

Dimensional Stability, Color Change, and Durability of Boron-MMA Treated Red Jabon (*Antocephalus macrophyllus*) Wood¹

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ABSTRACT

Boron compound had many advantages as wood preservative, but it was prone to leaching. Improving boron preservation was required to extend the service life of fast growing and low durability red jabon (*Antocephalus macrophyllus*) hardwood. This study aimed to evaluate the dimensional stability, color change and durability of modified red jabon wood by double impregnation with boron and methyl methacrylate (MMA) and heat treatment. Impregnation I used boric acid or borax, and impregnation II used MMA, while heat treatment used temperatures of 90 °C or 180 °C for 4 hours. The dimensional stability, leachability, water absorption, color change and decay resistance of modified red jabon wood were tested. The results showed that MMA impregnation increased the dimensional stability of red jabon wood, while the leaching and water absorption in the wood significantly reduced. Heating at 180 °C caused less water absorption and higher dimensional stability of the wood than that of heating at 90 °C. Impregnation with boric acid and MMA followed by heating at 90 °C resulted in the highest wood ASE, 89.9%. The color change (ΔE^*) of wood increased significantly after MMA impregnation and heating at 180 °C. Boric acid impregnation caused more resistant wood than borax impregnation against decay fungi and termites. Impregnation with boric acid and MMA followed with heating at 180 °C increased significantly the wood resistance against decay fungi and termites.

Keywords: decay fungi, fast growing wood, heat treatment, impregnation, leaching, dimensional stability, wood color

1. INTRODUCTION

Red jabon (*Antocephalus macrophyllus*) was a fast-growing hardwood with specific gravity and density of 0.48 and 0.45 g/cm³ respectively (Lempan, 2014; Cahyono *et al.*, 2015). This wood had a cylindrical

trunk, high branch-free area, and good veneer quality (Cahyono *et al.*, 2016). Red jabon wood according to Indonesian National Standard (known as Standar Nasional Indonesia/SNI) had strength class III (Cahyono *et al.*, 2012) and durability class IV (Halawane, 2015). Therefore, quality improvement of red jabon wood was

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needed especially in the resistance to wood destroying organisms. The quality of wood could be improved with wood modification, such as chemical impregnation and heat treatment.

Impregnation of wood preservative in a pressure tank had been a commercial method in producing durable wood (Hadi *et al.*, 2018). Boric acid and borax were boron preservatives commonly used in wood preservation (Salman *et al.*, 2014; Jebrane and Heinmaa, 2016). Boron compounds were good efficacy-preservative for protecting wood from fungi and insects (Kartal *et al.*, 2019). However, the main problem of boron preservative was prone to leaching in a wet environment (Lesar *et al.*, 2009). Various manipulations had been made to reduce this weaknesses, one of them was by adding the bulking agent. Methyl methacrylate (MMA) was bulking agent material and might be used to improve wood properties (Yalinkilic *et al.*, 1998; Ding *et al.*, 2011). MMA was a colorless and volatile liquid (Dorner *et al.*, 1998) that could improve the physical and mechanical properties of wood (Hadi *et al.*, 2013; Cahyono *et al.*, 2020).

Heat treatment was an environmentally friendly technique for wood quality improvement (Aydemir *et al.*, 2011; Sandberg *et al.*, 2017; Priadi *et al.*, 2019). The temperature of wood heat treatment was generally above 100 °C, or between 150-230 °C (Coto *et al.*, 2015; Romagnoli *et al.*, 2015). Heat treatment could improve the dimensional stability of wood (Lee and Lee, 2018; Priadi *et al.*, 2019), its resistance to decay fungi (Kim *et al.*, 2018; Park *et al.*, 2018) and reduce wood hygroscopicity (Tomak *et al.*, 2011; Chang *et al.*, 2019).

The objective of this research was to evaluate the dimensional stability, color change and durability of modified red jabon wood by double impregnation with boron and methyl methacrylate (MMA) and heat treatment.

2. MATERIALS and METHODS

2.1. Materials preparation

Red jabon (*Anthocephalus macrophyllus* Roxb. Havil) boards were cut from a 9-year-old tree, 3 trunks that had an average diameter at breast height of 40 cm. The woods were taken from the community forest in Bogor, Indonesia and highly selected to obtain the flawless trunks. The test sample was a board taken from a wood part with the distance of 50 cm above the ground and 8 cm from the wood pith. The boards were dried in an experimental drying kiln at 40 °C to gain moisture content of 10%. Then the boards were turned into different test samples. The sample size for dimensional stability, color, leaching, laboratory and field decay tests were $2.5 \times 2.5 \times 10 \text{ cm}^3$, $2.5 \times 2.5 \times 5 \text{ cm}^3$, $2 \times 2 \times 1 \text{ cm}^3$, $2.5 \times 2.5 \times 5 \text{ cm}^3$ and $2 \times 2 \times 37 \text{ cm}^3$ respectively. Each test used five replication of samples.

Boric acid, borax and methyl methacrylate (MMA) were used as impregnants. Boric acid and borax solutions were made at 5% concentration each for impregnation I. MEPOXE M (Methyl Ethyl Ketone Peroxide) was used as the catalyst for curing MMA, which was mixed at ratio 1:10 before impregnation II.

2.2. Impregnation and heat treatment

The impregnation treatment were in two stages then followed by heat treatments at temperatures of 90 °C or 180 °C for 4 hours, as described in Table 1. The impregnation was conducted in pressure wood treatment equipment at 5 atmospheric pressure for 4 hours. Before the impregnation II, the samples were oven-dried at $103 \pm 2 \text{ }^\circ\text{C}$. Heat treatments were conducted after impregnation II at temperatures of 90 °C or 180 °C for 4 hours in an electrical heating oven. The total amount of time required for the process of impregnation I, II and heat treatment was three days.

Table 1 Combination of double impregnation and heat treatments

| Impregnant I | Concentration (%) | Impregnant II | Heating |
|-----------------|-------------------|---------------|---------------|
| No Preservative | - | No MMA | 90 °C, 4 hrs |
| Boric acid | 5 | No MMA | 90 °C, 4 hrs |
| Borax | 5 | No MMA | 90 °C, 4 hrs |
| No Preservative | - | MMA | 90 °C, 4 hrs |
| Boric acid | 5 | MMA | 90 °C, 4 hrs |
| Borax | 5 | MMA | 90 °C, 4 hrs |
| No Preservative | - | No MMA | 180 °C, 4 hrs |
| Boric acid | 5 | No MMA | 180 °C, 4 hrs |
| Borax | 5 | No MMA | 180 °C, 4 hrs |
| No Preservative | - | MMA | 180 °C, 4 hrs |
| Boric acid | 5 | MMA | 180 °C, 4 hrs |
| Borax | 5 | MMA | 180 °C, 4 hrs |

2.3. Dimensional stability and wood color

Dimensional stability, water absorption and leaching tests were done by soaking wood samples in water for 2, 24 and 48 hours. Each soaking was followed by oven drying at 103±2 °C to a constant weight. Dimensional measurements were made before and after soaking using digital caliper. Weighing the samples were carried out before and after water soaking in both wet and dry conditions. Then the volumetric swelling from air dry to wet conditions were determined using Equation 1. The anti-swelling efficiency (*ASE*) was calculated with Equation 2. Dimensional shrinkage of wood in both tangential and radial directions were determined from wet to dry condition using Equation 3. The tangential-radial shrinkage ratio was calculated using Equation 4, while water absorption and leachant were calculated using Equations 5 and 6 respectively.

$$S = \frac{V_w - V_i}{V_i} \times 100 \quad (1)$$

$$ASE = \frac{S_c - S_t}{S_c} \times 100 \quad (2)$$

$$SH = \frac{D_w - D_o}{D_w} \times 100 \quad (3)$$

$$T/R = \frac{SH_t}{SH_r} \quad (4)$$

$$WA = \frac{W_{wf} - W_{oi}}{W_{oi}} \times 100 \quad (5)$$

$$L = \frac{W_{oi} - W_{of}}{W_{oi}} \times 100 \quad (6)$$

Where:

- S* = Swelling (%)
- V_i* = Volume before soaking (cm³)
- V_w* = Volume after soaking (cm³)
- ASE* = Anti swelling efficiency (%)
- S_c* = Swelling of untreated sample (%)
- S_t* = Swelling of treated sample (%)
- SH* = Shrinkage (%)
- D_w* = Dimension (tangential, radial) before oven drying (cm)
- D_o* = Dimension (tangential, radial) after oven drying (cm)
- T/R* = Ratio of tangential-radial shrinkage
- SH_t* = Tangential shrinkage (%)
- SH_r* = Radial shrinkage (%)
- WA* = Water absorption (%)
- W_{oi}* = Oven dry weight before soaking (g)
- W_{wf}* = Wet weight after soaking (g)
- L* = Leachant (%)
- W_{of}* = Oven dry weight after soaking (g)

Wood color evaluation began with scanning of a wood surface with CanoScan 4400F scanner (color dpi resolution of 4800 × 9600). Then ImageJ program was used to determine *L*a*b** values before and after

treatments at the same three positions on the wood surface image. In the CIELab system, the value L^* was a lightness parameter with values 0 (black) to 100 (white). The a^* chromatic value represents red (+) to green (-), while b^* chromatic value represents yellow (+) to blue (-) (Christie, 2014). The color change (ΔE^*) of wood was calculated based on Equation 7 and classified as negligible (0-0.5), slightly perceivable (0.5-1.5), noticeable (1.5-3.0), appreciable (3.0-6.0), very appreciable (6.0-12.0), and totally changed (more than 12.0) (Cui *et al.*, 2007, Hidayat *et al.*, 2017).

$$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (7)$$

Where:

ΔE^* = Color change

ΔL^* = L^* after – L^* before treatment

Δa^* = a^* after – a^* before treatment

Δb^* = b^* after – b^* before treatment

2.4. Decay test

The resistance of wood to decay fungi was tested based on SNI 7207: 2014 standard (BSN, 2014) with a slight modification, after determining oven dry weight, the samples were wetted to facilitate fungal growth by dipping shortly in sterile water. The wood samples were oven dried at 103 ± 2 °C until a constant weight. Furthermore in a laminar air flow, wood sample was put into a petri dish where decay fungi, *Schizophyllum commune* has been grown and covered potato dextrose agar (PDA) medium. After 12 weeks incubation at room temperature, the wood sample was cleaned and oven dried at 103 ± 2 °C until a constant weight to determine weight loss due to fungal attack using Equation 8.

The field test of wood resistance to termite was conducted at the Arboretum of Faculty of Forestry, IPB University. Test samples were oven dried at 103 ± 2 °C to a constant weight. According to ASTM D 1758-06 standard (ASTM, 2006), the sample was exposed in ground contact and buried up to 2/3 of its length. The

row and lane spacing between samples were 30 cm and 60 cm respectively. The sample placement points in the field were marked and then the samples were placed randomly on those marked points. After three months exposure, the samples were cleaned and oven dried at 103 ± 2 °C to a constant weight. The weight loss of wood sample was determined by Equation 8.

$$WL = \frac{W_i - W_t}{W_i} \times 100 \quad (8)$$

Where:

WL = Weight loss (%)

W_i = Dry weight of sample before test (g)

W_t = Dry weight of sample after test (g)

2.5 Data analysis

The effects of impregnation and heating on the dimensional stability, color change and durability of red jabon wood were analyzed statistically using a completely randomized factorial design. The experiment design included three variable factors of treatment with five replications. The factors were preservative (no preservative, boric acid and borax), monomer (no MMA and MMA), and heating temperature (90 °C and 180 °C). Analysis of variance was followed by Duncan test when there was a significant effect at the 95% confidence level. The data were processed using Microsoft Excel 2013 and IBM SPSS Statistic 22.0.

3. RESULTS and DISCUSSION

3.1. Leachability

According to Williams and Amburgey (1987), wood preservation with boron cannot be used in ground contact due to boron susceptible to leaching. Therefore, the use of boron preservative requires a leaching test to assess the stability of boron in wood. This research revealed that leachant in impregnated red jabon wood with borax was significantly higher than that of

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Treated Red Jabon (*Antocephalus macrophyllus*) Wood

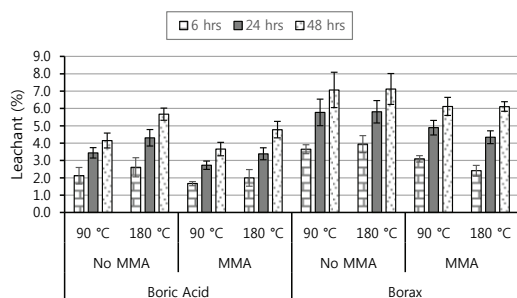


Fig. 1. Leachant of modified red jabon wood with impregnation and heating.

impregnated red jabon wood with boric acid, which means that borax preservative was more prone to leaching than boric acid. However, the use of MMA significantly reduced leaching of boron preservatives (borax and boric acid) in red jabon wood (Fig. 1). This could be caused by bulking agent effect of MMA in wood that inhibited the movement of boron compounds that had been previously impregnated into the wood. As the report of Hadi *et al.* (2013) that MMA entered cavities in wood experiences polymerization after heating and made wood denser. In addition according to Muberra

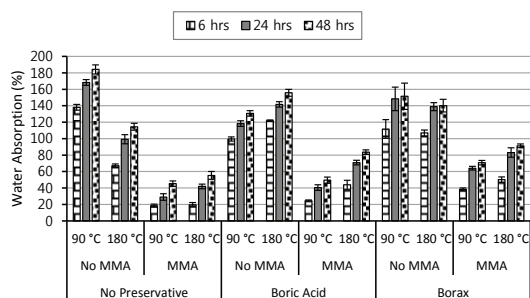


Fig. 2. Water absorption of modified red jabon wood with impregnation and heating.

(2016), a trans-esterification reaction could occur between polymethyl methacrylate to an aromatic boron compound.

3.2. Water absorption

Impregnation and heat treatment changed the water absorption of red jabon wood (Fig. 2). Analysis of variance and Duncan test revealed that water absorption of red jabon wood reduced significantly by boron (borax and boric acid) preservation and MMA impregnation (Table 2). The water absorption was also reduced by

Table 2. The Analysis of variance resume of the leachant (*L*), water absorption (*WA*), anti-swelling efficiency (*ASE*), tangential-radial shrinkage ratio (*T/R*), color change (DE^*), weight loss of decay test in laboratory (*WLL*) and field (*WLF*) scales.

| | Statistical Values | <i>L</i> | <i>WA</i> | <i>T/R</i> | AE^* | <i>WLL</i> | <i>WLF</i> |
|---------|--------------------|-----------|-----------|------------|-----------|------------|------------|
| B | F | 152.76 | 73.47 | 4.144 | 2.591 | 5.17 | 38.849 |
| | (P-value) | (0)** | (0)** | (0,022)* | (0,085)ns | (0,009)** | (0)** |
| MMA | F | 59.935 | 4562.387 | 0.063 | 487.252 | 0.001 | 467.826 |
| | (P-value) | (0)** | (0)** | (0.802)ns | (0)** | (0.977)ns | (0)** |
| H | F | 0.358 | 321.505 | 0.779 | 280.316 | 94.412 | 6.85 |
| | (P-value) | (0.552)ns | (0)** | (0.382)ns | (0)** | (0)** | (0.012)* |
| B×MMA | F | 4.588 | 23.755 | 1.395 | 2.751 | 1.008 | 2.714 |
| | (P-value) | (0.015)* | (0)** | (0.258)ns | (0.074)ns | (0.373)ns | (0.076)ns |
| B×H | F | 20.783 | 71.395 | 1.435 | 3.263 | 0.272 | 1.518 |
| | (P-value) | (0)** | (0)** | (0.248)ns | (0.047)* | (0.763)ns | (0.229) |
| MMA×H | F | 6.454 | 68.171 | 0.261 | 176.963 | 17.806 | 0.622 |
| | (P-value) | (0.014)* | (0)** | (0.612)ns | (0)** | (0)** | (0.434)ns |
| B×MMA×H | F | 3.568 | 140.401 | 0.546 | 3.361 | 0.025 | 0.527 |
| | (P-value) | (0.036)* | (0)** | (0.583)ns | (0.043)* | (0.975)ns | (0.594)ns |

Notes: B, MMA and H indicated boron preservatives, methyl methacrylate, and heat treatments.

* significant effect ($P < 0.05$); ** very significant effect ($P < 0.01$); ns, not significant effect.

heat treatment at 180 °C. This result was in accordance with Kartal *et al.* (2007) that water absorption of heat-modified wood decreases with increasing temperature and heating time. Li *et al.* (2011) also reported that wood moisture content reduced by high temperature heating.

The lowest water absorption in this research was achieved by MMA impregnation with heating at 90 °C. This corresponds to Hadi *et al.* (2013) that MMA treatment could reduce wood moisture content because of the formation of hydrophobic material in wood. A very low water absorption of red jabon wood was also achieved by boric acid and MMA impregnation followed by heating at 90 °C.

3.3. Dimensional stability

Dimensional stability was an important property of wood influenced by environmental conditions, especially temperature and humidity. Wood dimensional stability was evaluated based on anti-swelling efficiency (*ASE*). Fig. 3 showed that heat treated red jabon wood had positive *ASE* value, meaning its dimensional stability increased. In general, heating at 180 °C resulted higher *ASE* value of the wood than heating at 90 °C. This was consistent with Bekhta and Niemz (2003) that heated wood was more stable with increasing temperature and heating time. Kocaefe *et al.* (2008) also reported that heat treatment at 130 °C could modify the hydrophilic polymer of the wood that became more hydrophobic and improved wood dimensional stability.

ASE of red jabon wood increased after MMA impregnation, which might be caused by water inhibition by MMA polymer in wood. As Koubaa *et al.* (2012) reported that MMA impregnation filled voids in wood structures of fast-growing poplar clones. This was in accordance with Ding *et al.* (2011) that MMA impregnation could increase the density and dimensional stability of wood.

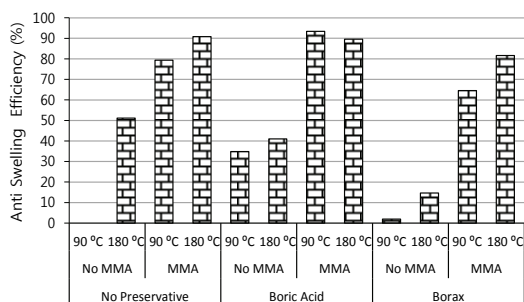


Fig. 3. Anti-swelling efficiency of modified red jabon wood with impregnation and heating.

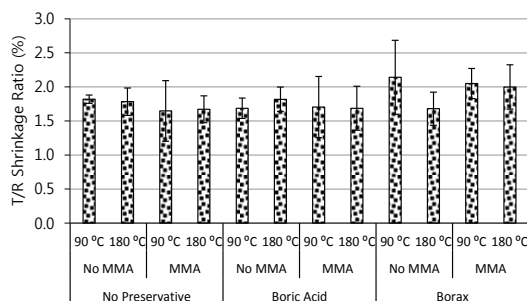


Fig. 4. Tangential-radial shrinkage ratio of modified red jabon wood with impregnation and heating.

Boron impregnation affected the tangential-radial shrinkage ratio (*T/R* ratio) of red jabon wood. The *T/R* ratio of wood impregnated with boric acid was lower than the wood impregnated with borax (Fig. 4). This meant that boric acid impregnation resulted better dimensional stability of wood than borax impregnation. *T/R* ratio closing to 1 showed better dimensional stability of wood, while Bowyer *et al.* (2007) stated that *T/R* ratio exceeding 2 indicated worse wood dimensional stability. MMA impregnation resulted in a lower *T/R* ratio compared to untreated red jabon wood. According to Hadi *et al.* (2013) MMA treatment had a bulking effect in wood shrinkage. In this research, the bulking effect showed to be a slight more in tangential direction than in radial direction of red jabon wood.

3.4. Wood color

Impregnation and heat treatment changed the color of red jabon wood. The color change mainly occurred after MMA treatment, while boric acid and borax preservatives did not cause color change significantly. Different heating temperatures also affected the color of wood (Fig. 5). This supported Hill (2007) that the color change of wood was affected by the temperature and time of heating. Heat treatment at 180 °C decreased brightness value (L^*) of red jabon wood, mainly those previously impregnated with MMA (Fig. 6). The reddish value (a^*) of modified red jabon wood was relatively unchanged, except a slight decrease of a^* after impregnation of borax and heating 90 °C. The yellowish value (b^*) was not clearly affected by boron preservation. The b^* value of MMA impregnated red jabon wood increased by heating at 90 °C, but decreased by heating at 180 °C.

The color change value (ΔE^*) of red jabon wood occurred significantly after MMA impregnation, especially those heated at temperatures of 180 °C (Fig. 7). Preservation with boric acid and borax caused only a

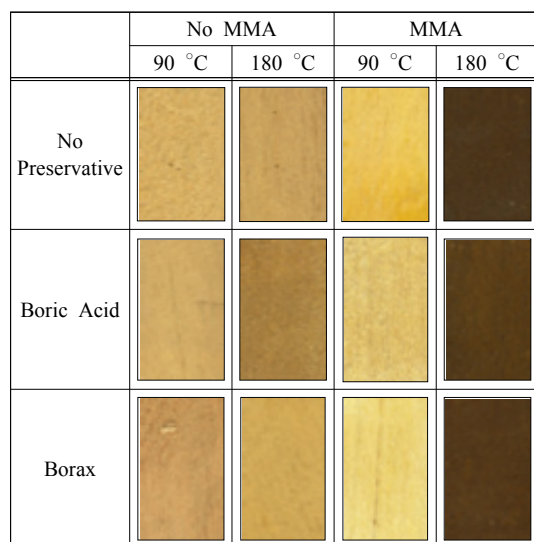


Fig. 5. The color of modified red jabon woods.

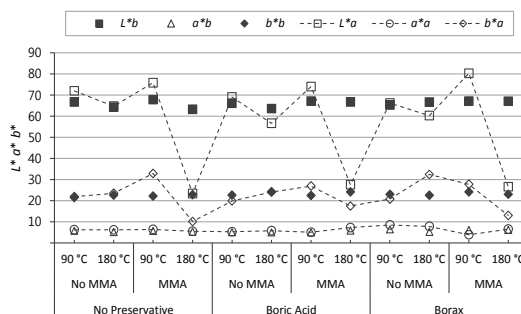


Fig. 6. The L^* , a^* , b^* values of modified red jabon wood with impregnation and heating. $L^*_{b_0}$, $a^*_{b_0}$, $b^*_{b_0}$: before impregnation and heating; $L^*_{a_1}$, $a^*_{a_1}$, $b^*_{a_1}$: after impregnation and heating.

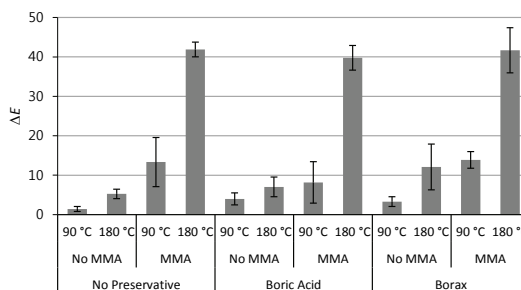


Fig. 7. Color change (ΔE^*) of modified red jabon wood with impregnation and heating.

slight increase in the ΔE^* value of red jabon wood, while heating at 180 °C on borax treated wood resulted in higher ΔE^* value than that on boric acid treated wood.

Color changes in wood due to heating could be related to the change or movement of extractives (Mohareb *et al.*, 2010), polysaccharides change (Sahin and Korkut, 2016, Gaff *et al.*, 2019, Cahyono *et al.*, 2020), or related to the formation of carbonyl groups from the oxidation of hydroxyl group in the phenolic components or the formation of quinoid structures from adjacent hydroxyl groups (Wei *et al.*, 2017).

3.5 The resistance to decay fungi

This research showed that boron preservative, MMA and heat treatment generally increased the resistance

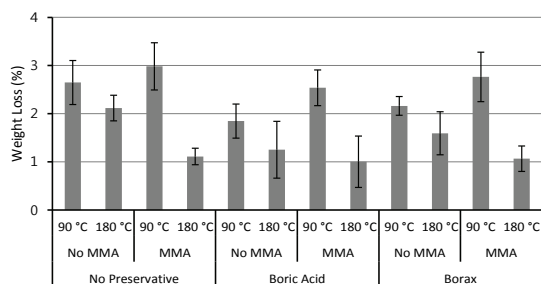


Fig. 8. Weight loss of modified red jabon wood in laboratory decay test.

of red jabon wood from decay fungi as indicated by a decrease in weight loss of the wood (Fig. 8). The analysis of variance and Duncan test at 95% confidence interval showed that weight loss of boron (boric acid and borax) impregnated woods were significantly lower than the control. A similar result was showed by Kartal *et al.* (2004) that boron treated wood was more resistant to brown-rot fungus *Fomitopsis palustris* and white-rot fungus *Trametes versicolor*. Moreover, this research showed that boric acid impregnation resulted more resistant wood against white-rot fungi, *Schizophyllum commune* than that of borax impregnation treatment. In addition, heating at 180 °C improved the resistance to fungi as indicated by a lower weight loss than that of heating at 90 °C. MMA impregnation followed by heating at 180 °C improved the resistance to fungi significantly, while those heated at 90 °C were not significantly different from the control. The lowest weight loss value occurred in red jabon wood that was impregnated with boric acid and MMA followed by heating at 180 °C.

3.6 Decay field test

Ground contact durability test of wood was evaluated based on its weight loss. According to Priadi (2011) termites and decay fungi were the main wood-degrading organisms that attack wood in ground contact.

This research revealed that boron impregnation,

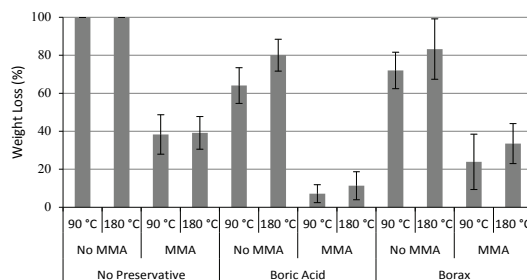


Fig. 9. Weight loss of modified red jabon wood in field decay test.

MMA impregnation or heat treatment improved wood durability that indicated by lower values of weight loss than that of the untreated wood (Fig. 9). The analysis of variance and Duncan test at 95% confidence interval also proved that the weight loss of impregnated red jabon wood with boric acid or borax was significantly lower than the control sample. In addition, MMA impregnated wood also has a prominent smaller weight loss than the wood without MMA impregnation. According to Kartal *et al.* (2004), MMA could maintain the presence of boron compounds in wood from leaching and become more resistant to the termites of *Coptotermes formosanus* Shiraki. Besides, according to Hadi *et al.* (2015), MMA treatment improved the resistance of jabon wood against *Coptotermes curvignathus* subterranean termites. MMA is not toxic to termites but can inhibit termite attacks on wood.

4. CONCLUSION

Boron impregnation, MMA impregnation as well as heat treatment resulted in dimensional stability and durability improvement of red jabon wood. MMA treatment with heating at 90 °C reduced water absorption and leaching of boron preservative from red jabon wood. Impregnation of boric acid and MMA with a heating temperature of 180 °C increased the dimensional stability of the wood with an *ASE* value of 81.71%. The hygroscopicity of wood also decreased, which was in-

icated by water absorption reduction to 43.86%. This treatment also increased the resistance of red jabon wood against decay fungi and subterranean termites.

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