Behaviour of a protective coating for bio-based materials used for energy rehabilitation regarding salt attack

Oumayma Bahou[1], Brahim Ismail[1], Naima Belayachi[1]

[1] *Univ-d'Orléans, Univ-Tours, INSA-CVL, LaMé-EA74494, 8 rue Léonard De Vinci, 45072 Orléans, France*

Abstract. This study focuses on the behaviour of three lime-based coatings against salts attack intended to protect straw biobased insulation composites as part of an energy retrofit of building. Salts are among the main causes of mortar degradation. Their sources are various between interior ones as mortar constituents and mixing water, and others external as groundwater in contact with gypseous soils, rainwater polluted by sulfur dioxide or di-icing salts that infiltrate the coating by capillary actions or by spaying. In this paper ageing tests are carried out by total immersion in salt solution to expose the samples to salts attack by using sodium sulfate and sodium chloride solutions. The studied mortars are composed by lime, calcareous sand and two additives used also in the biobased insulation materials. The reference mortar without additives is used to highlight the effect of the two additives (air entrainer and casein protein). The third mortar is used with the addition of a small percentage of fine straw about 7.5 %. This percentage allows the optimized mortar to meet all criteria for an insulation coating. The results show that the coating with straw and additives is more resistant than the one that is based on the reference mixture without additives, even when ageing tests are carried out with the most aggressive sodium sulfate solution. This resistance is linked to the induced porosity of the mortars which minimizes the crystallization pressure.

Keywords. Lime, straw, salt attack, biobased insulation materials, rehabilitation

I. INTRODUCTION

The energy rehabilitation of old buildings with a high quality of the building envelope are two important actions of the global and national strategy for the reduction of energy consumption of buildings. In response to this problem of energy rehabilitation and the reduction of environmental impact, the development of insulation materials and their protective coatings has been of particular interest. The challenge is to design lightweight materials with both exceptional insulation performance and durability against environmental and use conditions. Several biobased insulation materials have been proposed using local plant aggregates [1-5]. These bio-based insulators often need a coating both for aesthetic and long-term functionality of the building façade. They play the role of protectors of the buildings against external aggression and are therefore the most exposed to it. Several research works have been carried out to improve the performances of the different types of mortars by studying different parameters as the nature of the binder [6-7], the mortars composition and aggregates [8] and the incorporated additives used to improve the behaviour of the mortars facing specific degradations [9-10]. The facades coating are exposed to wetting-drying, freezing-thawing and salts crystallization. Therefore, the study of their durability against these aggressive conditions requires not only a good knowledge of the main degradation factors but also a good understanding of the resulting damage and its effect on the mortar properties [11-12].

In this experimental investigation, we are interested in studying the behaviour of three lime-based coatings against salt attacks. These coatings were developed to protect bio-based materials intended for the energy rehabilitation of buildings [2]. The studied mortars have been developed using additives that increase their porosity to ensure continuity of the hygrothermal transfer of the multi-layer wall. The addition of hemoglobin or a quantity of straw fibers leads to an increase in porosity of 45 % compared to the reference mortar [1]. For accelerated aging tests by salt attack in the laboratory, NaCl and Na2SO4 salts are used by total immersion. The porosity induced by the additives has significantly improved the resistance to chemical attack of the mortars.

II. MATERIALS AND METHODS

2.1. Sample preparation

The different studied mortars are optimized in the framework of the PHD thesis of Brahim Ismail [1]. The samples are prepared by using the preformulated lime (Tradical PF 70). It consists of 75 % aerial lime, 15 % hydraulic lime and 10 % pozzolana. The sand used in this study a fine calcareous sand 0/0.8 mm characterized by (D50=0.12 mm, Cu=1.5) and Solid density of 2710 kg/m^3. This sand was chosen with the company VEGA which is a partner in research project and specializes in the production of pre-formulated mortars. By using the spread test, the water/binder (W/B) mass ratio was set at 0.66 with a binder/sand mass ratio of 0.5. Two additives used to optimize the properties of the biocomposites were also added here to improve the properties of mortars which are hemoglobin (AE) and casein (C) [2]. The air entrainer was used to promote the porosity of the protective mortar, and the casein to improve their mechanical strength and their adhesion on the biobased substrates.

With the aim of selecting the appropriate percentages of the two additives for optimal interaction and create a high level of porosity of the mortar at fresh state, a hydrostatic weighing test was conducted after the mixture phase until stability. The general principle of this method proposed by Loukili et al. [13] consists to measure the change in volume of a sample immediately after the casting by hydrostatic weighing. The first step is to introduce a sample of mortar into a flexible, impermeable latex membrane, so that it can follow the volume deformations of the sample. The assembly is then immersed in a basin filled with water and connected to a balance (precision = 0.0001 g) by a nylon thread (Fig. 1). The change in volume is reflected by a change in the mass of the material immersed in water. For example, if the material swells, the buoyancy increases, and the balance registers a decrease in the apparent mass of the sample.

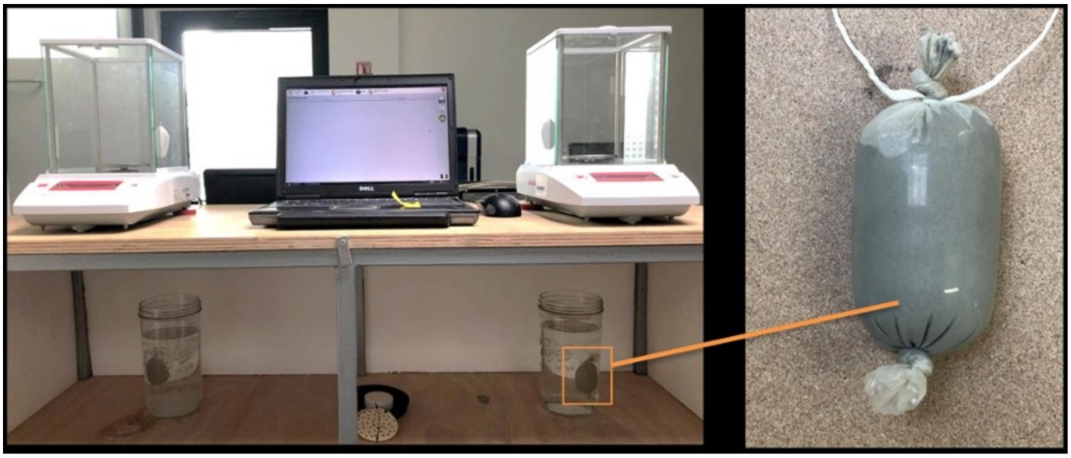
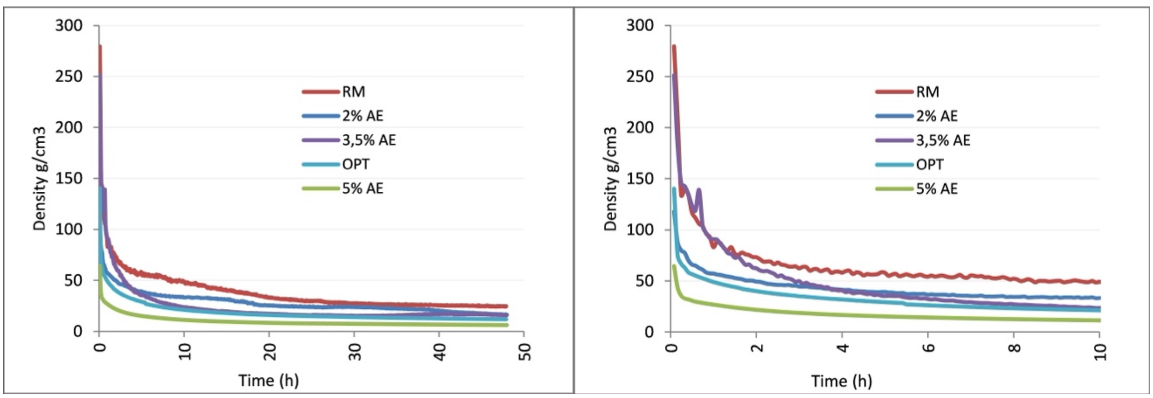


FIGURE 1. **Measurement of volume change by hydrostatic weighing.**

For the first additive, which is an air entrainer, we can see from (Fig. 2) its effect in creating porosity in the mortar, which is shown by the low density values compared to the reference mortar (RM). With 2% of AE, the reduction in density is 34% compared with the control mortar. This difference rises to 36% when the percentage is increased to 3.5% and to 75% with 5% of AE.

Furthermore, from the curves obtained, the mortars have a high initial density value, which means that the air entrainer did not affect the porosity during mixing with a behaviour similar to that of the control mortar. This contradicts the results found by Brahim Ismail (2020) [1] who found that the mortar with Air-Entrainer (AE) had a foam appearance after mixing. The mortar experienced a growth in porosity during its preparation. It is possible to explain this difference by the small quantity of mortar prepared, which did not allow generation of pores during mixing, or the high ambient temperature, which influenced the mixing process.



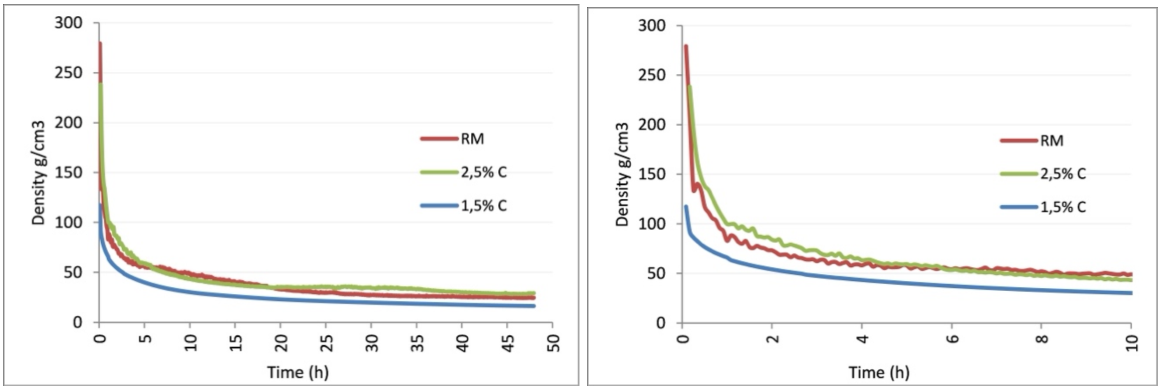


FIGURE 2. **Effect of the two additives (AE and Casein) on the evolution of mortar density for 48h and 10h.**

Figure 2 shows also that the second additive (casein: C) has a limited effect on the behaviour of the mortar. By adding 2.5% of this additive, the curve is the same for the reference mortar (RM). This means that the percentage of additive did not lead to any porosity in the mortar, on the contrary it caused a slight reduction in its volume. This reveals the fixative nature of this additive, which will subsequently increase the mechanical strength of the mortar and therefore achieve the desired function of its use.

After studying the effect of the different combinations of the two additives, the optimal mixture with the highest porosity (OPT) is the one with 5% of air entrainer (AE) and 2.5% casein (Fig. 2).

Table 1 presents the composition of the three mortars selected for this study (Table. 1). One without additives (Reference-RM), one with both additives by using a percentage of each additive allowing to benefit from the effect of hemoglobin and casein together (Optimal-OPT), and one with the additives and 7.5 % of fine straw (OPTF). The straw was crushed to obtain fibers with size less than 1 cm and added to improve the thermal conductivity of the coating. This cereal straw used was harvested in the north of the Region Centre Val de Loire. It is wheat straw that has hygroscopic behavior with an important water absorption [1]. Before his introduction in mixture, the straw is dried in the oven at 50°C until stability of mass.

Table 1. Studied mortars and their mixture proportioning

Mortar	W/B	B/Sd	AE/B	C/B	Straw (%)	Porosity (%)
RM	0.66	1	-	-	-	45
OPT	0.58	1	0,05	0,025	-	66
OPTF	0,58	1	0,05	0,025	0.075	68

W: water, B: Binder (lime), S: Straw, AE: air entraining agent, C: casein protein, Sd: sand

After the mixing protocol, samples of 4 x 4 x16 cm^3 were manufactured in clean and oiled molds (Fig.3). The mixing protocol was adjusted after some preliminary tests. It consists of adding the dry components (lime, sand, additives) and mixing for 3 minutes using a speed of 140 rpm. Then, the addition of water is done progressively in three times, mixing for 5 minutes at the same speed. Finally, the mortar is mixed for 3 minutes at a speed of 285 rpm. For mortar containing fine straw, the protocol is different. Only 80 % of the quantity of water is added in the first time. Then, adding the pre-wetted straw with the remaining 20 % of water. At the end, the mixing continues for 5 minutes at the speed of 140 rpm (Fig. 3).

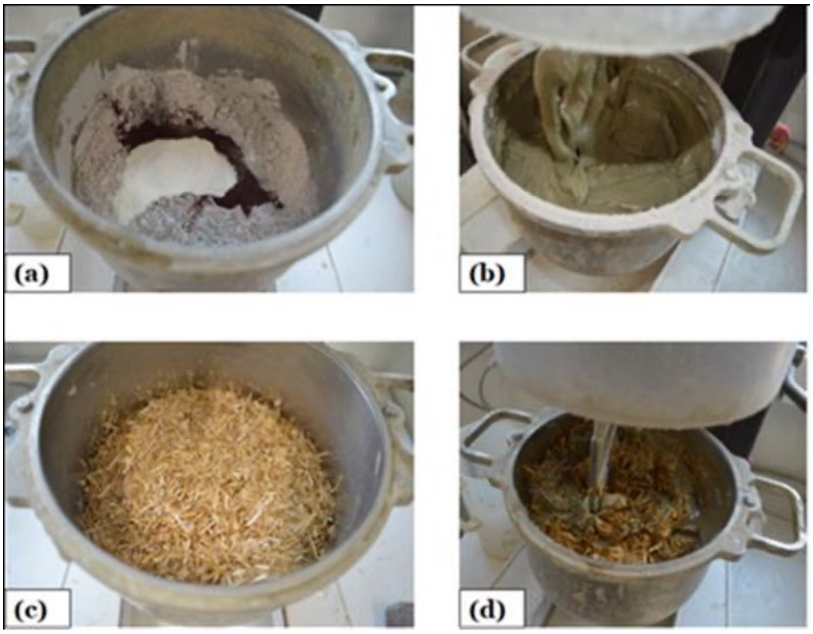


FIGURE 3. **Mixing protocol and sample preparation: (a) Introduction of lime and additives, (b) mixing of dry component with water, (c) prewetted straw, (d) Addition of straw and final mixing.**

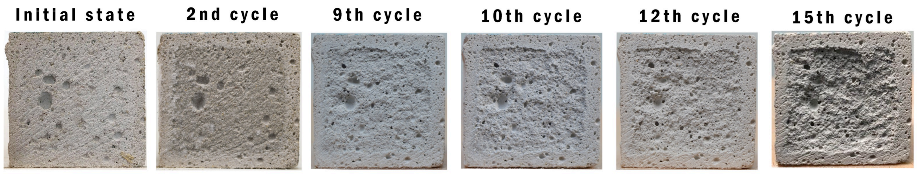
2.2. Salt attack test

To evaluate the mortar resistance to salts attack, the ageing protocol based on the total immersion of the cubic samples of 4 cm of side. Salt solutions of Na_2SO_4 and NaCl are prepared with a concentration of 14%. The choice of sodium sulfate is due to the important damage it causes to porous materials and its presence in significant quantities in soils and groundwater.

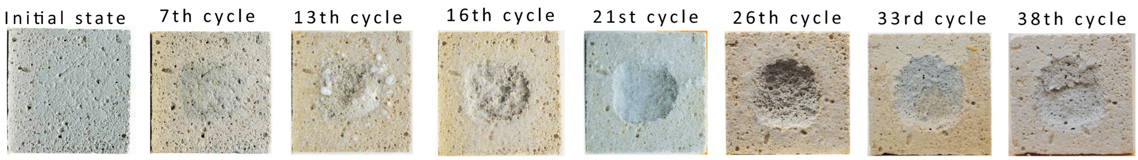
The test consists in immersing the samples in the two salt solutions according to the standard (EN 12370) [14-15]. The immersion/drying cycle is carried out by the immersion in the salt solution at 20°C for 2 hours, drying in an oven at 105°C for 20 hours and finally cooling to 20°C for 2 hours before the next cycle. The test is followed by weighing and photographic observation of the samples.

III. RESULTS AND DISCUSSION

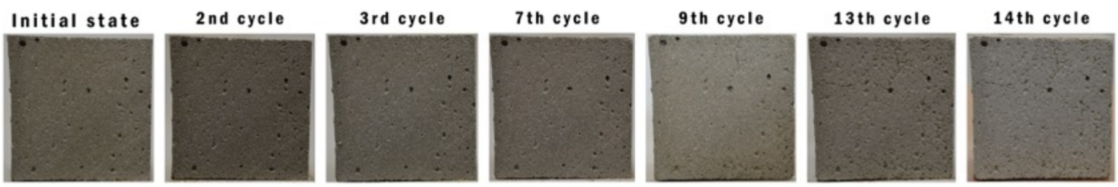
Fig. 4 shows the state of degradation of samples immersed in the NaCl solution for the three studied coatings. From this comparison, it is to be noted that the age of the samples or the curing time before ageing has considerably slowed down the cycle of appearance of degradation, especially for the optimal mortar with and without straw fiber. The cycle of appearance of the first degradation increased from 3 to 7 for the reference mortar with an important final degradation up to 15 and 40 cycles respectively after 1 and 3 months of cure. This behaviour is associated to the carbonation rate which evolves according to the age of the mortars. In fact, lime-based coatings are subject to the phenomenon of carbonation during their lifetime, which produces calcium carbonates that change the mineral structure of the mortar and consequently its mechanical performance and chemical resistance as discussed by other authors [12, 16, 17]. The two mortars with the additives (Optimal mortar) and with the fibers show a remarkable resistance with only some micro-cracks and a very slow degradation that begins on the edges of the samples, while the reference mortar subjected to a surface degradation that begins in the middle and propagates to the end with a loss of material. The behaviour of the optimal mortar has a better chemical resistance where we can hardly detect any changes in the external aspect. It seems that the additives have favored the carbonation process.



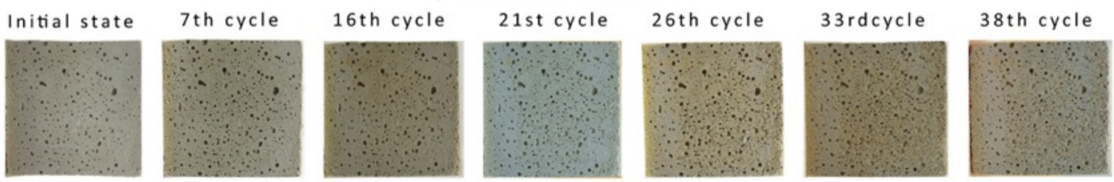
Reference mortar tested after 1 month



Reference mortar tested after 3 month



Optimal mortar tested after 1 month



Optimal mortar tested after 3 month

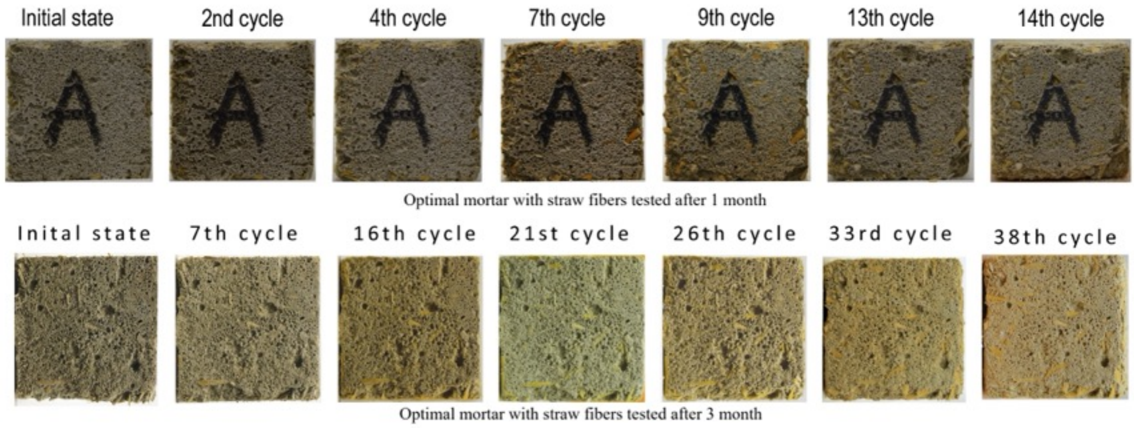


FIGURE 4. Effect of NaCl solution on the aspect of the different mortars

As noted above, we have in a second time studied the behaviour of mortar with a more aggressive salt the sodium sulfate. The evolution of the normalized mass (ratio between the mass of the sample and its initial dry mass) as a function of the number of immersion/drying cycles by using sodium sulfate is illustrated in Fig. 5.

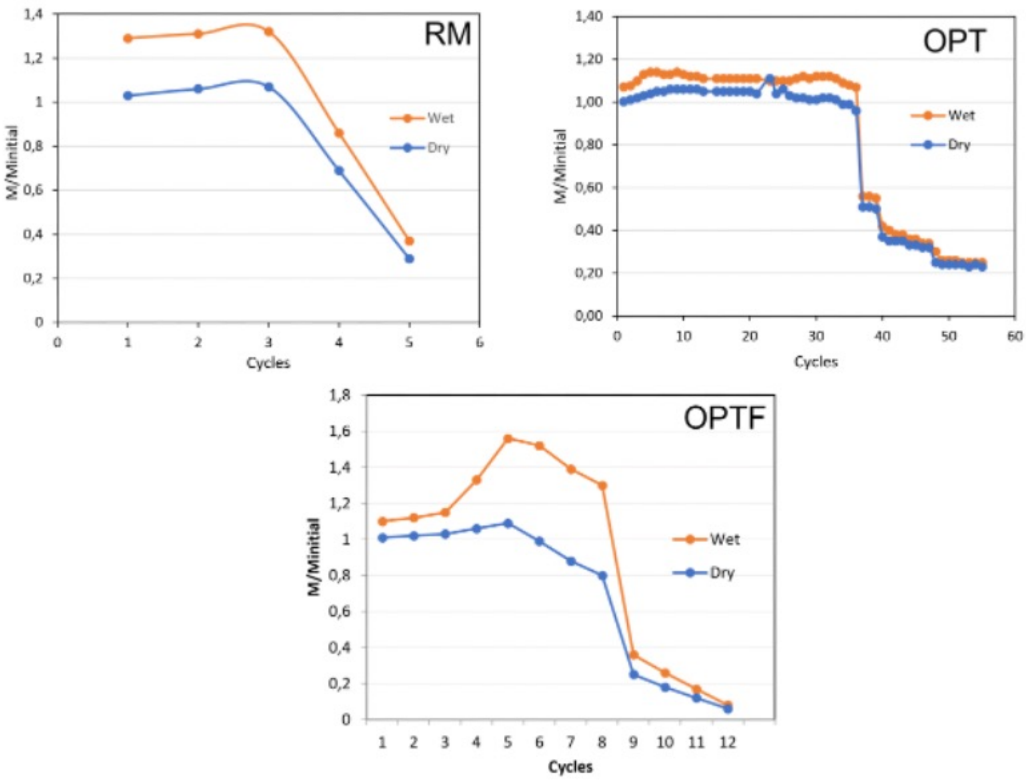


FIGURE 5. Mass loss evolution of the three mortars after immersion cycles in sodium sulfate for 1 month curing.

The results clearly show that among the studied mortars, the optimal mixture with additives has the best behaviour against salt attack. In fact, the admixture of the additives to the reference morta allowed on the one hand to slow down its mass loss until after the 37th cycle and on the other hand to increase its capacity to overcome the immersion cycles from 5 to 55. This behaviour is more than sufficient to meet the number of required cycles according to the standard EN 12370. We have therefore gone from a mortar that is not resistant to sodium sulfate to another that is more resistant to chemical attack.

This increase in chemical resistance can be associated to the action of the air-entrainer additive that generate porosity in the mortar. Therefore, the mortar became able to accumulate more salt without reaching great internal pressures that causing cracks. This improvement can also be explained by the effect of the second additive (the casein) that increased the mechanical strength of the mortar allowing it to support pressures generated by the crystallization of the salts.

The state of the material degradation shown in Figure 6, illustrate the aggressive action of sodium sulfate and the different effect on the three coating mortars. The addition of the fine vegetable fibers also shows a brittle behaviour despite the induced porosity. It should be noted here that this form of porosity does not promote the improvement of chemical resistance.

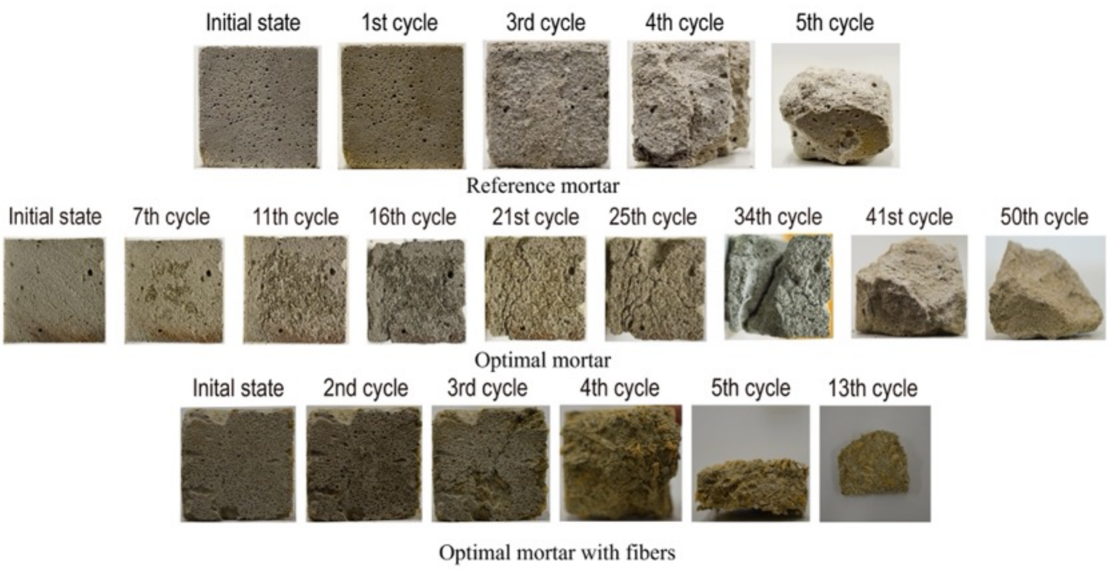


FIGURE 6. Effect of Na_2SO_4 **solution on the aspect of the different mortars after curing time of 1 month.**

IV. CONCLUSION

The objective of this investigation is to propose efficient and sustainable protective coatings for bio-based insulation materials. The effect of the porosity created by the additives show their potential to increase the chemical resistance. For the two used protocol, the results showed that:

- The sodium sulfate Na_2SO4 are more aggressive than the sodium chlorides NaCl, and this is confirmed by the considerable cracks and loss of mass and slight cracks caused by NaCL. The mortars studied are therefore more resistant to NaCL attack than to Na_2SO4.

- The effect of the additives was noticed by the high resistance of the optimal mortar to Na_2SO4 attack when compared to the reference mortar which was destroyed after 6 cycles. The addition of fine fibers is effective, but its effect remains low against Na_2SO4 by comparing with NaCl.

- The increase in chemical resistance after the addition of the additives can be explained by the porosity created by hemoglobin, which decreased the severity of the internal crystallization pressures. Also, this increase can be justified by the increase in the mechanical strength of the mortar by casein.

In the continuity of this work, the study of the system (coating and insulation material) will be carried out to prove the protective role of the developed mortars under climatic variations (wetting-drying cycles, freezing-thawing, high temperature).

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