HYDROPHOBIC PROTECTION OF FIBRE REINFORCED CEMENT BOARDS WITH SILICON-BASED MATERIALS

LECOMTE JEAN-PAUL[1]; SELLEY DAVID[1], MCAULIFFE TONY [1,2] and SPAETH VALERIE [3]

[1]Dow Corning, Rue Jules Bordet, B-7180 seneffe, Belgium, [2]current affiliation : SILSESQUI, Pty Ltd, Australia, [3]Université Libre de Bruxelles, Belgium

ABSTRACT

As manufacturers of FRC board move from using Asbestos to Cellulose as the reinforcement fibre in their products, a technical challenge has emerged that requires some innovative solutions. The KEY CHALLENGE is to maintain the types of performance expectations that the manufacturer (and the customer) had perceived as 'given' for the asbestos based fibrous cement technology. Life times can be difficult to predict with confidence but there is a widely accepted opinion that to achieve the same life in non asbestos boards, extensive modifications and additives need to be used. This is particularly true for reduction of water absorption.

What is required in the market place are systems that ensure the longest term possible performance under all types of weather conditions. One option to achieve this increased water exclusion performance might be by the use of silicone technology.

The paper will review the different strategies which can be used to reduced the water absorption of Fibre boards thanks to the use of silicon-based additives and illustrate them. Data showing the efficiency of silane/silicone based water repellent as post treatment or admixture are given. Impact of silane/silicone admixture is shown to be limited on cement hydration.

KEYWORDS:

Silane; silicone; integral water repellent.

INTRODUCTION

How to prevent the long term deterioration of cellulose fibre reinforced board due to water ingress?

Simply stated, cellulose board will generally be more susceptible to problems associated with water absorption than asbestos reinforced boards; and this can be a major problem to manufacturers of boards, as they move to replace asbestos by cellulose fibres (Johnston, 2001). Additives such as silica and other pozzolanic materials can be used to improve early strength build up and improved modulus of rupture. By their very nature of densifying the board, they will also provide some reduction in water uptake. Similarly some common additives, such as acrylics binders and metal soaps, will, as well as providing other properties such as strength and filler wetting, also have some positive benefit in reducing water uptake. BUT regardless of all these modifications, the cellulose boards can still absorb water at a higher rate than asbestos based boards. When boards absorb too much moisture, a myriad of potential problems can occur, such as:

- Increased warping in high rainfall areas or in very tropical (humid) conditions
- Reduced Freeze/thaw resistance
- Potential for white efflorescence salts affecting appearance
- Reduced protection from weathering even when post painted

- Increased unsightly mould and algae growth especially in humid or wet climates
- Reduced strength as the boards lose internal integrity (i.e. interlaminar bond)
- Weakening of fixings and fasteners long term due to shrinkage and cracking

Although still used in many countries, there is a major global shift away from using asbestos by replacing it with much safer fibres such as cellulose fibres. Some companies choose to move gradually away from asbestos by using asbestos/cellulose blends as they grapple with this changeover. Simply stated, if they move 100% to cellulose, then the water resistant properties can be reduced, unless specific precautions are taken. Formulations for FRC boards vary greatly throughout the world. The fibre source, the cement source and the type and amount of siliceous extenders vary dramatically from plant to plant. Additional manufacturers use many different cure processes and post treatments, which all mean that each plant will be different and may require slightly different solutions.

Possible Solution:

Although a relatively new concept; certain Silicone technologies can be used to significantly reduce the water absorption of cellulose reinforced FRC mouldings, sheets and boards. There are many long term examples in non FRC applications of silicones working in a unique way to provide optimized water resistance to building components. Principal amongst these being wet area gypsum boards where the advantages of using silicones as water proofing admixtures is well known and is widely practiced amongst key global manufacturers. In recent years we see many more innovative uses of silicones to reduce water uptake in construction applications. As example SILICONE Technology these days are being used as:

a) Impregnation or sealer additive to protect structural concrete from de-icing salts ingress and freeze-thaw damages.

b) Admixtures in Concrete pavers or concrete blockwork to reduce water uptake and hence reduce efflorescence staining.

c) Additives to improve water/weathering resistance of latex paints such as external acrylics

d) Binders in specialized external acrylic-based renders

e) As pre-priming sealers and also as admixtures for cement based panels such as FRC

Finishing systems for FRC

When FRC siding panels are manufactured, they will either be sold to the end user either ‘raw’, pre-primed or post finished:

- Raw: No post treatment in the factory. All coating takes place on site (or not at all).
- Pre-primed: Many manufacturers prime boards with ‘universal’ primers (typically acrylic based) before they leave the factory. They are then post coated to the desired colour after fixing on site.
- Sealed; Silicone based sealers can be used to protect the boards when raw cement appearance is desired.
- Sealed and pre-primed: Before painting, some manufacturers pre-seal with silicone penetrants before applying the acrylic prime coat, on the basis that the penetrating sealer provides long term protection to the boards
- Completely primed AND topcoated. In this case NO post treatment is required. It is becoming increasingly common for penetrating silicone based sealers to be used as the initial treatment prior to priming and coating.

The primary purpose for coating panels is for aesthetics reasons. As long as a coating is fully integral, then the water absorption of the board will be reduced. Once the coating fails, so does the protection. Coatings provide little or no protection against penetration of moisture at the edges or the rear surface of the board. By pre-sealing boards with silicone penetrants on the front and rear surfaces as well as the edges prior to coating, then far greater long term protection of the boards can be achieved. If treated in this way; the FRC boards and panels have long term protection against water penetration regardless of the life-time and/or quality of the paint treatment used.

Silicones Sealers for FRC:

It is relatively easy to demonstrate, that by presealing the boards with certain silicone based penetrating sealers, the water-uptake of the boards will be reduced. These penetrants work differently to primers. They can penetrate deeply into the matrix forming a barrier deep within the matrix while primers work at or near the surface. It is this deep penetrating effect that provides the long term protection for boards and clearly differentiates their mode of action to paints and paint primers.

While this type of application is the most widely accepted and used practice to date for manufacturers, who recognize the benefits of using silicone based penetrants, there is interest in other properties and methods of use of silicones, which may become more widespread in the future. For instance some manufacturers are looking to penetrating systems that also have the benefits of enhancing the bonding of paint films, and other again are experimenting with silicones being used as paint additives to improve water tightness of coatings.

The most novel of processes being currently investigated is the use of silicones as admixtures in the wet matrix. In this process the silicone will become an integral component of the board as it cures and therefore forms a water barrier throughout the entire board. One advantage of this approach would be that the FRC products would be protected from water absorption from the time they are initially produced and would not depend on a post-treatment or protective coating to provide improved water resistance.

Another useful, and valuable advantage of using an admixture to "build-in" water resistance will be the ability to cut FRC products at the location where they will be installed, and still maintain protection, that otherwise could be lost when using a post-treatment or coating. Extending this logic a bit, the FRC products will also retain protection even if sealers, coatings or edge sealants like caulking are damaged or not applied perfectly

The different application stage of the silicon-based hydrophobic material in the overall FRC production process is illustrated in the Figure 1.

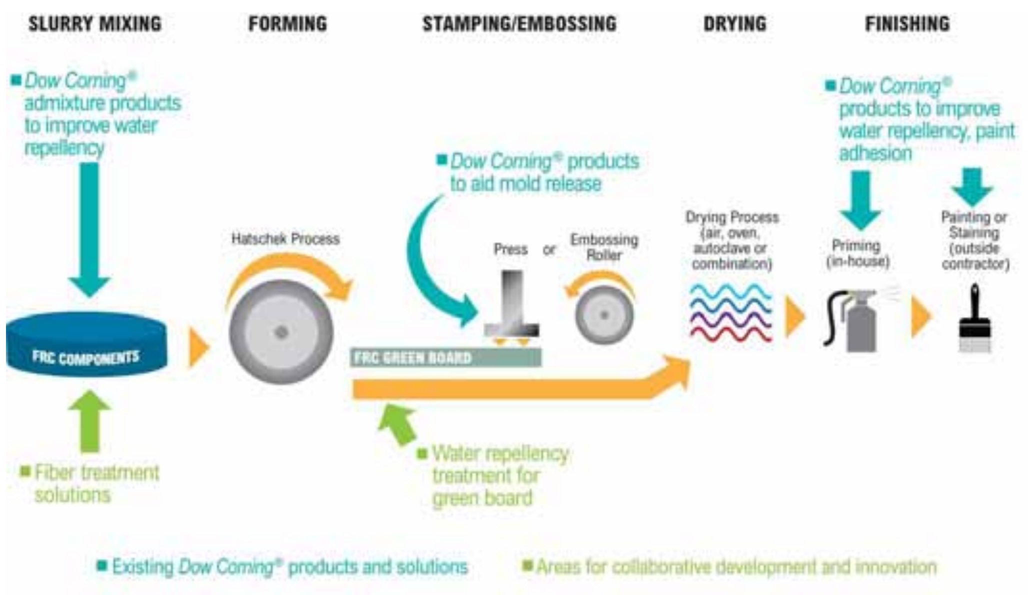

Figure 1 :schematic description of the process for the making of FRC boards which illustrates the different process steps where a silane or silicone sealers can be used. Additional potential other silicon-based additives are also illustrated.

Silane and siloxane.

Siloxane and alkoxysilanes have become a very important class of materials used for water-repellent treatment of masonry, where durability and minimal impact on substrate appearance are important.

Silicone is a generic term describing polymers encompassing a siloxane backbone (based on the repeating unit: Si-O-Si). Polydimethylsiloxanes or PDMS (illustrated in **Figure 2**) are the most common siloxane used worldwide, both in terms of volume and application.

Polydimethylsiloxane are available as linear fluids, cyclics, viscous polymer and even gums depending on their degree of polymerization and cross-linking. Terminated by a silanol group (as in **Figure 2**), they are reactive and can be anchored on appropriate substrates. The siloxane backbone can be partially functionalized with organic group like longer alkyl chain. They will be referred in a generic way as organofunctional siloxane in this document.

Silicones have many interesting properties that make them suitable for a wide range of applications. In the field of hydrophobic treatment, low surface tension, ability to react with cementitious material, improved UV resistance vs. organic polymer and high gas permeability are of most interest.

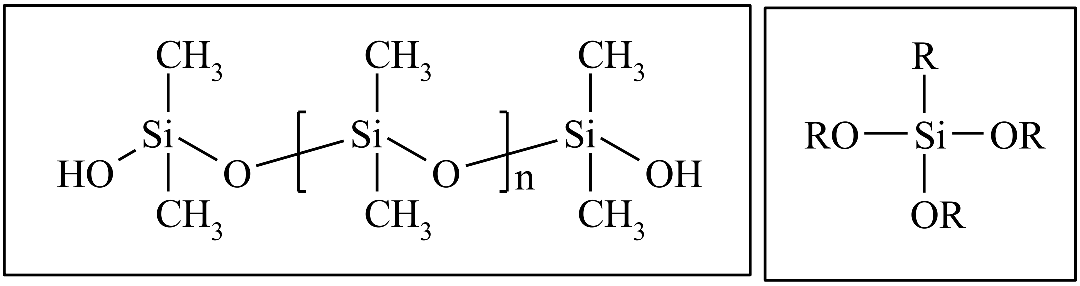

Figure 2 : polydimethyl siloxane and alkyl trialkoxysilane

Silanes are molecules based on one silicon atom which bears four substituents. Alkyl trialkoxy silane (as described in **Figure 2**) show a good reactivity towards construction material (reactions during which alcohol is released as leaving groups). They can be viewed as materials bearing latent silanol groups (Si-OH), yield by hydrolysis, which can bond covalently to masonry materials through condensation reactions with metal hydroxyl moieties. Silanol self-condensation also leads to cross-linking of the treatment, which leads to outstanding durability of water resistance. The R groups (most often, isobutyl or octyl chain) confer the hydrophobic character to the substrate to which the silane is anchored.

Figure 3 illustrates the need for the water–repellent material formulation (hydrophobic active + the delivery system around the hydrophobic active) to be adapted to the type of substrate and the expected incorporation stage. The water repellent active materials have to match the chemistry of the construction material. Cement-based materials are not reacting the same way as gypsum or wood. The chemistry of the water repellent has to be adapted to properly react with the construction material in order to provide the expected protection against water.

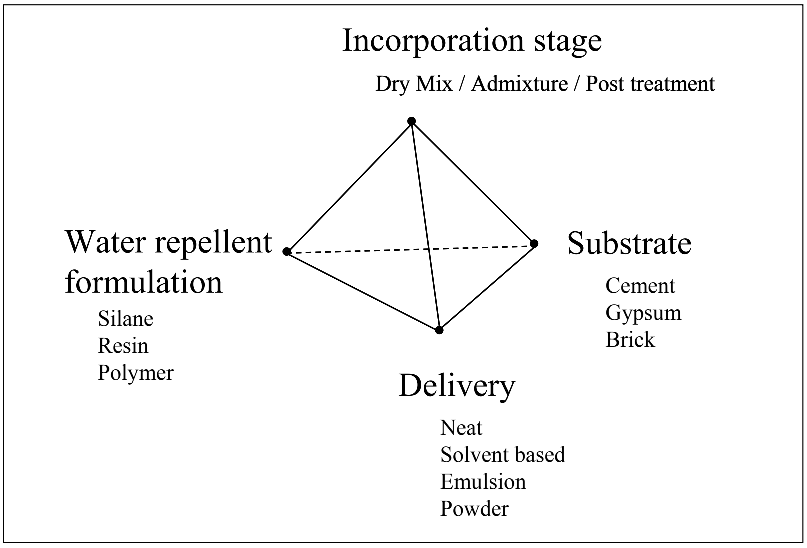

Figure 3 : The "hydrophobic tetrahedron".

The way the water repellent active material is formulated can be described as the *delivery system*. Water repellent can be used "neat" (as such), as solvent based solution, as emulsion or yet as powder. The delivery system has to be adapted to the requirement of the application method. For example, the water repellent can be added in a cement based wet slurry, after the cure of cementitious material in a post-application step (in this case, fibre reinforced cement boards) or even added before the addition of water, in what is called nowadays "dry mix". Water repellent can be also used by coating the cellulose fibres with it to avoid their interaction with the water. Several researches have been carried out on this subject (Telysheva et al., 1999, Bilba and Arsene, 2008). The delivery system (neat hydrophobing material, emulsion or powder) has then to be adapted to fit the stage of application of the water repellent.

The objective of this research is to illustrate the efficiency of silane/silicone water repellent on reducing the water absorption of fiber boards. Different application methods (post-application of neat silane or silane emulsion or addition of admixture) were used which all leads to effective reduction of water uptake by the modified boards. Impact of silicone admixture on the cement hydration process was studied as this application method leads to potential interaction of the silicone with the cement particles before and during the hydration process.

MATERIALS AND METHODS

FRC sample preparation and product application

All the boards were conditioned in a similar manner by drying them in oven at 40°C during 24 hours and let 1 day at Room Temperature before application of any sealers. Solid particles, dust at the surface of the sample were removed by compressed air.

Sealers were applied with a traditional paint brush. FRC samples were weighted before and after each application to calculate loading rate.

Boards tested for the water uptake immersion were coated on all 6 sides.

Treated boards were left at room temperature during 3 days to give enough time for the hydrolysis and condensation reaction to take place.

WATER UPTAKE BY IMMERSION: TEST METHOD

The test procedure included 7 steps:

1- After cure of the sealer, boards were initially weighted.

2- Treated boards were placed in a vat with the rear surface of the treated board down on an absorbent paper and covered with tap water to reach 2 cm coverage of water over the top surface of the treated board.

3- Periodically, samples were weighted at 2, 4, 6, 8 and 24 hours immersion time in water. For best repeatability, as the treated boards were removed from the water, excess water dripped off the surface and the sample was 'pat' dry with absorbent paper towel. As soon as all visible excess water was removed, the treated board was weighted and returned immediately to the water to continue test.

4- After results had been obtained for the 4 (or 5) time slots, results were recorded as water absorption:

% water absorption e.g. %WA (x hours) = (Wx – Wi)/ Wi X 100%

With Wx= Weight after x hours in water, Wi= Initial weight

Uncertainty on the Water Uptake Immersion results was measured to be +/- 0.6%.

ADHESION CROSS-CUT TEST

To test the adhesion of a paint layer on the top of the primer, two coats of acrylic were applied on the esthetic face with a traditional paint brush (sides were also covered in order to avoid discontinuity of the film).

Acryl Lak Indoor/Outdoor Satin WIT001 White manufactured by De Keyn was used in the paint adhesion test.

Two layers were applied at 7 hours interval. Then, the paint cured during 2 days at Room Temperature.

The adhesion cross-cut test was performed according to the ISO2409 standards.

The cross-cut was made on the sample of lines at right angles 1 mm with a special cross cut comb. The cut had to reach the substrate. A piece of tape Scotch 2525 was put on the cross (in the same way as it was made) and a slight pressure with the finger was applied. Then it was pulled off in one firm go. Adhesion was evaluated according to the quantity of coating missing on the squares (0 being perfect: with no coating gone and 5 very bad with all the coating removed).

RESULTS AND DISCUSSION

Benefit of treating FRC boards with silane or siloxane hydrophobic materials is illustrated hereafter. As discussed, there are different ways to apply hydrophobic material and both post application and admixture will be illustrated hereafter.

Sealing of FRC board by post-application

Octyltriethoxysilane was post added on an autoclaved board as a solvent based solution. This insured even application of the silane in these lab conditions. The 6 faces of the boards were coated.

After complete cure of the sealer (3 days), water uptake of the boards was assessed by immerging the treated board and the reference under water. Plot of water uptake (given as % of initial weight) as a function of time is given hereafter. Figure 4 and figure 5 as well clearly show that sealing of the board has a dramatic impact on reducing the amount of water absorption.

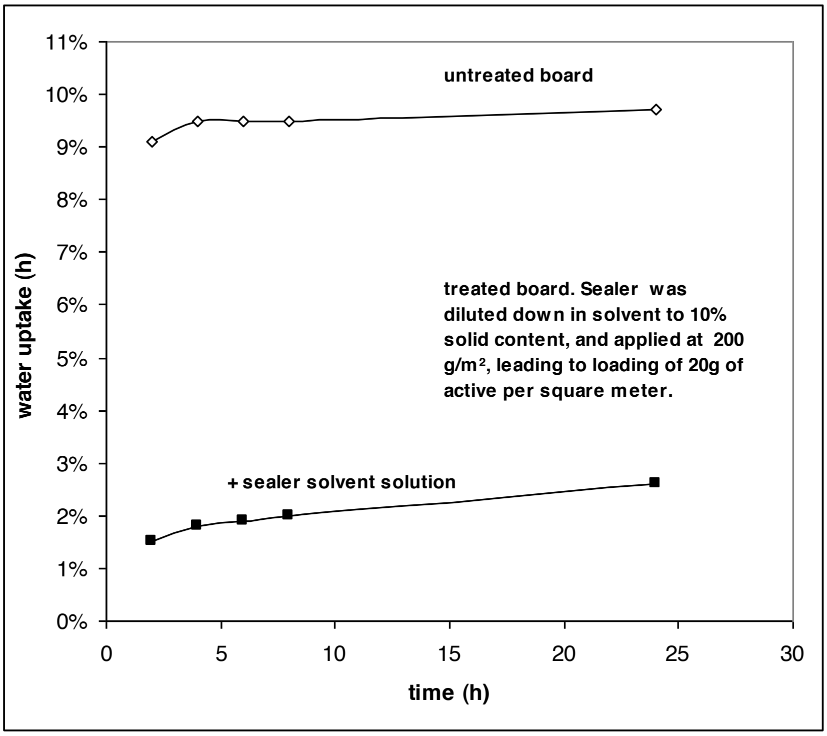

Figure 4 : water uptake (in terms of % of initial weight) of FRC board under immersion. Water uptake of reference board and board treated with a silane-based sealer (delivered as a solvent based solution of octyl triethoxysilane). Application condition given in the graph.

Sealed boards can be used as such but can be painted to provide the desired aesthetic. Paint adhesion cross-cut test were performed after 2 days of drying of the paint at room temperature, following the procedure as described. Although the sealing dramatically reduces the water absorption, it is observed that this preliminary step does not negatively impact paint adhesion (Table 1).

Table 1: Results of paint adhesion testing of un-treated board and board sealed with an octyl triethoxysilane solvent solution (application condition see Figure 4). Ranking given according to adhesion cross-cut test

	Paint adhesion
Reference board	2.5
Sealed board	2.5

In order to provide an environmentally friendly alternative to silane solvent solution, silane emulsion was tested as well as sealer for FRC board. Sealing of the board with the silane emulsion has the same dramatic impact on reducing the amount of water absorbed by the board (same testing procedure, see results in Figure 5).

Adhesion of paint used as a post coat was assessed as well and no detrimental impact of the silane emulsion on the paint adhesion was found.

Admixture

The hydrophobic additive can be also added in the raw material slurry (cement+cellulose fibres). This is expected to lead to treatment of the bulk of the board, which is very beneficial to avoid any edge effect or to avoid poorer hydrophobic treatment when boards are cut on the job site.

In order to evaluate the performance benefits of using silicone admixtures in FRC, a joint study was undertaken in 2007 between Dow Corning Corporation and the University of Sao Paulo in Brazil. One of the main objectives in this study was to find a suitable laboratory method for the incorporation and evaluation of water insoluble organosilicone admixtures. Using this process also enabled the evaluation of performance and physical properties of FRC boards made with and without these additives.

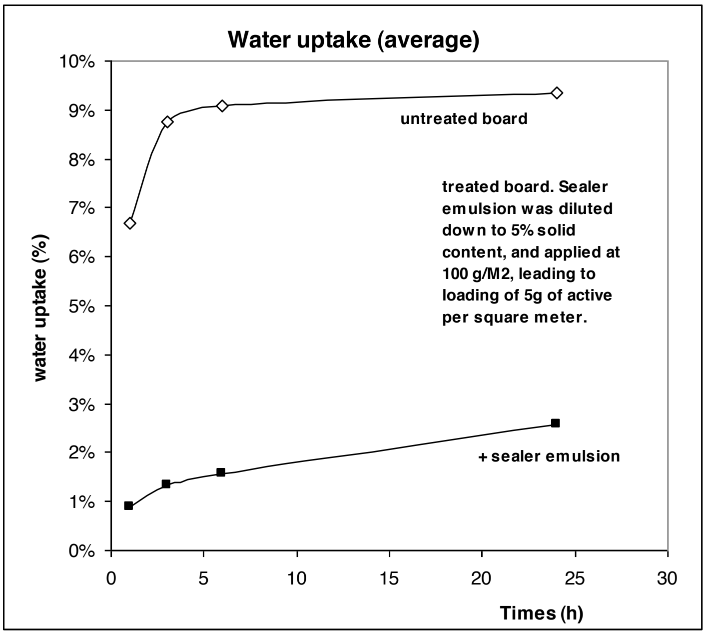

Figure 5 : water uptake (in terms of % of initial weight) of FRC board under immersion. Water uptake of reference board and board treated with a silane-based sealer (delivered as an emulsion of octyltriethoxysilane). Application condition given in the graph.

Boards were prepared in the lab by mixing the different raw materials (cellulose pulp, cement, limestone, silica fume and PVA fibres) with the water insoluble organosiloxane admixture. The wet slurry was filtered, compressed, cured in an oven at 65°C and further cured for 28 days at room temperature. Samples were then cut of the boards for testing of the resistance to water penetration.

Water uptake under immersion as a function of time for the reference and admixtured board are illustrated in **Figure 6** .

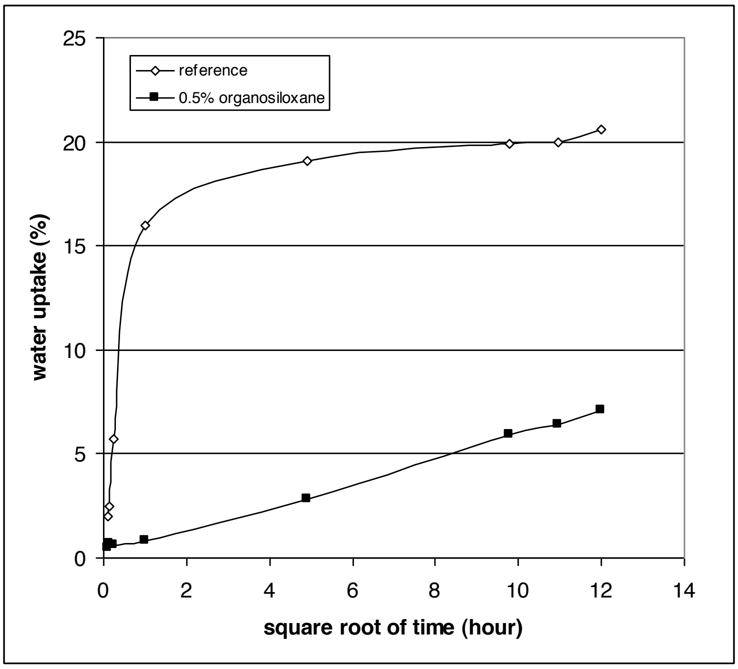

Figure 6 : water uptake (in terms of % of initial weight) of FRC board under immersion as a function of square root of time. Water uptake of reference board and board admixtured with 0.5% of an organosiloxane

Figure 6 illustrates that water resistance, as measured by weight gain during 24 hour immersion, was significantly lowered by using a silicone admixture.

As FRC are composite material, it can be questioned whether the silicone admixture is reacting with the cellulose reinforcement or with the cement matrix. It is also important to assess if silicone admixture has a negative impact on cement hydration and final strength of the cement matrix. Therefore, mortar blocks containing the organosiloxane were prepared and compressive strength were measured.

Effectiveness of hydrophobic admixture in cementitious matrix

The efficiency of the same organofunctional siloxane as hydrophobic admixture was assessed in a simplified system composed of a cement and mortar. 0.5% vs the weight of the dry mortar was added into the slurry and properly mixed in order to insure even distribution of the hydrophobing material into the cement matrix.

Mortar blocks were prepared. by mixing 450 g of CEM I 42.5 R cement, 1350 g of sand, 180 g of water and 0.5% of organosiloxane (vs dry composition weight). Mortar blocks (40x40x160mm) were cured for 28 days and then dried overnight at 40°C before testing.

Dry blocks were weighed and then placed in a water basin partially filled with water. Blocks were placed on rod (to enable free access of water) in a horizontal position. Water level is adjusted to have 3 cm of water above the top of the mortar blocks. The blocks were reweighed after 1, 3, 6 and 72 hours. Results in **Figure 7** are obtained by use of the following equation wherein:-

Percentage Water Pick Up (WPU %)= ((Wwet) - (Wdry))/ (Wdry)x 100

The same dramatic impact of organosiloxane addition on the water absorption is observed in this simplified system (Figure 7).

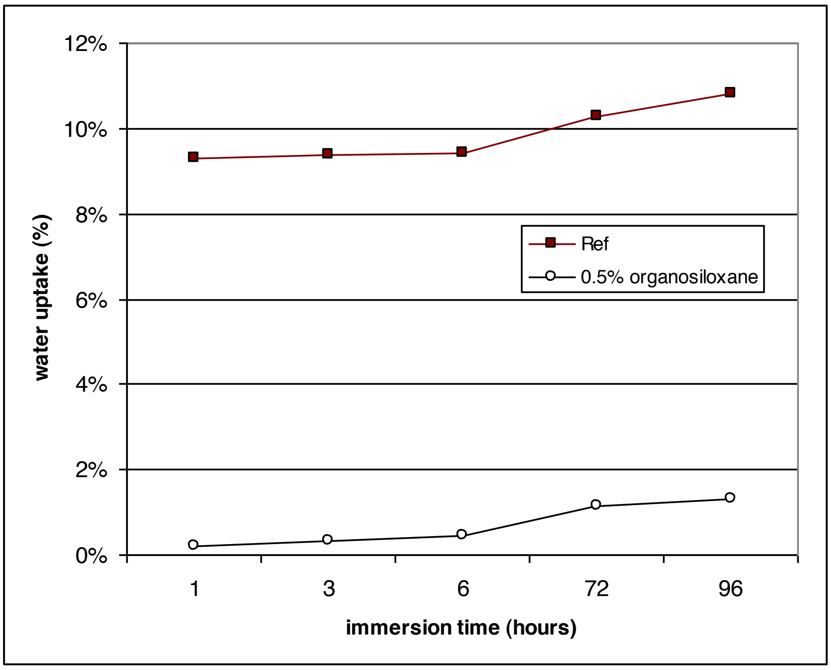

Figure 7 : Rate of water absorption of 196-1 reference mortar and 196-1 mortar admixtured with 0.5% organosiloxane.

Impact of the organosiloxane addition on cement matrix strength was assessed by preparing mortar blocks according to EN 196-1 norm. 450 g of CEM I 42.5 R cement was mixed with 1350 g of normalized DIN sand with 225 g of water and 0.20% and 0.5% of the same organofunctional siloxane. Mortar blocks (40x40x160mm) were cured for 28 days before testing for the compressive strength.

Results displayed in Table 2 show that only moderate impact on physical properties of mortar is observed. This set of experiments strongly suggests that the hydrophobic admixtures interact with the cement matrix of the FRC boards, protecting the composite boards against water ingress.

	Compressive strength after 7 days (MPa)	Compressive strength after 28 days (MPa)
Reference mortar block	32.0	48.9
Block with 0.5% organofunctional siloxane	28.4	43.1

Table 2 : compressive strength of admixtured mortar block prepared according to EN 196-1

Impact of silane or siloxane based admixture on cement hydration reaction.

A more fundamental study was initiated to better understand the impact of silane and siloxane (in this study, polydimethylsiloxane) based admixture on the cement hydration (Spaeth et al., 2008 and Spaeth et al., 2010). Detailed thermo-analyses, isotherm conduction calorimetry (ICC), XRD and SEM investigation demonstrated that the main processes involved in the overall cement hydration are not affected significantly.

Cement hydration reactions are mostly exothermic. Measurements of the heat release by ICC during hydration give an indication of the rate of hydration of cement which consists in five periods. [Odler, 1998, Taylor,1990 Stutzman, 1999, Chen and Odler, 1992]. Our experiments show that some periods of cement hydration are slightly accelerated or delayed depending on type of admixtures which leads to slight shifts in the setting time and workability of materials, while still maintaining final cohesion and mechanical resistance.

Thermal analyses combined with x-ray diffraction analyses were used to better understand the interaction between cement / admixtures and water. The study showed some impact of the silane and siloxane admixture on the growth of $Ca(OH)_2$ cristals. The presence of polymeric molecules at the solid-liquid interface are known to inhibit crystal nucleation and growth of hydrate products [Vikan and Hustnes, 2007, Jolicoeur and Simard, 1998]. Adsorption occurring on a nucleation center may prevent the nucleus from achieving a minimum critical size [Mailvagnum, 1979].

The polymer/cement ratio (P/C) determines the amount of polymer present in the pore solution and at the aggregate's surface. SEM experiment where carried out to study the impact of Si-based admixture on the morphology, size and orientation of hydration products. It was concluded that crystalline products precipitate from the modified pore solution progressively almost as usual. Only the addition of polydimethylsiloxane-based agent seems to encourage the growth of straight, parallel, sheet and hexagonal plate structures of portlandite. The fact that only limited impact on hydration production growth is observed can be rationalised on the basis that the polymer to cement ratio (P/C) is lower than 5%, small which is reported to lead to only tiny polymer bridges on limited number of spots [Beeldens et al.,2005, Knapen and Van Gemert, 2009].

The conclusion of this study is that despite incorporation of Si-based admixture, development and growth of usual hydrates were observed. At the same time, only small changes of physical properties were measured. The structure/microstructure of the modified cement matrix is such that it still provides good mechanical performances.

CONCLUSION

This paper is intended to review the different incorporation methods of silicone or silane sealers during the preparation of FRC boards and the impact of the board treatment on the reduction of water absorption by the treated boards.

Whether post-added on cured boards or incorporated as an admixture into the wet slurry, silicone or silane sealer addition leads to the same dramatic reduction of water absorption. Knowing that water absorption is a source of degradation for cellulose based boards, silicone or silane based sealers prove to be a solution of choice to extend boards lifetime.

Impact of admixture addition on the cement matrix was studied separately which demonstrated that hydrophobic treatment of the cement matrix provides an effective way to reduce water penetration. Cellulose fibre treatment will be studied separately in a next phase, in order to reduce even more the board affinity for water.

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