONENTS AND LIMESTONE CONTENT OF ADDITIONS CA, MA, CM

	Chemical components (%)							$CaCO_3$ (%)
	CaO	SiO_2	SO_3	Al_2O_3	Fe_2O_3	MgO	K_2O	
CA	80.45±0.13	1.60 ±0.13	3.50 ±0.12	3.60±0.60	0.06±0.01	1.95 ±0.30	---	97 ± 3
MA	2.60±0.01	55.52±0.53	0.10± 0.02	19.00±0.60	7.00±0.01	2.16± 0.36	1.65±0.02	12.6 ± 1.9
CM	14.27±0.04	38.72 ±0.43	0.82± 0.05	11.81±0.55	4.22±0.01	2.49± 0.30	2.24±0.02	30.6 ± 2.1

2.2. Preparation of cemented IER forms

Cemented IER forms are prepared with (OPC35) according to the determined experimental conditions developed in our previous work. Two used cementitious formulations are carried.

CONTENT OF CONSTITUENTS IN ONE KILOGRAM OF MORTAR-IER.

	Cement(g)	Sand(g)	Addition(g)	IER(g)	water(g)
Formulation without addition (A)	510	190	0	120	170
Formulation with Addition	510	150	40	120	180

2.3. ^{134}Cs leach tests

After cementing the resins loaded with ^{134}Cs, the leaching tests from the IER cemented form were performed at ambient temperature. The cemented waste form (0.55 Kg) was placed in a beaker containing 600 mL of distilled water so that the water forms a 2 cm cylinder around the package

2.4. Desorbed ^{134}Cs measurement and leaching parameters calculation

The desorbed ^{134}Cs activity was measured by Gamma spectrometry

^{134}Cs leaching rate Ri : $$R_i = \frac{\sum A_{des} \times V}{A_0 \times S \times \sum t} \quad \text{Eq.1}$$

The leaching index I is deduced from the diffusion coefficient De (Eq.3,4) :

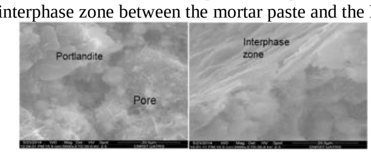
$$De = \frac{\pi}{t}\left[\frac{\sum A_{des}\ V}{A_0\ 2S}\right]^2 \quad \text{Eq.3} \qquad I = -\log(De) \quad \text{Eq.4}$$

where : Ri is the leaching rate (cm/s), Ades is the ^{134}Cs cumulative activities (Bq/Kg) in the residual aqueous phase, V is the total volume of the package (cm^3), A_0 is ^{134}Cs initially activities (Bq/Kg), S is the exposed surface area of the package (219.8 cm^2), T is the leaching time (s), De is the diffusion coefficient (cm^2/s), I is the leaching index (unitless).

3. Results and discussions

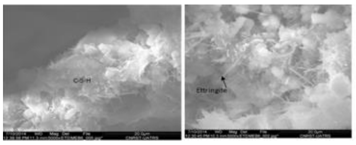
3.1. Morphological structure of cemented IER forms

SEM images of IER form A and CM (28d) show that morphological structure of cement hydration products namely C-S-H and Ettringite is not extensive. Portlandite crystals appear with a weakly developed size. The structure of waste forms produced with CA and MA is very rich in S-C-H-. This component responsible for the mechanical resistance and the adsorption power of cementitious materials is present, especially for the MA formulation, in abundance on the interphase zone between the mortar paste and the IER beads and around the mortar pores.

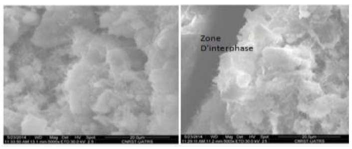


Formulation A (x20).

Formulation CA. (x10))



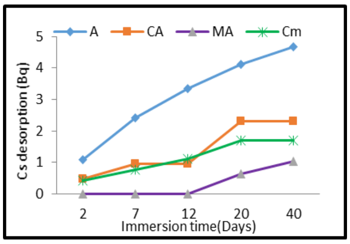
Formulation MA.(x20)



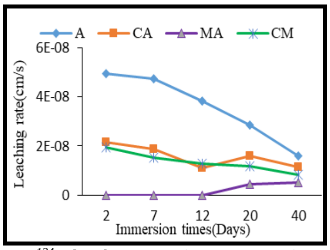
Formulation CM. (x20)

3.2. ^{134}Cs leaching results

Results of leached ^{134}Cs and leaching rate from IER forms prepared with the additions show that this work allowed a significant improvement in the Cs retention. Its retention capacity (MA > CM > CA) is due to the additions natures and structures. The formulation MA performance results from the role played by MA, rich in kaolinite and silica. In addition to their sorption capacity, local red clay promotes reactions for C-S-H production, which plays a decisive role in the cementing process: it improves the resistance of the package and leads to the reduction of its porosity. Furthermore MA is very water resistant thanks to its high iron content, which allowed it to resist water the longest while blocking the Cs inside these interlayers.



Variation of ^{134}Cs leached. (A_0= 59.73 Bq)



^{134}C leaching rate. (A_0= 59.73 Bq)

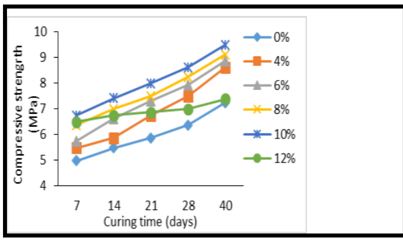
DIFFUSION COEFFICIENT AND LEACHING RATE OF THE FORMULATIONS A AND MA

Formulation	Diffusion coefficient (cm^2/s)	Leaching index
A	5.01 x 10^{-10}	9.30
MA	2.51 x 10^{-11}	10.60
CA	1.22 x 10^{-10}	9.91
CM	6.61 x 10^{-11}	10.18

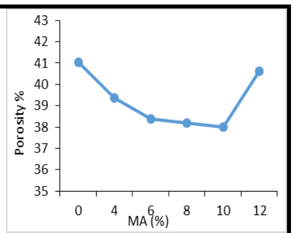
MA leaching index equal to 10.6 confirms the performance of the formulation developed to optimize the IER conditioning by cementation.

3.2. Optimization of red clay quantity introduced in the cementitious formulation.

The developed process performance is confirmed by: improvement of resistance by 137%, porosity by 14% and leaching index by 2.3. Furthermore, the comparison of raw material consumption for the cementing of 10 kg of spent IER with the two formulations), shows that the developed cementation formulation with 10% of MA makes it possible to reduce the cementing loads by minimizing the quantities of cement consumed by 31%, sand by 69% and mixing water by 36%. Thus, the final waste volumes are reduced by 30%**.**



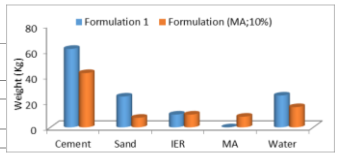
Variation of compressive strength of IER forms prepared with different amounts of MA as a function of curing time



Porosity variation of cemented IER forms as a function of MA amounts

	Formulation 1	Formulation (MA;10%)	Improvement
Strength (MPa)	4	9.50	137.80 %
Porosity (%)	44.21	38	14.05 %
Leaching index	8.30	10.60	2.30

FUNDAMENTAL CHARACTERISTICS OF IER FORMS PREPARED WITH FORMULATION 1 AND (MA; 10%) AT 40 DAYS OF HARDENING



Raw material consumption: formulations 1 and (MA;10%) for the containment of 10 kg of spent IER.

Conclusions:

The presented study results allowed to develop a new cementitious formulation which led to enhance the spent radioactive IER management sustainability and safety. The new developed formulation is composed of 510g of cement, 90g of sand, 100g of local red clay, 120g of spent IER and 190g of water. It reduces the cost of spent IER cementation process and decreases the produced final waste form by 30%, thereby it simplify its management during storage and final disposal.