Effects of Several Synthesis Conditions on Bond Strength of Plywood Adhered with Natural Rubber Latex – Styrene Adhesive

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Abstract

This report discusses the effects of synthesis conditions on bond strength of plywood adhered with natural rubber latex - styrene adhesive. Synthesis variables observed were catalyst (with and without catalyst), pre-stirring (0 and 3 hours) and heating time (1 and 2 hours). Three-ply plywood samples were prepared from Albizia (*Paraserianthes falcataria*) and Red meranti (*Shorea*, sp.) veneers and their gluability were evaluated in accordance to Indonesian Standard (SNI 01-2704-1992). The results revealed that synthesis conditions (i.e. catalyst, pre-stirring and heating time) did not significantly influence the bond strength of plywood. Natural rubber latex – styrene adhesive is very appropriate as adhesive for Red meranti plywood. Nevertheless, it can be used as limited interior application for Albizia plywood. An addition of 10% phenol formaldehyde (PF) in natural rubber latex - styrene adhesive slightly improved the exterior type bond strength of Red meranti plywood.

Key words: natural rubber, styrene, synthesis, adhesive, plywood, bond strength.

Introduction

The majority of wood panel adhesives currently used are based on four major synthetic thermosetting resins, phenol formaldehyde (PF), resorcinol formaldehyde (RF), urea formaldehyde (UF) and melamine formaldehyde (MF). These four resins are all derived from fossil fuels. Phenol and resorcinol are derivatives of benzene which is synthesize from oil; urea, melamine and formaldehyde are all derivatives of natural gas (Koch *et al* 1987). Efforts should be done in seeking alternative adhesives that might serve as substitutes for conventional fossil fuel-based adhesives.

Natural rubber (NR), which has long been known as wood adhesive, is one of alternatives to substitute fossil fuel-based adhesives. Unfortunately, natural rubber is characteristically low in adhesion and cohesion, has poor resistance to stress and heat and fair resistance to moisture (Marra 1992). On the other side, it has an excellent resistance to organisms and it can make instant bond with a relatively low pressure (Marra 1992). It is also quite cheap and abundantly available in Indonesia.

One of efforts to upgrade the natural rubber (NR) latex as wood adhesive is by copolymerization of NR latex with methyl methacrylate (MMA) or with styrene monomer. Heveaplus MG is a grafted copolymer based on natural rubber that has been available for many years. The products, symbolized as MG 30 and MG 49, are graft copolymers containing percentage amounts of methyl methacrylate indicated by the numbers (Pendle 1987; Gazeley and Wake 1990). By far the widest application of MG latexes is in adhesives. These graft latexes exhibit very good adhesion to a wide range of substrates including polyvinyl chloride, nitrile rubber, leather and poly propylene. On the other hand, there is not any report on the use of NR grafted with styrene as a commercial product available in the market. Majority of the reports are regarding the characteristics of the polymer. There were only few reports concerning the application of this adhesive in plywood (Hartoyo and Utama 1995; Santoso and Utama 1997; Hermiati *et al* 2000[a]; Hermiati et al 2000[b]). According to the reports, NR latex – styrene adhesive could only produce interior grade plywood. Besides, there is not very much information regarding optimum synthesis condition to produce NR latex – styrene, which has good bond strength quality as plywood adhesive.

Due to these facts, bond strength testing was conducted to be able to see if there was any effect of catalyst, pre-stirring and heating time on bond strength of the polymer as plywood adhesive. Besides that, there are some other factors that might affect bond strength of the polymer such as species of wood and the addition of phenolic compound to enhance the bond strength. Therefore, these factors were also investigated and discussed in this report.

Materials and Methods

The concentrated NR latex of 61~62% solid content was obtained from the Jalupang plantation of PTPN VIII, Subang, West Java. It was diluted with water to about 25% solid content. Styrene monomer used was technical grade, while potassium peroxodisulfate and ferro sulfate were analytical grade from E. Merck. Red meranti (*Shorea* sp.) veneers were from East Kalimantan, while Albizia (*Paraserianthes falcataria*) veneers were from Sukabumi, West Java. The research was conducted at the R & D Unit for Biomaterials-Indonesian Institute of Sciences, Cibinong from January until December 2004.

Polymer Synthesis

An emulsifier and styrene monomer were added to 250 g of the diluted NR latex. The mixture was then stirred at 550 rpm at room temperature for 0 and 3 hours. After that, the initiator (potassium peroxodisulfate) was added. Half of the samples were also treated with 0.6 ml of 1% ferro sulfate solution as a catalyst. The mixture was then heated at 65°C for 1 or 2 hours with continuous stirring at 550 rpm. Each treatment was conducted in three replications.

Adhesive Characterization and Formulation

The copolymer produced was measured for its total solid content (gravimetric method), pH, specific gravity (using picnometer) and viscosity (using Brookfield viscometer). Solidification method or hardening mechanism of the polymer as an adhesive was conducted using Seiko Instruments TG/DTA (Thermo gravimetry / Differential Thermal Analyzer). About 6 mg of sample was put in TG/DTA aluminum pan for the analysis. The temperature was programmed from 30 to 250°C and was raised 10°C/minutes. Nitrogen was used as purge gas.

There were two types of adhesives prepared, namely adhesive *A* and adhesive *B*. Adhesive *A* was the NR latex-styrene polymer only. Adhesive *B* was NR latex-styrene with 10% (w/w) of phenol formaldehyde.

Plywood Preparation and Bond Strength Testing

Plywood samples (3 replications) were prepared using veneers of Red meranti and Albizia. The veneers dimensions were 250 x 250 x 2.1 mm (core) and 250 x 250 x 1.5 mm (face and back). Adhesive *A* was applied on both veneers, while adhesive *B* on veneers of Red meranti. The adhesives were applied at the glue spread rate of 400 g/m^2 DGL (double glue line). The amount used was based on our previous study (Hermiati *et al* 2000[b]). The veneers with adhesive were cold pressed at 10 kg/cm^2 for 10 minutes and then hot pressed at 10 kg/cm^2, 120°C (Adhesive *A*) or 130°C (Adhesive *B*) for 5 minutes. The plywood produced was measured for its bond strength according to the Indonesian National Standard (SNI 01-2704-1992). There are 4 grades of plywood according to the standard, Exterior I, Exterior II, Interior I and Interior II. Standard testing procedure for Exterior I grade involves boiling plywood specimens for 4 hours, drying in an oven at 60°C for 20 hours, boiling for 4 hours and dipping in water for 20 hours, while that for Exterior II grade is the same except dipping in water only until the specimens reach ambient temperature. Standard testing procedure for Interior I grade involves dipping plywood specimens in water at 60°C for 3 hours, while that for Interior II grade is without any treatment. All the specimens (3 specimens for each treatment and each testing grade) were tested for bond strength using Shimadzu Universal Testing Machine. Bond strength value to pass the standard of each grade is $\geq$7 kgf/cm^2.

Statistical Analyses

Statistical analysis for bond strength data was conducted using SAS (Statistical Analysis System). F test (α = 0.05) was observed to see the effects of synthesis conditions on bond strength of the two types of adhesives and Duncan Multiple Range Test (α = 0.05) was observed to see the effects due to the difference of a certain synthesis condition levels.

Results and Discussions

Physico-chemical Properties of Adhesive

Total solid content, specific gravity and viscosity of NR latex-styrene produced from different synthesis conditions ranged from 39.56~39.93%, 0.99~1.00, and 31~36 cp, respectively. This suggested that properties of natural rubber latex-styrene were not affected by the differences of synthesis conditions (Table 1).

There are four types of hardening mechanism of adhesive, namely loss of solvent or liquid carrier, loss of heat, chemical reaction and combinations (Marra 1992). The TG line of the graph of TG/DTA for NR latex - styrene polymer (Figure 1) showed that the weight of the sample remained the same after all the solvent was evaporated. This suggests that hardening mechanism of NR latex-styrene was only due to loss of solvent or liquid carrier. This was in accordance with Marra (1992) who mentioned that natural rubber was one of solvent and water loss systems adhesives.

Table 1. Effects of catalyst, pre-stirring and heating time on physico-chemical properties of NR latex – styrene

Synthesis conditions		Total Solid (%)	Specific Gravity	Viscosity (cp)
Catalyst	w/o catalyst	39.93	0.99	36
	w/ catalyst	39.56	1.00	31
Pre-stirring	0 hour	39.58	1.00	35
	3 hours	39.90	1.00	32
Heating time	1 hour	3960	1.00	31
	2 hours	39.88	1.00	36

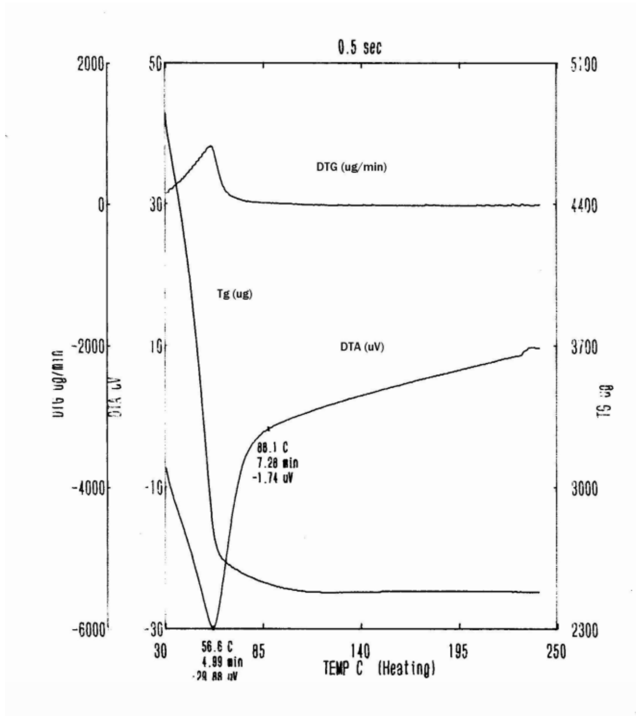

Figure 1. TG/DTA trace of NR latex – styrene.

Bond Strength of NR Latex-styrene as Plywood Adhesive

The results of Interior II Grade bond strength testing (Table 2) shows that addition of catalyst, prolonged pre-stirring and prolonged heating time increased bond strength value of Red meranti as well as that of Albizia plywood. However, the improvements were not statistically siginificant. The results of Duncan Multiple Range (DMR) test also confirmed that there was not any significant difference of bond strength due to different level of catalyst, pre-stirring or heating time. However, catalyst gave significant effects on bond strength of Red meranti and Albizia as a whole. The addition of catalyst significantly increased bond strength of plywood tested for Interior II grade. In Table 2 it can also be seen that all the polymers of different synthesis conditions and used as adhesives in this study could meet standard of bond strength as the Interior II grade plywood ($\geq$ 7 kgf/cm^2).

The results of statistical analyses for Interior I grade plywood (Table 3) was almost the same as that for Interior II grade. There was not any significant effect of either catalyst, pre-stirring or heating time on bond strength of Red meranti or Albizia plywood or Red meranti and Albizia plywood as a whole tested for Interior I grade. However, all of Red meranti plywood could pass the Interior I grade test, while Albizia plywood could not. Therefore, Albizia plywood was not tested for Exterior grade plywood.

Bond strength of Red meranti plywood at each treatment was significantly higher than that of Albizia plywood, either in Interior II or Interior I grade testing (Table 2 and Table 3). Wood species is one of so many factors that affects gluing. This might be because each species of wood has different properties, such as density, pH, moisture content and chemical contents. Even, those wood properties could be different at the same species. This is supported by Marra (1992) that stated the behavior of wood varies widely and is strongly consequential affecting the behavior of adhesives variously, even within individual pieces. Besides that, veneers of Red meranti and Albizia were obtained from different wood manufacturers. Eventhough both veneers have almost the same moisture content (9.82% - Red meranti and 10.41% - Albizia) and almost the same density (0.43 g/cm^3 - Red meranti and 0.42 g/cm^3 - Albizia), they might have different surface quality due to different cutting or veneering methods which further affect gluing quality.

The results of exterior grade bond strength testing of Red meranti plywood in Table 4 showed that there was not any significant effect of different adhesive synthesis conditions on bond strength of plywood. However, according to the SNI standard, there were some samples that could not pass the minimum standard value for bond strength, that is 7 kgf/cm^2. The samples are those that were produced using adhesives synthesized without catalyst, without pre-stirring or with shorter heating time. These samples could not pass the

Exterior II grade testing, but could pass the Exterior I grade testing. This is quite interesting since bond strength testing for Exterior I grade was longer and more severe than that for Exterior II grade. This contradiction was probably due to the characteristic of the adhesive, which was based on rubber, that has a poor resistance to heat (Marra 1992). In Exterior II grade bond strength testing, plywood specimens were tested right after they were boiled and then reach ambient temperature after soaking in water, while in that of Exterior I grade the specimens were tested after 20 hours soaking in water at ambient temperature. Therefore, in the last case the adhesive might have enough time to recover itself from heat effect during boiling, thus produce better bond strength.

Although most of plywood samples of Red meranti could pass the Exterior grade plywood standard, an addition of 10% phenol formaldehyde (PF) in NR latex-styrene would increase bond strength (Figure 2 and 3), thus ensure that the adhesive could pass either Exterior II or Exterior I grade standard. This positive effect of PF was probably due to cross-linking between reactive sites (aldehyde group) of PF and α–methyl or α–methylene group of polyisoprene chains. Blend of NR latex-styrene and PF also combine the strength, durability and adhesiveness of phenolics with the elastomeric properties of rubber, and produce a more tough, and durable bond.

Table 2. Duncan Multiple Range test of bond strength value (kgf/cm^2) for Interior II grade plywood (α = 0.05).

Variabel	Level	Red meranti	Albizia	Red meranti and Albizia
Catalyst	W/o catalyst	12.74	9.25	10.99
	With catalyst	14.72	10.31	12.52
Critical range		2.20	1.88	1.33
Pre-stirring	0 hr	12.64	9.60	11.12
	3 hrs	14.82	9.96	12.39
Critical range		2.20	1.88	1.33
Heating time	1 hr	14.39	8.56	11.48
	2 hrs	13.07	11.00	12.03
Critical range		2.20	1.88	1.33

Table 3. Duncan Multiple Range test of bond strength value (kgf/cm^2) for Interior I grade plywood (α = 0.05).

Variabel	Level	Red meranti	Albizia	Red meranti and Albizia
Catalyst	W/o catalyst	7.17	4.08	5.62
	With catalyst	9.11	4.43	6.77
Critical range		2.42	2.49	1.60
Pre-stirring	0 hr	8.37	3.79	6.08
	3 hrs	7.90	4.72	6.31
Critical range		2.42	2.49	1.60
Heating time	1 hr	9.12	4.77	6.94
	2 hrs	7.16	3.74	5.45
Critical range		2.42	2.49	1.60

Table 4. Duncan Multiple Range test of bond strength value (kgf/cm^2) for Exterior grade red meranti plywood (α = 0.05)

Variabel	Level	Exterior I	Exterior II
Catalyst	W/o catalyst	9.90	6.28
	With catalyst	7.08	7.83
Critical range		3.89	3.26
Pre-stirring	0 hr	8.58	6.80
	3 hrs	8.23	7.38
Critical range		3.89	3.26
Heating time	1 hr	9.38	6.48
	2 hrs	7.27	7.65
Critical range		3.89	3.26

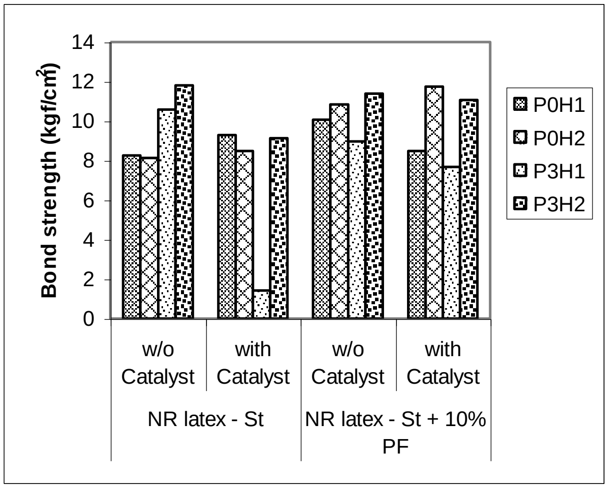

Fig. 2. Bond strength of Red meranti plywood (Ext. I grade testing), P = Pre-stirring; H = Heating time

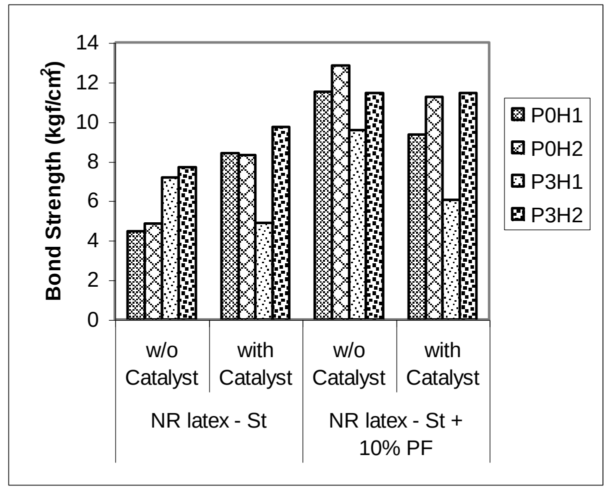

Fig. 3. Bond strength of Red meranti plywood (Ext. II grade testing), P= Pre-stirring; H= Heating time

Conclusions

Variables in synthesis conditions of NR latex-styrene, such as catalyst, pre-stirring and heating time, did not significantly affect bond strength of the polymer as plywood adhesive. Thus, it is suggested that the more simple, shorter and milder synthesis condition of NR latex-styrene, that is without catalyst, without pre-stirring and 1 hour heating can be used to produce plywood adhesive. Application of the adhesive on Albizia veneers could only produce plywood samples that could meet bond strength standard of Interior II grade, while that on Red meranti veneers could meet the standard of either Interior or Exterior grade. The addition of 10% phenol formaldehyde (PF) in NR latex styrene could slightly increase bond strength and ensure that all the red meranti plywood could pass Exterior I and Exterior II grades plywood.

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