

SYNTHESIS AND CHARACTERIZATION OF  
BIO-ADHESIVE USING NATURAL RUBBER  
LATEX AND BIO-BASED FILLERS FOR  
MEDIUM DENSITY FIBREBOARD  
MANUFACTURING

JAYSHREE THURAISSINGAM

MASTER OF SCIENCE

UNIVERSITI MALAYSIA PAHANG



## **SUPERVISOR'S DECLARATION**

We hereby declare that we have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Master of Science.

---

(Supervisor's Signature)

Full Name : DR. ARUN GUPTA

Position : PROFESSOR

Date :

---

(Co-supervisor's Signature)

Full Name : DR. BIJARAMI

Position : SENIOR LECTURER

Date :



## **STUDENT'S DECLARATION**

I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

---

(Student's Signature)

Full Name : JAYSHREE THURASINGAM

ID Number : MKC 15013

Date :

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## ABSTRAK

Pengeksploitasian berlebihan terhadap perekat sintetik dalam industri pembuatan komposit kayu telah menjadi satu ancaman terhadap alam sekitar dan kesihatan pengguna berikutan pembebasan wap formaldehid yang bersifat karsinogenik. Kajian ini berfokus pada penciptaan dan pengaplikasian bio-perekat sebagai alternatif bagi perekat sintetik. Kanji beras (RS), lignin industri (IL) dan lignin daripada hidrolisis enzim (EHL) digunapakai sebagai pengisi dan diperbadankan ke dalam susu getah asli (NRL) melalui kaedah pengadunan. RS dimodifikasi secara kimia menggunakan asid hidroklorik (HCl) dan natrium hidroksida (NaOH) sebelum diadun. Hidrolisis asid menggunakan 1M HCl pada 55°C selama 60 minit dan hidrolisis alkali menggunakan 2M NaOH selama 60 minit pada 45°C berjaya menghasilkan modifikasi RS yang memiliki kekuatan ricih masing-masing sebanyak 0.38MPa dan 0.43MPa. RS yang termodifikasi oleh asid dan alkali telah diadun dengan NRL mengikut kandungan peratusan berat yang berlainan (0%,25%,50%,75% dan 100%). Bio-perekat yang terhasil telah dicirikan secara fizikal bagi aspek masa gel, kelikatan dan kandungan pepejal dan semua formulasi bio-perekat diaplikasikan dalam pembuatan gentian kayu berketumpatan sederhana (MDF) melalui penekanan panas pada 180°C selama 4 minit. Kesemua spesimen gentian kayu diperincikan secara mekanikal dari segi kekuatan modulus kehancuran(MOR), kekuatan modulus kekenyalan(MOE), ikatan dalaman(IB) dan pengembangan ketebalan(TS).Kestabilan terma dianalisa melalui ujian termogravimetri manakala tindak balas kimia dianalisa melalui spektroskopi inframerah (FTIR) dan ujian mikroskopik pengimbasan elektron digunakan untuk analisa morfologi. Formula bio-perekat yang mengandungi pengisi RS yang terawat oleh hidrolisis asid dan alkali pada peratusan berat NRL:RS (75/25) berjaya mempamerkan MOR sebanyak 26.4 dan 36.7MPa masing-masing manakala bagi IB, nilai yang direkodkan ialah 0.39MPa dan 0.66MPa. Pengisi RS yang dirawat oleh alkali menunjukkan prestasi mekanikal yang lebih baik berbanding rawatan asid.Analisa terma membuktikan bahawa RS dalam peratusan berat 75/25 mencatatkan suhu kehilangan jisim yang lebih tinggi sekaligus menjelaskan bahawa perbadanan RS dalam NRL menambahbaik kestabilan terma dan pengoksidaan bio-perekat ini. Kajian ini juga merangkumi penciptaan dan pengaplikasian bio-perekat daripada NRL yang diperisi dengan EHL dan IL. Analisa awal menggunakan FTIR dijalankan untuk mengenalpasti keserasian kumpulan berfungsi yang wujud antara EHL dan IL. Pengadunan antara NRL, EHL dan IL dibuat menggunakan kandungan peratusan berat yang sama seperti RS dan perincian fizikal, mekanikal, terma dan morfologi yang sama turut diamalkan.Sampel gentian kayu yang diperbuat daripada bio-perekat yang mengandungi NRL-EHL menggambarkan kemerosotan pada MOR dan IB di mana pengisian EHL sebanyak 100% mengikut berat telah mengurangkan nilai MOR kepada 4MPa berbanding 15MPa yang tercapai melalui pengisian EHL sebanyak 25% peratus berat, berikutan ketidakserasian antara EHL dan NRL. Kaedah pengaktifan EHL melalui 3M NaOH telah dibuat bagi meningkatkan keupayaan bio-perekat ini. Formulasi bio-perekat yang mengandungi NRL dan EHL teraktif dalam nisbah yang sama (50/50) berjaya menjangkau nilai minima yang ditetapkan oleh piawai ASTM melalui penghasilan MDF dengan MOR sebanyak 36MPa dan IB sebanyak 0.75MPa. Perbadanan EHL teraktif memberikan prestasi keseluruhan yang lebih baik berbanding RS dan EHL tidak teraktif dan kajian ini berjaya membuktikan bahawa pengisi semulajadi boleh digunapakai dalam pembentukan bio-perekat berasaskan NRL yang lebih hebat dan selamat berbanding perekat sintetik secara meluas.

## ABSTRACT

The over-exploitation of synthetic adhesives in wood composite manufacturing industries has become a major threat to environment and consumers' health due to its carcinogenic formaldehyde emissions. In order to overcome this issue, this study has been focused towards the development of bio-based adhesive from natural rubber latex (NRL) incorporated with rice starch (RS) and lignin as the fillers, by blending technique. These fillers are vital in order to enhance the mechanical properties of NRL and thus, their respective compatibility with the base material upon blending is compared. Prior to the blending, RS were subjected to chemical modification using hydrochloric acid (HCl) and sodium hydroxide (NaOH). Acid hydrolysis of 1M HCl for 60 minutes at 55°C and alkaline hydrolysis using 2M NaOH for 60 minutes at 45°C yielded modified RS with the best working shear strength of 0.38MPa and 0.43MPa respectively. Both acid and alkali modified RS were blended with NRL in weight content of 0%, 25%, 50%, 75% and 100% accordingly. The bio-adhesives were characterized physically into gel time, viscosity and solid content. Consequently, all formulations were applied for medium density fibreboard (MDF) manufacturing via hot pressing at 180°C for 4 minutes and board specimens were characterized mechanically in terms of modulus of rupture (MOR), modulus of elasticity (MOE), internal bonding (IB) and thickness swelling (TS). Thermal stability is analyzed through thermogravimetric analysis (TGA) while the chemical interaction that resulted through incorporation of fillers with NRL is studied through Fourier transform infrared spectroscopy (FTIR) and the morphological analysis is observed through scanning electron microscopy (SEM). The results indicated that bio-adhesive formulation that contains the highest by weight content of acid and alkali modified RS exhibited outstanding MOR of 26.4MPa and 36.7MPa while IB of 0.39MPa and 0.66MPa respectively. As an overall, alkali modified RS exhibited greater mechanical adhesive performances compared to HCl modified RS. Moreover, thermal analysis proved that RS content by weight of 75% in bio-adhesive formulation exhibited significant weight loss at 361.73 °C, indicating that incorporation of RS actually enhances the thermal oxidative stability of bio-adhesive. The second part of this study revolves around the development and application of bio-adhesive from NRL, incorporated with EHL. Blending with NRL was performed by varying filler weight content (0%, 25%, 50%, 75%, 100%). Similar physical, mechanical, morphological and thermal analysis were investigated. Samples bonded with NRL-EHL bio-adhesives showed deteriorating effect on the MOR and IB values of MDF, where MOR declined from 15MPa to 4MPa when EHL loading was increased from 25wt% to 100wt% due to incompatibility between EHL and NRL matrices. Alkaline activation of EHL using 3M NaOH was applied to improve the performance of EHL based bio-adhesive. Consequently, bio-adhesive formulation with equivalent weight content of NRL and NaOH activated EHL exceeded ASTM standard requirement by yielding MDF with MOR of 36 MPa. The IB recorded was 0.75MPa which proved that alkaline modification of EHL enhanced the dispersion of filler into NRL matrix and caused better cross-linking between bio-polymers. Thoroughly, alkaline activated EHL incorporation into NRL formulation gave better physical, mechanical and thermal performance of the bio-adhesives compared to modified RS and EHL. As a conclusion, this study serves as a platform to prove that bio-polymers such as RS and EHL can be viable filler for NRL, with activated EHL to be the best option of fillers.

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## LIST OF ABBREVIATIONS

NRL	Natural Rubber Latex
RS	Rice Starch
MOR	Modulus of Rupture
MOE	Modulus of Elasticity
IB	Internal Bonding
TS	Thickness Swelling
SEM	Scanning electron microscopy
IR	Infrared Spectra
ASTM	American Standard of Testing Methods
MDF	Medium Density Fibreboards
EHL	Enzyme Hydrolysed Lignin
IL	Industrial Lignin
TGA	Thermogravimetry Analysis
cP	centiPoise

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