

1 Title: Preventive action of organosilicon treatments against disfigurement of wood under  
2 laboratory and outdoor conditions

3

4 Cite as: De Vetter, L., Van den Bulcke, J., De Windt, I., Stevens, M., Van Acker, J. (2009).  
5 Preventive action of organosilicon treatments against superficial attack and discolouration of  
6 wooden surfaces under laboratory and outdoor conditions. International Biodeterioration &  
7 Biodegradation. In press. DOI: 10.1016/j.ibiod.2009.05.010.

8

9 Liesbeth De Vetter\*, Jan Van den Bulcke, Imke De Windt, Marc Stevens and Joris Van Acker  
10 Ghent University, Faculty of Bioscience Engineering, Laboratory of Wood Technology  
11 Coupure Links 653  
12 B-9000 Gent, Belgium

13

14 \* Corresponding author.

15 Liesbeth De Vetter, Ghent University, Faculty of Bioscience Engineering, Laboratory of  
16 Wood Technology

17 Coupure Links 653, B-9000 Gent, Belgium

18 Tel.: + 32-92646125

19 Fax.: + 32-92646233

20 Email: [liesbeth.devetter@ugent.be](mailto:liesbeth.devetter@ugent.be)

21

## 22 **Scientific relevance**

23 Several authors state that organosilicons have potential to be used for treatment of wood  
24 under use class three applications, whether or not as part of more complex formulations.  
25 Most studies focus mainly on durability and stability of the obtained wood product, although  
26 also other parameters are important, especially when for example cladding is the final  
27 product. Moreover often only laboratory experiments are performed, while it is already  
28 demonstrated that real life exposure studies are indispensable to evaluate the treated wood  
29 properly. This research therefore examines the colour and fungal disfigurement of treated  
30 wood both under laboratory and outdoor conditions. Organosilicon treatment of wood is no  
31 real wood preservation, nor wood modification, nor the application of a surface coating. This  
32 study therefore tries to gain new insights in how to properly evaluate blue stain attack of  
33 organosilicon treated wood under laboratory conditions. Furthermore colour and fungal  
34 disfigurement of treated wood exposed outside are investigated. This research can therefore  
35 be regarded as a completion of existing work on organosilicon treated wood and tries to fill  
36 some of the gaps concerning the general appearance of organosilicon treated wood  
37 exposed outdoors.

38

39

## 40 1. Introduction

41 Wood is a building material that, due to its diversity, can be applied in a broad range of  
42 applications. Depending on the type of application specific demands of the wood are  
43 required ranging from architectural or technical specifications to customer-driven demands.  
44 For most customers appearance is the decisive factor in their choice between wood and  
45 synthetic, metal or mineral alternatives in favour of wood. Measuring customer preferences  
46 is however not straightforward. For outdoor wood applications fungal disfigurement and  
47 colour are two main factors determining the appearance (Salcã and Fotin 2007).

48 Organosilicons have proven effectiveness as hydrophobing agents in for example the textile,  
49 paper and building industry (Rochow 1987; Hager 1995; Lukowsky and Peek 1997; Roos et  
50 al. 2008). Their suitability to protect wood was suggested and aspects like durability,  
51 moisture stability etc. have been investigated (Hager 1995; Tshabalala et al. 2003; Mai and  
52 Militz 2004; De Vetter and Van Acker 2005). However, most studies found that treatment of  
53 wood with organosilicons can only lead to a significant improvement of the investigated  
54 property when applied at (very) high concentrations (Hill et al. 2004; Weigenand et al. 2007;  
55 De Vetter et al. 2009a). Treatments at lower, economically feasible concentrations lead to  
56 only modest improvements of the wood properties (Goethals and Stevens 1994; Mai et al.  
57 2005; De Vetter et al. 2009b). Nevertheless, most of these authors conclude that wood  
58 treatment with an organosilicon may contribute significantly to prolong the service life of the  
59 wooden element when applied as part of a more complex (preservative containing)  
60 formulation for use class three applications like exterior cladding (Mai et al. 2005; Donath et  
61 al. 2006; De Vetter et al. 2009a; b).

62 It was therefore the purpose of this study to evaluate both fungal disfigurement and colour  
63 change of organosilicon treated wood under use class three applications (EN 335-1 2006).  
64 Therefore a broad range of different organosilicons was used as test group. For a subgroup  
65 of specimens organosilicons were combined with biocides. Depending on the test set-up  
66 they were applied using dipping or vacuum impregnation. Both laboratory and outdoor field  
67 tests were performed investigating the performance of the applied systems under the  
68 specific circumstances.

69

## 70 2. Materials and methods

71

### 72 2.1. Products

73

74 Both organosilicon solutions as such, as well as combinations of an organosilicon with a  
75 biocide were used to treat the specimens. The organosilicons cover both solvent-based and  
76 water-based systems (De Vetter et al. 2009a; b). Basically two 100 % w/w active  
77 (conc\_active\_ingredient) solvent-based alkoxy-silanes were tested; the first product  
78 contained only N-octyltriethoxysilane (n-OTES) and the second was a combination of n-  
79 OTES and methyltrimethoxysilane (MTM). Furthermore a 50 % w/w active emulsion of  
80 methoxy-terminated dimethylphenylsiloxane (DMS) and n-OTES (DMS/n-OTES 1) and a  
81 100 % w/w active micro-emulsion of polydimethylsiloxane (PDMS) and triethoxysilane (TES)  
82 were included. Finally also a 40 % w/w active mixture of DMS and n-OTES (DMS:n-OTES 2)  
83 and a 60 % w/w active macro-emulsion of PDMS were used. All these products were  
84 developed at the laboratories of Dow Corning Corporation (Belgium), except for the 100 %  
85 PDMS/TES which was obtained from Wacker-Chemie GmbH (Germany). The solvent-based  
86 and water-based systems were diluted up to 5 % w/w (ai\_conc) with isopropylalcohol and  
87 water, respectively.

88

89 The same combinations of organosilicons and biocides as used in De Vetter et al. (2009b)  
 90 were applied. Briefly, a 5 % solution of the DMS/n-OTES 2 was combined with (1) 0.3 % 3-  
 91 iodo-2-propynyl-butyl carbamate (IPBC); (2) 0.3 % IPBC in combination with 0.6 % (±)-  
 92 (cis+trans)-1-(2-(2,4-dichlorophenyl)-4-propyl-1,3-dioxolan-2-yl-methyl)-1H-1,2,4-triazole  
 93 (propiconazole) and (3) 2 % 3-(trimethoxysilyl)propyldimethyloctadecyl ammonium chloride  
 94 (Si-Quat). As a solvent-based counterpart 5 % w/w MTM/n-OTES was combined with (1) 0.3  
 95 % IPBC and (2) 0.3 % IPBC in combination with 0.6 % propiconazole. Thirdly 10 % MTM/n-  
 96 OTES was combined with 1 % Si-Quat. Summarizing six organosilicons and six  
 97 combinations of an organosilicon and a biocide were applied. In addition to the treated  
 98 specimens, untreated Scots pine sapwood reference specimens were included in the tests  
 99 as well. Moreover, in the natural weathering test untreated heartwood of Scots pine,  
 100 Douglasfir (*Pseudotsuga* spp.) and larch (*Larix decidua*), having the same dimensions as the  
 101 Scots pine specimens, were added to the pine sapwood references.

## 102 103 104 2.2. Laboratory experiments

106 Both fungal disfigurement and colour change were evaluated on laboratory and semi-  
 107 industrial scale. Therefore pre-conditioned (12 % moisture content) straight grained Scots  
 108 pine (*Pinus sylvestris* L.) sapwood was used. For the laboratory experiments, the wood was  
 109 sawn to 320×40×10 mm, end-grain cross sections were sealed twice with a 2-component  
 110 water impermeable polyurethane system and dipped for a few seconds into the treating  
 111 solutions into four replicates. The specimens were weighed before and after dipping,  
 112 allowing calculation of the product and active ingredient retention of organosilicon and  
 113 biocide (Eq. 1 and 2). The specimens were placed into an Atlas UV2000 and artificially  
 114 weathered during six weeks using a test cycle as described in the new draft version of EN  
 115 152. Prior to weathering and in between two subsequent cycles, consisting of a wetter and  
 116 dryer subcycle, the colour was determined (see further).

$$118 \quad \text{Product}_{\text{retention}} (g / m^2) = \frac{m_{\text{after\_impr}}(g) - m_{\text{before\_impr}}(g)}{\text{area}(cm^2)} \times \frac{ai\_conc(\%) \times 10.000}{conc\_active\_ingredient(\%)} \quad (1)$$

$$119 \quad \text{Active\_ingredient}_{\text{retention}} (g / m^2) = \frac{m_{\text{after\_impr}}(g) - m_{\text{before\_impr}}(g)}{\text{area}(cm^2)} \times ai\_conc(\%) \times 10.000 \quad (2)$$

120  
121  
122 Afterwards the specimens were conditioned (at 20 °C and 65 % RH) and out of every  
 123 weathered specimen four smaller specimens were sawn for blue stain evaluation. Two had  
 124 the dimensions of standard EN 152 specimens (90×40×10 mm, 2003), while two smaller  
 125 specimens were sawn according to the recommendations for the reverse EN152 method  
 126 (50×40×10 mm, Van den Bulcke et al. 2006). The first method is the so-called wood  
 127 preservatives approach, while the latter is the wood coating approach.

128  
129 The EN 152 specimens were planed at the non-weathered side removing the applied  
 130 organosilicon (and biocide) leading to an untreated surface, easy accessible for fungi. In  
 131 contrast, half of the reverse specimens were sealed on all but the weathered side, forcing  
 132 the fungi to grow through the weathered organosilicon (and biocidal) layer. The other reverse  
 133 specimens were sealed on all but the non-weathered side, this time preventing fungal growth  
 134 from any side, except via the non-weathered organosilicon (and biocidal) layer. The sealant  
 135 was a translucent 2-component polyurethane system doped with dichlofluanid to prevent  
 136 staining. Resuming the sawing pattern lead to eight different standard EN152-specimens,  
 137 four reverse weathered and four reverse non-weathered specimens per treatment, each  
 138 exposed in a single jar to blue stain attack. Besides these treated specimens four artificially

139 weathered as well as non-weathered untreated Scots pine sapwood specimens were  
140 included in the test as reference specimens.

141

142 A spore suspension of *Aureobasidium pullulans* (de Barry) Arnaud P268 and *Sydowia*  
143 *polyspora* (Corda) V. Höhn S 231 was prepared as explained in Van den Bulcke et al.  
144 (2006). After the specimens were  $\gamma$ -sterilised (1.5 Mrad), they were shortly dipped in the  
145 spore suspension and put on a vermiculite substrate after which another 15 ml of spore  
146 suspension was poured over the specimens. After six weeks incubation at 22 °C and 70 %  
147 RH a visual, exterior assessment of the blue stain specimens, excluding the edges, was  
148 performed according to the rating scale as defined in EN 152: 0: not blue stained; 1:  
149 insignificantly blue stained; 2: blue stained; 3: strongly blue stained. These classes were  
150 further subdivided using 0.5 increments, as proposed by Van den Bulcke et al. (2006) to  
151 have more precise ratings. For the interior assessment the EN 152 specimens were cut  
152 parallel to the end faces at 30 mm from each end (EN 152), while the reverse specimens  
153 were sawn in half. The rating scale as proposed by Van Acker et al. (1998) was used for the  
154 evaluation: 0: no blue stain found; 1: few spots of blue stain; 2: small blue stained areas; 3:  
155 specimen is partly blue stained, but there are still areas free of blue stain; 4: the major part of  
156 the specimen is blue stained; 5: cross-cut of the specimen is completely blue stained.

157

158

### 159 2.3. Outdoor performance testing

160

161 For the semi-industrial scale or outdoor experiments Scots pine sapwood specimens of  
162 375×100×20 mm were used, which were also sealed at both end-grain cross sections in the  
163 same manner as for the laboratory scale tests. Half of these specimens were dipped into the  
164 treating solutions, while the other half was vacuum impregnated in the same solution using a  
165 pressure of 5 bars for 45 minutes. After releasing the pressure the specimens stayed  
166 submerged for another 15 minutes at atmospheric pressure and were finally removed from  
167 the tank and allowed to drip for 15 minutes. Four replicates per treating solution and  
168 treatment procedure were used. Mass of each specimen was measured prior to and after  
169 treatment allowing calculation of the organosilicon (and biocide) product retention (Eq. 3) as  
170 well as weight percent gain (WPG, Eq. 4) for the impregnated specimens. The treated  
171 specimens were then dried at 60 °C until they reached constant mass.

172

$$173 \text{Product}_{\text{retention}} (\text{kg} / \text{m}^3) = \frac{m_{\text{after\_impr}} (\text{g}) - m_{\text{before\_impr}} (\text{g})}{\text{volume}(\text{cm}^3)} \times \frac{ai\_conc(\%) \times 1.000}{\text{conc\_active\_ingredient}(\%)} \quad (3)$$

174

$$175 \text{WPG}(\%) = \frac{m_{\text{after\_impr}} - m_{\text{before\_impr}}}{m_{\text{after\_impr}}} \times ai\_conc (\%) \quad (4)$$

176

177

178 After conditioning at 20 °C and 65 % RH the specimens were weighed again, inspected  
179 visually and mounted outdoors on a rack having an inclination of 45° and facing south-  
180 southwest (EN 927-3 1996). The rack was located at the outdoor weathering site of the  
181 Laboratory of Wood Technology in Belgium. The specimens were not inoculated with any  
182 spore suspension.

183

184 Donath (2004) already demonstrated that visually evaluating the back, and thus non-  
185 weathered side (facing north) of such outdoor exposure specimens reflects well the  
186 resistance against moulds of a product. Therefore, for the evaluation of fungal disfigurement  
187 the specimens were visited every season and evaluated external on their back for fungal  
188 disfigurement. Although most fungi were moulds, other fungi were not disregarded. The

189 following rating scale was drawn up and used to classify each specimen: 0: no fungal  
190 disfigurement; 1: small spots of fungi are detected; 2: fungi in a small band at the upper part  
191 of the specimen; 3: fungi scattered in broader bands over the surface of the specimen; 4:  
192 specimen' surface completely overgrown with fungi.

193

194 For the colour evaluation the specimens were yearly removed from the rack and conditioned  
195 for seven days at 20 °C and 65 % RH. Afterwards the colour was measured at the front side,  
196 facing south-south west.

197

198

#### 199 *2.4. Colour evaluation*

200

201 For the colour evaluation use was made of a Spectrophotometer Konica Minolta CM-2600d  
202 and the obtained colour was expressed as a CIE\*Lab-value. Per specimen five colour  
203 measurements were performed, which were averaged to a mean value of L\*, a\* and b\*. L\*  
204 represents the lightness of the sample and ranges from black (0 %) to white (100 %) while a\*  
205 and b\* are chromaticity values representing the red to green and yellow to blue colour,  
206 respectively. It is plausible to assume that for a customer once he has chosen for a certain  
207 wood product, not the colour as such but the colour change over time is of major importance  
208 in the appreciation of the wood product. Therefore preference was given to evaluate the  
209 colour difference dE of each specimen and this considering both the application of a  
210 treatment product as well as the time of weathering. Therefore dE was calculated as the  
211 colour difference between each specimen at a certain time t compared to the colour of  
212 untreated and non-weathered Scots pine sapwood (Eq. 6). The reference values for L\*, a\*  
213 and b\* were the average values of all untreated and non-weathered Scots pine sapwood  
214 specimens included in the artificial weathering test.

215

$$216 \quad dE = \sqrt{(L_t^* - L_{ref}^*)^2 + (b_t^* - b_{ref}^*)^2 + (a_t^* - a_{ref}^*)^2} \quad (6)$$

217

#### 218 *2.5. Statistics*

219

220 Since for fungal disfigurement only a limited number of replicates was used and the rating is  
221 nonlinear anyhow the median value was preferred over the average value as to minimise the  
222 impact of outliers. Furthermore it is not the purpose to evaluate products but to retrieve  
223 information whether organosilicons as a group can decrease fungal disfigurement. Therefore  
224 the obtained rating values were not interpreted as such, but used to make clusters of  
225 products performing the same as, better than or much better than untreated Scots pine  
226 sapwood. To lower the impact of outliers the Partitioning Around Medoids (PAM) cluster  
227 analysis was preferred. The analysis was performed for each testing protocol separately  
228 (EN152, reverse weathered and reverse non-weathered) and for combinations of tests. The  
229 number of clusters to retain was determined using scree analyses. Prior to acceptance of  
230 each clustering it was checked whether they could explain at least 80 % of the variability  
231 between the treatments. The clusters fulfilling this requirement were then compared with  
232 each other.

233

234 For the colour evaluation of both tests, using artificial or natural weathering, first a two-way  
235 analysis of variance (ANOVA) with fixed factors was performed. The dependent variable was  
236 the colour difference dE of each specimen compared to untreated Scots pine sapwood prior  
237 to weathering and the independent variables were treatment and time. If significant  
238 interaction between the independent factors was found, meaning that dE depends on the  
239 combination of treatment and time, the two-way ANOVA could not be further interpreted.  
240 Therefore a one-way ANOVA was performed with dE as dependent variable and a new  
241 factor Group as independent variable. Group contains all possible combinations of treatment

242 and time. Consecutive post-hoc analyses using Scheffé-tests revealed which groups differed  
243 significantly from each other.

244  
245

246  
247

### 3. Results

248  
249

#### 3.1. Product retention

250  
251

Table 1 gives a schematic overview of the product compositions, product codes, product  
252 retentions, active ingredient retentions and WPGs for the different test set-ups.

253  
254

Table 1

255  
256

#### 3.2. Laboratory experiments

257  
258

##### 3.2.1 Fungal evaluation

259  
260  
261  
262  
263  
264  
265  
266  
267  
268  
269  
270  
271  
272  
273  
274

Clustering the EN 152 data was not possible for the exterior evaluation data and was not  
satisfactory for the interior EN 152 evaluation data. Nearly all specimens were completely  
blue stained and had therefore the same exterior rating (Fig. 1), while less than 80 % of the  
variability of the interior evaluation could be explained by the treatment (Fig. 2). Although  
slight differences in fungal disfigurement were observed, they could not be extracted from  
the analysis. Furthermore the biocides used have proven anti blue stain effectiveness  
(Isquith et al. 1972) at the concentrations applied (Valcke 1989). Therefore it seems that the  
wood preservatives approach is not fully suitable for evaluating organosilicon treated wood.  
Indeed, the coating approach as defined by Van den Bulcke (2006) is more appropriate for  
evaluating the blue stain resistance of organosilicon treated wood. For all reverse data,  
whether exterior or interior, weathered or not, three clusters were obtained. Untreated Scots  
pine sapwood and most treatments comprising only an organosilicon belong to the same  
cluster, the one of worst performing treatments. The cluster with the best performing  
treatments contains those treatments where IPBC with propiconazole is involved. The other  
biocide containing treatments most often belong to the intermediate cluster. However, in  
case of interior evaluation these treatments might also belong to the best performing cluster.

275  
276

Figure 1

277  
278

Figure 2

279  
280

##### 3.2.2 Colour evaluation

281  
282  
283  
284  
285  
286  
287  
288  
289  
290  
291  
292  
293  
294  
295  
296

The average  $L^*$ ,  $a^*$  and  $b^*$  values of untreated and non-weathered Scots pine sapwood  
specimens were 82.7, 4.6 and 24.3 respectively and were used both in the artificial and  
natural weathering tests to calculate the colour difference  $dE$ . For  $dE$  significant interaction  
( $p < 0.05$  for both artificially and naturally weathered wood) between treatment and time was  
found. The consecutive one-way ANOVA revealed significant differences between the  
groups (both  $p < 0.05$ ), leading to Scheffé-tests to discover which groups differ significantly  
from each other. The mean colour difference  $dE$  and the corresponding standard deviation  
are presented in function of the treatment in Fig. 3 for artificially weathered wood. The figure  
shows that dipping of untreated Scots pine sapwood in solutions containing DMS/n-OTES 1  
or MTM/n-OTES + Si-Quat induce a significant colour difference compared to untreated  
Scots pine sapwood. The other treatments do not lead to a significant colour difference.  
Furthermore the figure shows that, as expected, colour difference of untreated Scots pine  
sapwood increases with time. However, there is no significant colour difference between  
specimens exposed for the same time period. After the third weathering cycle  $dE$  increases  
again, leading to significant colour differences compared to the previous cycle (cycle 2). It  
can therefore be summarized that among specimens exposed for the same exposure period  
no major colour differences are present. After one weathering cycle only a small number of

297 treatments have a significant different dE compared to the non-weathered wood, whereas  
298 this number increases after two cycles and is valid for all treatments after three cycles.

299  
300

### Figure 3

301

302 Although the total colour difference is of major importance to consumers, dE does not  
303 indicate the direction of colour change. Therefore, a closer look was taken on its  
304 components L\*, a\* and b\* (data not presented). None of these values on its own was  
305 significantly different from each other or untreated Scots pine sapwood, indicating treatment  
306 did not influence the colour parameters. The weathering procedure itself however did  
307 influence the colour change, making all specimens discolour in the same manner. The  
308 specimens became brighter and less red until the second weathering cycle after which they  
309 became darker and redder, while yellowing continued. Summarizing it can be said that  
310 dipping of Scots pine sapwood into an organosilicon solution does not have a significant  
311 effect on any of the colour components, while artificial weathering does have a significant  
312 effect on the colour.

313

### 314 3.3. *Outdoor performance testing*

315

#### 316 3.3.1 Fungal evaluation

317 Also for the natural weathering test clustering into three groups was the best option (data not  
318 shown). Regardless the exposure period untreated Scots pine sapwood and half of the  
319 dipping treatments in organosilicons are grouped as worst performing. The other half of the  
320 in organosilicon dipped treatments, along with impregnations with organosilicons and dipping  
321 in solutions containing Si-Quat are clustered in the intermediate performing group. To the  
322 best performing group belong, besides all reference wood species, impregnations containing  
323 biocides IPBC and IPBC + propiconazole. The remaining treatments not yet mentioned, are  
324 impregnations with an organosilicon and the biocide Si-Quat and dipping including IPBC and  
325 IPBC + propiconazole. After one year natural weathering they belong to the best group,  
326 while they lose effectiveness over time resulting in a shift towards the medium group after  
327 two (and three) years weathering.

328

329

#### 330 3.3.2 Colour evaluation

331 For the colour evaluation of naturally weathered wood, only Scots pine sapwood was  
332 evaluated, whether treated or not. The colour difference dE seems far more complicated  
333 (Fig. 4 and 5) and doesn't change so uniformly as for artificially weathered specimens. Since  
334 the colour data prior to weathering are missing, no conclusions concerning the influence of  
335 each treatment on the colour can be made. Although the standard deviations are much  
336 greater than for artificially weathered specimens, after one year natural weathering it is clear  
337 that there is no significant difference in dE value of dipped specimens and untreated Scots  
338 pine sapwood, except when the biocides IPBC or IPBC + propiconazole are included (Fig.  
339 4). However, these differences fade away with longer exposure time. Specimens  
340 impregnated with DMS/n-OTES 1 (Fig. 5) have the largest colour difference compared to  
341 untreated and non-weathered Scots pine sapwood, while specimens impregnated with  
342 combinations of an organosilicon with IPBC or IPBC + propiconazole have the smallest  
343 colour difference. This means that the colour of these last treatments resemble the longest  
344 to untreated and non-weathered Scots pine sapwood. After two and three years the colour of  
345 these specimens approach that of untreated and weathered Scots pine sapwood. Generally,  
346 dE differences become comparable for all treatments after two years weathering and  
347 increases slightly for all specimens when weathering is continued for another year

348

349

### Figure 4

350

### Figure 5

351

352 Trying to retrieve which colour component is most influenced, L\*, a\* and b\* values were  
353 compared with each other (data not presented). The data can be separated into a group  
354 containing untreated Scots pine sapwood and Scots pine treated with solely an organosilicon  
355 and a group containing specimens treated with a combination of an organosilicon and a  
356 biocide. While the L\* value does not significantly differ within the first group, specimens  
357 belonging to the second group are much brighter, especially when treated with IPBC or IPBC  
358 + propiconazole. The trends in redness and yellowness are comparable for all specimens in  
359 that way that all values are comparable to each other regardless the exposure period, except  
360 for impregnation treatments with DMS/n-OTES 1 and impregnations involving IPBC and  
361 IPBC + propiconazole, which have slightly higher a\* and b\* values after the first exposure  
362 year. It can therefore be concluded that mainly the L\* value is responsible for the bigger  
363 variation in colour among all specimens, whereas also a\* and b\* contribute to the colour  
364 difference treatments with DMS/n-OTES 1 and the biocides IPBC and IPBC + propiconazole  
365 have compared to all other treatments and untreated Scots pine sapwood.

366  
367  
368

## 4. Discussion

### 369 4.1 Fungal evaluation

370 Although both laboratory and semi-industrial experiments evaluate fungal disfigurement,  
371 they cannot just be compared with each other. In the first experiment blue stains are the  
372 fungi leading to the disfigurement, while in the second experiment multiple factors influence  
373 the disfigurement, i.e. fungi (mainly moulds) and dirt.

374 Nevertheless are the poor results of organosilicons as preventive agents of blue stain and  
375 mould supported by the results found by Weigenand et al. (2006) and Ritschkoff et al.  
376 (2003). While better performance of outdoor specimens treated with a biocide containing  
377 solution is not astonishing, better performance of with organosilicons impregnated  
378 specimens is remarkable. Knowing that the presence of mould fungi indicates the availability  
379 of nutrients at the wood surface (Block 1953), it can be assumed that the organosilicons,  
380 when impregnated, protect the wood surface from fast release of nutrients. This, on its turn,  
381 might be attributed to the influence organosilicons have on the moisture dynamics of the  
382 treated wood (Tshabalala et al. 2003; Donath et al. 2006).

383 The diminishing effect of dipped specimens can probably be ascribed to weakening and  
384 subsequent degradation of the wood below the surface due to weathering (Banks and Evans  
385 1984), leading to a reduced effect of the superficially applied organosilicon. It can be stated  
386 that, except for the organosilicon plus biocide dipped specimens, the clustering of visual  
387 ratings after one year weathering already gives a good idea of the clustering after three  
388 years exposure.

389

### 390 4.2 Colour evaluation

391 Artificial weathering is supposed to imitate natural weathering in a fast and uniform way,  
392 trying to obtain reliable results which are easily reproducible on a standardized method. Up  
393 to now no such method uniting all these parameters has been found. Therefore care must be  
394 taken when comparing artificial and natural weathering with each other.

395

396 Concerning the initial darkening and subsequent lightening of the wood during artificial  
397 weathering, this can be explained because UV light (340 nm) induces the formation of free  
398 radicals and lignin is broken down while absorbing the UV light, in that way darkening the  
399 wood. However, afterwards reaction products are leached out by which the wood becomes  
400 brighter again (Donath et al. 2007). Lightening of naturally exposed specimens however,



401 seems to depend on some extra parameters, since the biocide presence influences the  
402 lightness of the treated wood considerably. Two main reasons are probably the presence of  
403 dirt and discolouring fungi at the specimens' surface. Since the specimens were washed with  
404 clear lukewarm water prior to colour measurements, it is assumed that the effect of dirt is  
405 minimised. Basically it is assumed that due to the presence of a biocide the wood surface is  
406 not so vulnerable to colonisation of discolouring fungi, leading to less darkening of the wood.  
407 The fact that also the biocide treated specimens become darker with time support this  
408 hypothesis, as it might be expected that the biocide becomes less effective due to  
409 weathering. This is further supported because darkening after one year exposure is far more  
410 distinct for dipped specimens than for impregnated specimens

411  
412 The redness and yellowness values are much lower for the naturally weathered specimens  
413 after one year exposure than for the artificially weathered specimens after six weeks.  
414 Moreover they reach for nearly all treatments a constant value, supporting that most  
415 discolouration happens shortly after exposure. Certain impregnated specimens have slightly  
416 higher  $a^*$  and  $b^*$  values after one year exposure, indicating they are somehow protected  
417 from fast degradation, leaching or evaporation of wood components (Sjöström 1992; Grekin  
418 2007; Salcă and Fotin 2007), or from other processes induced by UV radiation (Hon 1979)  
419 which usually lead to colour change. This effect is however only temporarily and minor  
420 compared to the impact of the  $L^*$  value on the total colour difference.

421  
422

#### 423 *4.3. General appearance*

424

425 The general appearance of weathered specimens is greatly influenced by fungal  
426 disfigurement and UV induced colour variations. Because in the laboratory experiment both  
427 parameters were investigated separately from each other, it is not straightforward to give an  
428 impression of the general appearance of the specimens. Nevertheless do both the fungal  
429 and colour evaluation suggest that only when a biocide is involved in the treatment process,  
430 significant different appearance of the specimens can be expected. The natural weathering  
431 test is far more interesting, since disfigurement of both kinds was happening simultaneously  
432 on the same specimens. The test showed that the general appearance depended on several  
433 factors. The presence of a biocide influenced both parameters positively, leading to a  
434 brighter and more uniformly discoloured specimen compared to untreated Scots pine  
435 sapwood. Slight differences in colour and fungal disfigurement were present between  
436 specimens treated with different organosilicons. Regardless the composition of the treatment  
437 solution, both discolouration and fungal disfigurement were less pronounced for impregnated  
438 specimens compared to their dipped counterparts.

439

440

#### 441 **5. Conclusion**

442 Treatment of wood with an organosilicon cannot be regarded as a form of wood modification  
443 in a strict sense, wood preservation or the application of a coating. Therefore it is not self-  
444 evident to find a method for evaluating the performance against blue staining of  
445 organosilicon treated wood whether or not in combination with a biocide. This study showed  
446 that the coating approach is more suited than the wood preservatives approach, since it is  
447 more discriminating. Secondly this research proved that under laboratory conditions an  
448 organosilicon as such is not able to protect the wood sufficiently, but combinations with  
449 biocides have good perspectives. However, outdoors the organosilicons show better  
450 resistance against fungal disfigurement than untreated wood. Obviously, the addition of a  
451 biocide enhances this effect. The discrepancy between laboratory testing and outdoor  
452 performance testing is stressed. While the former was not able to distinguish products with  
453 good perspectives from those with fewer perspectives, real outdoor performance proved  
454 significant differences are present between specimens depending on the treatment product

455 and application technique and was more hopeful for the potential of organosilicons as part of  
456 formulations designed to protect wood surfaces under use class three conditions.  
457

## 458 **Acknowledgements**

459 The authors owe their gratitude to the European Commission for funding the research  
460 project 'Improvement of Wood Product Properties by Increased Hydrophobicity Obtained by  
461 the Use of Silicon Compounds' (HYDROPHOB-QLK5-CT-2002-01439), which was the  
462 framework for this study. Furthermore we would like to thank Dow Corning Corporation for  
463 supplying the organosilicons.

464

## 465 **References**

466 Banks, W.B., Evans, P.D., 1984. The degradation of wood surfaces by water. IRG/WP/3289.  
467 The International Research Group on Wood Preservation, Stockholm.

468  
469 Block, S.S., 1953. Humidity requirements for mold growth. *Applied Microbiology* 1, 287-293.  
470

471 De Vetter, L., Stevens, M., Van Acker, J., 2009a. Fungal decay resistance and natural  
472 durability of organosilicon treated wood. *International Biodeterioration and Biodegradation*  
473 63, 130-134.  
474

475 De Vetter, L., Stevens, M., Van Acker, J., 2009b. Potential contribution of organosilicon  
476 compounds to reduced leaching of biocides in wood protection. *Annals of Forest Science* 66,  
477 209.  
478

479 De Vetter, L., Van Acker, J., 2005. Standard testing of organosilicon compounds as wood  
480 modification agents. In: Militz, H., Hill, C. (Eds.), *Proceedings of the Second European*  
481 *Conference on Wood Modification*, pp. 232-241.  
482

483 Donath, S., 2004. Treatment of wood with silanes, PhD thesis, University of Göttingen,  
484 Göttingen, Germany.  
485

486 Donath, S., Militz, H., Mai, C., 2006. Creating water-repellent effects on wood by treatment  
487 with silanes. *Holzforschung* 60, 40-46.  
488

489 Donath, S., Militz, H., Mai, C., 2007. Weathering of silane treated wood. *Holz als Roh- und*  
490 *Werkstoff* 65, 35-42.  
491

492 European Committee for Standardization EN 152, 2003. Test method for wood  
493 preservatives. Laboratory method for determining the protective effectiveness of a  
494 preservative treatment against blue stain in service. European Committee for  
495 Standardization, Brussels.  
496

497 European Committee for Standardization EN 335-1, 2006. Durability of wood and derived  
498 materials. Definition of use classes. Part 1: General. European Committee for  
499 Standardization, Brussels.  
500

501 European Committee for Standardization EN 927-3, 1996. Paints and varnishes. Coating  
502 materials and coating systems for exterior wood. Part 3: Natural weathering test with water  
503 trap. European Committee for Standardization, Brussels.  
504

505 Goethals, P., Stevens, M., 1994. Dimensional stability and decay resistance of wood upon  
506 modification with some new type chemical reactants. IRG/WP/94-40028. The International  
507 Research Group on Wood Preservation, Stockholm.  
508

509 Grekin, M., 2007. Color and color uniformity variation of Scots pine wood in the air-dry  
510 condition. *Wood and Fiber Science* 39, 279-290.  
511

512 Hager, R., 1995. Waterborne silicones as wood preservatives. IRG/WP/95-30062. The  
513 International Research Group on Wood Preservation, Stockholm.  
514

515 Hill, C.A.S., Farahani, M.R.M., Hale, M.D.C., 2004. The use of organo alkoxysilane coupling  
516 agents for wood preservation. *Holzforschung* 58, 316-325.  
517

518 Hon, D.N.S., 1979. Photooxidative degradation of cellulose: Reactions of the cellulosic free  
519 radicals with oxygen. *Journal of Polymer Science - Polymer Chemistry* 17, 441-454.  
520

521 Isquith, A.J., Abbott, E.A., Walters, P.A., 1972. Surface-bonded antimicrobial activity of an  
522 organosilicon quaternary ammonium chloride. *Applied Microbiology* 24, 859-863.  
523

524 Lukowsky, D., Peek, R.D., 1997. Water-based silicones on wood. IRG/WP/97-30144. The  
525 International Research Group on Wood Preservation, Stockholm.  
526

527 Mai, C., Donath, S., Weigenand, O., Militz, H., 2005. Aspects of wood modification with  
528 silicon compounds: material properties and process development. In: Militz, H., Hill, C.  
529 (Eds.), *Proceedings of the Second European Conference on Wood Modification*, pp. 222-  
530 231.  
531

532 Mai, C., Militz, H., 2004. Modification of wood with silicon compounds. Treatment systems  
533 based on organic silicon compounds - a review. *Wood Science and Technology* 37, 453-  
534 461.  
535

536 Ritschkoff, A.C., Mahlberg, R., Suomi-Lindberg, L., Viikari, L., Nurmi, A., 2003. Properties of  
537 wood treated with hydrophobisation agents. In: Van Acker, J., Hill, C. (Eds.), *Proceedings of*  
538 *the First European Conference on Wood Modification*, pp. 267-271.  
539

540 Rochow, E.G., 1987. Silicon and silicones. About stone-age tools, antique pottery, modern  
541 ceramics, computers, space materials and how they all got that way. Springer-Verlag,  
542 Germany.  
543

544 Roos, M., König, F., Stadtmüller, S., Weyershausen, B., 2008. Evolution of silicone based  
545 water repellents for modern building protection. In: De Clercq, H., Charola, A.E. (Eds.),  
546 *Proceedings of the Hydrophobe V. Fifth International Conference on Water Repellent*  
547 *Treatment of Building Materials*, pp. 3-15.  
548

549 Salcă, E.-A., Fotin, A., 2007. Colour changes occurred on veneer surfaces under indoor  
550 exposure. In: *Bulletin of the Transilvania University of Braşov*, pp. 359-366.  
551

552 Sjöström, E., 1992. *Wood chemistry: Fundamentals and applications*. Academic Press, New  
553 York.  
554

555 Tshabalala, M.A., Kingshott, P., Van Landingham, M.R., Plackett, D., 2003. Surface  
556 chemistry and moisture sorption properties of wood coated with multifunctional alkoxysilanes  
557 by sol-gel process. *Journal of Applied Polymer Science* 88, 2828-2841.  
558

559 Valcke, A., 1989. Suitability of propiconazole (R 49362) as a new-generation wood-  
560 preserving fungicide. IRG/WP/3529. The International Research Group on Wood  
561 Preservation, Stockholm.  
562

563 Van Acker, J., Stevens, M., Brauwiers, C., Rijckaert, V., Mol, E., 1998. Blue stain resistance  
564 of exterior wood coatings as a function of their typology. IRG/WP/98-20145. The  
565 International Research Group on Wood Preservation, Stockholm.  
566

567 Van den Bulcke, J., Van Acker, J., Stevens, M., 2006. Assessment of blue-stain resistance  
568 according to the EN 152 and a reverse method using visual and computer-aided techniques.  
569 International Biodeterioration and Biodegradation 57, 229-238.  
570

571 Weigenand, O., 2006. Wood modification with different types of silicon compounds, PhD  
572 thesis, University of Göttingen, Göttingen, Germany.  
573

574 Weigenand, O., Millitz, H., Tingaut, P., Sebe, G., de Jeso, B., Mai, C., 2007. Penetration of  
575 amino-silicone micro- and macro-emulsions into Scots pine sapwood and the effect on  
576 water-related properties. Holzforschung 61, 51-59.  
577  
578  
579  
580

581 **Figure captions**

582

583 Figure 1: Median, minimum and maximum exterior rating (0-3) of specimens dipped into an  
584 organosilicon (and biocidal) solution, weathered during six weeks in an Atlas UV2000 and  
585 subsequently blue stained according to (1) the standard EN 152 method, (2) EN 152 reverse  
586 method or (3) EN 152 reverse method, but at the non-weathered side.

587

588 Figure 2: Median, minimum and maximum interior rating (0-5) of specimens dipped into an  
589 organosilicon (and biocidal) solution, weathered during six weeks in an Atlas UV2000 and  
590 subsequently blue stained according to (1) the standard EN 152 method, (2) EN 152 reverse  
591 method or (3) EN 152 reverse method, but at the non-weathered side.

592

593 Figure 3: Averages and standard deviations of the colour parameter dE for untreated Scots  
594 pine sapwood (Control) and Scots pine sapwood dipped into an organosilicon (and biocidal)  
595 solution. The diamond symbols represent the values prior to weathering while the circle,  
596 square and triangle symbols represent the values after one, two and three consecutive  
597 artificial weathering cycles, respectively.

598

599 Figure 4: Averages and standard deviations of the colour parameter dE for untreated Scots  
600 pine sapwood (Control) and Scots pine sapwood dipped into an organosilicon (and biocidal)  
601 solution. The circle, square and triangle symbols represent the values after respectively one,  
602 two and three years natural weathering at the outdoor exposure site of the Ghent University,  
603 Belgium.

604

605 Figure 5: Averages and standard deviations of the colour parameter dE for untreated Scots  
606 pine sapwood (Z) and Scots pine sapwood impregnated with an organosilicon (and biocidal)  
607 solution. The circle, square and triangle symbols represent the values after respectively one,  
608 two and three years natural weathering at the outdoor exposure site of the Ghent University,  
609 Belgium.

610

611











