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Study of the Durability of a Mineral Bilayer Material: Case of Granito Coated Tiles

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Abstract: Granite coated tiles offer excellent mechanical properties for cladding applications. But, as with any application in the housing field, the question of their durability arises. The durability study and the alkaline degradation process of the cementitious medium were undertaken in this work to evaluate the effect of granite and marble coating on the substrate. The mixture of the constituents with different characteristics results in a material whose properties will vary depending on the density of each constituent. For this purpose, the bilayer materials were subjected to different chemical attacks such as concentrated solutions of strong and weak acids and strong bases and the resistance to chemical attack of the samples immersed in each of these solutions is evaluated according to ASTM C 267-96. The results obtained show that before 7 days of immersion, the bilayer materials and the mortar increase their capacity to resist the attack solutions. As for 35 days of immersion, the mass losses are 9.49%, 40.63%, 3.48% for the mortar; 3.81%, 18.51%, 1.07% for the granite bilayer materials and 10.44%, 22.62% 2.94% for the marble bilayer materials in HCl solutions. This study also highlights the alkaline degradation affecting the interface of bilayer materials and it is found that hydrated cement releases basic substances, which react with acidic solutions to give salts easily soluble in water.

Keywords: Granite Coated Tiles, Durability, Attack Solutions, Alkaline Degradation

1. Introduction

Granite coated tiles offer excellent mechanical properties for cladding applications. But, as with any application in the field of housing, the question of their durability arises. In all situations where building materials may come into contact with water or moisture, a prerequisite for the development of life, they may be subject to the action of microorganisms [1].

The mechanical properties generated by a granite and marble double-layer material are defined by the ability of the material to maintain its physical characteristics and thermo-mechanical performance under satisfactory safety conditions throughout the life of the structure, taking into account the environmental conditions to which it is exposed [2-6]. Several factors impact the durability of granite and marble bilayer materials. These include:

1) the high alkalinity of the cementitious medium responsible for its alkaline degradation, which not only affects the mechanical and interface properties of cement/granite bilayers, but also reduces their service life [7-11].
2) the high basic oxide content of marble exposes this material to chemical attacks, especially in acidic environments, which dull the shine and beauty of this material in the long term [12, 13].
3) the aging of two-layer materials under the action of

water and chemical agents, especially when used for the exterior cladding of seaside walls [13, 14].

4) the nature and concentration of the solution to be used for finishing work after the installation of granitos.

The bibliographic reference on the study of the durability of bilayer materials, especially granite, suffers from many gaps. Nevertheless, there is some work on cement-based composite materials. Current research work aims to develop different solutions to inhibit or reduce microbial growth on construction materials and substrates [15-20]. For example, many studies have focused on the durability of recycled concrete [21, 22] or concrete containing natural pozzolan [23] or modified with styrene-betadiene latex [24]. A study on the effectiveness of an exterior coating to improve the durability of concrete was conducted by Almussam A. A. et al [25]. The study presented in [26] is a contribution of mineral additions on the durability of mortars. The study of the durability of reinforced concrete systems in marine environment conducted by Abderrahman Soufi [27] confirmed that two-component mortars have a better physical barrier for the protection of reinforcement. In their work, Badreddine Bessa and Bertron A. in [28, 29] were interested in the durability of cementitious materials subjected to acid attacks. However, knowledge in the field of durability of bilayer materials, especially granite, is still limited. This is why we wish to contribute to a better understanding of the behavior of these materials with respect to the aggressive environments to which they will be exposed. The present study is part of this logic and aims to study the effect of the coating of the mineral layer in granite and marble on the durability of these materials.

2. Materials & Methods

2.1. Materials Used

The materials used to make the bilayers are: sand, cement, water and granite.

2.1.1. Sand

The sand used, composed of 20% of coarse sand and 80% of medium sand according to the work of [30], comes from the region of Agamè, commune of Adjohoun; region located in the south of Benin, in the department of Ouémé. This lagoon sand washed with tap water, is dried in the oven at 105°C until stabilization of the mass and then sorted by sieving into two different granular classes: The medium sand and coarse sand composed of grains, sizes ranging respectively from 0.4 mm to 2 mm and 2 mm to 6.3 mm.

2.1.2. The Cement

The portland cement type CEM II/B-LL.42, 5R of NOCIBE is used because of its relatively fast setting.

2.1.3. The Granites

The granites used are crushed granite and marble respectively from the town of Zangnanado and Abomey, two regions of southern Benin located in the department of Zou.

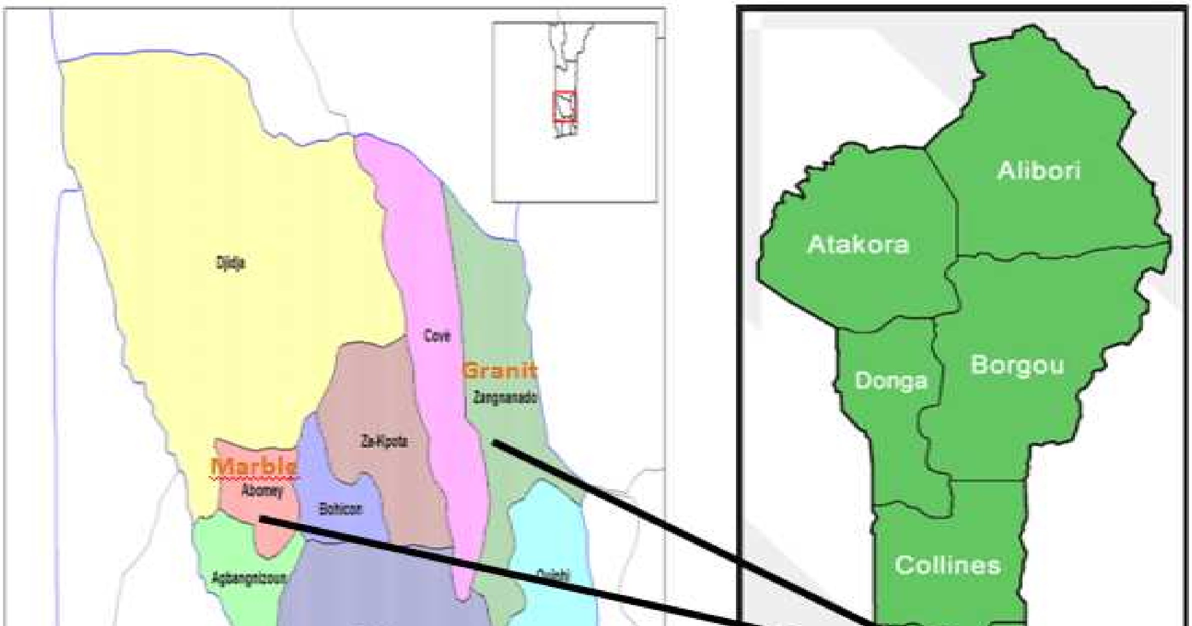

Figure 1. *Geographic location of rock sampling sites.*

2.1.4. Water

The mixing water used is that of the SONEB collected at the UAC and supposedly free of impurities.

2.2. Methods

2.2.1. Preparation of the Samples

The substrates of dimensions 10cm 10 cm 2cm were made by respecting the ratios of the dosage:

$$\frac{M_C}{M_S} = \frac{1}{3} \quad (1)$$

$$\frac{M_E}{M_C} = \frac{1}{2} \quad (2)$$

After this step, the granite coating is placed in such a way as to have a final bilayer sample of 4 cm thickness. Once the bilayer was obtained, we immersed it in water for 28 days. After 28 days, the surface of the sample removed from the water was sanded (figure 2).

Figure 2. *Granite bilayers.*

2.2.2. Chemical Attacks

The specimens of size 10, are partially immersed (1cm from the coating) in different solutions all molar (C) obtained by dissolving soda tablets and by diluting commercial solutions (concentrated solutions) of acids. These prepared solutions are:

1) Hydrochloric acid solution (HCl): strong monoacid;
2) Sulfuric acid solution (H_2SO_4): supposedly strong diacid;
3) Acetic acid solution (CH_3COOH): weak acid;
4) Sodium hydroxide (N_aOH): strong monobase.

Hydrochloric acid and sulfuric acid are inorganic acids, and acetic acid (COOH) is an organic acid [31].

The resistance to chemical aggression of samples immersed in each of these solutions is evaluated according to ASTM C 267-96 [32]. The specimens, removed from the solution, are cleaned three times with distilled water to remove mortar and/or weathered rock and then left to dry in an oven for one hour to remove the adsorbed solution. The samples are then weighed using a 0.1g precision balance. This operation is carried out after 7, 14, 21, 28 and 35 days after immersion. The etching solution is renewed every 7 days.

The chemical resistance is evaluated by measuring the loss of mass of the specimen given by the formula

$$\text{Loss of mass (\%)} = \frac{M_1 - M_2}{M_1} \times 100 \quad (3)$$

With M1 and M2 the masses of the test piece before and after immersion respectively.

The calculation of the mass losses is recorded in Table 1.

Table 1. *Values of mass losses in (%).*

Duration (days)	HCl			H_2SO_4			N_aOH			CH_3COOH		
	BG	BM	M	BG	BM	M	BG	BM	M	BG	BM	M
7	0,46	1,95	0,77	1,24	1,45	3,75	0	0	0	0	0	0
14	1,07	4,42	1,79	5,29	5,17	11,88	0	0	0	0,31	0,35	0,29
21	1,37	6,19	3,08	11,82	12,6	23,75	0	0	0	0,46	1,04	1,16
28	2,13	8,49	5,90	14,77	18,74	31,25	0	0	0	0,76	1,9	2,32
35	3,81	10,44	9,49	18,51	22,62	40,63	0	0	0	1,07	2,94	3,48

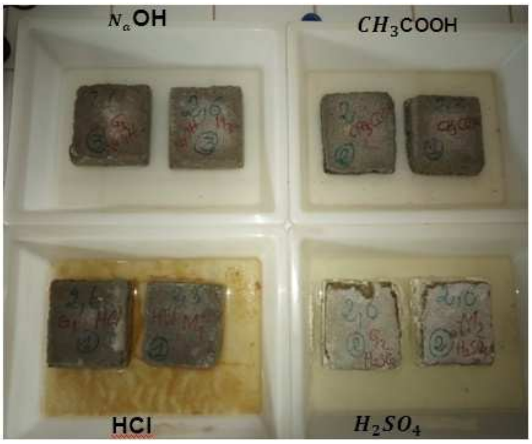

Figure 3. *Samples immersed in the solutions.*

3. Results

3.1. Attacks on Mortars and/or Bilayers by Different Solutions Acid and Basic Solutions

Figures 4, 5 and 6 show the variation of the mass loss of the specimens as a function of the immersion period in the molar solutions of HCl, H_2SO_4 and CH_3COOH respectively.

Before 7 days of immersion, the bilayer materials and the mortar increase their ability to resist the attack solutions. After this period, these materials begin to be weakened by these acid solutions, except for the BG which continues to oppose a very remarkable resistance in HCl and CH_3COOH.

The losses of masses are enormous in the solution of sulfuric acid and that of hydrochloric acid while they are very weak in the solution of acetic acid. This observation would be related to the chemical classification of acetic acid as a weak acid and hydrochloric and sulfuric acid as strong acids. Moreover, we notice for all the specimens, that the losses in mass due to the sulfuric acid are largely superior to those due to the hydrochloric acid. This would be due to the fact that at equal concentration, sulfuric acid (supposedly strong diacid) releases more hydronium ion (H_3O^+) than hydrochloric acid (strong monoacid).

The granite coating reduces the mass losses, compared to the control mortar, by 5.68% in HCl, 22.13% in H_2SO_4 and 2,41% in CH_3COOH. However, in the case of the marble bilayers, it is difficult to evaluate the difference in mass losses due to acid solutions, compared to the control mortar. Thus, instead of having only a loss of mass of the substrate, there is also a dissolution of the mineral layer of marble in the solutions of attack, leaving a materials. This precipitate is the product of an acid-base reaction between the oxides, especially calcium and magnesium oxide (basic oxides), which are mainly present in marble, and the acid solutions.

Moreover, during the whole test period, no loss of mass is recorded with the sodium hydroxide solution (N_aOH)). This remark excludes a chemical reaction between two bases, but confirms on the other hand, that the chemical attacks taking place in the different reaction media, are purely acid-base reactions (proton transfer H^+).

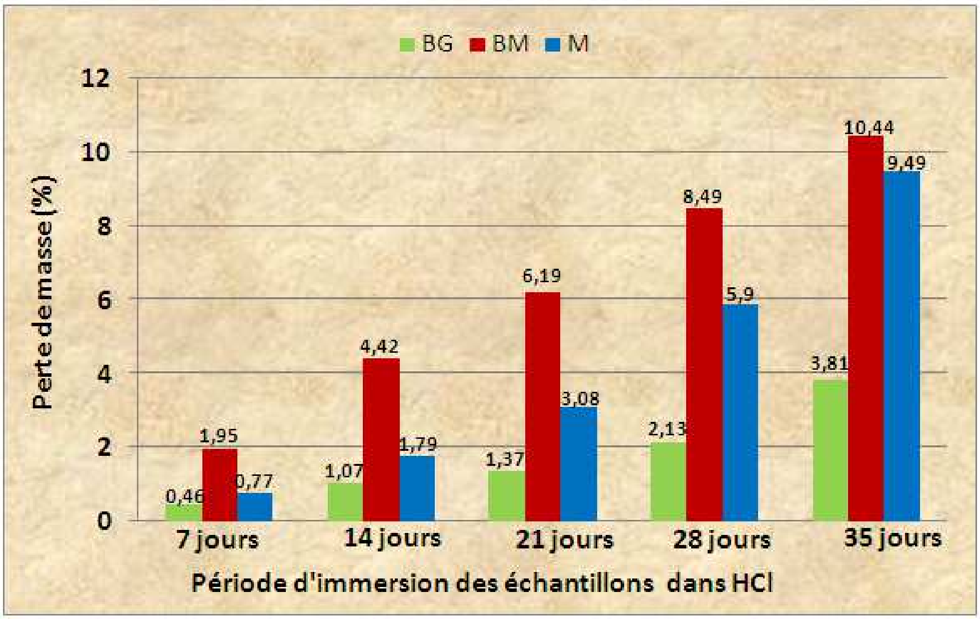

Figure 4. *Variation of mass loss as a function of immersion period in hydrochloric acid solution.*

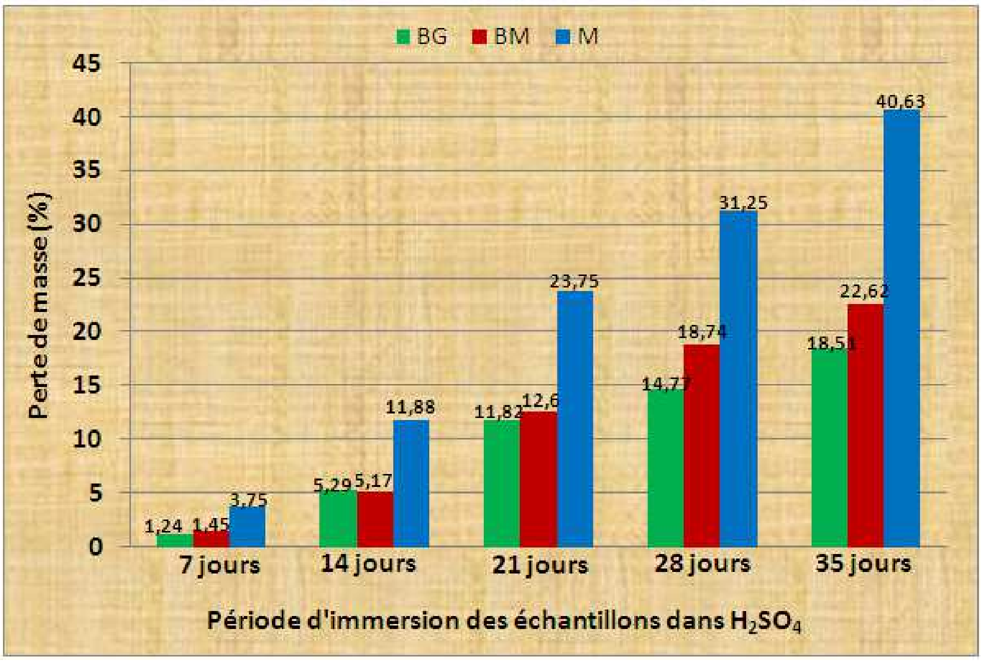

Figure 5. *Variation of mass loss as a function of immersion period in the sulfuric acid solution.*

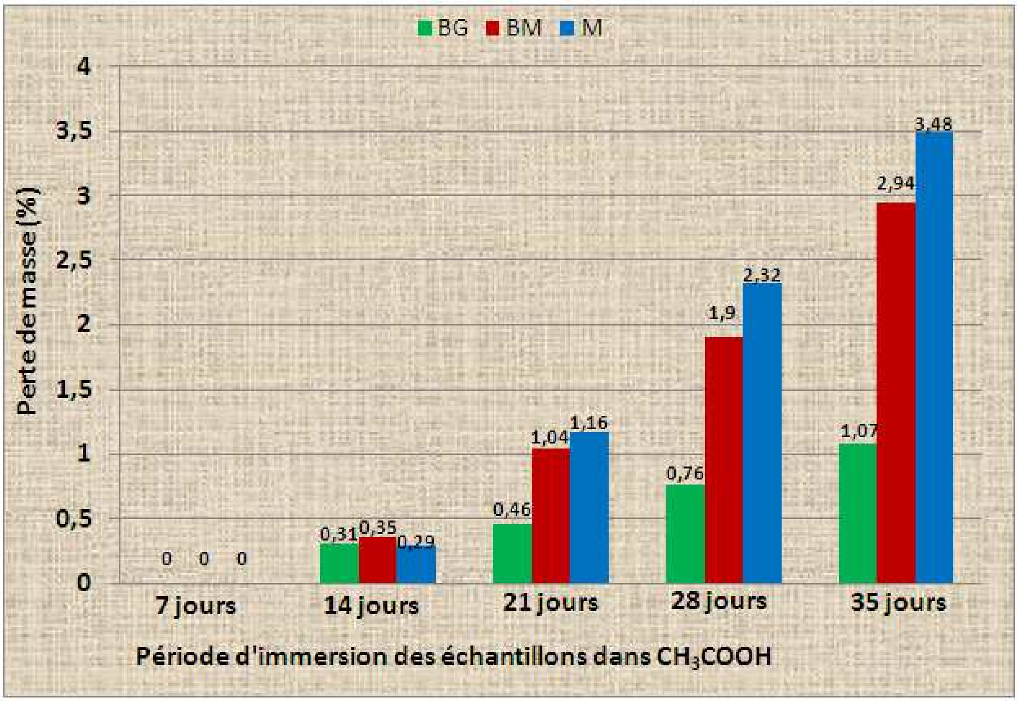

***Figure 6.** Variation of mass loss as a function of immersion period in acetic acid solution.*

Comparing the results of mass loss at 35 days, mortars and/or composites immersed in acetic acid solution, compared to other results, we note that the loss of mass due to acetic acid is about one third and one tenth ($\frac{1}{10}$) of the loss of mass due to the hydrochloric acid solution and sulfuric acid respectively (Figure 7).

According to figure 7, the chemical attacks depend more or less on the concentration, but also and especially on the nature of the acids in the most aggressive order, H_2SO_4 then HCl and finally CH_3COOH.

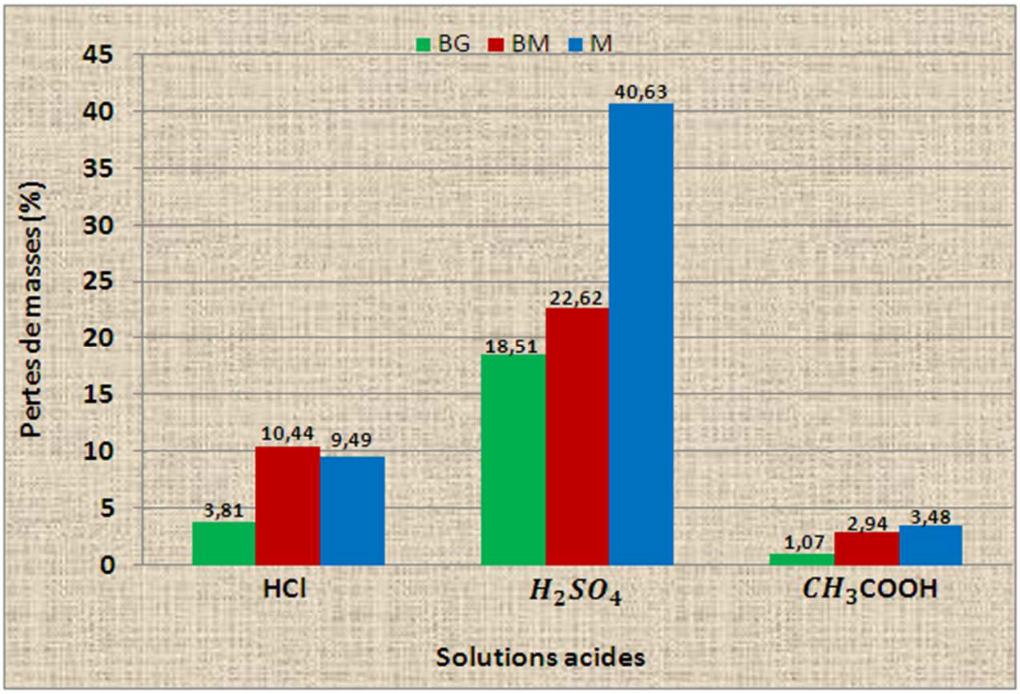

***Figure 7.** Variation of mass loss at 35 days of immersion in different acid solutions of mortars and mineral bilayers.*

3.2. Visual Examination of the Results

Figure 8 shows the state of the samples before their partial immersion in the aggressive solutions, and Figure 9 shows the change in surface state of these samples subjected to attacks in the different aggressive media.

Figure 8. *Surface condition of the specimens before immersion immersion in the etching solutions.*

Figure 9. *Surface condition of the samples after 35 days of immersion in HCl,, H_2SO_4, CH_3COOH and N_aOH and (from left to right).*

The loss of mass of the specimens recorded at the level of the mortar, would be due to a dissolution of the hydrates leading to porous mortars. This is justified by the fact that Portland cement, after hydration, releases an important part of calcium hydroxide which $Ca(OH)_2$, in contact with an acid solution, is leached inside the material. This phenomenon (leaching) is more accentuated with strong inorganic acids than with organic acids. For the mortar in contact with the sulfuric acid solution, we observe the deposition of gypsum ($CaSO_4$), resulting from the reaction between this attack solution and the calcium hydroxide released by the Portland cement.

The precipitated gypsum layer is easily leached out, resulting in a considerable loss of mass [33, 34].

The chemical attack of marble bilayer materials reveals two chemical reactions taking place simultaneously in the same reaction medium. Indeed, in addition to the dissolution of the hydrates contained in the cement paste, we also witness an acid-base attack between the basic oxides, in particular the oxides of magnesium and calcium mainly present in the marble. Therefore, mass losses are recorded not only at the substrate level, but also and especially at the marble coating level. These losses are enormous in the sulfuric acid solution (figure 9, second column) where white precipitates of calcium sulfate (gypsum) and magnesium sulfate ($MgSO_4$) are obtained. On the other hand, with the acetic acid solution, we note a strong ability of the marble coating to resist the chemical attack caused by this solution. The loss of mass in this case is due to a destruction of the cement paste coating the marble granites.

In contrast, chemical attack at the granite bilayer materials affected mainly the cementitious paste encasing the granite crushes and the substrate. In all three acid solutions, no loss is recorded at the granite coating, which protects a large part of the substrate from any chemical attack. Nevertheless, the chemical attacks taking place, caused a progressive destruction of the cementitious paste, thus weakening the bond between the granitos which end up progressively detaching from the coating, exposing the substrate (Figure 9, 3rd row and 1st column). This observation was more noticeable in the hydrochloric and sulfuric acid solutions than in the acetic acid solution, which seems to be non-aggressive to these materials.

In addition to these different observations, a thick whitish layer appears in the acetic acid solution (Figure 10). In addition, we observe an intense effervescence during five minutes following the introduction of the test tubes in the hydrochloric acid solution and the appearance of a yellow coloration.

Figure 10. *Surface appearance of specimens immersed in CH_3COOH.*

In order to identify the origin of the effervescent gas and the yellow color obtained, we introduced some pieces of granite, marble and substrate, in three beakers containing the hydrochloric acid solution used.

Only in the beaker containing the piece of marble (Figure 11c), for about five minutes, was there a strong effervescence. The marble is a limestone or dolomite recrystallized by heat and pressure. Thus, the observation made, would be attributed to a chemical reaction between the hydrochloric acid solution and the calcium carbonate (CO_3) present in the marble. It results among other products, the release of carbon dioxide (CO_2) according to the equation:

$$CaCO_3 + 2HCl \rightarrow CaCl_2 + CO_2 + H_2O \quad (4)$$

In the beaker containing the granite and the substrate (figure 11a and b respectively), we note the appearance of a yellow coloration two minutes after immersion in the beaker containing the substrate and three days later in the beaker containing the granite, which gradually intensifies as time passes. This finding could be related to the contact surface offered by the material to the etching solution. Thus, the substrate being realized with cement which is directly in contact with the solution of hydrochloric acid while the granite crushed which are naturally compact materials, thus difficult to penetrate by the solution of attack. This could justify the delay observed in the appearance of the color at the level of the beaker containing the granite.

Nevertheless, the yellow color observed would be the result of a chemical reaction between the hydrochloric acid solution and the iron sesquioxide, also called ferric oxide ($F_{e2}O_3$) contained in the granite and the cement, to form iron III chloride (F_eCl_3), which in solution releases iron III ions (F_e^{3+}) responsible for the yellow color. This ion thus imposes its color to the solution of the reaction mixture.


(a) (b) (c)

Figure 11. *Reaction of hydrochloric acid with the different materials.*

On the other hand, the comparison of the test results with those of the literature seems difficult. This can be justified by the differences in the raw materials used (cement, nature of the material and coating), the dosage made, the concentrations of the chemical attack solutions and the test methods.

4. Conclusion

The study of durability has allowed us to understand and provide an explanation for the alkaline degradation of cement/granite bilayer materials and the aging of these materials under the action of external chemical agents. This study has also allowed to review the behavior of granite and marble pavements towards different aggressive environments. In general, the granite bilayer materials develop a better resistance than the marble bilayer materials against the different chemical attacks.

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