Dry wood termite resistance of acetylated and polymerized tributyltin acrylate (TBTA) Indonesian and USA wood

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Effect of acetylation and polymerized Tributyltin Acrylate on dry wood termite *Cryptotermes cynocephalus* (Light) resistance of Indonesian Pine (*Pinus merkusii* Jungh et de Vriese.), Indonesian Jabon (*Anthocephalus sinensis* Lamk A. Rich ex Walp.) and USA southern yellow pine (*Pinus taeda*) had been investigated. Indonesia and USA wood was either acetylated with acetic anhydride at 120°C for 24 hours or polymerized with tributyltin acrylate (TBTA) and trimethylolpropane trimethacrylate (TMPTM) as the crosslinker for five minutes with vacuum and 30 minutes without vacuum And then the wood samples had been exposure to dry wood termite *Cryptotermes cynocephalus* (Light) for 10 weeks in laboratory test.

The acetylated wood samples had an average Weight Percent Gain (WPG) was 27 % for Indonesian Pine, 17 % for Indonesian Jabon and 21 % for USA southern yellow pine. The TBTA wood samples had an average WPG was 12 % for Indonesia Pine, 11 % for Indonesia Jabon and 10 % for USA southern yellow pine. Infra red analysis showed esterification reaction occur between wood samples and acetic anhydride. Atomic absorption Spectroscopy showed increasing percentage of tin for each TBTA treated wood samples.

The result indicated that treated wood samples were effective against degradation by Cryptotermes cynocephalus (Light) with indication decreasing weight loss of wood samples and increasing mortality of termite. Average value weight loss was 11.38 % for untreated wood samples, 1.78 % for acetylated wood samples and 1.56 % for TBTA treated wood samples. Average mortality of termite was 97.1 % for acetylated samples, 100 % for TBTA treated samples and only 40.89 % for untreated.

Keywords:dry wood termite [*Cryptotermes cynocephalus* (Light)], acetylation, polymerization, tributyltin acrylate (TBTA), wood preservation.

Introduction

Termite degradation causes extensive destruction in wood in Indonesia as well as many parts of the world. Existing wood treatments for protection against biodegradation are mainly based on toxicity. The toxicity of wood preservative in wide commercial use have been the main subject to public criticism, because they are the subject of growing environmental concern.

New environmentally friendly technologies to protect wood and wood based products from biodegradation are needed. Product need which are not only environmentally friendly, but also they are not hazardous to human beings as well. Efforts to develop non-toxic methods for preserving wood have been intensified all over

the world. That will be more important in the future because of its effect on the resistance to termites attack of the end products in use without a hazard to health and the environment.

Wood modification techniques are currently receiving interest for the prevention of biodeterioration. Chemical modification of wood is potentially a good alternative to conventional wood preservation. One of modification system which has received considerable attention is acetylation. Chemically modifying wood by acetylating the hydroxyl groups of the wood components are reacted with chemical reagents, resulting in stable, covalently bonded group attachment [7].

Another technology involves polymerization of bioactive polymers in the lumens of the wood. The mechanism of effectiveness may still be toxicity, but it is envisioned as a controlled release system, therefore holding the toxicant in place longer [4]. Conventional wood treatments disperse the preservative in the wood without chemical anchoring, therefore having a greater leaching rate. With bioactive polymer the duration of protection and optimum release of toxicant can be controlled by the polymer matrix properties. Thus, the environmental hazard can be minimized.

The present research carries out to evaluate dry wood termites resistance of acetylated and TBTA wood. The ultimate goal is to treat woods to obtain a high resistance against dry wood termite.

Materials and Methods

Materials. Wood of Indonesian Pine (*Pinus merkusii Jungh et de Vriese*), Indonesian Jabon (*Anthocephalus chinensis Lamk A. Rich. ex Walp*) and USA Southern yellow pine (*Pinus taeda*), which had relatively poor durability were used for specimens.

Oven dried wood samples were weighed and then they had immersed in acetic anhydride for two weeks. Subsequently wood samples were transferred to a stainless steel vessel and reacted with acetic anhydride. Acetylation was carried out for a required period 24 hours at 120°C. Then, after unreacted acetic anhydride and the by-product acetic acid were recovered, the samples were rinses in water until the smell of acetic acid was removed. Then the acetylation level was indicated by weight percent gain based on the oven-dried of wood samples. One sample acetylated wood was used for further infra red analysis using Spectro fotometre.

The ASTM D 1413 – 1976 standard method was followed for treating TBTA wood samples with a few modifications. Oven-dried wood samples placed in a treating chamber and the system was evacuated for one hour with a water aspirator (30 mm Hg). The selected solution (Tributyltin Acrylate (TBTA), 2,4-dimethylvaleronitrile as the catalyst, Trimethylolpropane Trimethacrylate (TMPTM) as the crosslinker, and aceton solvent) was admitted into the treating chamber until the solution covered all the samples, and held for five minutes. Samples were held in place with a glass weight to prevent floating. The vacuum was released and the chamber was brought to atmospheric pressure. The samples were allowed to soak for 30 minutes in the solution, removed, wiped of excess solution, wrapped immediately in aluminum and then placed in 52°C oven. Then the samples were flushed with nitrogen and left for 24 hours to allowed polymerization. The foil was removed and then obtain at weight percent gain based on the oven-dried of wood samples. One sample was used for further tin analysis by using atomic absorption Spectroscopy (Perkin Elmer 5100PC).

Test Methods. Acetylated, TBTA treated and untreated wood samples (19 mm x 19 mm x 10 mm) were tested against the dry wood termite *Cryptotermes cynocephalus* (Light) in laboratory testing [2,3]. Fifty healthy and active nymphae of *Cryptotermes cynocephalus* (Light) were put into each glass box (inside dimensions 50 x 45 x 35 mm), and those boxes were put in a dark room at an average temperature of 20°C to 32°C and 81 to 89 percent relative humadity (RH) for 10 weeks. The 10 weeks test duration was based on a starvation test, so during this time starvation test reached 100% mortality. At the end of the test duration, the test specimens were then removed, cleaned the surface by washing and subsequently oven dried and weighed. Nymphae mortality and weight loss were determined. The weight loss percentage of each wood samples was determined by using the following formula:

$$WL = \frac{W_b - W_a}{W_b} X\ 100\ \%$$

where :

WL = weight loss percentage

W_b = wood sample weight prior to the test at oven dried condition

W_a = wood sample weight after the test at oven dried condition

Results and discussions

Weight Percent Gain through Acetylation and TBTA. After treatment the acetylated woods had an average WPG was 27 % for Indonesian Pine (IP), 17 % for Indonesian Jabon (IJ) and 21 % for USA Pine (USP). The WPG range may be due to the variability of the wood structure and chemical components. Infra Red of each wood samples are presented in Fig. 1 , Fig.2, and Fig.3.

The absorption at 1730 cm^{-1} assigned to the C=O stretching vibration of carboxyl and acetyl groups, much increased with acetylation. The absorptions at 1380 and 1240 cm^{-1} are due to CH_3 bending vibration and C-O stretching vibration, respectively. Both of them are derived from acetyl groups and increased with acetylation too.

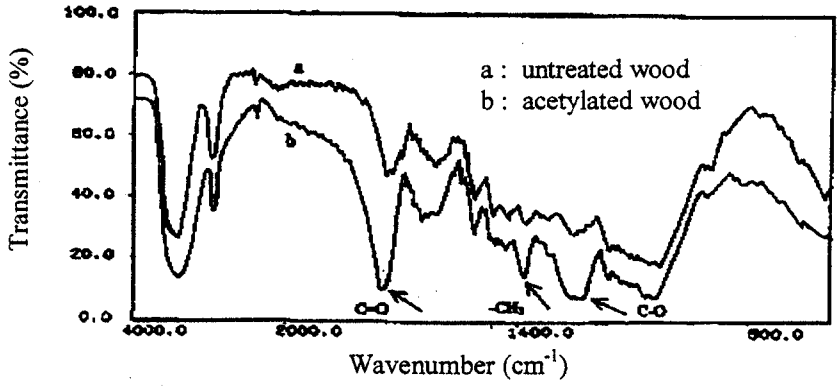

Figure 1. Infra Red Spectra of Acetylated Indonesian Pine

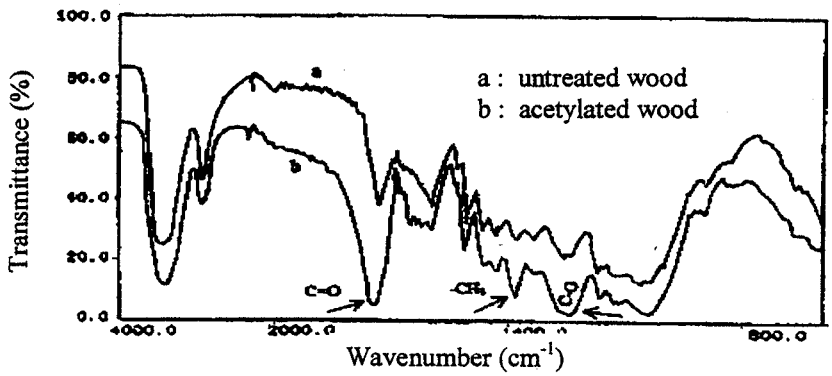

Figure 2. Infra Red Spectra of Acetylated Indonesian Jabon

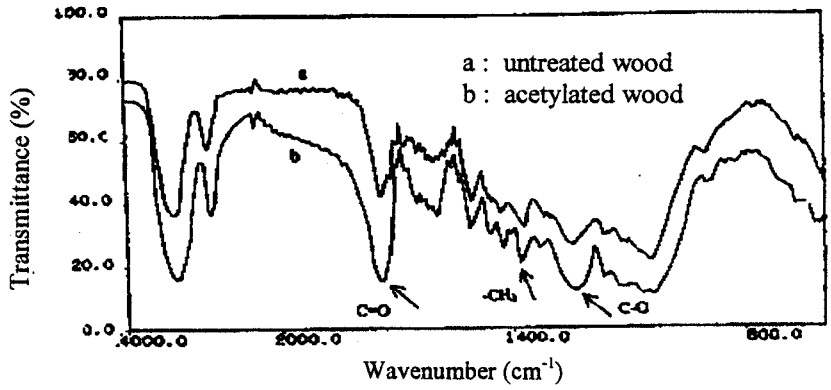

Figure 3. Infra red spectra of acetylated US pine

The TBTA treated woods had an average WPG was 11 % for IP, 12 % for IJ, and 10 % for USP. The polymerization for IP and USP woods followed the procedure, but after polymerization in IJ there was polymerized material on the surface samples that was brushed off before weighing. This phenomenon perhaps was due to the structure of the IJ wood that was have very narrow lumens. Percentage of tin for each wood samples are shown in Table 1.

Dry Wood termite resistance. Weight loss of wood samples and dry wood termite mortality after 10 weeks of testing are shown in Table 2. Both the acetylated and TBTA treated wood samples gave better resistance to dry wood termite attack than the untreated wood. On the other hand, the weight loss of the acetylated and TBTA wood samples were lower than the untreated wood. The untreated wood samples was 11.4 %; the acetylated wood samples was 1.8 %; and the TBTA treated wood samples was 1.6 %. As well as the weight loss, both the acetylated and TBTA wood treated samples had higher termite mortality than the untreated wood samples. The acetylated wood samples reached 97.3 % termite mortality; the TBTA treated wood samples reached 100 % termite mortality; and the untreated wood samples only reached 40.7 %.

Table 1. Tin percentage of TBTA treated wood samples

Wood Species	Treatment	Avg. Wt. Gain (%)	Sn (%)
Indonesian Pine	Untreated	0	0.03
Indonesian Pine	TBTA	10.47	0.17
Indonesian Pine	TBTA	9.09	0.23
Indonesian Pine	TBTA	12.865	0.19
Indonesian Jabon	Untreated	0	0.01
Indonesian Jabon	TBTA	11.22	0.32
Indonesian Jabon	TBTA	11.27	0.37
Indonesian Jabon	TBTA	13.56	0.48
USA-Southern Pine	Untreated	0	0.01
USA-Southern Pine	TBTA	9.13	0.19
USA-Southern Pine	TBTA	10.90	0.15
USA-Southern Pine	TBTA	10.22	0.18

Table 2. Resistance of acetylated and TBTA wood against wood termite of *Cryptotermes cynocephalus* light

Wood Species	Treatment	Weight Loss (%)	Mortality (%)
Indonesian Pine	Untreated	9.31	42
	Acetylated	1.85	98.67
	TBTA	1.46	100
Indonesian Jabon	Untreated	16.45	31.33
	Acetylated	1.88	94.67
	TBTA	1.37	100
US Pine	Untreated	8.39	49.33
	Acetylated	1.60	98
	TBTA	1.85	100

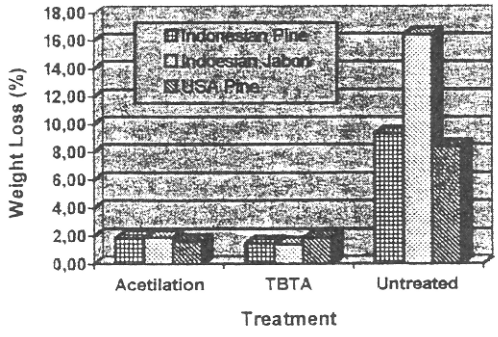

Figure 4. Weight loss of wood samples after exposure to dry wood termite

Figure 4. and Figure 5. show the difference decreased weight loss of wood samples for each treatment after 10 weeks exposure to dry wood termite.

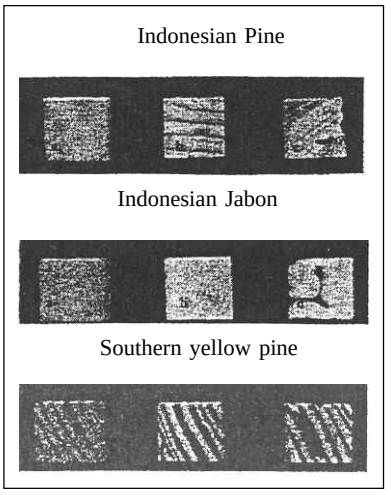

Figure 5. The wood samples conditions for each treatment after 10 weeks exposure to *Cryptotermes cynocephalus* light. a : Acetylated wood; b : TBTA treated wood; c : untreated wood

As shown Fig. 4. and Fig. 5., although *Cryptotermes cynocephalus* Light was able to attack acetylated and TBTA treated wood, but loss in volume of wood was significantly smaller than untreated wood. These are caused by any chemical reaction between the hydroxil groups of principal wood components and single chemical reagent. Hydroxil groups are the most abundant reactive sites in the wood cell wall polymers, and therefore provide the biological enzymatic reaction sites.

As well known termites have a very specific enzyme system capable of degrading wood polymers into digestible units. When the substrate for this system is chemically changed, this enzymatic action cannot take place. Therefore, the chemicals used for modification need not to be toxic to the wood-deteriorating organism. It is caused by their action which renders the substrate unrecognizable or indigestible as a food source to support biological growth [6].

Meanwhile, the TBTA treated wood have small loss in volume, it is possible that chemicals are toxic while still bound to the cell wall but the toxicity is more likely due to release of the chemical into the surrounding wood structure [5].

Conclusions

Three wood species (Indonesian Pine, Indonesian Jabon, and USA southern yellow pine) are treated with acetic anhydride and polymerized with TBTA. As the result, both treatments showed resistance to dry wood termite *Cryptotermes cynocephalus* Light. After exposure to dry wood termite, the weight loss of wood treated samples were decreased and the mortality of termites were increased. For that reason, the chemical modification of wood is very important for the prevention of wood biodeterioration which are environmentally friendly and also not hazardous to human beings.

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Sustainable Utilization of Forest Products : Socio-Economical and Ecological Management of Tropical Forests

PROCEEDINGS OF THE THIRD INTERNATIONAL WOOD SCIENCE SYMPOSIUM

November 1-2,2000, Uji, Kyoto, Japan

JSPS-LIPI Core University Program in the Field of Wood Science

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