Development of an *in situ* penetration test for the uptake of preservatives in applied wood

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**ABSTRACT:** This paper concerns the study of biocides application in old timber structures of maritime pine (*Pinus pinaster Ait.*), previously impregnated with other products. A method was developed in laboratory to determine *in situ* the penetration depth of a product applied superficially. As initial treatment, three traditional products for sawn timber for buildings were used and, for new treatment, two newer, more environmentally benign products were used. Their ability to penetrate the pre-treated surfaces was evaluated after 1, 2 and 3 applications at 24 hours intervals and the results obtained are presented. Finally, the applicability of the developed test to the in-situ evaluation of timber structures is also discussed.

1 INTRODUCTION

Among the anomalies most currently observed in buildings timber structures those due to the presence of xylotrophic organisms stand out, such as rot fungi and subterranean termites, as well as dry-wood termites and wood-worms. The first two occur in timber with high moisture content and the second two in dry timber.

Both the maintenance of a good conservation state of the timber and the curative treatment when there is an infection or infestation normally resort to the use of chemical preservation products. These should be duly registered after application.

As for the type of products it can be referred that until the early 90’s of the XX century the use of CCA (chromated copper arsenate) compounds was generalized, as well as active substances such as pentachlorophenol, copper, tin or lindane applied in organic solvents (LOSP). Although some of these formulations were very efficient in extending the life of wood, the health hazard of operators and the risk of environmental impact in soil and landscape has to be seriously questioned (Savluchinske-Feio et al., 2007). Therefore, in the last decades severe restrictions to the use of many of the substances mentioned above were imposed in Europe (Directive 2003/2/CE) and for instance in the USA, the wood preservative manufacturers voluntarily withdraw CCA products from the market.

Also frequently applied there are some traditional products based in ancient practices, such as linseed oil, adequate as a water repellent finishing, and used motor oil that though with very arguable effectiveness, is often used in Portugal in the preventive treatment of wood.

In this context present maintenance interventions in ancient buildings will necessarily lead to the use of newer and more environmentally benign products for existing timber. However, the application of a curative/preventive measures is often impaired by the presence of previous treatments or finishes not always well documented and difficult to characterize.

Within the development of a PhD project concerning the rehabilitation of degraded timber structural elements due to biological agents, through laboratory and in situ analysis, the need arise to develop a penetration test that would allow a swift evaluation of the possibility to re-treat with a certain new wood preservative with minimum disturbance to the structure under rehabilitation.

Three types of products were considered as representative of previous treatments of old structures, namely, oil-borne preservatives (in the present case the “traditional” used motor oil), light organic solvent...
preservative (LOSP) using white spirit as the solvent carrier to deliver the actives into timber and arsenic copper combinations such as chromate copper arsenate (CCA). The later was not considered relevant under the present study as it usually performs well when applied in roof timber structures and re-treatments are seldom needed.

This paper describes the test developed and presents the results of its calibration for untreated maritime pine (Pinus pinaster, Ait.) and for maritime pine treated with either an oil carrier formulation or two typical LOSP formulations, one that has disappeared from the market due to the active ingredients restrictions already mentioned and one that has been marketed for a very long time.

Two products were chosen for the re-treatment, one of them a water borne product (boron based) with better toxicological and environmental characteristics and a second product (LOSP) of common use at the present time, easily obtained for the curative and preventive treatment of timber structures. Their ability to penetrate the pre-treated surfaces was evaluated after 1, 2 and 3 applications at 24 hours intervals and the results obtained are presented.

These products and their sequence of application are represented schematically in Figure 1. In Table 1 the active ingredients and the solvents of each of the products applied are presented.

2 WOOD TREATMENT

In historical terms, industrial preservation of timber started in 1838 when John Bethell registered a patent for timber treatment with creosote in autoclave, using the so-called Bethell of filled-cell process, which is even today the most frequently used preventive in depth treatments even if coupled with other products. Much later the empty-cell impregnation process came along using creosote (Rueping in 1902 and Lowry in 1906), a method still used with the same product 90 years later.

Table 1. Description of the products used in the test.

<table>
<thead>
<tr>
<th>Product</th>
<th>Type</th>
<th>Active ingredients (a.i.)</th>
<th>Solvent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial treatment products</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>A</td>
<td>LOSP</td>
<td>5,0% – Pentachlorophenol</td>
<td>White spirit</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0,9% – Lindane</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>0,5% – Diclofluanide</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>Oil-borne</td>
<td>Used motor oil – a.i. unknown</td>
<td>unknown</td>
</tr>
<tr>
<td>C</td>
<td>LOSP</td>
<td>13,75% – Copper naftanate</td>
<td>White spirit</td>
</tr>
<tr>
<td>New treatment products</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Water</td>
<td></td>
<td>–</td>
<td>Water</td>
</tr>
<tr>
<td>White spirit</td>
<td></td>
<td>–</td>
<td>White spirit</td>
</tr>
<tr>
<td>D</td>
<td>Aqueous Solution</td>
<td>Sodium oxide 14,70%</td>
<td>Water</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Boric oxide 67,10%</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>LOSP</td>
<td>Cypermethrin 0,17%</td>
<td>White spirit</td>
</tr>
<tr>
<td></td>
<td></td>
<td>I.P.B.C 0,15%</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Propiconazole 0,15%</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>Tebuconazole 0,15%</td>
<td></td>
</tr>
</tbody>
</table>
Creosote remained until the 30’s of last century as the only recognized timber preservative product. In 1931 pentachlorophenol was patented and in 1933 the first product containing copper salts, chromium and arsenates, commonly known as CCA, followed suit. The domination of these three substances was only slightly shaken by the introduction in the 50’s of boron in treatments by immersion – diffusion and especially of products in light organic solvent, some of which were 10 years later top sellers, mostly in the market segment so-called “general public” due to the versatility and ease of application (Nunes, 2001).

The last two decades of the XX century dramatically changed the panorama of an area that seemed to be consolidated, environmental concerns, disposal issues and general public perceptions questioned the use of traditional active substances (Goodell et al., 2003).

Research related with the search of new active raw materials for timber treatment has been a field of intense work in the last years. Copper (e.g. alkaline copper quat and copper azole) stood out as fungicide agent, applied in formulations without chromium and arsenate, both in organic solvent and in aqueous basis. Products based on copper are therefore positioned as the predominant solution in the near future (Humar et al., 2007) for the preventive long term treatment of wood. However, rot fungi that are resistant to copper have been known for some time and most of the environmental concerns that were expressed for other active ingredients may also be called against copper use.

The use of boron should also be considered whenever the risk of leaching is not a conditioning factor, since it possesses good fungicide and insecticide characteristics (Nunes, 1997). Laboratory and field tests have demonstrated that timber treatments with boron are very effective in the prevention of brown and white rot as well as insect attack (Jorge et al., 2004) and wood modification together with boron might be able to help the recognized problem of leaching (Kartal et al., 2007).

New multi-components biocide systems to inhibit the growth of moulds, rot fungi and termites are also under development and show promising results (Clausen & Yang, 2007).

The use of natural products, such as rosin derivatives and other extractives and modified wood, are also possibilities to take into account (Rowell, 2006; Savluchinske-Feio et al., 2007).

3 STUDY DESCRIPTION

3.1 Objective

The objective of the present study is the development of a method to be used in situ to determine the impregnation depth achieved by a new generation biocide product, when applied on timber from an ancient building treated with a non-documented product difficult to characterize without extensive and eventually expensive analysis.

3.2 Development

For laboratorial simulation of real situations, controlled quantities of the products chosen for the initial treatment were applied in a first step and later on new products of prevention and/or treatment were similarly applied.

To quantify in situ the penetration depth, a method was devised, very little intrusive and easy to apply to any building timber structure. It consists on drilling holes parallel to the face under treatment, at distances from it successively bigger, in which cotton swabs testimonies are inserted before the application of the new treatment product. The testimonies absorb the liquid impregnated into the timber when it reaches the depth that corresponds to its position.

4 MATERIALS AND METHODS

4.1 Test specimens

For this purpose, 50 × 7 × 5 cm specimens were used, cut from dry maritime pine (Pinus pinaster Ait.), with various ring dispositions, from pith and sap, with some knots and some sapstain. The aim was simulate non-exhaustively the diverse real situations that can be found in ancient buildings timber structures. These specimens were divided in four groups, to three of which a pre-treatment was applied. The remaining one, without treatment, was used for control purposes.

The preparation of the test specimens occurred five and a half months after the application of the pre-treatment and according to the procedure described next. Four holes (diameter = 0,45 cm) were drilled parallel to the face under treatment and at distances successively greater from it (0,5, 1,0, 1,5, and 2,0 cm), as shown in Figures 3 and 4. Six plastic rings were then glued over the larger face that had fewer defects, as represented in Figures 2 and 4, to limit the application of the new product. A transparent epoxy adhesive was used, uniformly distributed in the border and around the ring, in order to confine the absorption of the product to the area inside the ring. The compatibility between the glue and the solvents was tested experimentally.

After the preparation, the specimens were left to stabilize for 1 week in a conditioned chamber with constant relative humidity (65 ± 5%) and temperature (20 ± 2°C). The moisture content just prior to the tests was as presented in Table 2. This value was obtained using a needle humidimeter.
4.2 Initial treatment products

As referred three types of products were used for the initial treatment, considered representative to simulate real situations. Products A and C are both light organic solvent preservatives (LOSP) and product B is an oil-borne product obtained from an auto repair-shop and is a mix of various used motor oils, whose composition is unknown. Table 1 presented the active substances in percentage of mass and the solvents of each of the products used.

The pre-treatment products were applied with a brush on one of the larger faces of each specimen (7 × 50 cm). From each of the 4 initial samples four specimens with the highest levels of absorption of the three products referred were chosen (Table 3). Therefore, 16 specimens were subjected to testing.

4.3 New treatment products

To verify the method under development two products (D and E) were used, as described above, as well as their solvents, water and white spirit.

To make the presence of the products and solvents more easily recognizable a dye was added before treatment. After various preliminary selection tests, it was decided to use Neutral Red as a marker for water and the boron-based product. For organic solvents, the dark blue dye Ceresblau was selected. The concentrations, measured in mass, were as follows:

- Water + 0.5% Neutral Red;
- Water + 5.0% Product D + 0.5% Neutral Red;
- White spirit + 0.5% Ceresblau;
- Product D + 0.5% Ceresblau.

With the help of a pipette, 1 ml of product was applied over the area defined by the interior of the pre-glued rings, measuring 22 ± 0.8 cm². The density of each product and the quantity introduced are presented in Table 4.
4.4 Methodology

The new treatment products were applied in three coats with 24 h intervals between them. Before the application, the testimonies were positioned within the specimens. After the application, the rings were covered with a membrane in order to stop the loss of product by evaporation.

Six replicas of application were performed for each of the new treatment products for each situation of pre-treatment, randomly distributed over the specimens. A total of \(6 \times 4 \times 4 = 96\) product application locations were tested for each of the three coats.

After each period of 24 h the testimonies were removed and the respective presence or absence of color registered.

In Figure 5 (from a. to d.), the sequence of non-destructive laboratory test actions is illustrated.

5 RESULTS

5.1 Results from the non-destructive analysis

Readings of the testimonies were visual for water, white spirit and Product E. In the case of product D, the presence of boron was also verified by a colorimetric qualitative method as the testimonies were sprayed with curcumin reagent which is considered adequate for boron detection (Nunes, 1997). The reagent consisted of 0.25 g of curcumin and 10 g of salicylic acid dissolved in 10 ml of concentrated hydrochloric acid diluted with 100 moles of ethanol.

In the situations where the dyed product involved the cotton of the testimony in at least \(\frac{3}{4}\) of its cross-section it was considered that it had reached the axis of the hole, i.e. the depth of 0.5, 1.0, 1.5, or 2.0 cm.
Figure 7. Results obtained using the destructive analysis for the various situations studied: (a) water over pre-treatment with product B; (b) white spirit over pre-treatment with product C; (c) product D over pre-treatment with product C; (d) product E over pre-treatment with product C.

The average absorption depths obtained for each coat are presented in the graphs of Figure 6.

5.2 Results from the destructive analysis

To check the results obtained previously some of the specimens were cut longitudinally in half and in a quarter of the cross-section. It was then possible to register through visual analysis the penetration depth of the two products in organic solvent and in water.

It was confirmed that the products were uniformly absorbed by the timber, leading to a continuous surface of penetration in the cut cross-section, for the white spirit and Product E, and an irregular surface for the water and Product D.

The two products in organic solvent penetrated in some cases more than 2.5 cm as illustrated in Figure 7.

6 DISCUSSION

The objective of the method under development, in laboratory, is to be used, in future, to determine in situ the penetration depth of a product applied over an already treated surface.

The main results obtained, so far, can be summarized as follows:

- The penetration of the organic solvent preservative and the organic solvent alone was good, independently of the product used as initial treatment;
- The same was observed for the water borne product, the initial treatment does not seem to influence the results;
- The penetration of the organic solvents is, as expected, much better (around × 1.5) than the water borne ones and would most probably be adequate for a curative/preventive treatment;
- Also, for the organic solvents alone each application seems to increase the depth at which the product penetrates (around 0.5 cm);
- The number of applications does not seem to especially influence the final result, for water and the water borne product.

The applicability of the developed test to the in-situ evaluation of timber structures was not yet fully studied but the results obtained are quite promising as a good picture of the distribution of the products inside the wood was obtained just by the insertion of easy to obtain cotton swabs in pre-bored small holes.

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REFERENCES


