EFFICIENCY EVALUATION OF NEEM (AZADIRACHTA INDICA) OIL AND COPPER-ETHANOLAMINE IN THE PROTECTION OF WOOD AGAINST A SUBTERRANEAN TERMITE ATTACK

OCENA UČINKOVITOSTI NIMOVEGA OLJA (AZADIRACHTA INDICA) TER BAKER-ETANOLAMINSKIH PRIPRAVKOV PRI ZAŠČITI LESA PRED NAPADOM PODZEMNEGA TERMITA

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Abstract: The demand for environmentally safe preservatives for wood is increasing all around the world. This study is focused on the evaluation of the ability of Neem (Azadirachta indica) seed oil and copper-ethanolamine to protect Sydney blue gum (Eucalyptus saligna) and Ayous (Triplochiton scleroxylon) against termites. Wood blocks were impregnated with solutions of Neem oil in acetone and/or an aqueous solution of copper-ethanolamine. Impregnated woods with a retention rate of up to 108.3 kg/m³ were obtained. A leaching experiment was used to determine the fixation of preservatives in wood. The termicidal effect of the preservatives was studied through exposure in a termitarium. Neem oil was less efficient for Sydney blue gum but showed significant protection for Ayous at a higher retention rate. At the concentration investigated, the treatment of both wood species with copper-ethanolamine solution was not suitable. However, a combination of copper-ethanolamine and Neem oil showed an improvement in the protection of Sydney blue gum wood. Nevertheless, the impregnated woods still suffered an increased termite attack after leaching.

Keywords: Neem oil, copper, ethanolamine, wood, preservation, leaching, termite
pentachlorophenol or naphtenates were once also widely used for preservation of wood. However, these products are now subjected to increasing restrictions or even banned in many countries around the world due to their potential health hazards and adverse environmental effects (the liberation of toxic carcinogen metals such as chromium and arsenate, emissions of organic non-biodegradable pollutant as creosote) (Hingston et al., 2001; McMahon & Chen, 2001; Tobia et al., 1994). To address these issues, copper alternatives with reduced environmental impact have been developed using nitrogen-based organic mediators such as azole or ethanolamine to facilitate the fixation of copper in wood and thus prevent leaching (Freeman & McIntyre, 2008; Humar et al., 2006; Thaler et al., 2013; Zhang & Kamdem, 2000). Owing to the increasing demand for more and more environmentally friendly, sustainable alternative products all over the world, natural organic products such as plant-extracts from wood or barks, vegetable oils, and essential oils have been investigated as alternative solutions (Abedeji et al., 2018, Dev & Naufiyal, 2004; Gilmara et al., 2013; González et al., 2015; Hyvönen et al., 2005; Muktarul et al., 2009, Saheb & Mouhouche, 2016; Singh & Singh, 2012; Venmalar and Nagaveni, 2005). Moreover, Subbaraman and Brucker (2001) examined formulations containing Azadirachtin extracted from Neem tree with a lignin or tannin compound added as a binding agent and usable as wood preservatives against termites.

This work reports the ability of Neem oil, copper-ethanolamine or a combination of both in the protection of wood against termites. For this purpose, two wood species from Cameroon were tested: Sydney blue gum (Eucalyptus saligna) and Ayous (Triplochiton scleroxylon). Sydney blue gum was the main targeted wood because of its use for electricity poles for the distribution of power in Cameroon and neighbouring countries. Untreated wooden poles are rapidly eaten by termites, leading to a short service life (5-10 years). Previous research (Adebawo et al., 2015; Fatima & Morell, 2015; Himmi et al., 2013; Machado et al., 2013; Sotannde et al., 2011; Syofuna et al., 2012) on the application of Neem oil as a wood preservative against termites were performed at lower retention rates and showed the ability of the oil to cause the mortality of termites, although with an undesirable weight loss of wood. In this work, the impregnation parameters were set to attain a higher oil retention rate.

2 MATERIALS AND METHODS
2 MATERIALI IN METODE
2.1 MATERIALI

Blue gum wood was collected from a local timber market in Bamenda in the North-West region of Cameroon while Ayous was obtained in a local timber market in Yaounde in the Central region. Sydney blue gum (Eucalyptus saligna) is an exotic wood species mainly cultivated in the high mountains of West and North-West Cameroon, while Ayous (Triplochiton scleroxylon) comes from the natural forests located in the Center, South and East regions. Heartwood was used for both species. Neem oil was extracted from Neem seeds by a cold mechanical process. The copper chloride, ethanolamine and acetone were reagent grade and used without further purification.
2.2 WOOD SPECIMENS

2.2 VZORCI LESA

Wood blocks with the dimensions of (6 x 2 x 2) cm³ were cut from each wood species and dried in the laboratory till constant mass was obtained (after about one month). The moisture content of the wood was determined by drying some dedicated samples in an oven at 103 °C till constant mass was achieved. The moisture content was 11.9(±0.3)% and 11.1(±0.6)% for Sydney blue gum and Ayous, respectively.

2.3 IMPREGNATION OF WOOD WITH PRESERVATIVE SOLUTIONS

2.3 IMPREGNACIJA LESA Z RAZTOPINAMI ZAŠČITNIH SREDSTEV

Homogeneous preservative solutions were prepared and used for impregnation of wood blocks under reduced pressure. Neem oil was diluted with acetone. The mass ratio of Neem oil (NO)/acetone of different solutions was 10:90 (NO 10%), 30:70 (NO 30%), 50:50 (NO 50%) and 70:30 (NO 70%). Copper-ethanolamine solution (Cu-EA) was prepared using distilled water as a solvent. The mass concentrations of copper and ethanolamine in the solution were 0.5% copper chloride and 1.4% ethanolamine. A combining treatment was carried out by impregnation of wood with Cu-EA, drying the wood for 24h, and finally a second impregnation with NO 50%. Xylamon® (Syl), a commercial product still sold in Cameroon for wood protection, was used as a reference. Xylamon is a mixture of permethrin or cypermethrin, tebuconazole and benzalkonium chloride according to the former certification CTP-P+ 2002 of CTBA (today FCBA) (CTBA, 2002). Impregnation process was carried out under reduced pressure in a 5 L round bottom flask. Care was taken to keep all wood blocks completely immersed in the impregnation solution. Twelve wood blocks were used for each impregnation, and the impregnation was performed for 30 min using a tap water pressure reduction system. Afterwards, any excess solution on the surface of the impregnated wood samples was mopped up and the mass of the sample determined. The retention rate (%) was calculated using Equation 1 (Sen et al., 2009):

\[ R(\%) = \frac{(m_1-m_0) \times C}{V} \times 100 \text{ (kg} / \text{ m}^3) \]  

(1)

Where \((m_1-m_0)\) is the amount of impregnation solution retained by the wood in g, \(C\) the mass concentration of the solution in (%) and \(V\) the volume of wood block in cm³. \(m_1\) and \(m_0\) are the wood masses after and before impregnation, respectively.

The impregnated wood blocks were air dried till constant mass was obtained (after two weeks).

2.4 LABORATORY LEACHING EXPERIMENTS

2.4 LABORATORIJSKI POISKUZI IZPIRANJA

The impregnated wood blocks were divided into two groups: the first consisted of six blocks that were used for the leaching experiment, while the second six blocks were kept as the non-leached samples. The leaching experiment was performed according to the standard NF X41-565 with a slight modification. Wood blocks were introduced in glass containers with 5 to 10 times their equivalent volume in distilled water. The glass containers were placed on a mixing mechanical table (Edmund Bühler GmbH SM-30) and kept under agitation for four cycles of leaching (1h, 2h, 4h and 8 h). The leaching water was changed after each time period. The leached wood blocks were air dried till constant mass was achieved (after two weeks) and then weighed. The percentage weight loss (PWLL) due to leaching was determined by Equation 2:

\[ PWLL = \frac{m_1-m_2}{m_1-m_0} \times 100 \]  

(2)

Where \(m_1\) and \(m_0\) are the formal masses of wood after and before impregnation respectively, and \(m_2\) the mass of wood after leaching.

2.5 ACTIVITY OF TERMITES – FIELD TESTS

2.5 DEJAVNOST TERMITOV – TERENSKI POISKUS

Termites are highly destructive wood agents in many tropical regions, particularly in sub-Saharan Africa. The efficiency of preservative products against a subterranean termite was assessed by a field test. Non-impregnated, impregnated and leached wood samples were placed in a termitarium located on the university campus. Wood samples were covered with a black polyethylene sheet to be in the dark. The test was performed for 16 weeks (see Figure 1).
After the defined time, wood (or residual wood) was collected, cleaned and dried in an oven for 72h at 103 °C. Percentage weight losses (PWLT) due to termite attack were determined according to the equation 3.

\[
PWL\% = \frac{m_0 - m_3}{m_0} \times 100
\]  

3.1 RETENTION RATE

The retention rates of the different solutions are presented in Figure 2. The retention rate gives the amount of active preservative substance introduced in a wood block. As expected, Ayous showed a greater retention capacity than Sydney blue gum. This difference in behaviour is attributed to the difference in density, as the Sydney blue gum and Ayous apparent densities were 0.79(±0.06) g/cm\(^3\) and 0.36(±0.04) g/cm\(^3\), respectively. Blue gum is thus less porous, and it is more difficult for the solution to ingress towards the depths of the wood. The results showed that rising concentration of Neem oil in the Neem oil-acetone mixtures significantly increased the retention rate even though the solutions were slightly more viscous at higher Neem oil concentrations. Retention of Neem oil varied from 11.0 kg/m\(^3\) to up to 108.3 kg/m\(^3\) under the conditions investigated, and these results are consistent with those in the literature (Can & Sivrikaya, 2016; Naveri et al., 2017). It is well recognized that retention depends on the concentration of active substances in the solution, as well as other parameters such as impregnation process and time. An increase in the concentration of preservative generally increases the retention (Naveri et al., 2017). Can and Sivrikaya (2016) reported that, for impregnation of wood with tall-oil in similar experimental conditions, the retention rate followed the same order. A combination of copper-ethanolamine and NO 50% showed a retention rate (67.0 kg/m\(^3\) for Sydney blue gum and 90.7% for Ayous) closer to that of the solution NO 50% (69.5 kg/m\(^3\) for Sydney blue gum and...
88.3% for Ayous). For comparison, the retention rates for CCA preservative are around 4kg/m³ for surface applications and around 40 kg/m³ for underground applications or those in salt water (American Wood-Preservers Association, 1997). For Alkaline-Copper-Quaternary (ACQ), retention is around 2.4 kg/m³ for above-ground treatments and 6.4 kg/m³ for underground wood (American Wood-Preservers Association, 2006). It can be noticed that the retention rates obtained in this study were significantly higher compared to the traditional preservative requirements for underground applications of wood.

3.2 LEACHING

3.2 IZPIRANJE

The percentage weight losses caused by leaching (PWLL) of wood impregnated with Neem oil or copper-ethanolamine are shown in Figure 3.

Leaching can extract non-fixed preservatives, but also some extractives naturally present in wood. The extractive contents (ethanol-toluene and hot water) obtained from those wood species were 3.4% for Ayous (non-published research work) and 5.0% for Sydney blue gum (Chokouadeu Youmssi et al., 2017). The PWLL obtained for almost all the treatments was higher than the extractive content.
of the woods and showed that Neem oil as well as copper-ethanolamine were both leached. The PWLLs were generally lower with Ayous than with Sydney blue gum. This result can be explained by the depth of penetration and amount of oil in the wood. Sydney blue gum, being denser, had less in-depth impregnation and underwent more leaching. The higher oil content in Ayous could also confer to this wood a certain hydrophobicity limiting the ingress of leaching water. Wood impregnated with oil exhibited reduced water absorption, as reported in the literature (Can & Sivrikaya, 2016). The PWLL decreased for Ayous as retention increased. Copper-ethanolamine was significantly leached from the woods. Thaler and Humar (2014) also reported a mass reduction of the copper content of around 3-5% for wood impregnated with copper-ethanolamine, based on laboratory leaching tests. Sylamon® was the most leached product for both wood species.

3.5 EVALUATION OF TERMITE ACTIVITY
3.5 OCENA AKTIVNOSTI TERMITOV

The percentage weight losses caused by termite activity (PWLT) after exposure of wood blocks to field termites are shown in Figure 4.

Some degraded wood blocks at the experimental site are shown in Figures 5 and 6.

Figure 4. Percentage weight losses due to termite (PWLT) attack for Sydney blue gum (Eucalyptus saligna) and Ayous (Triplochiton scleroxylon) impregnated with different preservative solutions.

Slika 4. Izguba mase (%) zaradi napada termitov (PWLT) lesa vrst evkalipta (Eucalyptus saligna) in sambe (Triplochiton scleroxylon), impregniranega z različnimi raztopinami zaščitnih sredstev.

Figure 5. Ayous (Triplochiton scleroxylon) wood impregnated with NO 10% (a) and NO 30% (b) after 16 weeks in a termitarum.

Slika 5. Les sambe (Triplochiton scleroxylon), impregniran z NO 10 % (a) in NO 30 % (b), po 16 tednih v termitarju.
Non-treated wood was completely consumed by termites, and thus the PWLTs were 100% for both wood species. Neem oil was globally less efficient in the preservation of wood against the investigated termites. For Sydney blue gum approximately 100% of wood was degraded at retentions lower than 69.5 kg/m³, but a significant reduction in PWLT was noticed at the retention of 96.6 kg/m³. Ayous with a retention value of 108.3 kg/m³ showed the most significant resistance to termites. Many research reports have been published on the efficiency of Neem oil for the preservation of wood with different experimental conditions and retention rates. It is shown that Neem oil and other vegetable oils (linseed oil, jatropha) are toxic to termites and can cause 100% death in laboratory tests, but with a certain loss of wood weight (degradation) (Ahmed et al., 2020; Fatima & Morell, 2015; Himmi et al., 2013; Machado et al., 2013). Tests performed in field conditions (a graveyard) also revealed that wood treated with Neem oil was susceptible to termite attack, but the level of resistance was better than that seen with untreated wood (Adebawo et al., 2015; Sotannnde et al., 2011). Given the higher retention rates tested compared to the values generally used with traditional commercial products for efficient preservation, it can be concluded that Neem oil could not be applied alone for the protection of wood against termites.

At the concentration investigated, the treatment of the wood blocks with copper-ethanolamine solution was not suitable. Copper is mostly a fungicide and has insufficient termiticide or insecticide activity. The addition of co-biocides (e.g. boron) to copper amine could improve the efficacy against termites, as reported by Kalawate (2013), who found no damage after 24 months and less than 5% average weight loss after 30 months for wood impregnated with copper-ethanolamine-boron formulations and exposed to a subterranean termite attack in a field test/graveyard test. Xylammon showed significant activity with Ayous but much less with Sydney blue gum. Sydney blue gum was surprisingly (according to general perceptions) more sensitive to termites than Ayous. A combination of copper–ethanolamine and Neem oil was also tested in this study. The resistance of Sydney blue gum was significantly improved by the mixed preservatives. Further studies are under consideration to understand the synergetic effects and explain the results.

The percentage weight losses due to termite infestation of leached impregnated woods are presented in Figure 7. The samples of leached Sydney blue gum for all the treatments were almost completely decomposed by termites. The effect of leaching with Ayous was mitigated, probably because of the lower leaching rate in some samples. Ayous blocks impregnated with copper-etha-

nolamine followed by Neem oil showed the highest resistance to termites after leaching. Leaching generally removes part of active preservatives introduced in wood and increases wood biodegradation. The significant difference between non-leached and leached Sydney blue gum treated with the combined solutions suggested the loss of synergy between both preservatives.

4 CONCLUSIONS

4 ZAKLJUČKI

Neem oil applied as a natural preservative showed a low efficiency in the protection of wood against termites in the field test. An increase in the retention increased the resistance to termite attack. Copper-ethanolamine used alone was also less suitable in the protection of wood at the experimental conditions investigated. A combination of Neem oil and copper-ethanolamine showed a promising improvement in the protection of wood. A reduction of the percentage weight loss due to termite activity from 100% (Sydney blue gum impregnated with copper-ethanolamine or with Neem oil) to 7.4% (Sydney blue gum impregnated with combined copper-ethanolamine and with Neem oil) was observed. Nevertheless, the impregnated wood samples were more exposed to termite attack after leaching. Further research efforts are under consideration to understand the synergetic effects between these two products, increase the fixation of preservatives into wood and modulate their leaching.

5 SUMMARY

5 POVZETEK

Anorganska in organska zaščitna sredstva so že dolgo v uporabi za zaščito lesa pred biološko razgradnjo, ki jo povzročajo glive, termiti in drugi insekti ter morski lesni škodljivci. Tradicionalna zaščitna sredstva na osnovi spojin bakra, ki vsebujejo strupe-ne spojine kroma in arzena ali organski sredstvi krezot in pentaklorofenol (PCP), so podvržena vse večjim omejitvam in so v mnogih državah po svetu celo prepovedana. V iskanju okolju prijaznejših izdelkov kot zanimive alternative preučujejo baker-aminske pripravke in različne naravne organske proizvode, kot so na primer rastlinski izvlečki iz lesa ali skorje dreves in grmov, rastlinska olja in eterična olja. Ta študija je osredotočena na oceno možnosti uporabe olja nima (*Azadirachta indica*) in baker-etanolaminih sredstev za zaščito lesa evkalipta (*Eucalyptus saligna*) in sambe (*Triplochiton scleroxylon*) pred napadom zemeljskega termita. Omenjeni evkalipt je pomembna lesna vrsta, ki se uporablja za električne drogove v Kamerunu in sosednjih državah. Nim (*Azadirachta indica*) je drevesna vrsta družine Meliaceae - melijevke, ki raste v tropskih in subtropskih območjih. Vrsta izvira z indijske podceline, danes pa je razširjena v Afriki v Avstraliji. Ta vrsta je znana že vsaj dva tisoč let, ker imajo izvlečki iz njenih listov, skorje in semen fungicidne, protibakterijske in insekticidne lastnosti in se uporabljajo za zdravila. Predhodne raziskave o uporabi nimovega olja za zaščito lesa ter mitom so bile izvedene z manjšimi navzemi in so pokazale, da olje lahko povzroči smr-

ZAHVALA

This work was supported by the University of Yaoundé 1 and the Ministry of Higher Education of Cameroon through the “Programme d’appui à la recherche”. The authors are also grateful to the Slovenian National Research Agency (ARRS) and University of Ljubljana for financial support of the project “Silwoodcoat”.

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