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Effect of the Chemical Composition of Building Materials on Algal Biofouling

Philippe GROSSEAU¹, Estelle DALOD², Alexandre GOVIN³, Christine LORS⁴, René GUYONNET⁵ and Denis DAMIDOT⁶

¹Professor, SPIN-EMSE, CNRS:UMR5307, LGF, Ecole Nationale Supérieure des Mines de Saint Etienne, France

² PhD Student, SPIN-EMSE, CNRS:UMR5307, LGF, Ecole Nationale Supérieure des Mines de Saint Etienne, France and LGCgE- GCE, Ecole Nationale Supérieure des Mines de Douai, France

³ Assistant Professor, SPIN-EMSE, CNRS:UMR5307, LGF, Ecole Nationale Supérieure des Mines de Saint Etienne, France

⁴ Professor, LGCgE- GCE, Ecole Nationale Supérieure des Mines de Douai, France

⁵ Professor, SPIN-EMSE, CNRS:UMR5307, LGF, Ecole Nationale Supérieure des Mines de Saint Etienne, France

⁶ Professor, LGCgE- GCE, Ecole Nationale Supérieure des Mines de Douai, France

Abstract: The main cause of aesthetical deterioration of outdoor exposed building materials is the colonization by microorganisms. This phenomenon depends on factors such as geographical situation, environmental conditions and surface state of the substrate. Several researches have been devoted to the study of the effect of porosity, roughness and surface treatment on the biofouling of building materials. However, none of them has addressed the influence of cement composition. The main objective of this study is thus to highlight the influence of the composition of the material on its biocolonization by algae. The green alga *Klebsormidium flaccidum* was chosen because of its representativeness in France. It is indeed the species the most frequently identified and isolated from samples taken on sites. In order to characterize the influence of the composition of building materials on their biofouling, the behavior of mortars prepared with two types of Portland cement and two types of calcium aluminates cement is studied. The biofouling is followed by measuring the covering rate thanks to image analysis. This work is realized both on samples exposed outdoor and on samples tested in a laboratory bench. Obtained results prove that the composition of cementitious materials is a determining factor.

Keywords: biodeterioration, algae, mortars, Portland cement, Calcium Aluminate Cement.

1. Introduction

The colonization of building facades by microorganisms is the main cause of their aesthetical deterioration. The ageing of facades causes a change of the state of surface which favors the development of these species, in particular in the place where the rainwater flows and where run-outs appear. These latter are made of an association of organic particles and microorganisms called biofilm.

The main colonizing microorganisms are microalgae, fungi and bacteria (1,2,3). Barberousse (1) identified the micro-algae *Klebsormidium flaccidum* as the major microorganism developed on facades in France. *Klebsormidium flaccidum* is considered as a ground species. It is a green algae with a wide ecological amplitude (4). It can be found in the ground, on rocks, stone walls, and bark of trees. It is known for its wide presence in temperate regions (5,6,7).

The environment conditions the type of microorganism which colonizes a façade (3,8). The climate which includes temperature and moisture determines which microorganism will mainly grow. The rain and the wind favor the transport of microorganisms. The orientation of facades also impacts the biological development. An exposure toward North will be characterized by higher humidity and an absence of sun. These conditions favor the biofouling (1,9,10).

Organic materials such as paints and polymers and mineral materials such as mortar, concrete, natural stones or clay bricks are sensitive to biodeterioration. Barberousse (1) showed that the organic materials resist generally better to the colonization by algae and Cyanobacteria than mineral materials. The microorganisms observed are more diversified and more plentiful on mineral materials than on organic substrates.

Cementitious materials are heterogeneous, porous and their surface roughness can vary within a wide range. All these properties define the bioreceptivity of a material (2,11,12). Previous studies have

determined the influence of mortars' physical characteristics on their biofouling. Among them, the roughness of the surface (3,5,8,9,13) and the porosity (8,12,14) seem to be the most important ones.

Cementitious matrices partially weather because of the presence of carbon dioxide in the atmosphere. The natural carbonation of the mineral matrix leads to a decrease in surface pH and favors the microbiological colonization (8,10) and an alkaline surface pH can totally inhibit the colonization of a material by microorganisms. Indeed the pH affects the microorganisms because it regulates the ionization mechanism of metabolites (15).

The solutions developed to prevent the biocolonization of facades by microorganisms must take into account the preservation of the environment and the understanding of the interactions between microorganisms and materials is thus necessary for this purpose.

The main objective of this study is to highlight the influence of the chemical composition of the substrate on the biofouling, comparing the behavior of mortars prepared with cements of different mineralogy.

2. Materials and methods

2.1 Mortars formulation

In order to study the influence of the mineral chemistry on the biofouling of cement mortars, four types of cement were selected; two Portland cements and two calcium aluminate cements. The materials were a white Portland cement CEM I 52.5 N CE CP2 NF "SB" provided by Italcementi (coded CEMB), a grey Portland cement 52,5N-PM-ES-CP2 provided by Lafarge (coded CEMG) and two calcium aluminate cements provided by Kerneos, a white one (coded CACB) and a grey one (coded CACG). The chemical compositions obtained by X-Ray fluorescence are given in Table 1.

Table 1 : Chemical Composition of the Cements CAC B, CAC G, CEM B and CEM G and the Siliceous Sand (%wt)

Sample	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MnO	MgO	Na ₂ O	K ₂ O	P ₂ O ₅	LOI
CAC B	4.98	1.89	49.99	1.70	38.61	0.06	0.58	0.39	0.26	0.15	0.47
CAC G	0.25	0.38	69.8	0.18	30.61	0.03	0.46	0.18	0.42	0.12	0.56
CEM B	21.70	0.16	4.25	0.41	69.53	0.01	0.79	0.32	0.07	0.05	2.79
CEM G	22.40	0.15	2.87	2.22	69.34	0.05	0.91	0.19	0.18	0.07	1.65
Sand	97.44	0.04	1.15	0.1	0	0.07	0.15	0.39	0.53	0.01	0.2

The mortars were prepared with either CAC B, CAC G, CEM B or CEM G and siliceous sand (Sifraco DURANCE DU 0.1-0.35 with $d_{50}=254\mu\text{m}$). The formulation was 30% cement, 70% sand and the ratio w/c was 0.57 for the CAC B based mortars, 0.54 for CAC G based mortars and 0.5 for the CEM based mortars, in order to obtain similar consistencies.

The dry mix was added in water in a standard mixer. The mixing time was chosen accordingly to the procedure of the NF-EN-196-1 standard. The fresh mortar was then poured into polystyrene moulds of 50x50x1 cm (LxWxH). The air bubbles were eliminated by means of a vibrating table and of a ruler. For a higher roughness, the roughcast roller was applied during the setting. The arithmetical roughness was determined with an optical profilometer CHR-150-L. The mortars were stored 7 days in 100 % of relative humidity (RH) before being cut. The final dimensions of the samples were of 8x20x1 cm (LxWxH).

Accelerated weathering of mortar was carried out in a cell under pure CO₂ at 20°C and 65% RH. Carbonation reaction and decrease of surface pH were thus speeded up (16).

The pH-decrease was monitored with a surface electrode (WTW Sentix Sur). When the surface pH reached 9, carbonation was stopped and the mortars chemical composition was estimated by X-ray diffractometry and thermogravimetric analysis. The carbonation time was 30 days for CEM cement based mortar and 38 days for CAC cement based mortar.

The mortar porosity was measured via mercury intrusion porosimetry (Micrometrics Autopore IV 9400) after drying 24 hours at 60°C.

2.2 Algal culture

The algal culture of *Klebsormidium flaccidum* requires an appropriate culture medium: the Bold's Basal Medium, according to NF-EN-15458 standard. This medium contains macro-elements such as carbon, oxygen, phosphorus, nitrogen and hydrogen as well as some mineral salts (or trace elements).

Strains were supplied by the Museum National d'Histoire Naturelle (MNHN, Paris, France). The cultures were made in batches exposed to an artificial light for photoperiods of 12 hours. Two fluorescent lamps (Fluora Osram) provided a light intensity of $20\mu\text{mol}/\text{m}^2/\text{s}$ PPF (Photons Photosynthetic Flux) which corresponds to a power of 30 W. This type of lamp was chosen because the light emitting spectrum matches with the chlorophyll light absorbing spectrum. Temperature and hygrometry were regulated at 20°C and 60 % RH respectively. Algal growth was followed thanks to various microbiological techniques such as cellular counting, optical density via UV-visible spectrophotometer, and determination of dry mass via algal suspension filtration. The ionic concentration was measured by ionic chromatography. These techniques were also used to characterize the algal growth in the biofouling laboratory bench.

2.3 Biofouling laboratory test

The system consisted of a glass chamber of dimensions 100x50x50 cm in which were settled two stainless steel supports inclined at 45 ° (Figure 1). The mortar samples were exposed to the same conditions of light ($20\mu\text{mol}/\text{m}^2/\text{s}$ PPF, photoperiods of 12 hours) and temperature (20°C) as for the algal growth. The temperature was maintained with a thermoregulator. The box was filled with 50L of microalgal suspension, (concentration of 4mg/L dry mass) which was pumped and sprinkled on the top of mortar samples. The watering was made by means of a full holes banister of 2 mm in diameter, every 1 cm. The flow was set to 26 ± 1 L/h. The sprinkling was carried out for 90 minutes every 12 hours and enabled to reproduce the phenomenon of colonization of a facade: microalgae were transported by the solution and had the possibility of hanging on and of adhering to the surface according to the characteristics of the mortar.

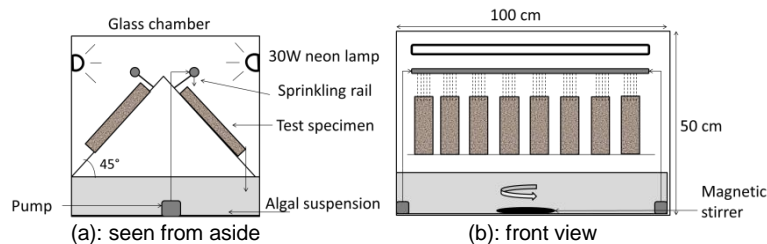


Figure 1. Laboratory bench for the colonization of mortars by streaming of algal suspension

2.4 Biofouling in-situ test

The in situ bench is set up at the "Ecole Nationale Supérieure des Mines de Douai" situated in Douai (North of France). The bench test on which are exposed 60 samples is constituted by a stainless steel structure (Figure 2). The angle of inclination is fixed to 45° which allows exposing samples to the weather conditions (period of sunshine, streaming, gusts of wind). Rows of samples are situated at least 1 meter above ground level, to avoid spatters phenomena. Samples are arranged so that the flow from one sample cannot contaminate the sample situated below and are spaced out by 5 cm to avoid contaminations. The site of exposure is a flat ground, distant from the traffic and close to some trees (situated at about 15 m from the bench)

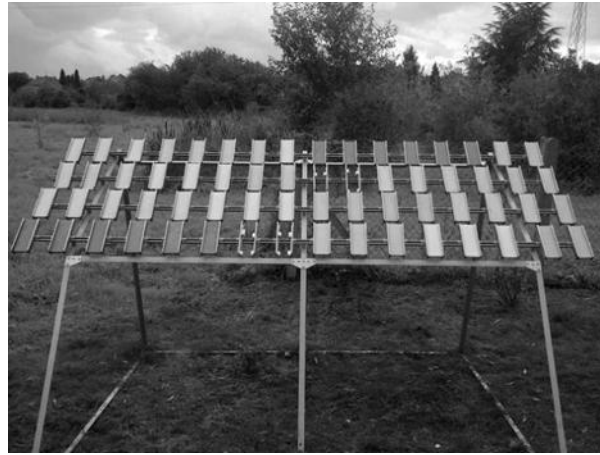


Figure 2. In-Situ bench for the colonization of mortars

2.5 Biofouling evaluation

The biodeterioration intensity was measured by image analysis. Thanks to an office scanner, the surface of sample mortars was digitized at different times. These images were then treated with the software Aphélon[®] which calculates a covering rate (noted $X(t)$) of samples by microalgae (13). The initial RGB color space was converted in YIQ color space in order to identify easily the green algae on mortars.

The Q channel was used to quantify the number of colonized pixels after thresholding and segmentation. The colonization rate was given by the ratio of colonized area to the total surface. The different steps of image analysis are detailed on Figure 3. After segmentation the binary image represents in red the fouled area and in black the unfouled one.

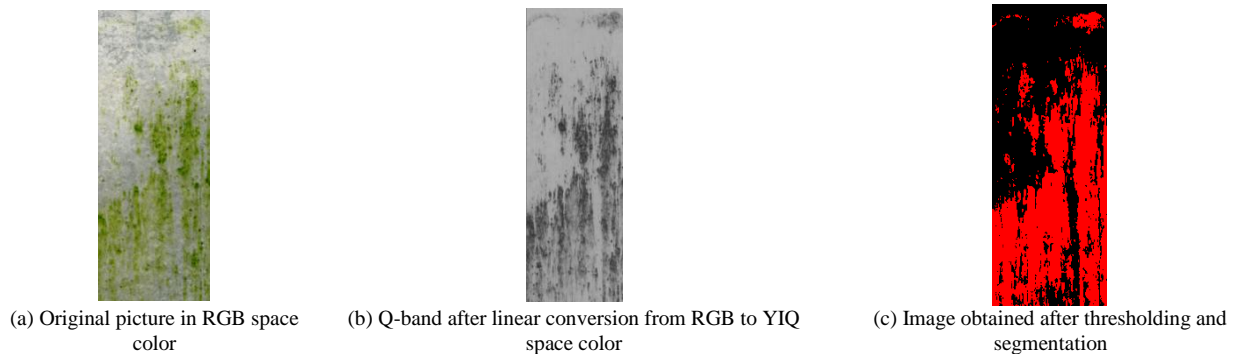


Figure 3. Example of image analysis of a specimen after 9 days test

3. RESULTS AND DISCUSSION

3.1 Characterization of the materials

Results represented on Figures 4, 5 and 6 show that roughness, surface pH and porosity are not significantly different for all samples, around a value of about $220 \mu\text{m}$ for roughness, 8.8 for surface pH and 14% for the porosity. This is an important result since it was already proven that these characteristics have a strong influence on the kinetics of biocolonization of mortars (4,5,9,12,13,14). As the physical characteristics of presently studied mortars are similar, we will thus be able to compare the effect of the chemical composition on the resistance to biofouling.

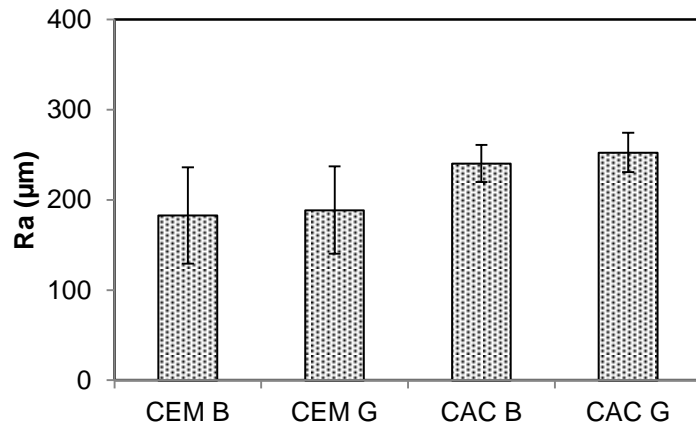


Figure 4. Roughness of studied materials

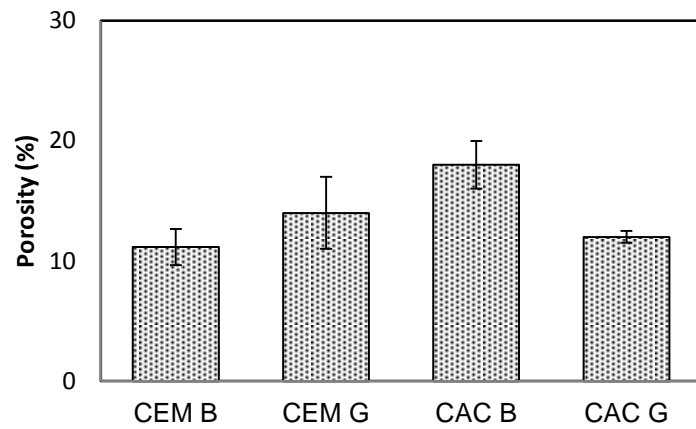


Figure 5. Porosities of studied materials

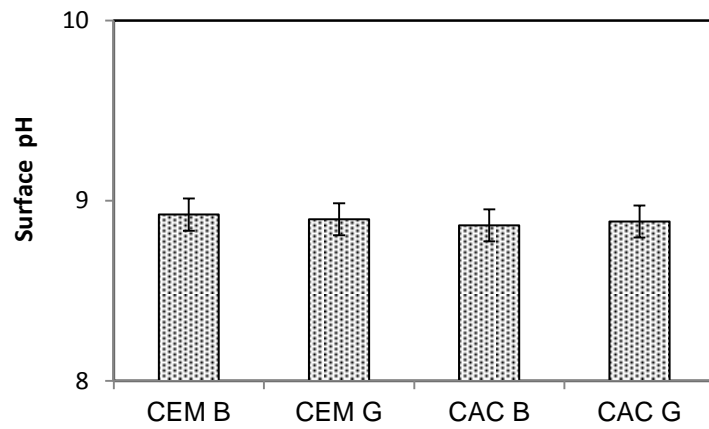


Figure 6. Surface pH of studied materials

3.2 Biofouling study in laboratory bench

The pictures of Figure 7 show the evolution of the surface of the specimens during the test period for Portland cement mortars and CAC mortars and we can thus notice that CAC based mortars were much less quickly colonized than Portland cement based mortars.

This phenomenon is fully confirmed by the evolution of the colonization rates of the specimens which is shown on Figure 8. The colonization of Portland cement based mortars both started earlier and was completed in a shorter time than the colonization of CAC based mortars.

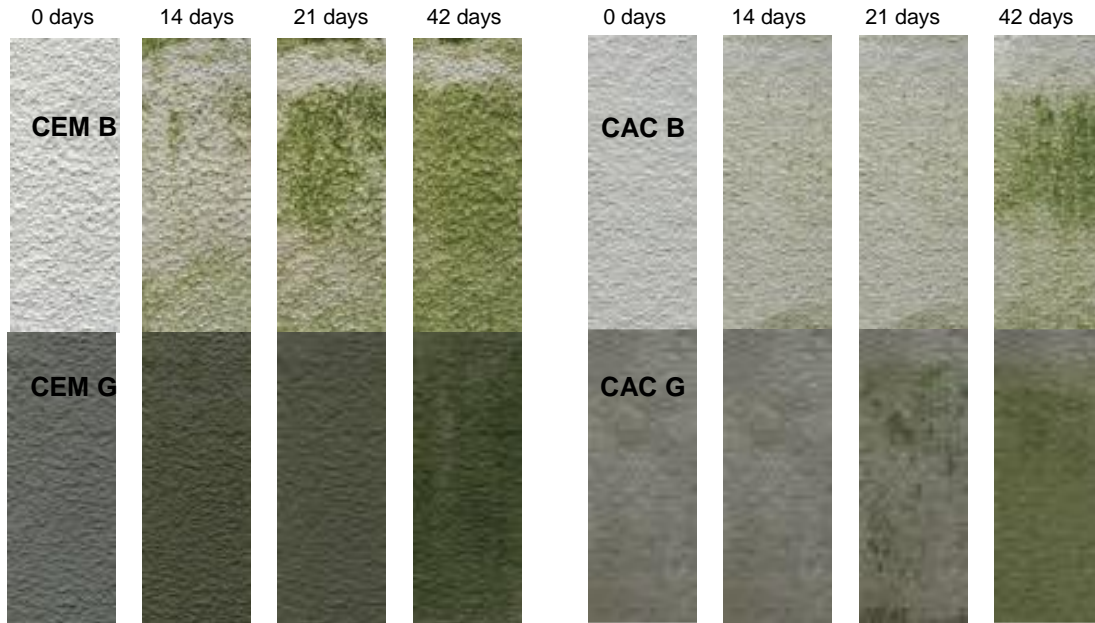


Figure 7. Evolution of the biocolonisation of the samples during accelerated laboratory test

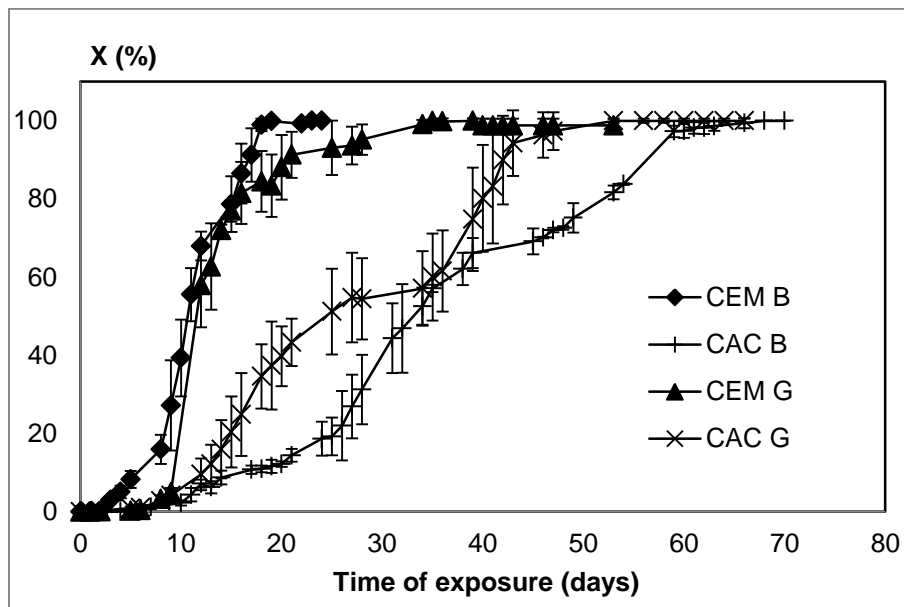


Figure 8. Evolution of the fraction of colonized area during accelerated laboratory test

3.3 *In-situ study of the biocolonization of mortars*

This part of the work was realised only with white mortars CEM B and CAC B. The samples were exposed from March 2012, at the beginning of spring. In a similar manner to what happened in laboratory tests, we can again notice that mortars containing CAC cement were more slowly colonized than the mortars which contain Portland cement (Figures 9 and 10).

On Figure 10, one can notice a decrease of the colonization rate after about 550 days and 900 days of exposure. This phenomenon was due to winter conditions which implied a detachment of algae from the material.

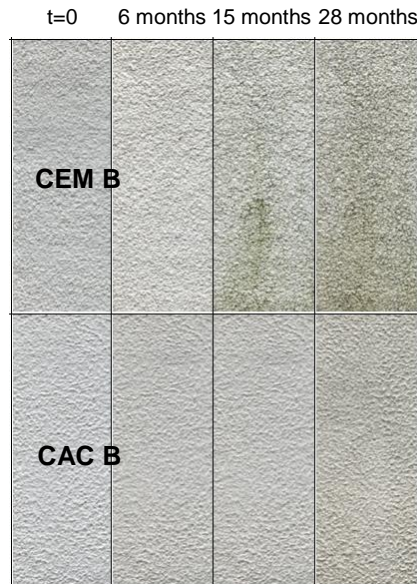


Figure 9. Evolution of the biocolonisation of the samples during outdoor exposition

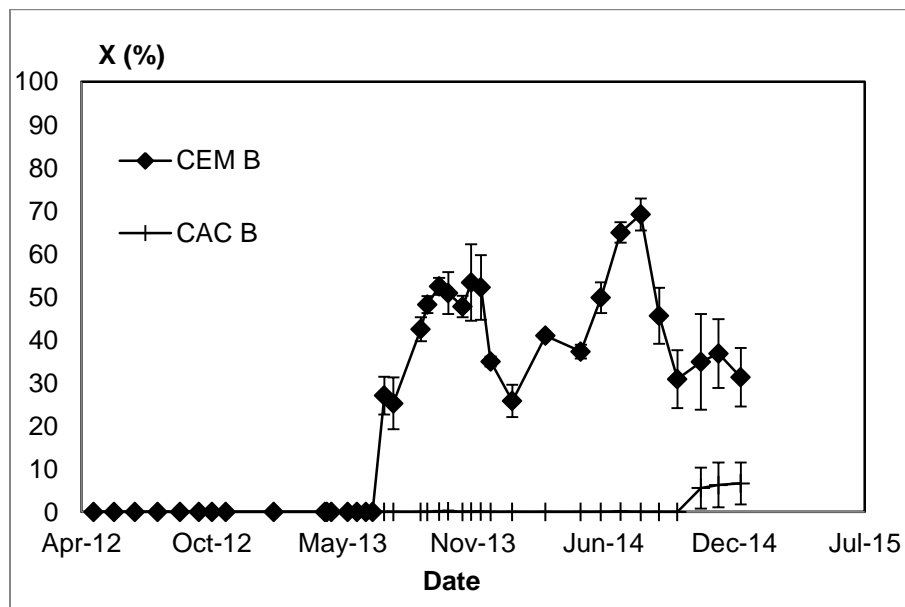


Figure 10. Evolution of the fraction of colonized area during outdoor exposition

3.4 Discussion

In order to verify that the observed phenomena were not due to a difference in the alkalinity of the studied mortars, we followed the evolution of surface pH of the samples and of the pH of the algal suspension all along the laboratory test. The results are shown in Figures 11 and 12. It is quite clear that there are no significant differences between the different materials tested, which is of course consistent with the fact that all samples are carbonated.

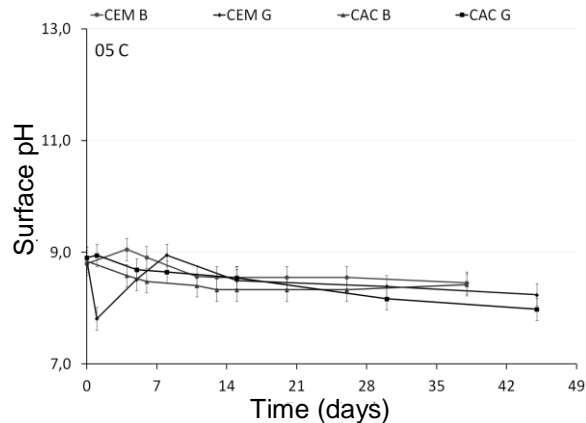


Figure 11. Evolution of the surface pH of samples during laboratory test

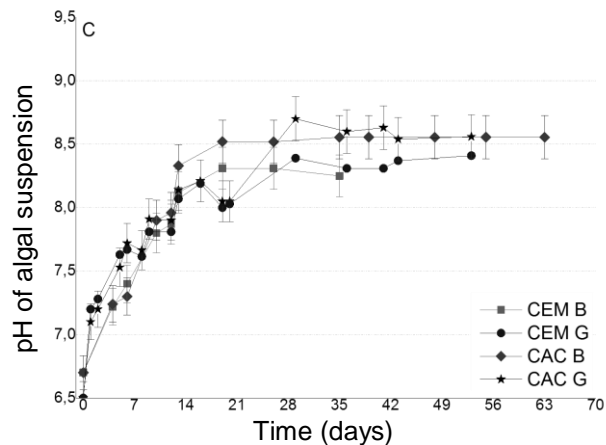


Figure 12. Evolution of the pH of algal suspension during laboratory test

Lindemann et al.(17) emphasize that aluminum toxicity is known in acidic pH and highlight a slowdown in the growth of algae at neutral pH (~ 6.5), pH range in which aluminum hydroxide $Al(OH)_3$ is predominant. A bacteriostatic effect of hydrated alumina gels formed by mild acid attack in sewage pipes is also proposed to explain the durability of aluminous cements for such applications (18).

It is thus likely that species containing aluminum such as the gel of $Al(OH)_3$, present in significant amounts in carbonated aluminates cements (19,20), possess a bacteriostatic effect at the origin of the particularly high durability of CAC based mortars with respect to the algal biocolonization.

4. Conclusions

In this study, we have shown, both by accelerated laboratory tests and by outdoor exposition, that aluminate cement-based mortars present a resistance to algal biocolonization much higher than Portland

cement mortars. This study was performed using mortars with similar physical characteristics (roughness, porosity, surface pH) and which do not influence the pH of their environment in different ways. We can thus conclude that this effect is mainly due to mineralogical differences. Previous studies indicate that this phenomenon might be caused by a bacteriostatic effect of aluminum hydroxide. We plan to continue this study and verify this hypothesis by additional tests on the interaction between $\text{Al}(\text{OH})_3$ and microorganisms.

5. References

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