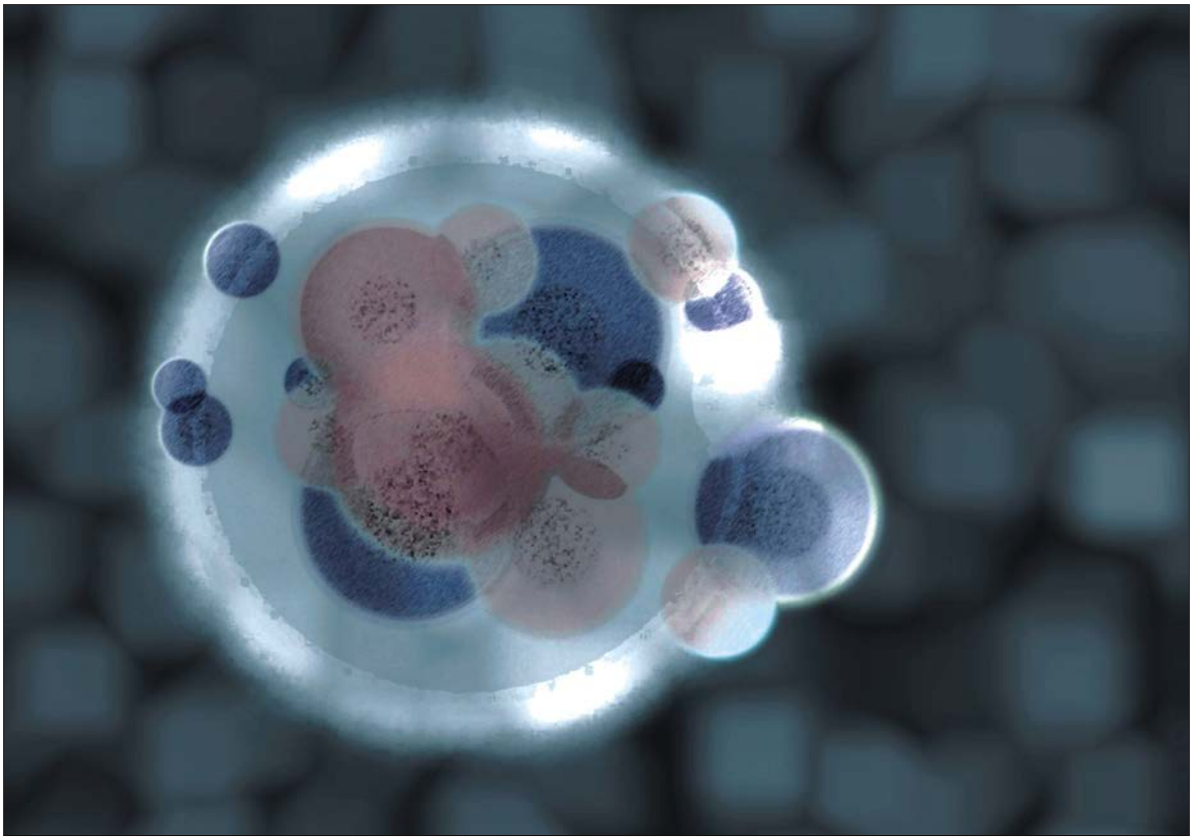

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# *Trans*-1.4-Isoprene Rubber as Hot Melt Adhesive

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**Abstract:** The use of maleic anhydride (MAH) modified and unmodified natural and synthetic *trans*-1.4-isoprene rubber (TIR Gutta Percha and TIR Synthetic) as hot-melt adhesives for plywood manufacturing under various glue spread levels was investigated and their gluability and durability in accordance to SNI 01-5008.2-99 standard were evaluated. TIR modified with MAH (MTIR) either natural (MTIR Gutta Percha) or synthetic (MTIR Synthetic) adhesives could reveal a satisfactory waterproof gluability. This is considered to be due to an enhanced adhesion caused by an occurrence of esterification between the hydroxyl groups of wood and the acid anhydride groups of MTIR during hot pressing. Infra red spectroscopic analysis showed both TIR Gutta Percha and TIR Synthetic have almost similar spectra. In the spectra of MTIR's after purification, the peak at 1724 and at 1716 $cm^{-1}$, respectively for MTIR Synthetic and MTIR Gutta Percha existed, indicated the characteristic for C=O of MAH. Differential thermal analysis showed that before and after modified with MAH, the melting and decomposition temperatures of MTIR Gutta Percha and MTIR Synthetic adhesives were quite different from TIR Gutta Percha and TIR Synthetic adhesives indicated that chemical modification of TIR's with MAH were occurred. Either MTIR Gutta Percha or MTIR Synthetic adhesives are very appropriate for wood as hot melt adhesives.

**Key words:** Modification, TIR Gutta Percha, TIR Synthetic, maleic anhydride, hot-melt adhesive, plywood

## INTRODUCTION

Adhesive commonly used for bonding wood based panel products such as plywood, particleboard, Oriented Strand Board (OSB), fiberboard etc is mainly thermosetting resins (i.e., urea formaldehyde, phenol formaldehyde and melamine formaldehyde). Besides derived from petroleum which is a non renewable resource and limited available, these resins are also not environmentally friendly material (i.e., formaldehyde release). It has been known that formaldehyde is a probable human carcinogen when inhaled or ingested. In fact: Breathing even small amounts of formaldehyde may increase the risk of contracting lung and nasal cancer. Furthermore, chronic formaldehyde exposure can cause menstrual disorders and pregnancy problems in women workers exposed to higher levels. Short-term inhalation exposure can result in eye, nose and throat irritation and respiratory symptoms. The level of formaldehyde emission from any new product is time dependant.

On the other hand, the hot melt adhesives (HMAs), which have the merit of high performance of adhesion, are used widely in various fields of industries, for example packaging, book binding, wood working, plywood manufacturing and so forth (Han, 1990). The base polymers used in HMAs are polypropylene, polyamide, polyester and ethylene-vinyl acetate copolymer. More over, the used of a block copolymer (i.e., styrene-butadiene-styrene, styrene-isoprene-styrene and styrene-ethylene/buthylene-styrene) as HMA has led to enhancement of adhesion. The graft polymerization of monomers which have polar groups onto these block copolymers gives improved adhesion obviously. Furthermore, the graft polymerization of unsaturated carboxylic acids (i.e., acrylic acid, maleic acid, maleic anhydride and so forth) onto homopolymers and block copolymers has been revealed as an effective method which improves adhesion. Han (1990) had been evaluated polypropylene (PP) and PP modified with maleic anhydride (MAH) so called MPP as HMAs for wood. It

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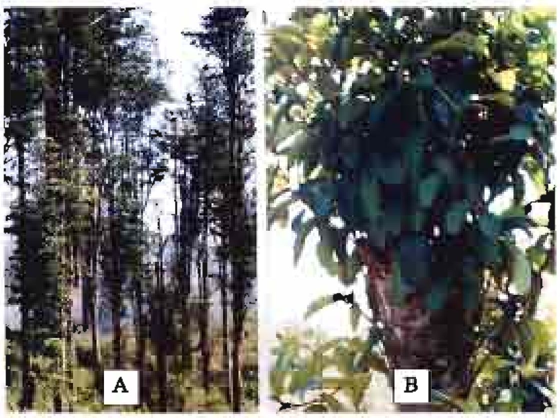


Fig. 1: (A) Trees and (B) Leaves of *Palaquium oblongifolium*

was proved that MPP could reveal a satisfactory waterproof gluability.

One of the environmentally friendly elastomer materials is *trans*-1.4-isoprene rubber (TIR). Natural TIR is biodegradable in nature and it is known as Balata or Gutta Percha. Gutta Percha was obtained from Palaquium and Payena trees through extraction of the leaf or tapping of the stem, particularly from *Palaquium gutta* and *Palaquium oblongifolium* trees (Fig. 1). Alfa *et al.* (1997) reported that Gutta Percha contained of 92% gutta, 1.5% dammar and 6.5% water. The basic color of Gutta Percha is white, but it can be turned to grayish or brownish when expose to the air (Benu *et al.*, 1979). TIR has a high level of crystallinity which leads to good strength properties at room temperature and it is easily to be processed at a temperature near or above the crystalline melting point. Furthermore, TIR is vulcanizable due to the presence of unsaturated C = C bonds. The raw polymer compounded with conventional compounding ingredients may be used without vulcanizing agents in a wide variety of applications where the strength at ambient temperature is important. Such application includes medical casts, adhesives and moldable sheets.

Modification of TIR Synthetic with MAH without initiator can be done easily in the kneader (Toyo Seiki Labo Plastomill) under certain conditions to produce MAH-modified TIR so called MTIR. The resulted product was then used as a compatibilizer for woof flour (WF) -TIR composites. The WF-TIR composites with addition small amount of MTIR showed excellent physical and mechanical properties (water absorption, thickness swelling, tensile strength, breaking elongation and Young's modulus). This was due to esterification between the hydroxyl groups of wood and the acid anhydride groups of MTIR (Febrianto *et al.*, 2001, 1999). The idea behind the present work is to apply analogous reactions as described in the previous publications using a biodegradable natural TIR (TIR Gutta Percha) and TIR Synthetic resins as hot melt adhesives for plywood manufacturing. The effect of adhesive type and glue spread level on the shear strength of Keruing (*Dipterocarpus* spp.) plywood was explored in accordance to Indonesian standard (SNI 01-5008.2-99).

## MATERIALS AND METHODS

**Materials:** Gutta Percha was supplied by PTPN VIII Cipetir West Java Indonesia, while TIR Synthetic was supplied by the Japan Synthetic Rubber Co., Ltd. The glass transition and melting temperatures of TIR Synthetic are reported to be 60 and 74°C, respectively (Nielsen, 1985). MAH was purchased from PT. Frisconina Indonesia and Benzoyl Peroxide (BPO) was purchased from Nacalai Testque Inc., Japan. Keruing veneer (*Dipterocarpus* spp.) with the size of 20×20×0.16 cm was supplied by PT. Sumalindo Lestari Jaya, East Kalimantan, Indonesia. The research was carried out in Laboratory of Bio-Composites, Forest Products Department, Faculty of Forestry, Bogor Agricultural University, Indonesia from January to July 2003. Prior to be used for plywood manufacturing, several veneers were randomly chosen to evaluate their moisture content, thickness and density. The moisture content, thickness and density of the veneer were in the range of 4.47-6.04%, 1.59±0.016 mm and 0.70 to 0.76 g $cm^{-3}$, respectively (Fig. 2-4).

**Modification of TIR Gutta Percha and TIR Synthetic with MAH:** Modification of TIR Gutta Percha and TIR Synthetic with 5% MAH were done in Labo Plastomill at 150 and 30 rpm for 3 min and continued at 70 rpm for 7 min. The obtained results TIR-modified- MAH (MTIR) were milled into a powder adhesive. Benzoyl peroxide (BPO) was added to the MTIR Synthetic in the glue spread process. The whole process for adhesive preparation and plywood manufacturing was presented in Fig. 5.

**TIR Gutta Percha, TIR Synthetic and MTIR analysis:** For infrared spectroscopic measurements and differential thermal analysis, a Shimadzu FTIR 8600 PC and Shimadzu DT-30 apparatus were used, respectively.

**Plywood manufacturing:** Three-ply plywood was prepared from Keruing veneer (*Dipterocarpus* spp.). TIR Gutta Percha and TIR Synthetic with or without modification were applied as adhesive with various glue spreads ranging from 175-250 g $m^{-2}$ (175, 200,

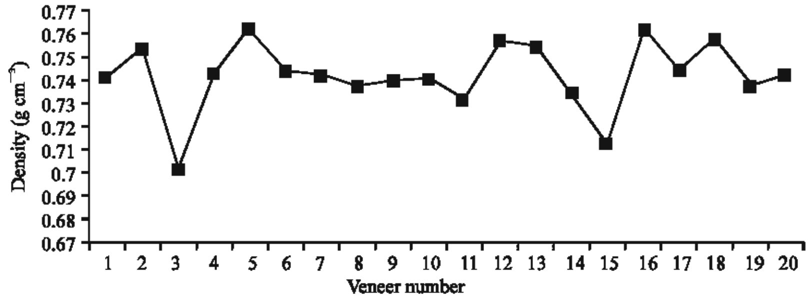


Fig. 2: Density (g cm$^{-3}$) of Keruing wood (*Dipterocarpus* spp.) veneer

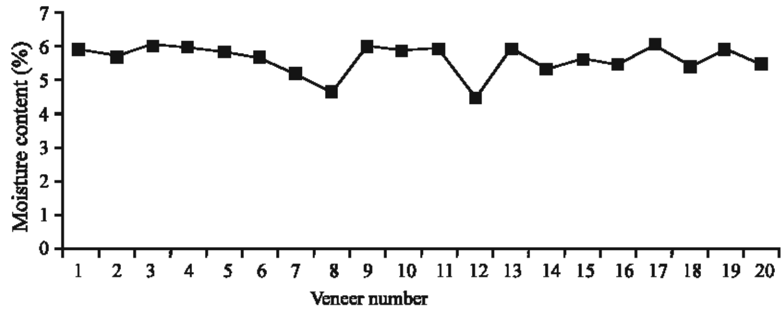


Fig. 3: Moisture content (%) of Keruing wood (*Dipterocarpus* spp.) veneer

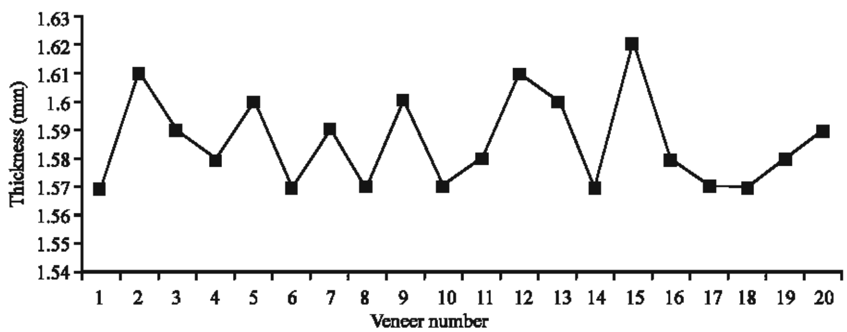


Fig. 4: Thickness (mm) of Keruing wood (*Dipterocarpus* spp.) veneer

225, 250 g m$^{-2}$). The pressing conditions were 120°C, 0 to 20 kg cm$^{-2}$ pressure for 10 min. The first 3 min was pressed without pressure and the remained 7 min with 20 kg cm$^{-2}$ pressure. Then, the plywood was conditioned in room temperature for 1 week prior to evaluation.

**Plywood properties evaluation:** Evaluation of plywood properties (moisture content, density and bonding strength) were measured in accordance to Indonesian Plywood Standard (SNI 01-5008.2-99). Evaluation of shear strength and wood failures were done in dry and wet conditions. Four types of plywood were evaluated namely interior II, interior I, exterior II and exterior I. The procedures are as followed:

**Interior II type:**

- Shear strength test was carried out in dry conditions without any treatments.

**Interior I type:**

- Samples were immersed in hot water at 60±3°C for 3 h
- Samples were dipped in cold water until reached room temperature
- Shear strength test was done in wet condition

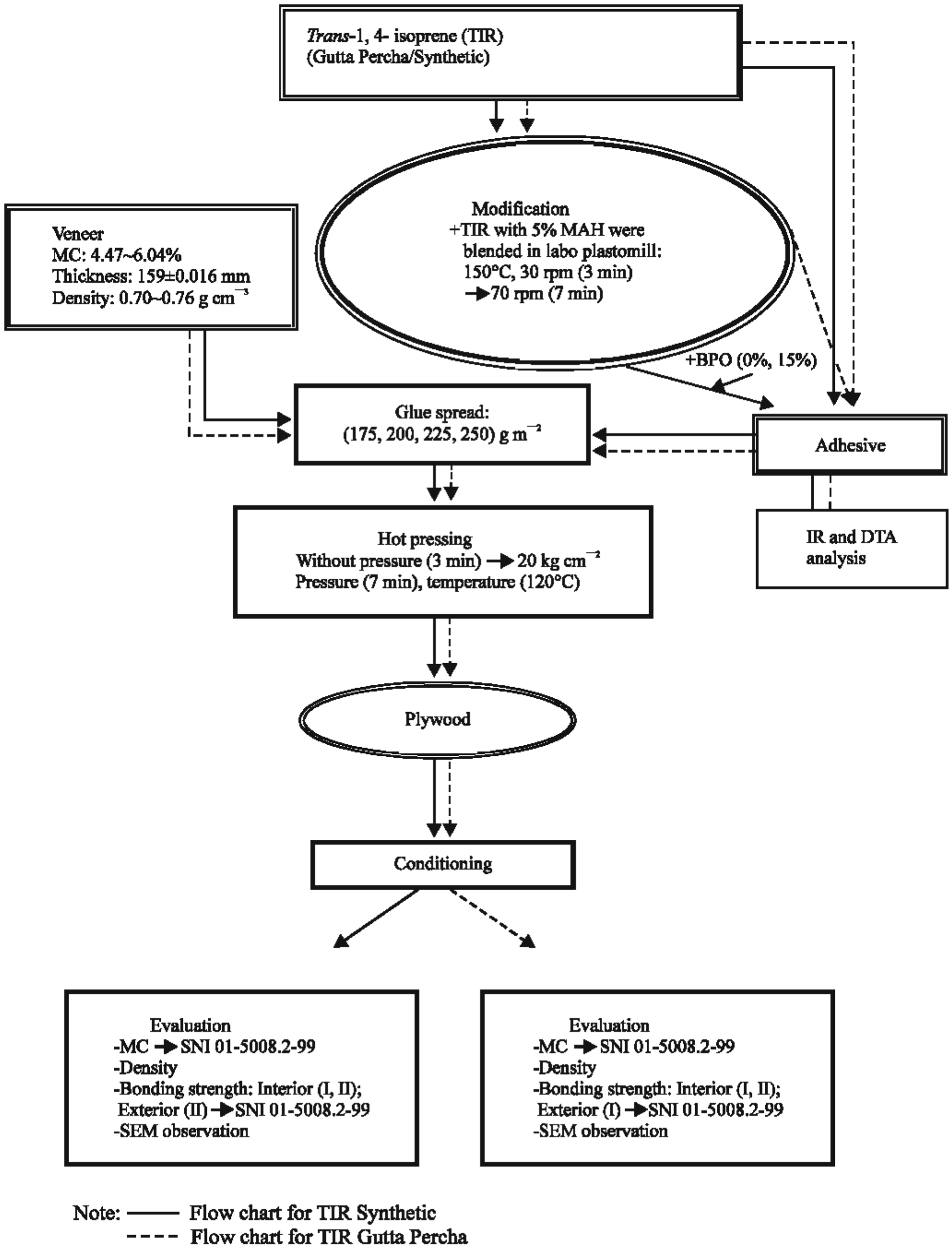


Fig. 5: Flow chart for plywood manufacturing using modified TIR Gutta Percha and TIR Synthetic as adhesive

**Exterior II type:**

- Samples were immersed in boiling water for 4 h.
- Samples were then oven dried at 60±3°C for 20 h
- Samples were immersed in boiling water for 4 h.
- Samples were dipped in cold water until reached room temperature
- Shear strength test was done in wet condition.

**Exterior I type:**

- Samples were immersed in boiling water for 4 h.
- Samples were then oven dried at 60±3°C for 20 h.
- Samples were immersed in boiling water for 4 h.
- Samples were immersed in cold water for 20 h.
- Shear strength test was done in wet condition.

**SEM observation:** The morphology of the sample fractures after shear strength evaluation was studied by using a JEOL JSM-T330A scanning electron microscope. SEM photographs were taken under the following conditions: working distance 15 mm and accelerating voltage of 10 kV.

## RESULTS AND DISCUSSION

**Characteristics of TIR and MTIR:** The appearance of the TIR Gutta Percha and TIR Synthetic before and after modified with 5% MAH were presented in Fig. 6. The color of TIR Gutta Percha and TIR Synthetic are white color (No. 1 and 3). The color of MTIR Synthetic, MTIR Gutta Percha and MTIR Synthetic with BPO are creamy (No. 4), brown (No. 2) and creamy to yellowish (No. 5) colors, respectively.

Figure 7 showed the infra red spectra of TIR Gutta Percha and TIR Synthetic before and after modified with 5% MAH. The spectra from TIR Gutta Percha and TIR Synthetic showed almost similar spectra. After they were modified with MAH (MTIR) and purified in which the unreacted of MAH was removed, a new peak occurred both in MTIR Gutta Percha and MTIR Synthetic at 1716 and 1724 $cm^{-1}$, respectively. These peaks indicated the C = O of MAH. Similar result has been reported that the C = O of MAH occurred at 1790 $cm^{-1}$ from the xylene unextractable residue of WF-MTIR-TIR composites (Febrianto *et al*., 1999). It was reported that the peak of C = O occurred at the wave length of 1800-1650 $cm^{-1}$ (Creswell *et al*., 1982).

Table 1 showed the melting and decomposition temperatures of TIR Gutta Percha and TIR Synthetic before and after modified with 5% MAH. The melting and decomposition temperature of TIR Gutta Percha was somewhat lower compare to TIR Synthetic. The value of the melting and decomposition temperature for TIR Gutta Percha and TIR Synthetic were 72.64, 482.23, 74.14°C and 490.50°C, respectively. After they were modified with MAH with or without BPO, the melting and decomposition temperatures of TIR Synthetic decreased. Conversely, for TIR Gutta Percha adhesive addition of MAH resulted in increasing the melting temperature and decreasing decomposition temperature. This is probably due to TIR Gutta Percha contained dammar. Alfa *et al*. (1997) reported that Gutta Percha contained of gutta (92%), dammar resin (1.5%) and water (6.5%). In all cases, after modified with 5% MAH, the melting and decomposition temperatures had changed. These results indicated that chemical reaction took placed when TIR Gutta Percha or TIR Synthetic adhesives were modified with MAH resulted in MTIR as a new substance.

**Plywood properties:** Evaluation of the plywood properties (i.e., moisture content, density and bonding strength) was done under various adhesive types and various level of glue spreads. TIR Gutta Percha, TIR Synthetic, MTIR Gutta Percha, MTIR Synthetic and MTIR Synthetic with BPO were used as adhesives. Glue spread was varied from 175 to 250 g $m^{-2}$ (i.e., 175, 200, 225 and 250 g $m^{-2}$). The

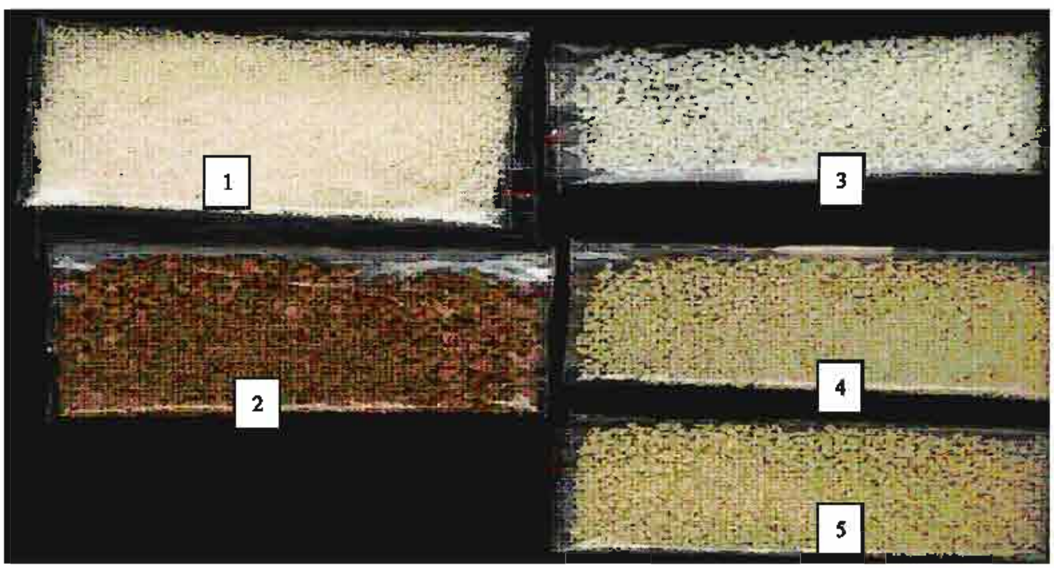


Fig. 6: Appearance of MAH modified and unmodified TIR. (1) TIR Gutta Percha ; (2) MTIR Gutta Percha; (3) TIR Synthetic; (4) MTIR Synthetic and (5) MTIR Synthetic + BPO

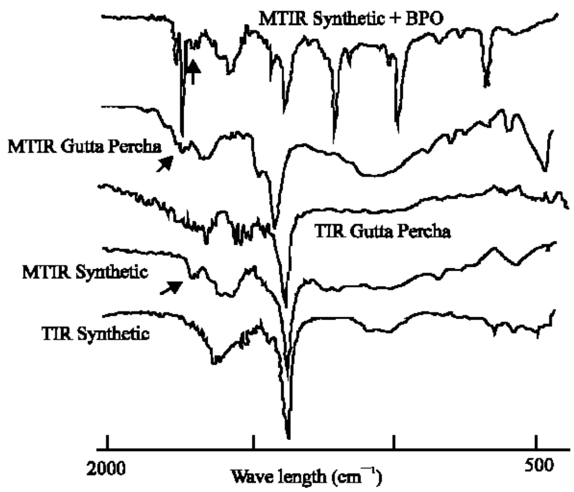


Fig. 7: IR spectra of TIR Gutta Percha and TIR Synthetic before and after modified with MAH

Table 1: Melting and decomposition temperatures of TIR Gutta Percha and TIR Synthetic before and after modified with MAH

| Adhesives | Melting temperature (° C) | Decomposition temperature (°C) |
|---|---|---|
| TIR Gutta Percha | 72.64 | 482.23 |
| MTIR Gutta Percha | 79.03 | 469.47 |
| TIR Synthetic | 74.14 | 490.50 |
| MTIR Synthetic | 71.36 | 488.53 |
| MTIR Synthetic + BPO | 70.87 | 482.56 |

pressing conditions were as followed: pressing temperature was 120°C and pressing time was 10 min (the first 3 min without pressure and 7 min. with pressure of 20 kg cm$^{-2}$).

**Moisture content:** The moisture content of the plywood bonded with TIR Gutta Percha and TIR Synthetic adhesives with or without modification with MAH under various glue spreads varied from 6.91 to 8.07%. The moisture content of plywood with TIR Synthetic adhesive with or without modification varied from 6.91 to 7.66%, while the moisture content of plywood with TIR Gutta Percha with or without modification varied from 7.42 to 8.07%. Statistical analysis showed that adhesive types and glue spread treatments resulted in no significant difference of the moisture content values. It is interesting to note from data displayed in Table 2 that modification of TIR Gutta Percha or TIR Synthetic with MAH tend to resulted in lower moisture content compared to unmodified TIR adhesives. Similar results occurred when the glue spread of adhesive was increased from 175 to 250 g m$^{-2}$. Furthermore, the moisture content of plywood with TIR Synthetic adhesive with or without modification with MAH is lower compared to TIR Gutta Percha adhesive. In all cases, the moisture content of the obtained plywood are met the SNI 01-5008.02/1999 standard.

**Density:** The density of plywood adhered with TIR Gutta Percha and TIR Synthetic adhesives with or without modification with MAH under various glue spreads varied from 0.86 to 0.91 g cm$^{-3}$ (Table 2). The density of plywood with TIR Synthetic adhesive with or without modification varied from 0.87 to 0.91 g cm$^{-3}$, while the density of plywood with TIR Gutta Percha varied from 0.86 to 0.91 g cm$^{-3}$. Statistical analysis showed that adhesive types and glue spread treatments resulted in no significant difference among density values. However, the density of plywood tend to increase with the rise of glue spread level. The density of the plywood is higher compared to the density of the veneer. This is might be due to addition of adhesive and pressure applied during plywood manufacturing.

**Bonding strength:** The adhesive strengths of the specimen in the dry state (interior II type plywood) which were hot pressed at 120°C showed significantly difference among the adhesive types and glue spread levels (Fig. 8). The shear strength of plywood bonded with TIR Gutta Percha and TIR Synthetic adhesives showed almost similar value. The values were in the range of 14.71 to 19.68 kg cm$^{-2}$ and 13.44 to 19.08 kg cm$^{-2}$, respectively for shear strength of plywood prepared with TIR Gutta

Table 2: Moisture content and density of plywood adhered with various types of adhesives and various level of glue spreads

| | Glue spreads (g m$^{-2}$) | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | 175 | | 200 | | 225 | | 250 | |
| Adhesives | MC (%) | Density (g cm$^{-3}$) | MC (%) | Density (g cm$^{-3}$) | MC (%) | Density (g cm$^{-3}$) | MC (%) | Density (g cm$^{-3}$) |
| TIR Gutta Percha | 8.07 | 0.87 | 7.89 | 0.88 | 7.98 | 0.88 | 7.59 | 0.91 |
| MTIR Gutta Percha | 7.94 | 0.86 | 7.77 | 0.88 | 7.67 | 0.88 | 7.42 | 0.90 |
| TIR Synthetic | 7.66 | 0.87 | 7.51 | 0.88 | 7.30 | 0.89 | 7.19 | 0.91 |
| MTIR Synthetic | 7.39 | 0.87 | 7.31 | 0.88 | 7.20 | 0.88 | 6.91 | 0.89 |
| MTIR Synthetic+BPO | 7.29 | 0.86 | 7.07 | 0.87 | 6.99 | 0.89 | 6.92 | 0.90 |

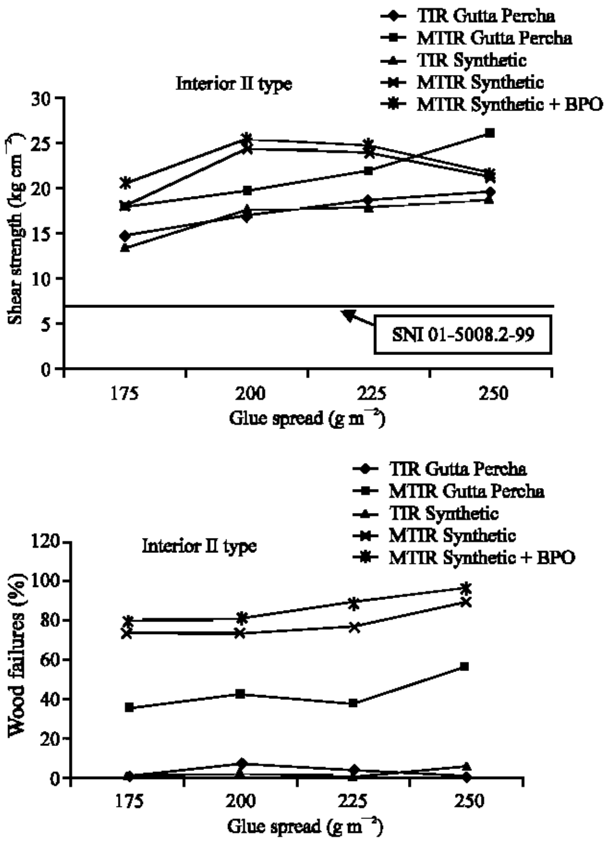


Fig. 8: Shear strength (kg $cm^{-2}$) and wood failures (%) of interior II type plywood adhered with various adhesive types and glue spreads

Percha and TIR Synthetic adhesives. The wood failure values for the specimen prepared from both TIR Gutta Percha and TIR Synthetic adhesives were in the range of 0 to 6.8%. The shear strength value of plywood adhered with TIR Gutta Percha or TIR Synthetic adhesives tend to increase with the raise of glue spread level. It is remarkably the shear strength and wood failure values of plywood adhered with MTIR Gutta Percha and MTIR Synthetic adhesives were much higher compared to the shear strength and wood failure values of plywood adhered with unmodified TIR adhesive. It is noteworthy, at lower glue spread level the shear strength of plywood with MTIR Synthetic adhesive was higher than that of MTIR Gutta Percha adhesive and reached its maximum value at 200 g $m^{-2}$ glue spread. Beyond this point it tends to decrease with increasing glue spread. On the other hand, the shear strength of plywood adhered with MTIR Gutta Percha adhesive showed steadily increasing with the rise of glue spread and reached its maximum value at 250 g $m^{-2}$. Wood failure values data also support these findings. The value of wood failures increased with the rise of shear strength. The maximum value of shear strength of plywood adhered with MTIR Gutta Percha and MTIR Synthetic adhesives were 26.00 and 24.56 kg $cm^{-2}$, respectively. At these points the values of their wood failure were 56.0 and 74.0%, respectively for MTIR Gutta Percha and MTIR Synthetic adhesives. Although the wood failure value of specimen behaved almost similar trend i.e., it increased with the rise of glue spread level, the wood failure for specimen adhered with MTIR adhesive showed much greater than those for the specimen adhered with TIR adhesive. This effect is considered to have been due to the strong bonding of MTIR adhesive to the wood. Furthermore, addition of BPO onto MTIR Synthetic adhesive did not improve the shear strength of plywood. However, compared to MTIR Synthetic adhesive at the same glue spread addition of BPO onto MTIR Synthetic adhesive resulted in higher wood failure value. All the bonding strength of the interior II type plywood prepared under various adhesives and glue spread levels met the requirement of SNI 01-5008.02/1999 standard.

Figure 9 to 11 showed the adhesive strengths and wood failure of the specimen in the wet state after immersed in hot water (interior I Type), after the cyclic boil test (exterior II type) and after the cyclic boil and followed by immersed in cold water (Exterior I). For interior I type plywood, the values of shear strength and wood failure of specimen were in the range of 6.92 to 17.16 kg $cm^{-2}$ and 0 to 74.3%, respectively. The shear strength and wood failure values were varied with the adhesive type and glue spread level. For unmodified TIR adhesive, increasing the glue spread level linearly increased the shear strength of the plywood either prepared from TIR Gutta Percha or TIR Synthetic adhesives. However, the values of wood failure were very small and did not show similar trend with shear strength parameter. The values of shear strength and wood failure of plywood prepared with TIR adhesive at various glue spread level were in the range of 6.96 to 15.28 kg $cm^{-2}$, 0 to 5% and 6.92 to 12.16 kg $cm^{-2}$, 0 to 13.5%, respectively for TIR Gutta Percha and TIR Synthetic adhesives. Furthermore, the data presented in Fig. 9 also clearly showed that at the same glue spread level addition of MAH onto either TIR Gutta Percha or TIR Synthetic adhesives greatly improved the shear strength and wood failure values compared to unmodified TIR adhesive. The higher the value of wood failure indicated that the failures mostly occurred on the veneer rather than on the glue line, vice versa. The shear strength and wood failure values of plywood adhered with MTIR Synthetic adhesive were higher than that of MTIR Gutta Percha adhesive. The shear strength and wood failure values of plywood adhered with MTIR Gutta Percha and

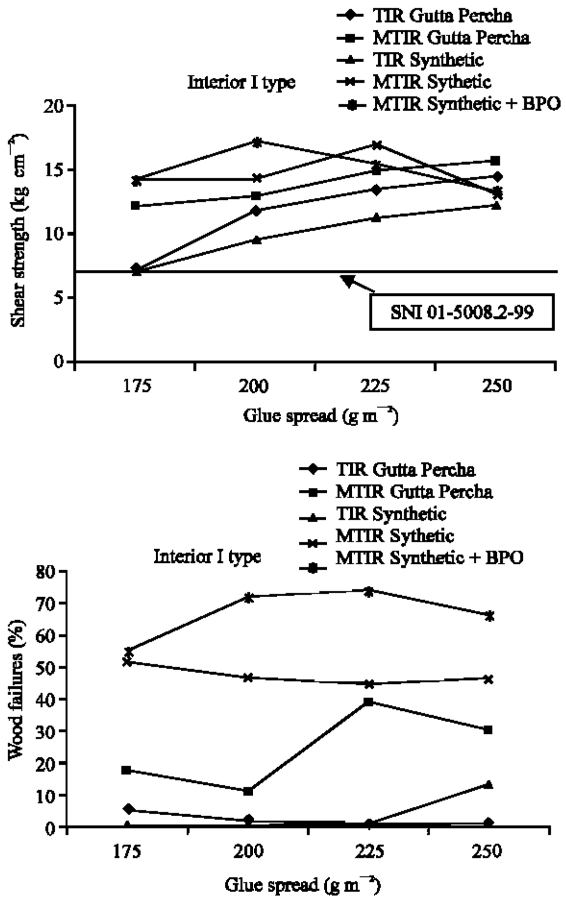


Fig. 9: Shear strength (kg cm$^{-2}$) and wood failures (%) of interior I type plywood adhered with various adhesive types and glue spreads

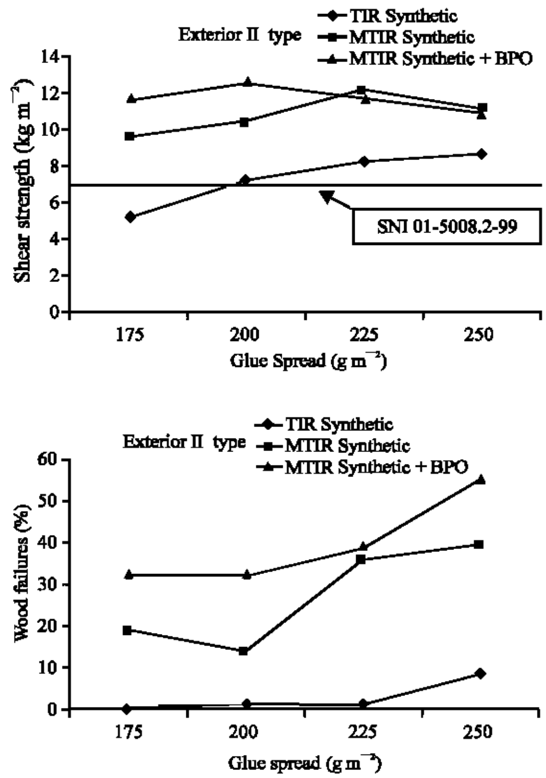


Fig. 10: Shear strength (kg cm$^{-2}$) and wood failures (%) of exterior II type plywood adhered with various adhesive types and glue spreads

MTIR Synthetic adhesives were 11.76 to 13.92 kg cm$^{-2}$, 11.30 to 39.20% and 13.00 to 16.98 kg cm$^{-2}$, 44.5 to 51.8%, respectively. The highest value of shear strength of specimen adhered with MTIR Gutta Percha adhesive was achieved at 200 g m$^{-2}$, while for MTIR Synthetic was at 225 g m$^{-2}$. Addition of BPO onto MTIR Synthetic did not significantly influence the shear strength parameter. However, the results of wood failure values showed that plywood adhered with MTIR Synthetic + BPO adhesive indicated better performance compared with MTIR Synthetic. The values of shear strength and wood failure of plywood adhered with MTIR Synthetic + BPO were in the range of 13.22 to 17.16 kg cm$^{-2}$, 55.1 to 74.3%, respectively. All the bonding strength of the interior I type plywood adhered with various type of adhesives and glue spread levels met the requirement of SNI 01-5008.02/1999 standard.

The shear strength and wood failure of the specimen in the wet state after the cyclic boil test (exterior II type) were presented in Fig. 10. In this experiment we evaluated the performance of TIR Synthetic adhesive with or without modification with MAH. The shear strength and wood failure values of plywood adhered with TIR Synthetic adhesive were in the range 5.16 to 8.60 kg cm$^{-2}$ and 0 to 8.4%, respectively. They increased with the rise of glue spread level. On the other hand, the shear strength and wood failure values of specimen adhered with MTIR Synthetic adhesive was higher compared to that of unmodified TIR Synthetic adhesive. The values of shear strength and wood failure of plywood adhered with MTIR Synthetic were in the range of 9.55 to 12.16 kg cm$^{-2}$ and 13.9 to 39.8%, respectively. The highest shear strength was achieved at 225 g m$^{-2}$ glue spread. Addition of BPO onto MTIR Synthetic resulted in similar value in term of shear strength parameter, but wood failure value was increased. The values of shear strength and wood failure of plywood adhered with MTIR Synthetic + BPO adhesive were in the range of 10.91 to 12.60 kg cm$^{-2}$ and 32.3 to 55.2%, respectively. The highest value of shear strength was achieved on the specimen at 200 g m$^{-2}$ glue spread. All the bonding strength of the Exterior II type plywood specimen, except the plywood adhered with TIR Synthetic

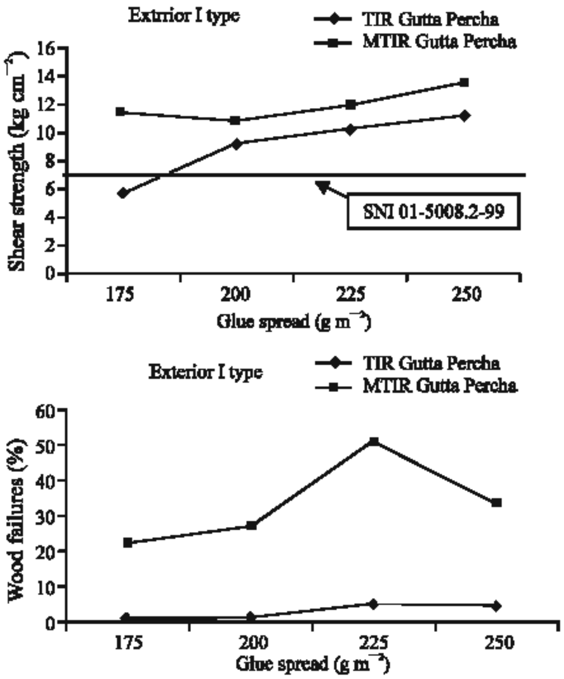


Fig. 11: Shear strength (kg cm$^{-2}$) and wood failures (%) of exterior I type plywood adhered with various adhesive types and glue spreads

adhesive at 175 g m$^{-2}$ glue spread met the requirement of SNI 01-5008.02/1999 standard.

Figure 11 exhibited the shear strength and wood failure of the specimen adhered with TIR and MTIR Gutta Percha adhesives in the wet state after the cyclic boil and followed by immersing in cold water (exterior I type). It is exactly clear that the shear strength and wood failure of specimen adhered with MTIR Gutta Percha adhesive were superior compared if they were adhered with unmodified TIR Gutta Percha adhesive. The shear strength and wood failure values of plywood adhered with TIR Gutta Percha adhesive were in the range of 5.68 to 11.28 kg cm$^{-2}$ and 1 to 4.4%. They increased with the rise of glue spread level. The highest value was achieved at 250 g m$^{-2}$ glue spread. Meanwhile, the shear strength and wood failure values of plywood adhered with MTIR Gutta Percha adhesive were in the range of 10.80 to 13.44 kg cm$^{-2}$ and 22.0 to 51.00%. The highest value was achieved at 250 g m$^{-2}$ glue spread. When compared to shear strength and wood failure values of plywood adhered with MTIR Gutta Percha in dry state, the value of shear strength of specimen after cyclic boil and followed by immersing in cold water was much lower, but its wood failure was almost similar. The decrease in the adhesive strength means that the cohesive failure of wood adherend occurred more easily than in the glue line. That is, the cyclic boil and measurement in wet state weakened the cohesive strength of wood adherend. All the bonding strength of the exterior I type plywood specimen, except the plywood adhered with TIR Gutta Percha adhesive at 175 g m$^{-2}$ glue spread met the requirement of SNI 01-5008.02/1999 standard.

The difference in shear strength and wood failure values between plywood adhered with MTIR Gutta Percha or MTIR Synthetic and TIR Gutta Percha or TIR Synthetic adhesives either in the dry or wet conditions is due to chemical bonding of the applied adhesives. Figure 12 showed the shear strength and sample failure of the plywood adhered with TIR Gutta Percha adhesive with and without MAH modified. For unmodified TIR Gutta Percha adhesive, the failure of plywood occurred on the glue line (Fig. 12A). On the contrary, on the MAH-modified Gutta Percha adhesive, the failure occurred on the veneer (adherend) (Fig. 12B). Thus, it can be recognized that adhesion with MTIR (Gutta Percha or Synthetic) adhesives gave stronger interface bond strength than the cohesive strength of wood. At the same time, the interface bond is quite waterproof. Occurrence of esterification between the hydroxyl groups of wood and MTIR can support these findings. It has been reported in number of literatures that specific anhydrides allow the esterification of wood surfaces for different matrices; acetic anhydride for cellulose butyrate (Glasser *et al.*, 1999), phthalic anhydride for polypropylene (Maldas and Kokta, 1990), maleic anhydride grafted polypropylene (MAPP) for polypropylene (Felix and Gatenholm, 1999; Kazayawoko *et al.*, 1999; Albano *et al.*, 2001; Van De Velde and Kiekens, 2001), maleic anhydride grafted styrene-ethylene-butylene-styrene (Hedenberg and Gatenholm, 1995; Oksman *et al.*, 1998) or maleic anhydride grafted polyethylene (Kim *et al.*, 1997) for polyethylene and maleic anhydride grafted *trans*1.4-isoprene rubber for

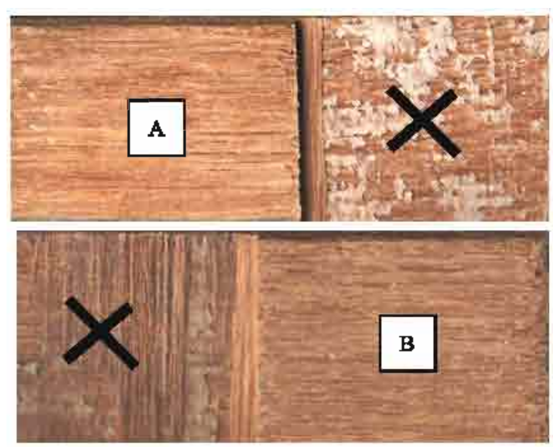


Fig. 12: Shear strength sample failures (X). Unmodified Gutta Percha adhesive (A); MAH modified Gutta Percha adhesive (B)

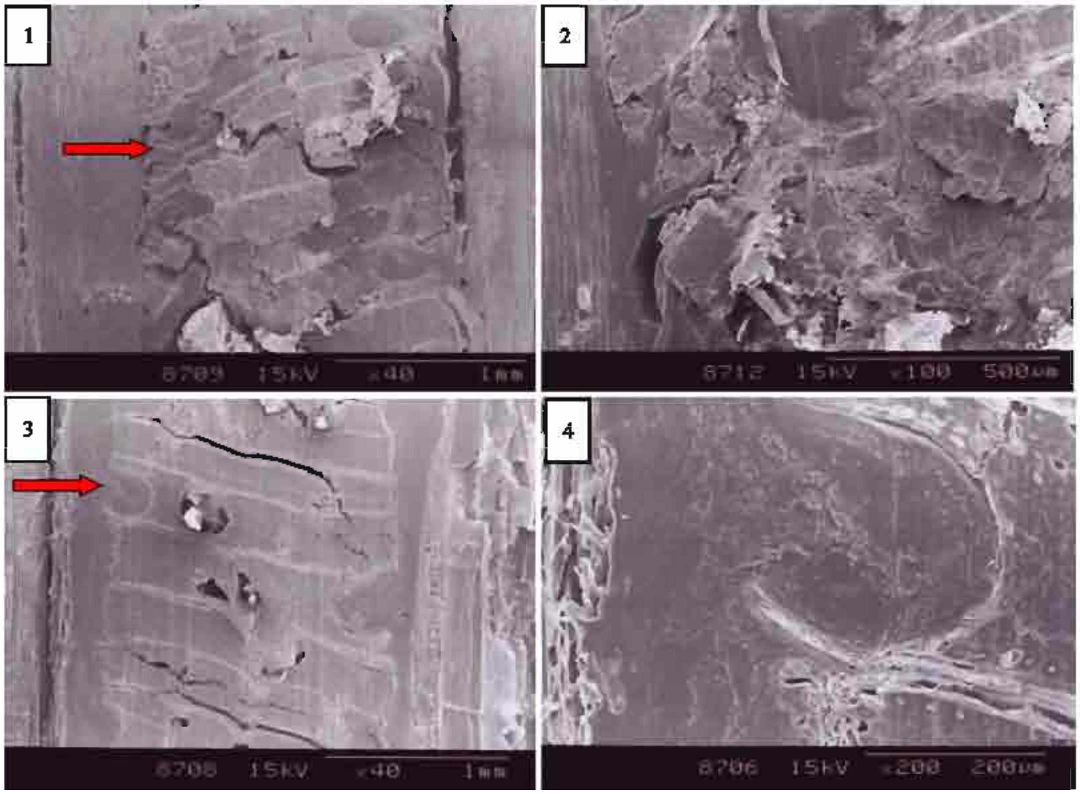


Fig. 13: SEM micrograph of the shear strength sample fractures of plywood with Gutta Percha adhesive. 1) Non Modified Gutta Percha adhesive (40x); 2) Non modified Gutta Percha adhesive (100x); 3) MAH modified Gutta Percha adhesive (40x): 4) MAH modified Gutta Percha adhesive (200x). Arrow lines showed the plywood glue line

*trans*-1.4-isoprene rubber (Febrianto *et al*., 1999). FTIR spectroscopy revealed that MAPP was covalently bonded to the fiber surface in WF-MAPP-PP composites (Kishi *et al*., 1989; Han, 1990; Felix and Gatenholm, 1991) and MTIR was covalently bonded to the fiber surface in WF-MTIR-TIR composites (Febrianto *et al*., 1999).

It has been discussed that TIR adhesive produce almost comparable adhesive strength to that of MTIR. For obtained maximum wetting and spreading, melted non-polar TIR has a greater tendency to spread and penetrate into the pores of the veneer adherend and achieving interfacial contact, than does MPP. It can be seen from Fig. 13, lack of adhesive presence in the glue line when plywood was adhered by unmodified TIR adhesive. This mechanical spreading is the first step in the fulfillment of mechanical and specific adhesion of wood in gluing. In other woods, the low levels of polarity of adhesives can guarantee mechanical and specific adhesion in the cases of hot-melt adhesion. It was reported that wood gluing with hot-melt adhesive of non polar polypropylene is dominated by mechanical adhesion rather than the specific adhesion (Han, 1990).

According to the polar theory of adhesion, strong adhesion can only be produced if the molecules of both contacting bodies have polar functional groups or if the molecules of both the adhesive and the adherend are non polar. The adhesion between a non polar and a polar substance are weak. After wetting and spreading stages, the stage of formation of molecular bonding comes. In this stage, quality of bonding becomes important as shown above. In this sense, MTIR adhesive should become superior to TIR adhesive. Just as with the molecular bonding, the chemical bonding such as esterification is strong and desirable. The formation of esterification, as was proved during preparation of wood flour-MTIR composites (Febrianto *et al*., 1999, 2001) is also expected in this hot-melt gluing with MTIR. The findings of stable waterproof adhesiveness for MTIR adhesive system would support the formation of esterification. Similar phenomenon was reported by Han (1990) when using polypropylene and maleated-modified polypropylene as hot melt adhesive. Based on the above findings, it can be therefore concluded that either MTIR Gutta Percha or MTIR Synthetic adhesives are very appropriate for wood as hot melt adhesives for interior and exterior applications.

Peroxide was added to the rubber with the aim of improving the cross-linked density (Hofmann, 1989). However, in this experiment addition of peroxide (benzoyl peroxide) on the MTIR Synthetic adhesive in amount of 15% based on the MAH weight did not significantly affect the shear strength of plywood. It is noteworthy, at the same level of glue spread addition of peroxide tends to result in higher shear strength and wood failure values.

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