

Weathering and Termite Resistance of Woodflour-Recycled Polypropylene Composites in Tropical Region*¹

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ABSTRACT

Wood flour (WF)-recycled polypropylene (RPP) composites composed of 50% WF of *Eucalyptus deglupta* Blume species, 50% RPP, various contents of maleic anhydride (MAH) modifier (0; 1; 2; 5; 5; 7.5; and 10%), and 15% dicumyl peroxide (DCP) initiator (based on MAH weight) were subjected to weather for 1 year and subterranean termite (*Coptotermes curvignathus* HOLMGREN) and dry wood termite (*Cryptotermes cynocephalus* LIGHT) for 3 and 4 weeks, respectively. WF-RPP composites with 2.5% MA modifier had tensile strength, breaking elongation and Young's modulus about 2.2, 2.3, and 1.2 times, respectively higher compared to MAH-free composites. The WF-RPP composites with or without MAH modifier had 5.5 times higher resistance to weather compared to RPP film alone. The color of the WF-RPP composites with or without MAH modifier became lighter after exposures to the weather. The WF-RPP composites with or without MAH modifier are resistant to subterranean termite *Coptotermes curvignathus* HOLMGREN and dry wood termite *Cryptotermes cynocephalus* LIGHT under the experimental condition adopted.

Keywords : Wood flour, recycled polypropylene, composite, modifier, weathering, and termite

1. INTRODUCTION

Wood plastic composites (WPC) are a new group of materials that are generating interest in many applications. The term WPC covers an extremely wide range of composite materials using both virgin and recycled plastics ranging from polypropylene (PP) to poly vinyl acetate (PVA) and fillers ranging from wood flour

(WF) to agricultural fibers.

The application of wood fiber-thermoplastic composites are enormous, for examples, automobile, constructions component, frames, shipping crates, pallets, storage bins, toys, games, household goods, outdoor furniture, and recreational boats etc.

Thermoplastic also bring a serious problem to environment after service because of difficulty

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of its decomposition under natural conditions. Accordingly, thermoplastic wastes cause serious environmental pollution all over the world every year. For example the amount of plastics annually consumed in the United States is approximately 100 kilograms per person. Among plastics, the big four plastic products (Polyethylene/PE, poly propylene/PP, poly vinyl chloride/PVC, and polystyrene/PS) were accounted about 92% by volume of the whole plastics consumption in the United States. Therefore, in order to reduce the negative impact of thermoplastics waste to environment, there is urgent need to utilize the waste plastics as a matrix resin for WPC (Chernier, 1992). On the other hand, one of the disadvantages of wood is susceptible to deterioration caused by organisms such as fungi, termite, beetle, and marine borers, and to environment such as weather and fire etc. (Stark and Berger, 1997; Feist and Hon, 1984). Furthermore, thermoplastics resins are polymeric in nature; they are susceptible to environmental stresses.

Since wood plastic composites are relatively new product, up to now, there is very few information regarding their resistance against bio-deterioration agents and weathering, particularly in tropical region. It was reported that wood-PP composites were resistant to brown-rot and white rot fungi and turned to lighten within 400 to 600 hours of UV exposure in laboratory tests (Caulfield *et al.*, 2005). In this publication, we investigated the durability of the WF-RPP composites against weather and termite in tropical region. The objectives of this study are to 1) to evaluate the performances (color change and tensile properties) of WF-RPP composites with or without MAH modifier after exposure to the weather at elevated time; and 2) to observe the resistance of WF-RPP composites against subterranean termite (*Cryptotermes curvignathus* HOLMGREN) and dry wood ter-

mite (*Cryptotermes cynocephalus* LIGHT).

2. MATERIALS and METHODS

2.1. Materials

First recycled polypropylene (RPP) was used as matrix resin which was collected from Bogor district, West Java, Indonesia. WF of *Eucalyptus deglupta* Blume species was use as filler with the size of less than 120 mesh. Maleic anhydride (MAH) and dicumyl peroxide (DCP) were used as a modifier and an initiator, respectively. WF-RPP composites composed of 50% WF and 50% RPP and various content of MAH ranging of 0 and 10% and 15% DCP (based on MAH weight).

2.2. Compounding the Composites

RPP was placed into the kneader at 170°C and 10 rpm for 1 min. After charging the RPP, MAH and DCP were added into the molten RPP within 1 min. After further reaction for 1 min, WF was added within 3 min at 30 rpm. The rate of rotation was increased at 50 rpm and kneading was continued for 9 min. The total kneading time was 15 min, and the amount of RPP and WF were 48 grams. For MAH-free composite, after charging the RPP for 1~2 min at 170°C and 10 rpm, WF was added for 3 min at 30 rpm. The rate of rotation increased to 50 rpm and kneading was continued for 10 min.

2.3. Preparation of Composites Sheets

Kneaded samples were molded into sheet by hot pressing. The prescribed amount of kneaded samples (7~10 grams) was placed between a pair of polyester film with 0.3 mm thick spacer. The temperature of hot press was 180°C and the samples were subjected to 0~100 kgf/cm² for

1~2 min. After subsequent cold pressing at the same pressure for 30 sec, the sheets were then cooled at room temperature.

2.4. Preparation of Specimen for Tensile Test

Strip samples were prepared from the composite sheets. The average values of tensile strength, breaking elongation, and Young's modulus were obtained from 5 repeated measurements.

2.5. Weathering Test

The samples were exposed to weather for 1 year in Bandung, West Java, Indonesia. The latitude and longitude of exposure place were 6°54'SL and 107°36', respectively (Subowo *et al.*, 2000).

2.6. Color Difference

Color difference was observed visually using portable color difference meter model CDX 105 according to JIS Z.8729.

2.7. Scanning Electron Microscopy (SEM) Observation

The morphology of the tensile fractures and the surface of the WF-RPP composites were studied by using a scanning microscope (A JEOL JSM-T330). The samples were dried before coating to remove the absorbed water. SEM photographs were taken under the following conditions: working distance 15 mm and accelerating voltage of 10 Kv.

2.8. Termite Test

The samples were subjected to dry wood termite (*Cryptotermes cynocephalus* LIGHT) and subterranean termite (*Coptotermes curvignathus*

HOLMGREN) accordance to JWSA standard No. 12-1992. Termite mortality and weight loss of samples were measured at the end of testing.

3. RESULTS and DISCUSSIONS

3.1. Effect of Weather on Color Change of WF-RPP Composites

The mean color change of WF-RPP composites with or without MAH modifier after exposure to weather at elevated time was presented in Table 1. The L* values of the WF-RPP composites with or without MAH modifier increased with increasing of exposure time. The WF-RPP composites tend to lighten over time. The color of WF-RPP composites changed from dark brown to white pale (Fig. 1). Similar phenomena were reported by Falk *et al.* (2002). In the first and second month's exposure times, the color change of WF-RPP composites free-MAH is much faster to become white pale compared to WF-RPP with MAH modifier. Caulfield *et al.* (2005), reported that in laboratory weatherometer tests, wood-PP composites lightened within 400 to 600 hours of UV exposure. After UV and water exposure of 2,000 hours, specimens were white and the white color continued throughout the 10 mm specimen. This in-depth color change may be due to stabilization of the free radicals formed from ultraviolet radiation by the lignin in the wood, allowing the free radicals to penetrate deeper into the wood. Weathering degraded and solubilized lignin, so the white pale color of composites after exposure to weather indicated that the remaining filler consisted of nearly chemically pure cellulose. These changes occur only to a depth of 0.05~2.5 mm and are a result of sunlight, particularly UV light, which initiates photo degradation (Feist and Hon, 1984; Rowell, 1984).

Table 1. Average values of color changes (E*ab) of the WF-RPP composites with or without MAH modifier at after exposure to weather at elevated times

MAH content (%)	Exposure time (month)	L*	a*	b*	ΔE^*_{ab}
0	0	12.02	10.56	13.20	0
	1	54.88	1.72	8.28	44.04
	2	68.24	0.46	6.10	57.60
	3	73.78	0.40	4.48	63.20
2.5	0	12.66	21.16	16.90	0
	1	44.84	7.82	7.56	36.07
	2	61.12	2.62	5.34	53.16
	3	71.96	1.34	3.18	64.01

L*: Lightness; a*: red; b*: yellow

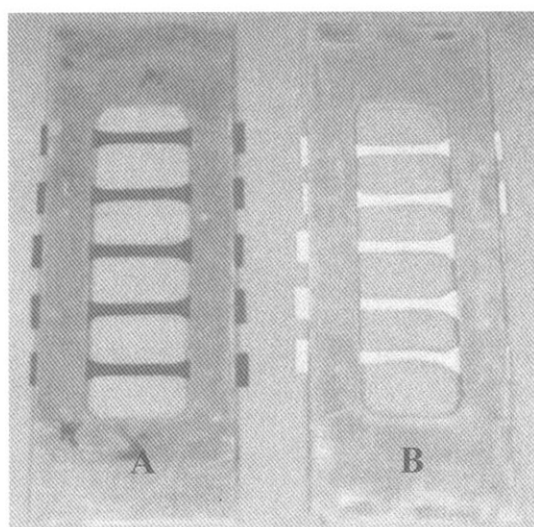


Fig. 1. Color change of WF-RPP composites after exposure time of 3 months; A: Before exposure; B: After exposure.

3.2. Effect of Weather on Tensile Properties of WF-RPP Composites

The mean values of tensile strength, breaking elongation and Young's modulus of WF-RPP composites with or without MAH modifier after exposure to weather for 11 months were presented in Fig. 2~4 and Table 2. It is obvious that the addition of MAH modifier together with DCP initiator improved the tensile pro-

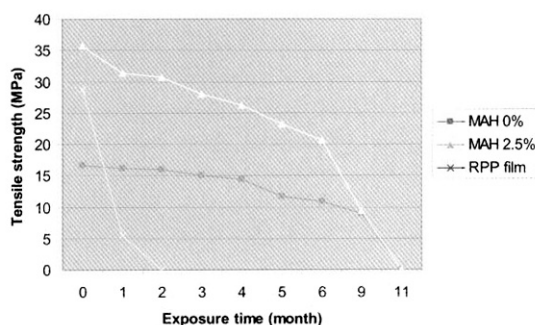


Fig. 2. Tensile strength of WF-RPP composites with or without MAH modifier after exposure to weather at elevated time.

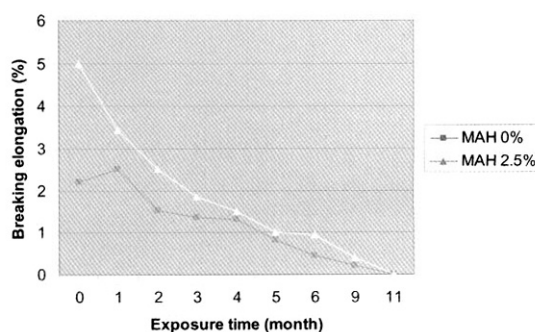


Fig. 3. Breaking elongation of WF-PP composites with or without MAH modifier after exposure to weather at elevated time.

erties of WF-RPP composites reaching its maximum value at 2.5% MAH addition (Table 1). The improved composites showed tensile

Table 2. Tensile strength of WF-RPP composites after exposure to weather at elevated times under various MAH content

Time (month)	MAH content (%)						
	RPP film	0	1	2.5	5	7.5	10
0	28.88	16.63	30.48	34.74	32.64	31.59	31.02
1	5.43	16.22	28.78	31.40	30.79	30.31	29.74
2	0	15.94	26.85	30.79	27.45	27.47	27.44
3		15.11	23.74	27.96	25.94	25.91	25.74
4		14.47	20.12	26.19	24.04	23.83	24.50
5		11.71	18.22	23.13	21.08	21.21	22.09
6		10.90	15.26	20.60	19.44	19.56	18.73
9		8.88	nd	9.44	nd	nd	nd
11		0	nd	0	nd	nd	nd

nd: not determined

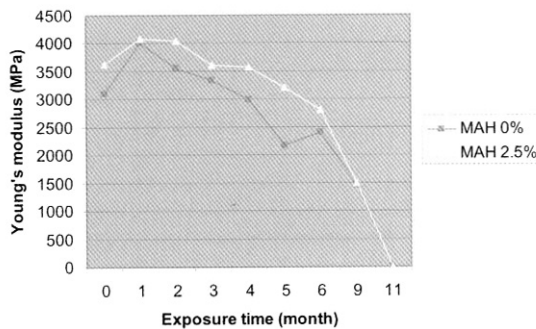


Fig. 4. Young's modulus of WF-PP composites with or without MAH modifier after exposure to weather at elevated time.

strength, breaking elongation and Young's modulus about 2.2, 2.3, and 1.2 times higher, respectively compared with MAH-free composites. Beyond this point, addition of MAH over 2.5% MAH modifier resulted in gradual decreases in the tensile strength, breaking elongation, and Young's modulus. From these observations, the reason for the positive role of MAH addition in the presence of a free radical initiator (DCP) is apparent and attributable to the enhanced formation of MRPP (MAH-modified RPP). The MRPP is grafted onto WF by succinic half-ester bridge resulted in enhancement of interfacial adhesion between RPP and WF (Fig.

5-B). It was reported that addition small amount of MPP onto WF-PP composites (Kishi *et al.*, 1989; Takase and Shiraishi, 1989; and Han *et al.*, 1989), and MTIR (maleic anhydride modified *trans*-1.4-isoprene rubber) onto TIR-WF composites (Febrianto *et al.*, 1999) greatly enhanced the tensile properties of the composites.

Furthermore, the possible reasons for its negative role which appeared when excess MAH (over 2.5% MAH) (Table 2) was used presumably due to 1) the increased occurrence of low molecular weight substances within the composites with the excess addition of MAH (MAH remained and its reactants with other additives); 2) the reduction of initiator caused by possible reaction with excess MAH; 3) the degradation of WF and RPP resulting from kneading and molding at high temperature in the presence of excess acidic MAH, and so forth (Han *et al.*, 1989; and Febrianto *et al.*, 1999).

The tensile strength, breaking elongation, and Young's modulus of WF-RPP composites with or without MAH modifier decreased linearly after exposures to weather at elevated time. The longer exposure time resulted in the greater reduction of tensile strength, breaking elongation, and Young's modulus values (Figs. 2, 3,

and 4). The presence of various MAH modifier combined with DCP initiator did not improve the decreasing rate of tensile properties of WF-RPP composites (Table 2). However, it is exactly clear from Figs. 2~4 that up to exposure time of 6 months the WF-RPP composites with MAH modifier showed better performance, compared to MAH-free composite. Beyond this point, after exposure time of 9 months the WF-RPP composites with or without MAH modifier showed similar resistance to the weather. The WF-RPP composites with or without MAH modifier totally decreased their strength after exposure time of 11 months. In contrast, the RPP film alone totally lost its strength after exposure time of 2 months. This is presumably due to (1) the location of exposure was set at high latitude. The decreasing rate of strength of polypropylene film exposed at high latitude is faster compared at low latitude, and (2) the fact that PP is much easier to degrade in the nature, compared to other polyolefin's (polyethylene/PE and polystyrene/PS) (Subowo and Satoto, 1993; Subowo *et al.*, 2000). The polymeric materials usually contain chromophore as impurities produced by polymerization or thermal processes. The chromophore is able to absorb UV light at a certain wave length and becomes excited. The excited chromophore enables the induction of polymer radicals formation. The present of oxygen in air causes free-radical photo oxidation and further processes which will degrade the polymer chain leading to the deterioration of mechanical properties of materials (MC Kellar and Allen; 1979; Randy and Rabek, 1975).

The surface and tensile fracture surfaces of the WF-RPP composites with or without MAH modifier before and after exposure to weather were presented in Fig. 5. Before exposure to weather, the addition of MAH modifier together with DCP initiator improved adhesion between

WF filler and RPP matrix. It is exactly clear in the MAH-free composites that holes and spacing are commonly formed along the fiber and the fibers pulled out, indicating insufficient adhesion, low compatibility, poor contact, and inferior stress transfer between the phases (Figs. 5-A and 5-C). In contrast, the MRPP prevents hydrogen bonds among WF from being formed and renders the properties of WF surfaces and matrix more similar (Figs. 5-B and 5-D). The woody fillers are covered by matrix resin and it indicated that chemical interaction occurred in the composites. After exposure to weather for 3 months, due to degradation of PP matrix caused by weathering, the WF fillers are separated from the PP matrix, and it can be easily recognized (Figs. 5 E and 5F). The surface of the WF-RPP composites with or without MAH modifier become rough and also crack occurred (Figs. 5G and 5 H). However, degradation condition of WF-RPP composites is more severe compared to WF-RPP composites with MAH modifier. From these findings, it can be concluded that WF-RPP composites with or without MAH modifier showed 5.5 times higher resistance to weather, compared to RPP film alone in tropical region. Up to 6 month exposure to the weather the WF-RPP composites showed better resistance to weather compared to MAH-free composites. Beyond this point WF-RPP composite with or without MAH-modifier showed similar resistance to weather.

3.3. The Resistance of WF-RPP Composites Against Termite

In tropical region due to warm and humid climate conditions, deterioration of wood and wood products caused by termites was very severe. It was reported by Nandika *et al.*, (2003) that in the last 2 decades, 70% of buildings in four big cities in Indonesia were

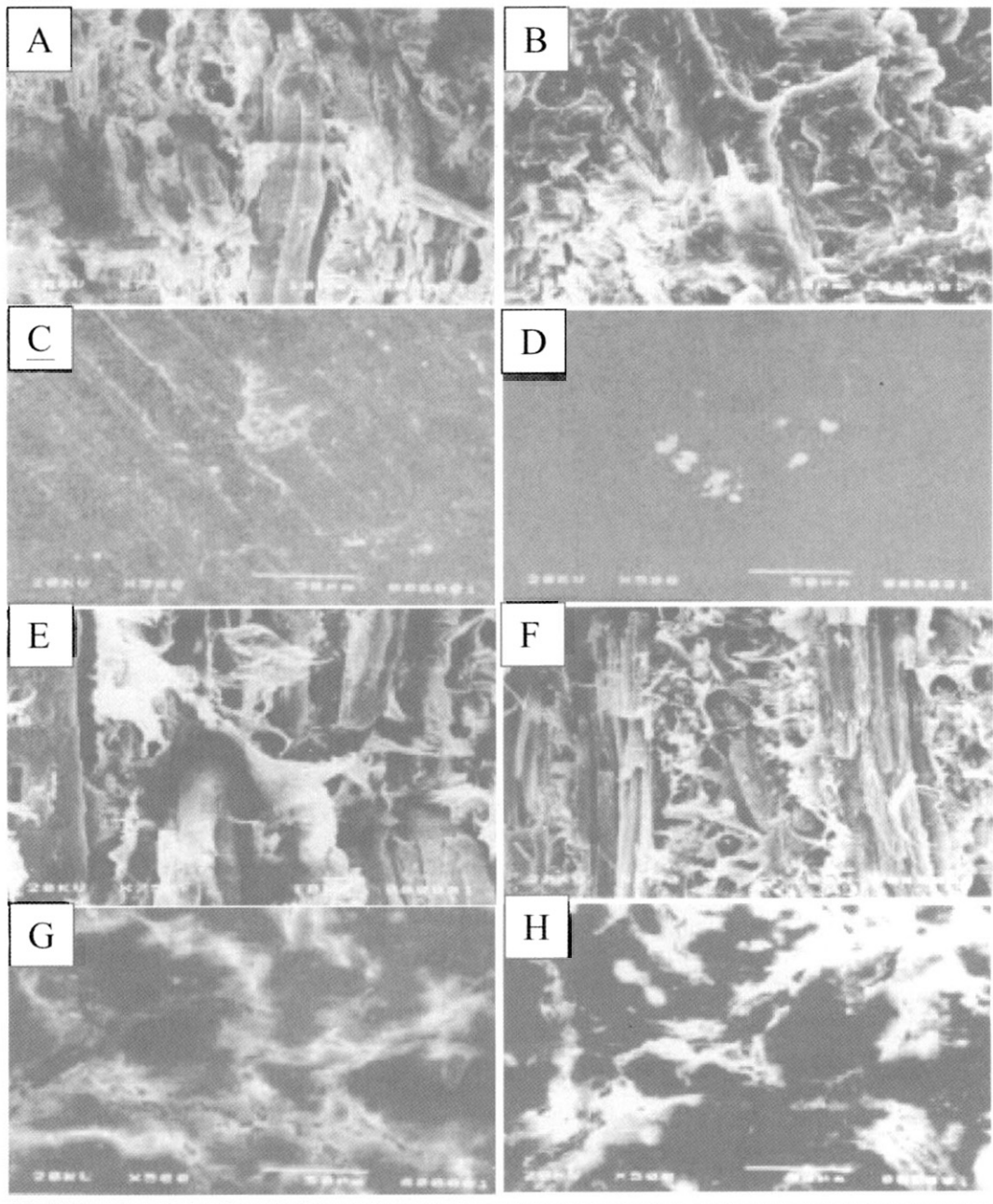


Fig. 5. SEM micrograph of tensile fracture and surface sample of composites of WF-RPP with or without MAH before (A, B, C and D) and after (E, F, G, and H) exposure for 5 months to weather. Tensile fracture WF- RPP composite without MAH (750X); B) Tensile fracture WF-RPP composite with 2.5% MAH (750X); C) Surface of WF-RPP composite without MAH; D) Surface of WF-RPP composite with 2.5% MAH; E) Tensile fracture WF-RPP composite without MAH (750X); F) Tensile fracture WF-RPP composite with 2.5% MAH (750X); G) Surface of WF-RPP composite without MAH; H) Surface of WF-RPP composite with 2.5% MAH.

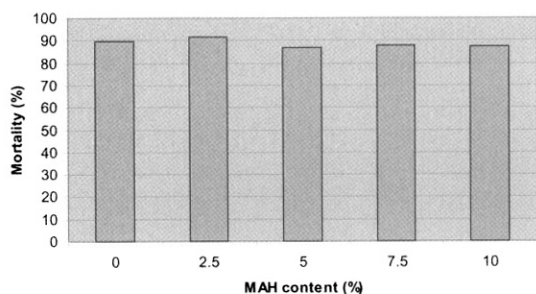


Fig. 6. Termite mortality of *Coptotermes curvignathus* HOLMGREN under various MAH contents.

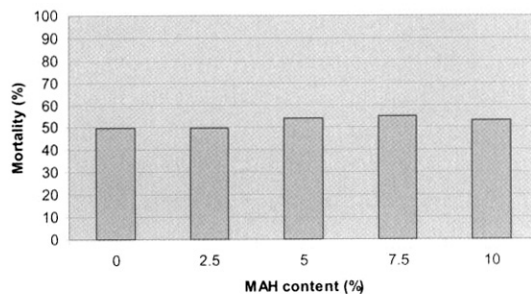


Fig. 8. Termite mortality of *Cryptotermes cynocephalus* LIGHT under various MAH contents.

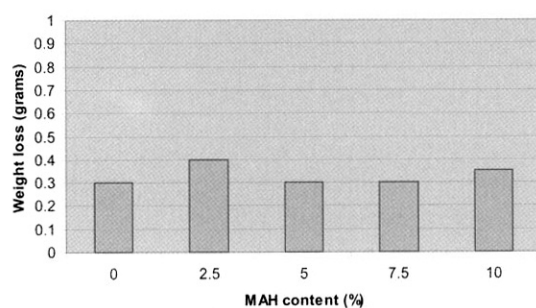


Fig. 7. Weight loss of samples after baited to *Coptotermes curvignathus* HOLMGREN under various MAH contents.

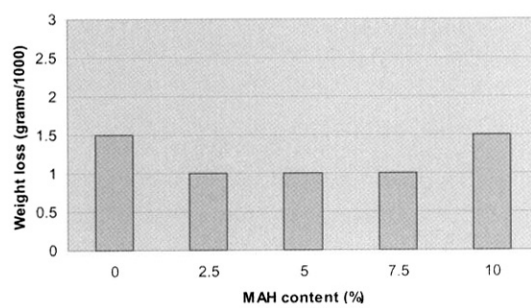


Fig. 9. Weight loss of samples after baited to *Cryptotermes cynocephalus* LIGHT under various MAH contents.

attacked by termites. It was also reported that the most hazardous termite in Indonesia is *Coptotermes curvignathus* HOLMGREN that belongs to subterranean termite and *Cryptotermes cynocephalus* LIGHT that belongs to dry wood termite. Hence, in this part of experiment we evaluated the resistance of WF-RPP composites against subterranean termites (*Coptotermes curvignathus* HOLMGREN) and dry wood termites (*Cryptotermes cynocephalus* LIGHT) under various MAH modifier contents after baited for 3 weeks, and 4 weeks, respectively. The parameters measured were termite mortality and weight loss of samples.

Figs. 6~9 exhibit the effect of MAH modifier content on the termite mortality and the weight loss of WF-RPP composites samples. The termite mortality and the weight loss of WF-RPP composites samples after baited to

Coptotermes curvignathus HOLMGREN and *Cryptotermes cynocephalus* LIGHT termites were about 87~90.5%, and 0.003~0.04 grams; 50~55%, and 0.001~0.0015 grams, respectively. It was clear that the survival rate of *Cryptotermes cynocephalus* LIGHT termite was higher than *Coptotermes curvignathus* HOLMGREN termite. However, the weight loss of samples after baited to *Cryptotermes cynocephalus* LIGHT termite was lower than weight loss of samples after baited to *Coptotermes curvignathus* HOLMGREN. It was presumably due to the size of *Cryptotermes cynocephalus* LIGHT termite was much smaller than *Coptotermes curvignathus* HOLMGREN termite. It was also clear that termite mortality and weight loss of WF-RPP composites prepared under various MAH modifier content both subjected to *Coptotermes curvignathus* HOLMGREN and *Cryptotermes*

cynocephalus LIGHT termites were similar. It means that chemical interaction between WF filler and RPP matrix resulted from addition of MAH modifier together with DCP initiator did not enhance the resistance of the resulted composites against *Coptotermes curvignathus* HOLMGREN dan *Cryptotermes cynocephalus* LIGHT under the experimental condition adopted. Based on the above results it can be concluded that WF-RPP composites with or without MAH modifier are resistant to subterranean termite *Coptotermes curvignathus* HOLMGREN and dry wood termite *Cryptotermes cynocephalus* LIGHT.

4. CONCLUSIONS

1) The addition of MAH modifier together with DCP initiator improved the tensile properties of WF-RPP composites reaching its maximum value at 2.5% MAH addition. The improved composites had tensile strength, breaking elongation and Young's modulus about 2.2, 2.3, and 1.2 times, respectively higher compared to MAH-free composites.

2) The color of the WF-RPP composites with or without MAH modifier turned lighter after exposures to the weather.

3) After exposure time of 11 months, the WF-RPP composites with or without MAH modifier showed 5.5 times higher resistance to weather compared to RPP film alone in the tropical region. The WF-RPP composites in the presence or in the absence of MAH modifier exhibited similar resistance to weather.

4) The WF-RPP composites with or without MAH modifier had similar resistance to subterranean termite *Coptotermes curvignathus* HOLMGREN and dry wood termite *Cryptotermes cynocephalus* LIGHT and categorized as termite resistance composites under the experimental condition adopted.

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