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Section 3 **Wood protecting chemicals**

Assessment of the biological durability of wood treated with organosilicon compounds

Magdalena Kutnik [(1)], Mathilde Montibus [(1)], Sarah Derocker [(2)], Sabrina Salivati [(2)], Jean-Paul Lecomte [(2)]

(1) FCBA Technological Institute, Biology Laboratory, Allée de Boutaut, BP 227
33028 Bordeaux Cedex, France
(2) Dow Corning Europe S.A., rue Jules Bordet Parc Industriel Zone C, 7180 Seneffe, Belgium

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IRG SECRETARIAT
Box 5609
SE-114 86 Stockholm
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Magdalena Kutnik (1), Mathilde Montibus (1), Sarah Derocker (2), Sabrina Salivati (2), Jean-Paul Lecomte (2)

[1] FCBA Technological Institute, Biology Laboratory, Allée de Boutaut, BP 227

33028 Bordeaux Cedex, France magdalena.kutnik@fcba.fr and mathilde.montibus@fcba.fr

[2] Dow Corning Europe S.A., rue Jules Bordet Parc Industriel Zone C, 7180 Seneffe, Belgium

ABSTRACT

The European wood construction market is in need of environmentally friendly wood-based products whose service life meets the expectations of end users. Non-biocidal silicon-based water repellents, which can be applied to different materials, help minimize their vulnerability/susceptibility to liquid water. Consequently, they have the potential of increasing wood's resistance against decay fungi by reducing their ability to absorb water. Moreover, by filling the pores of wood, they may reduce its vulnerability to wood-boring insects and thus improve its durability. In order to determine their suitability as wood protectors, three silicon-based water repellents were applied at different concentrations to samples of Scots pine, beech and oak. The application was either superficial (dipping) or by deep (vacuum impregnation). Durability tests were then performed in order to determine the resistance of the treated wood samples against moulds, blue stain and basidiomycete decay fungi, as well as against *Reticulitermes* subterranean termites. Additionally, eco-toxicological tests were run in order to select the product which had the least environmental impact. The results demonstrate that the three tested organosilicons exhibit different levels of toxicity and improvement of the resistance of the tested wood species against biological threats. The anti-fungal resistance is lower than that provided by traditional biocidal wood-preservatives, but it is still adequate in applications where short-to-average service-life is acceptable and where maintenance and/or replacement is possible. Termite behaviour and feeding preferences in situations where choosing between treated and untreated wood was possible demonstrated that insects tend to avoid consuming wood specimens impregnated with organosilicons. Wood-based materials with levels of biological resistance which are intermediate between wood's natural (inherent) resistance and that improved with the help of biocides as well as wood-protection products and processes which allow reaching these levels should be appreciated more. They increase the range of possible end-uses for wooden commodities made of European non-durable wood species such as Scots pine and beech both with regard to the material's properties and its expected performance.

Keywords: organosilicons, silicone, wood durability, moulds, blue stain, decay fungi, termites

1. INTRODUCTION

The SILEX project titled "Improving sustainability of construction materials using innovative silicon based treatment" is a Life+ project with a LIFE+11 ENV/BE/1046 designation. Initiated in April 2013, it will continue through August 2016. One of its major objectives is to demonstrate that a new class of silicon-based compounds, characterized by low toxicity compared to traditional biocidal products used for decades, provide silicon-based solutions which can be used in the wood protection sector as multifunctional water repellents which help improve wood's dimensional stability (by reducing moisture absorption), aesthetics, resistance to decay organisms, service life and fire resistance in outdoor applications.

Wood is subject to water infiltration by both liquid and vapour, its high hygroscopicity being one of the major drawbacks of wood-based materials. As the moisture content increases, wood will swell until it reaches its maximum dimension at its fiber-saturation point (about 30% moisture). Rapid dimensional variations resulting from changes in the level of bound water cause the wood to crack and split. These cracks will then allow moisture to penetrate easily and quickly into wood. Depending on the moisture content level and how long the moisture is present as free water, wood is susceptible to biological (mainly fungal) attacks.

Many silicone-based treatments, using various compounds (silico-fluorides, colloidal silicic acid solutions combined with various metal compounds or boric acid) have been tested with regards to their ability to act as preservatives or biocides. Past studies demonstrated that silicon-based water repellents increase the hydrophobicity of wood. Investigating impregnation with silicones to give hydrophobic properties to wood, Tshabalala *et al.* 2003 and Donath *et al.* 2006 focused on organosilicons, such as silanes and siloxanes. However, most studies (Hill *et al*. 2004; Weigenand *et al*. 2007; De Vetter *et al*. 2009) have shown that treatments of wood with organosilicons are effective only when they are applied at very high concentrations.

The aim of this project is to investigate the potential of silicon-based water repellents to reduce moisture uptake when used outdoors and to improve the natural resistance of wood against biological agents such as fungi and insects. The long-term objective is to find substitutes for biocides applied to wood to obtain the desired levels of protection for the expected service life or reduce the quantity needed to obtain such levels. In fact, wood protective chemicals sometimes "over-protect" wood as determining accurately their effectiveness over time is based on laboratory tests combined with artificial ageing and field tests, whose duration is much shorter than the expected service life of wooden commodities. Regarding wooden commodities which can withstand degradation for short-to-average periods of time and those whose durability can be extended through maintenance and replacement of parts in case of failure (such as fences, furniture, structures providing protection against weather, wooden elements used in house building, etc.), we believe that priority should be given to technical solutions aiming at moderate but acceptable levels of protection such as wood of average natural durability or of wood-based materials whose durability has been enhanced by means of products and/or processes with a low environmental impact.

At the initial stages of the project, various silicon-based hydrophobers were applied, using different application methods (dipping, brushing, and vacuum impregnation) to test their ability to penetrate the matrix of different wood species (beech, oak and Scots pine) and reduce the water uptake of samples subsequently submerged into water. The first results, reported in Malassenet et al. 2014, Van den Bulcke et al. 2014, Lecomte et al. 2015, demonstrated that treatments with selected organosilicon compounds minimized wood's water absorption without affecting the rate of water desorption.

In this study, we report the environmental profile and the effectiveness of three silicon-based products applied to improve the natural resistance of wood against biological degradation caused by disfiguring fungi (blue stain and moulds), decay fungi and subterranean termites.

2. EXPERIMENTAL METHODS

2.1. Organosilicon products investigated in the study

Three silicon-based products ("Si-products"), presented in Table 1, were investigated in this study. They were used either in concentrated form (for the eco-toxicological tests) or diluted with water to reach an active content of 5% in the dilute solutions (for biological tests).

Table 1: Silicone-based solutions used in this study.

Product	Description
1	Emulsion of silane, siloxane and resin
2	Water soluble silane
3	Cationic emulsion of organofunctional siloxane

Diluted products were applied to samples of beech, oak and Scots pine by dipping or by vacuum impregnation, the samples being then submitted to different laboratory tests in order to determine their characteristics with regard to biological durability.

Chemical analyses were also performed in order to determine the uptake and the distribution of Si compounds in the treated samples (not shown here).

2.2. Determination of the efficacy of organosilicons against blue stain and moulds

Scots pine sapwood, oak sapwood and beech wood without visible defects were selected and cut into samples of the following dimensions: 110 (±0.5) mm x 40 (±0.5) mm x 10 (±0.5) mm (measured at 12% relative humidity [RH]). The samples were then dipped in the tested solutions (5% of active content) and held submerged for 5 minutes with tweezers. The Si-solutions were expected to penetrate into the wood samples only by capillarity as no pressure was applied. Once removed from the Si-solutions, the samples were allowed to drain on a grid in climatic chambers at 20 (± 2) °C with air RH of 70 (± 5) % prior to testing.

2.2.1 Testing under laboratory conditions (in vitro tests)

Samples were inoculated either with a conidia suspension of blue stain fungi or a conidia suspension of moulds. For each condition, 4 replicates were tested. Samples were then incubated for 6 weeks at 22°C and 70% of relative humidity to allow fungi development. Fungi growth and intensity of development were then assessed according to the EN 16492 rating scheme as presented in Table 2.

Table 2: The rating scheme used to evaluate the development of mould growth on the surface of the treated wood specimens

Rating	Percentage area of growth	Intensity of visible growth
0	No growth on the surface	Unchanged
1	Up to 10% growth on the surface	Very slight
2	Between 10 and 30% growth on the surface	Slight
3	Between 30 and 50% growth on the surface	Moderate
4	More than 50% growth on the surface	Considerable
5	-	Strongly marked

2.2.2 Testing under conditions of natural exposure

In April 2015, treated samples were placed outdoors at FCBA premises located in Bordeaux (see Figure 1) and remained exposed to the elements for one year. For each condition (wood species/Si-based solution), 3 replicates were tested. Ratings meant to assess fungal growth and intensity of development were performed on wood specimens after 3 and 6 months of outdoor exposure, according to EN 16492 as presented in Table 2.

Figure 1: Specimens treated with the three silicon-based solutions and exposed outdoor.

2.3. Determination of the efficacy of organosilicons against decay fungi under laboratory conditions

Previous tests carried out according to the EN 113 protocol were found to be inconclusive. Negative mass losses were recorded in the treated samples, which meant that they were heavier after being exposed to decay fungi than before. These observations indicated that the test protocol as such was not suitable for Si-based solutions and had to be modified by adding control samples to allow for monitoring both the moisture content and mass variation.

Consequently, the resistance of treated wood was determined by setting up a new test protocol, adapted from two standards, EN 113 (used to determine the efficacy of wood preservatives against Basidiomycetes) and CEN/TS 15083-1 (used to determine wood's natural durability). This test protocol is an internal FCBA method and is referenced as FCBA-BIO-M-014.

Samples of Scots pine and beech of 50 mm x 25 mm x 15 mm were introduced in a crystalliser placed under a vacuum vessel. A vacuum of 7 mbars was applied for 15 minutes. Si-based solutions at 5% of active content were added under vacuum. Air was then released into the vacuum vessel until atmospheric pressure was reached. The samples were kept in the crystalliser for 2 hours. Next, they were placed on a grid for 2 weeks to allow drying.

Treated wooden blocks and untreated control specimens were exposed to two wood destroying Basiodiomycete fungi, *Coniophora puteana* and *Coriolus versicolor*. After 16 weeks of exposure in a climatic chamber under constant climatic parameters (22°C, 70% RH), the specimens were cleaned and dried and their mass loss due to fungal degradation was determined.

2.4. Determination of the efficacy of organosilicons against subterranean termites under laboratory conditions

Samples of Scots pine and beech of 50 mm x 25 mm x 15 mm were treated with the three organosilicon products by vacuum impregnation (7 mbars vaccum applied for 15 minutes) and conditioned at 65% RH and 20 °C prior to testing. Treated and untreated specimens were then exposed to groups of European subterranean termites *Reticulitermes flavipes*.

Two series of parallel tests were performed:

- “force-feeding” tests, based on the test protocol described in the EN 117 standard

and

- “choice” tests, based on an adaptation of the EN 117 test protocol

In the “force-feeding” tests, ten samples of treated wood and three control specimens (untreated Scots pine sapwood) were individually placed in test devices (glass containers filled with sand) and exposed to subterranean termites. In these tests, wood impregnated with organosilicons was the only source of food given to termites.

In the “choice” tests, one sample of untreated Scots pine (reference) and one sample treated with Si-based solution were simultaneously exposed to subterranean termites in the same test container.

The samples were exposed to termite attack for eight weeks in a climatic chamber with constant climatic parameters (27°C, 75% RH). At the end of the test, the survival rate of termites and the degree of attack recorded in wood specimens were evaluated and classified according to the rating scale shown in Table 3. In addition to visual rating, each sample was oven dried at 103 °C to a constant mass before and after exposure to termites in order to determine the amount of wood consumed by termites.

Table 3: Rating scale used for visual assessment of termite attack according to EN 117

Rating	Description
0	no attack
1	attempted attack: i. superficial erosion of insufficient depth to be measured on an unlimited area of the test specimen; or ii. attack to a depth of 0,5 mm provided that this is restricted to an area or areas not more than 30 mm2 in total; or iii. combination of i) and ii)
2	slight attack: i. erosion of 1 mm in depth limited to not more than 1/10 of the surface area of the test specimen; or ii. single tunnelling to a depth of up to 3 mm; or iii. combination of i) and ii)
3	average attack: i. erosion of < 1 mm in depth over more than 1/10 of the surface area of the test specimen; or ii. erosion of > 1 mm to < 3 mm in depth limited to not more than 1/10 of the surface area of the test specimen; or iii. isolated tunnelling of a depth > 3 mm not enlarging to form cavities; or iv. any combination of i), ii) or iii)
4	strong attack: i. erosion of > 1 mm to < 3 mm in depth of more than 1/10 of the surface area of the test specimen, or ii. tunnelling penetrating to a depth > 3 mm and enlarging to form a cavity in the body of the test specimen, or iii. combination of i) and ii)

2.5. Assessment of the environmental impact of the tested formulations

The effects of the investigated organosilicons on aquatic organisms were examined in carefully controlled studies in compliance with the recent EC directive and OECD guidelines. Laboratory studies were conducted with algae and daphnids using pure products. Additionally, in order to get a better understanding of the possible environmental impact of wood treated with organosilicones, wood samples were leached and leachates were put in contact with daphnids.

2.4.1 Growth inhibition tests on algae

Growth inhibition tests were performed according to EC OECD 201 guidelines. Progress in growth was measured, with the number of cells, the growth rate, the yield and the biomass as the observation parameters.

For each pure product, three concentrations (1, 10 and 100 mg/L) were prepared in an algae test medium. The exponentially-growing cultures of *Pseudokirchneriella subcapitata* were then exposed to these concentrations. The algae growth inhibition in relation to the control was evaluated over a period of 72 hours. Concentrations inhibiting 50% of the average growth rate and of the average yield rate were evaluated and linked to the CLP (i.e. Classification, Labelling, Packaging) criteria classification.

2.4.2 Accute immobilization test on daphnids

The tests were performed according to the EC OECD 202 guidelines. For each pure product, three concentrations (1, 10 and 100 mg/L) were prepared, and 10 mL of each solution were incubated with 5 daphnids *Daphnia magna*. Moreover, the effect of leachates obtained from wood specimens treated with the 3 products at 5% of active content and subjected to several rainfall simulation events (effect of leachates after 1,3,5,8,10,12,15,17 and 19 days of rainfall events) was assessed.

The tests were performed in the dark between 18 and 22°C. Observations of the daphnids' mobility were recorded after 24 and 48 hours. Concentrations immobilizing 50% of the daphnids exposed were evaluated after 24 and 48 hours and linked to the CLP criteria classification.

3. RESULTS AND DISCUSSION

3.1. Protection against blue stain and moulds

3.1.1 In vitro tests

Treated samples were inoculated either with a conidia suspension of blue stain fungi or a conidia suspension of moulds. For each condition, 4 replicates were tested. Samples were then incubated for 6 weeks at 22°C and 70% of relative humidity to allow fungi development. Fungi growth and intensity of development were then assessed (according to Table 2).

The results obtained for blue stain growth are presented in Table 4, the ones obtained for mould growth in Table 5.

Table 4: Ratings for blue stain growth on test wood specimens, after *in vitro* experiments.

Wood species	Product	Retention (kg/m^3)	Percentage area of growth	Intensity of visible growth
Scots pine	Cationic emulsion	52.44	1.75	2
	Emulsion of silane	51.14	3.25	3.25
	Water soluble silane	45.63	2.75	3
	untreated	-	2.5	3.5
Beech	Cationic emulsion	61.02	2.25	3.25
	Emulsion of silane	47.84	4	5
	Water soluble silane	45.57	3.5	5
	untreated	-	4	5
Oak	Cationic emulsion	51.65	3	4.5
	Emulsion of silane	33.81	4	5
	Water soluble silane	36.70	3.75	5
	untreated	-	4	5

Table 5: Ratings for mould growth on test wood specimens, after *in vitro* experiments.

Wood species	Product	Retention (kg/m^3)	Percentage area of growth	Intensity of visible growth
Scots pine	Cationic emulsion	50.97	1.25	0.75
	Emulsion of silane	46.99	3.5	2
	Water soluble silane	47.95	2.25	1.5
	untreated	-	4	2
Beech	Cationic emulsion	63.86	2	3
	Emulsion of silane	61.25	4	3
	Water soluble silane	46.42	4	2.75
	Untreated	-	4	3
Oak	Cationic emulsion	43.64	4	3.5
	Emulsion of silane	30.17	4	5
	Water soluble silane	30.80	4	3.5
	untreated	-	4	4.75

For blue stain (Table 4), ratings recorded on treated samples ranged between 1.75 (10 to 30% of growth) and 4 (more than 50% of growth) for the percentage area of fungal growth on wood's surface and between 2 (slight) and 5 (strongly marked) for the intensity of fungal growth.

For mould growth (Table 5), ratings recorded on treated samples ranged between 1.25 (10 to 30% of growth) to 4 (more than 50% of growth) for the percentage area of fungal growth on wood's surface and between 0.75 (very slight) and 5 (strongly marked) for the intensity of fungal growth.

Results showed that when applied to all three species, but especially Scots pine and beech, the cationic emulsion prevents blue stain growth to some degree and limits its intensity when compared to untreated wood (1.75 and 2.25 when treated, 2.5 and 4 for control). Interestingly, in all three cases the retention of the cationic emulsion in the wood was higher than the retention of the two other products.

The cationic emulsion and water soluble silane were the most efficient in reducing mould growth on wood's surface, while the emulsion of silane showed close to no effect. The cationic emulsion reduced the percentage of mould growth on Scots pine and beech (1.25 and 2) as well as the intensity of growth on Scots pine and oak sapwood (0.75 and 3.5), compared to untreated wood. Water soluble silane reduced the percentage of mould growth on Scots pine and slightly lowered the intensity of mould growth on the three wood species (see Figure 2).

Figure 2: Blue stain growth on beech samples treated with water soluble silane (on the left) and untreated (on the right).

3.1.2 Outdoor exposure

The results obtained for blue stain fungi development after 3 and 6 months of natural weathering in Bordeaux are presented in Table 6. Ratings were performed only for blue stain fungi growth as no moulds were observable on the wood specimens' surface after 6 months of exposure.

Table 6: Ratings for blue stain growth on test wood specimens, after 3 and 6 months of outdoor exposure.

Wood species	Product	Retention (kg/m^3)	After 3 months		After 6 months	
			Percentage area of growth	Intensity of visible growth	Percentage area of growth	Intensity of visible growth
Scots pine	Cationic emulsion	54.70	4	4	4	4
	Emulsion of silane	60.98	3.7	4	4	4
	Water soluble silane	55.53	3.7	3.3	4	3.3
	untreated	-	4	3.3	4	4
Beech	Cationic emulsion	49.02	4	5	4	5
	Emulsion of silane	52.42	4	5	4	5
	Water soluble silane	50.30	4	5	4	5
	untreated	-	4	5	4	5
Oak	Cationic emulsion	38.33	4	3.7	4	4.3
	Emulsion of silane	37.20	4	5	4	5
	Water soluble silane	34.09	4	5	4	5
	untreated	-	4	4.7	4	5

Ratings recorded on treated samples ranged between 3.7 (between 30 and 50% of growth) and 4 (more than 50% of growth) for the area of fungal growth and between 3.3 (moderate) and 5 (strongly marked) for the intensity of fungal growth.

The observations recorded after 3 and 6 months of exposure demonstrated that the treatments did not enhance the performance of the treated wood specimens irrespective of the considered treatment or wood species. However, the cationic emulsion slightly reduced the intensity of visible growth on oak sapwood, both after 3 and 6 months of exposure (4.3 for treated, 5 for control).

Thus, based on the *in-vitro* tests and the outdoor exposure we may assume that the cationic emulsion seems to have the potential for delaying and lowering the appearance of blue stain growth on wood. Higher concentrations or retention rates of the cationic emulsion should be tested in the future in order to confirm this hypothesis.

3.2. Resistance against basidiomycetes fungi

Resistance against decay fungi was evaluated by assessing mass losses of the test specimens after 16 weeks of exposure. Ten replicates per condition were tested. Results are presented in Table 7.

Table 7: Mass losses recorded on treated and control samples exposed to *Coniophora puteana* and *Coriolus versicolor*. MC = moisture content.

Wood species	Si-based product	*Coniophora puteana*			*Coriolus versicolor*		
		Mean retention (kg/m^3)	Mean final MC (%)	Median mass loss (% m/m)	Mean retention (kg/m^3)	Mean final MC (%)	Median mass loss (% m/m)
Scots pine	Water soluble silane	746.7	33.4	1.36	708.0	32.9	1.76
	Emulsion of silane	758.8	32.5	16.00	743.0	32.2	5.01
	Cationic emulsion	744.8	32.1	0.92	744.6	31.3	1.49
	untreated	**-**	**47.5**	**32.04**	**-**	**38.0**	**12.24**
Beech	Water soluble silane	681.2	38.6	14.30	683.5	33.5	6.57
	Emulsion of silane	680.4	40.9	40.70	683.6	32.2	7.54
	Cationic emulsion	689.6	38.4	8.71	690.7	32.5	2.32
	untreated	**-**	**47.1**	**41.6**	**-**	**34.7**	**15.2**

The fungus *Coniophora puteana* was more aggressive on beech wood compared to Scots pine, both in the case of treated and untreated wood (mass losses ML Scots pine < ML beech). The same behaviour was observed with *Coriolus versicolor*; however, because the virulence (i.e. mass losses on untreated) of this species was lower than expected (<20% ML on untreated specimens), the results have to be interpreted cautiously.
All three Si-based products lowered fungal degradation and reduced moisture uptake during the experimentation, as the final moisture content was lower on the treated samples compared to the untreated ones. However, differences were observed between products in terms of their ability to improve the resistance of wood to fungal degradation.
Median mass losses estimated for Scots pine samples impregnated with water soluble silane and the cationic emulsion were very low with both fungal species (<2% ML). Mass losses estimated for beech samples treated with these two products were higher than the ones recorded on Scots pine, but still much lower than was the case with the untreated controls. Because retention was lower in beech (between 680 and 690 kg/m^3) than in Scots pine (between 700 and 760 kg/m^3), we may assume that a dose-response effect may occur, which would explain why Scots pine is better protected by both Si-based products.
The emulsion of silane did not protect either Scots pine or beech from fungal degradation as high mass losses were recorded (up to 40.7% on beech with *C. puteana*, which is not different from what was observed in untreated wood).

3.3. Resistance against subterranean termites

At the end of the test, the survival rate of termites, the degradation rate of wood samples (ranging from 0 = no attack to 4 = strong attack according to the scale presented in Table 3) and the mean amount of wood consumed were evaluated for "forced-feeding" and "choice" tests. The results are presented in Table 8.
Retention rates measured for wood impregnated with organosilicons are indicated for your information in an attempt to correlate retention with the observed degradations. The retention rates of the emulsion of silane and the cationic emulsion were found to be higher on Scots pine than on beech.

Table 8: Mean retention of organosilicon products in impregnated wood, termite survival rates, quantity of wood consumed and degradation ratings recorded at the end of the tests.

product	wood species	mean retention (kg/m^3)	survival %		mean amount of wood consumed (g)		number of samples per rating (off of 5 replicates)									
							forced-feeding					choice test				
			forced-feeding	choice test	forced-feeding	choice test	0	1	2	3	4	0	1	2	3	4
Water soluble silane	beech	646.9	10.8	34.5	2.60	1.94	-	-	-	-	5	-	5	-	-	
	Scots pine	629.7	36.9	71.6	2.35	1.65	-	-	-	-	5	-	4	1	-	
Emulsion of silane	beech	647.6	0.2	42.8	2.67	2.17	-	-	-	-	5	-	1	2	2	
	Scots pine	717.7	0.0	59.8	1.59	1.68	-	-	-	5	-	1	1	-	3	
Cationic emulsion	beech	644.0	0.0	31.6	2.35	1.84	-	-	1	4	-	5	-	-	-	-
	Scots pine	703.1	0.0	56.9	1.66	1.43	-	1	3	1	-	3	-	1	1	-
untreated	beech	-	41.1				-	-	-	1	8					
	Scots pine	-	65.9				-	-	-	-	9					

3.3.1 Termite mortality

In the "forced-feeding" tests, termite mortality was total with the emulsion of silane and the cationic emulsion (survival% = 0.2 or 0), suggesting a possible toxicity of these two products when used as the sole source of food by termites. Water soluble silane caused lower reduction of termites than the two other products as living specimens were found at the end of the test (10.8% of survival on beech and 36.9% on Scots pine). However, survival was heavily reduced compared to the controls (41.1% of survival on beech and 65.9% on Scots pine).

In the "choice" test, termites which fed upon both treated and untreated wood survived to an extent equivalent to the controls, which indicates that the ingestion of wood impregnated with organosilicons is not lethal as such for termites, provided that other sources of food are available.

3.3.2 Termite degradation of organosilicon-treated wood

In situations where wood treated with organosilicons was the only available source of food, termites were able to degrade wood samples to an extent ranging from attempted attack (rating 1) to strong attack (rating 4). The emulsion of silane and water soluble silane did not prevent termite attack, as all samples were rated 3 or 4 (equivalent to controls). However, with the cationic emulsion, Scots pine blocks, impregnated with higher levels than blocks of beech, were slightly less degraded (ratings 4 recorded on Scots pine and ratings 3 recorded on beech), which may suggest a dose-response effect. Samples treated with the cationic emulsion were less degraded, Scots pine samples being the less attacked ones (mean rating of 2, slight attack) while beech samples, treated at a lowest retention rate, were moderately degraded (mainly ratings 3 recorded). This observation suggests that a dose-response effect may exist for this product as well.

Degradation rates recorded on treated samples in situations where termites had the choice between two sources of food clearly demonstrated that termites preferentially tend to feed on untreated wood. The total amount of treated wood consumed by termites was lower in "choice" test situations compared to the "forced-feeding" for the majority of tested parameters. The consumption rate was found to be equivalent only for the emulsion of silane and the cationic emulsion when impregnated into Scots pine wood, which can be explained by the smaller extent of attack recorded (lowest ratings).

4.1 Ecotoxicological tests

The data obtained in the two tests were translated into the CLP classification presented in Table 9.

Table 9: CLP classification of the three tested products.

Product	Water soluble silane	Emulsion of silane	Cationic emulsion
Classification	Chronic toxicity Category 2	Chronic toxicity Category 2	Acute toxicity Category 1
GHS pictogram			
Signal word	No signal word	No signal word	Warning
Hazard statement	H411 Toxic to aquatic life with long lasting effects	H411 Toxic to aquatic life with long lasting effects	H400 Very toxic to aquatic life

The obtained levels of classification demonstrated that among the three tested pure products the cationic emulsion is the less environmentally friendly one, which is consistent with the results of the biological tests, suggesting that it may have a slight biocidal effect on the tested organisms.

However, the tests performed on leachates originating from wood treated with organo-silicones and exposed to ageing procedures simulating rainfall events (lasting from 1 to 19 days) demonstrated that leachates, and thus treated wood, have a very low impact. For the water soluble silane and the emulsion of silane, the leachates showed no impact on daphnids. For the cationic emulsion, leachates had a slight impact on daphnids after 1 day and 3 days of leaching, then no effect is observed.

4. CONCLUSIONS

Protection against wood damaging fungi

All three organosilicons tested in this study failed in providing long-term protection of wood against blue stain in real-use situation (outdoor exposure). However, the results obtained for *in vitro* experiments demonstrated that the products had an effect on the development of moulds and blue stain fungi, slightly reducing their growth. Further investigations are needed in order to determine if the use of higher concentrations can be considered, ideally by focusing on the potential of the cationic emulsion.

The tests aiming at determining protective efficacy against basidiomycete decay fungi were the most conclusive. Among the three tested products, the cationic emulsion and water soluble silane significantly reduced mass loss caused by fungal degradation compared to the untreated controls. Because the mode of action of the two products is not fully understood at this stage of the study (water repellency limiting moisture uptake, repellent/toxic effect of the product itself), it is hard to properly select the tools classifying the obtained level of resistance. By analogy to the classification of wood's natural durability based on the standards CEN/TS 15083-1 and EN 350-1 (durability class DC from "1 – Resistant" to "5 – Not resistant"), the median mass losses (ML) obtained with *C. puteana* (the fungus causing the strongest damage) on wood samples impregnated with the cationic emulsion would allow for classifying treated Scots pine as Durability class 1 (median ML <5%) and treated beech as Durability class 2 (5% <median ML < 10%). In the case of water soluble silane, Durability class 1 classification could be assigned to Scots pine and Durability class 3 classification (10% <median ML < 15%) to beech. However, the better protection given to Scots pine may be due to a higher retention of the product in this wood species and could possibly be improved.

In conclusion, water soluble silane and the cationic emulsion were both found to improve the resistance against fungal decay of Scots pine and beech to an extent comparable to the natural durability of European oak (Natural durability class ranging from 1 to 3, Brischke et al. 2013). Further artificial weathering tests are required in order to evaluate the suitability of the two organosilicons as wood protection products for outdoor above-ground use.

Protection against subterranean termites

In artificial situations of forced feeding, termites appeared to attack wood impregnated with organosilicons out of necessity, but could not sustain themselves and died after a few weeks. In situations of choice between different sources of food, which are more representative of real-life situations encountered by subterranean termites, they tended to avoid organosilicon-treated wood.

Among the three tested products, water soluble silane and the cationic emulsion can be considered as good choices among products protecting wood from termite attack. Water soluble silane is especially interesting if priority is given to products exhibiting a non-biocidal effect. Treatment with this product resulted in high damage of wood but low mortality in the "force feeding" test, which means that the product exhibits low or no toxicity. This observation is also supported by the ecotoxicological tests, which demonstrated that water soluble silane has a better environmental profile than the cationic emulsion. By contrast, little damage and low mortality were recorded in the "choice" tests, demonstrating that termites were poorly attracted by this product, which in turn resulted in indirect protection of the treated samples. On the other hand, the cationic emulsion could be regarded as a good choice if a biocidal effect is acceptable, as treatment with this product resulted in low damage of wood and high mortality of termites for both test protocols.

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