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ADHESION AND BONDING PROPERTIES OF MALAYSIAN HARDWOOD SPECIES AFTER ALKALINE COPPER QUATERNARY TREATMENT FOR CROSS LAMINATED TIMBER

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By

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Chairman: Paridah Md Tahir, PhD
Institute: Tropical Forestry and Forest Products

The bonding and strength properties of ACQ-treated cross laminated timber (CLT) made of four Malaysian hardwood species, i.e. Batai, Sesendok, Rubberwood and Kedondong were evaluated. The present study was carried out to produce ACQ-treated CLT manufacturing process with good and excellent bonding properties, without compromising their true strength. Three layers of single- and mixed-species CLT were fabricated at three different glue spread rates, i.e. 200 g/m^2, 250 g/m^2, 300 g/m^2, and two different clamping pressure, i.e. 0.7 N/mm^2 and 1.4 N/mm^2. Each CLT combinations were treated with ACQ at 2 % concentration and compared with untreated group. Block shear strength and delamination tests were carried out in compliance with BS EN 16351: 2015 to examine the bonding performance of the CLT. Two-way analysis of variance (ANOVA) and Least Significant Difference (LSD) test were conducted to evaluate the effects of ACQ and species on density, surface roughness, contact angle of wettability, block shear strength and delamination percentage. The result revealed that the density of each hardwood species were improved after ACQ treatment and notable increments up to 48 % were observed on their surface roughness value. The contact angles of ACQ-treated samples resulting from water and ACQ treatment were low, less than 44°, suggest high wettability rate. Generally, ACQ did not significantly affect the bonding properties of CLT produced. Clamping pressure give no significant effect on bonding properties of CLT while glue spread rate give a notable results between the species. Single-species CLT with a higher glue spread rate, i.e. 300 g/m^2 glue spread, provided a stronger mean shear bond strength. Among the ACQ-treated single-species CLTs, Rubberwood has the highest mean shear bond strength of 9.06 N/mm^2, followed by Kedondong (6.26 N/mm^2), Sesendok (5.52 N/mm^2) and the weakest was Batai (4.30 N/mm^2). The fabrication of mixed-species CLT has improved the overall performance of the CLT. The combination of ACQ-treated Rubberwood-Sesendok-Rubberwood provided the highest mean shear bond strength of 8.05 N/mm^2. None of the CLT delaminated after subjected to the delamination test

indicates that the behaviour of CLT samples was not influenced by both clamping pressure and glue spread rate. Among the examined species, Rubberwood exhibited the best bonding performance for untreated control and ACQ-treated samples, followed by Kedondong, Sesendok, and Batai. These combined data suggest that under the conditions tested, this study has met the requirements stated in BS EN 16351:2015 standards. By using ACQ, the bonding strength between CLT members was not significantly affected and shows overall better bonding performance compared to untreated group.

Keywords: Density, surface roughness, contact angle of wettability, shear bond strength, delamination, clamping pressure, glue spread rate

Abstrak tesis dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

SIFAT LEKATAN DAN IKATAN SPESIS KAYU KERAS MALAYSIA SELEPAS RAWATAN KUARTAL TEMBAGA ALKALI UNTUK KAYU BERLAPIS SILANG

Oleh

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Sifat ikatan dan kekuatan kayu berlapis silang (CLT) yang dirawat Alkaline Copper Quaternary (ACQ) yang diperbuat daripada empat spesies kayu keras Malaysia iaitu Batai, Sesendok, kayu getah dan Kedondong telah dinilai. Kajian ini dijalankan untuk menghasilkan proses pembuatan CLT yang dirawat ACQ dengan sifat ikatan yang baik dan cemerlang, tanpa menjejaskan kekuatan sebenar mereka. Tiga lapisan CLT spesies tunggal dan campuran telah dihasilkan pada tiga kadar sebaran gam yang berbeza, iaitu 200 g/m^2, 250 g/m^2, 300 g/m^2, dan dua tekanan pengapitan yang berbeza, iaitu 0.7 N/mm^2 dan 1.4 N/mm^2. Setiap kombinasi CLT dirawat dengan ACQ pada kepekatan 2% dan dibandingkan dengan kumpulan yang tidak dirawat. Ujian ricihan blok dan peleraian telah dijalankan menurut BS EN 16351:2015 untuk menguji prestasi ikatan CLT. Analisis dua hala varians (ANOVA) dan Ujian Perbezaan Paling Ketara (LSD) dijalankan untuk menilai kesan ACQ dan spesies pada ketumpatan, kekasaran permukaan, sudut sentuhan kebolehbasahan, kekuatan ricihan blok dan peratusan perlekangan. Keputusan kajian mendedahkan bahawa ketumpatan setiap spesies kayu keras telah meningkat selepas rawatan ACQ dan kenaikan yang ketara sehingga 48% telah diperhatikan pada nilai kekasaran permukaan mereka. Sudut sentuhan sampel yang dirawat oleh ACQ yang terhasil daripada rawatan air dan ACQ adalah rendah, kurang daripada 44°, mencadangkan kadar kebolehbasahan yang tinggi. Secara amnya, ACQ tidak menjejaskan sifat ikatan CLT yang dihasilkan dengan ketara. Tekanan pengapit tidak memberi kesan yang ketara ke atas sifat ikatan CLT manakala kadar sebaran gam memberikan perbezaan yang ketara antara spesies. Kadar sebaran gam yang lebih tinggi iaitu 300 g/m^2 memberikan purata kekuatan ricihan ikatan yang lebih kukuh bagi spesies tunggal CLT. Antara spesies tunggal CLT yang dirawat ACQ, Rubberwood mempunyai purata kekuatan ricihan ikatan tertinggi iaitu 9.06 N/mm^2, diikuti Kedondong (6.26 N/mm^2),

Sesendok (5.52 N/mm^2) dan Batai adalah yang paling lemah (4.30 N/mm^2). Penghasilan spesies campuran CLT telah meningkatkan prestasi keseluruhan CLT. Gabungan kayu getah-Sesendok-kayu getah yang dirawat oleh ACQ memberikan purata kekuatan ricihan ikatan tertinggi iaitu 8.05 N/mm^2. Tiada satu pun CLT yang terlekang selepas ujian perlekangan, menunjukkan bahawa tingkah laku sampel CLT tidak dipengaruhi oleh tekanan pengapit dan kadar penyebaran gam. Antara spesies yang diperiksa, kayu getah mempamerkan prestasi ikatan terbaik untuk kumpulan sampel yang tidak dirawat dan sampel yang dirawat ACQ, diikuti Kedondong, Sesendok, dan Batai. Data gabungan ini menunjukkan bahawa di bawah syarat-syarat yang diuji, kajian ini telah memenuhi keperluan yang dinyatakan dalam piawaian BS EN 16351:2015. Dengan menggunakan ACQ, kekuatan ikatan antara ahli CLT tidak terkesan dengan signifikan dan menunjukkan prestasi ikatan yang lebih baik secara keseluruhan berbanding dengan kumpulan yang tidak dirawat.

Kata kunci: Ketumpatan, kekasaran permukaan, sudut sentuh kebolehbasahan, kekuatan ricihan ikatan, perlekangan, tekanan pengapit, kadar penyebaran gam

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

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LIST OF ABBREVIATIONS

ACQ	Alkaline Copper Quaternary
ANOVA	Analysis of Variance
ASTM	American Society for Testing and Materials
BS	British Standards
CLT	Cross Laminated Timber
FRIM	Forest Research Institute Malaysia
h	Hours
Min	Minutes
Mpa	Mega Pascal
N	Newton
PRF	Phenol Resorcinol Formaldehyde
RH	Relative Humidity
SEM	Scanning Electron Microscopy
SPSS	Statistical Product and Service Solutions
UPM	Universiti Putra Malaysia
WPG	Weight Percent Gain
g/m^2	Grams per square metre
N/mm^2	Newton per square millimetre

CHAPTER 1

INTRODUCTION

1.1 Background of Study

Malaysia has been endowed with abundant and great varieties of timber species and has been used as raw materials for structural applications in the form of engineered wood products for years. Malaysian hardwood timbers have been grouped by Malaysian Standard (MS 544: Part 2: 2001) into seven strength group which are Strength Group (SG) 1, 2, 3, 4, 5, 6 and 7. SG1 are considered strong and often utilised as structural timber while SG7 are considered the weakest and mostly used for non-structural applications. In order to upgrade Malaysian hardwood timber species; especially timber from SG5-SG7, prefabricated multi-layer engineered products or also known as cross laminated timber (CLT) may be the option. Cross laminating process improves dimensional stability, potentially cost-competitive and one of wood-based solution that may consist both light frame and heavy timber options. CLT also offered virtually unlimited flexibility in both size and shapes. The laminations in CLT allows control over the placement of different timbers grades within its cross section which makes it highly cost-effective.

In Malaysia, any timbers, especially non-durable timbers that exposed to outdoor environments are normally treated with wood preservatives to prevent deterioration and prolong their service life According to MS 544: Part 10: 2003, timbers employed for structural purposes has to be treated to reduce the risk of degradation and consequences of failure, especially non-durable timber species such as Batai, Sesendok, Rubberwood and Kedondong. These timbers species have low resistant to biological degradation agents. Thus, there is a need to treat the timbers and the best way is through chemical treatments. Chemical preservation maintains timbers quality and ensures a longer service life. The success of timbers treatment is defined by the choice of preservative used. Preservative selected should commercially available, environmental friendly, possess fungicidal and insecticidal properties and will not affect much the properties of treated hardwood. Alkaline Copper Quaternary (ACQ) preservative become one of most promising preservative with the absence of chromium and arsenic in the formulation. Some of the ACQ compound that have been formulated into preservative, use different compound as their quaternary and are available in market under different type names. In earlier study, ACQ was reported to give negligible effect on mechanical strength of most of laminated timber tested. Moreover, ACQ pre-treatment by most researchers found no delamination occurs after the test which indicates good bonding properties on laminated wood. Since there is a potential usage of Batai, Sesendok, Rubberwood and Kedondong in lamination form, the adhesion and bonding characteristics of these laminates should be investigated.

1.2 Problem Statement and Justification

With regard to understand the efficiency of stress transfer across the adhesive bond, it is important to study the areas between the substrate and adhesive. A series of chain link which follows from outermost link 8 and 9 (wood substrate) and then link 6 and 7 (wood interphase); links 4 and 5 (typical interface); links 2 and 3 (adhesive interphase regions) and link 1 (bulk adhesive layer) are visualised by Marra for better understanding. As Marra noted, failure in bonding was found seldom at the interface unless poor bond formed. Poor bonding leads to wood failure, high delamination as well as low shear bond strength. The most possible reasons of poor bonding for treated wood are the preservative interference towards curing of adhesive, physical blockage and reduction in wettability of wood. On the other hand, high roughness value of wood surface also may cause decreasing in bond strength of laminated wood. Poor wettability may leads to unsufficient glue penetration. A good penetration of the adhesive is highly promoted by excellent wood-to-adhesive-surface interaction as well as excellent adhesive mobility. Type of preservative, preservative retention and interaction with the surface was reported as a highly significant factors that affecting the shear bond strength of glue bonds.

As ACQ treatment will change the interface between adhesive and cohesive bonding, it is crucial to ensure a stable glueline formed within the interface layer at link 4 and 5 (typical interface) and also link 2 and 3 (adhesive interphase regions) to avoid glue failure. A preliminary study was conducted to evaluate the ACQ retention value after pressure treatment. The results showed that all hardwood species able to retained > 5.6 kg/m^3 ACQ solution at 2 % concentration which has fulfil the minimum preservative retention required by MS 360:2006 and able to impart some degree of durability for the end product. Specifically, hardwood species used in this study were treated with ACQ prior CLT fabrication in order to evaluate the possibilities of ACQ preservative interference between wood and adhesive at interface layer. It is also to evaluate the probability of ACQ preservative to significantly reduced the shear bond strength of each hardwood species after treatment.

ACQ chosen in this study due to its availability, low toxicity and give good bonding properties on previous laminated timber products. The methods of preservation were modified according to pressure chamber used in this study and results obtained in preliminary test. Density can be a great indicator to estimate the bond ability of variety of wood species. Thus, different hardwood species at different density ranges were chosen to understand their adhesion and bonding behaviour. According to adhesive manufacturer recommendations, glue spread rate for Malaysian hardwood were ranged from 200 to 350 g/m^2. As specified in this study, three different glue spread rate (200, 250 and 300 g/m^2) and two different pressure (0.7 and 1.4 N/m^2) were chosen according to density range of hardwood used (220-620 kg/m^3) and adhesive manufacturer recommendations. Evaluations on two different cramping pressure are crucial to

give good bonding properties and compatibility when different hardwood species were mixed for production of mixed-species CLT. Besides, the effects of ACQ preservatives on wood surface properties and adhesive curing after lamination might vary the penetration depth between untreated and treated wood.

In the spirit of upgrading timber with superior properties, timber from SG 5 (Rubberwood and Kedondong) and SG 7 (Batai and Sesendok) were used in this study. They have potential as new alternative raw materials for structural construction, relatively good strength and working properties and also available throughout the year. Based on previous research regarding engineered wood products, a satisfactory bonding strength was obtained which allow in exploring more potential from these timbers species. Plus, these timbers were hard to obtain in large diameter and length, i.e except for Kedondong, thus processing it into CLT become an important way to improve the utilization rate of logs. For efficient use of timbers, the CLT layup for mixed-species was arranged using different strength grade combinations where lower grade laminations; i.e. Batai and Sesendok, were placed in the inner layer while the outer layers consist of better grades; i.e. Rubberwood and Kedondong, to resist higher stress. Mixing the species were said to provides better bonding performance especially for timber with low grades. Moreover, mixed-species combination in CLT allows dispersion of timber defects throughout the CLT cross section.

Although extensive research has been conducted on CLT, limited studies have been conducted to investigate adhesion and bonding properties of ACQ-treated CLT using Malaysian non-durable timbers. In this study, a series of species combinations and working parameters were studied and optimized. By understanding the bonding between adjacent layers of ACQ-treated single- and mixed-species, the possibilities of shear bond failure and delamination when CLT is in service were minimized. From the information and data collected, this study provides important information for the benefit of the industry intending to explore the future potential of CLT in building construction.

1.3 Research Aim and Objective

The aim of this study is to evaluate the characteristics of four species of Malaysian hardwoods after Alkaline Copper Quaternary (ACQ) treatment and the bond integrity of CLT made from these hardwoods. The four Malaysian hardwood used were Batai, Sesendok, Rubberwood and Kedondong. The specific objectives of the study are:

a) To study density, surface roughness and wettability of four Malaysian hardwoods after ACQ treatment.

b) To evaluate the effects of ACQ treatment on shear bond strength, percent wood failure and percent delamination of CLT made from single- and mixed-species Malaysian hardwoods.

1.4 Organizational of Thesis

This thesis is organized into six chapters where the first chapter gives the overview, problem statement and the objectives of study. Chapter 2 reviews the selected and relevant literatures on Malaysian hardwood species, CLT, ACQ preservatives and PRF adhesive used to manufacture CLT. This chapter also discussed the methods used to preserve wood as well as the type of preservatives used throughout the process. The selection of raw materials and adhesives were also further elaborated in this chapter.

Chapter 3 describes the experimental work for the study which includes the selection of wood raw material, treatment/impregnation process, wood density, contact angle of wettability and surface roughness of the lumber after treated with ACQ. Fabrication of CLT, effects of ACQ on the shear bond strength of single- and mixed-species CLT, testing and data analysis were evaluated and discussed in Chapter 4. Chapter 5 concludes the study findings and relates the results from all chapters. It also provides some recommendations for future work.

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BIODATA OF STUDENT

Nur Amira binti Adnan was born on 11th March 1994 at Hospital Bhar Maternity Pulau Pinang. She is the fourth child in the family and grew up at Teluk Kumbar, Pulau Pinang. She loves to read and do adventurous activities such as mountain hiking, waterfall hunting and travelling. She started her primary education at Sekolah Kebangsaan Seri Bayu then continued her secondary education at Sekolah Menengah Sains Tun Syed Sheh Shahabudin. She developed her interest in science and technology during her secondary school and often became a school representative in a few programmes.

After completing her matriculation program at Kolej Matrikulasi Pulau Pinang, Kepala Batas, she enrolled in Universiti Sains Malaysia for her Bachelor's Degree in 2013. She took an Industrial Technology program with major Bioresource, Paper and Coatings. Then, she obtained her Bachelor's Degree in Technology in 2017. Nur Amira has been not just an ordinary student. She was very active during her student life and used to be a player and manager of the USM Ladies Hockey team. She also became the Head of Exhibition of Novel Research and Innovation Competition 2015 and a few positions during her student year.

She is now pursuing her Master's Degree of Science at the Institute of Tropical Forest and Forest Products (INTROP) in Universiti Putra Malaysia. Her research is specifically on Malaysian hardwood species and preservative treatment methods. Besides, her research also into structural design and evaluating performance of laminated timbers.

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