BIODEGRADATION BEHAVIOUR OF PARTICLEBOARD BONDED WITH MODIFIED PVOH/OIL PALM STARCH AND NANO SILICON DIOXIDE

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ABSTRACT

The main objectives of this study were to evaluate the biodegradation behaviour of experimental particleboard panels bonded with modified 30 % of PVOH, 70 % of Oil palm starch and 3 % of nano silicon dioxide (SiO_2). Boric acid at 2 % was also added as a cross-linker to PVOH in modified oil palm starch to enhance decay resistant for the samples. All the particleboards were evaluated by soil burial, borer and fungal strains test. The samples were then compared with board bonded with native oil palm starch and commercial urea formaldehyde (UF). The results indicated that particleboards bonded with modified PVOH/Oil palm starch were more resistant than particleboard bonded with their native starch and can be used as a potential binder for green particleboard in future. SiO_2 also showed a significant effect towards modified oil palm starch as compared to their native oil palm starch and commercial binder.

Keywords: Biodegradation; oil palm starch; particleboard; PVOH; SiO_2.

INTRODUCTION

Approximately 60% of the end used products production was used for human utilization, meanwhile, 33% used for animal feed and 7% for different industries such as textile, paper, food and adhesives as reported by Pandey, 2000 [1] . Adhesive is defined as any substance that is capable of holding materials together in a useful way by surface attachment that will prevent it from separation [2]. Particleboards can be produced from any lignocellulosic materials, as long as they can contribute a proper physical, mechanical and biological resistance [3]. Phenol-formaldehyde (PF) resins and urea-formaldehyde (UF) resins are commonly main binders in production of particleboard due to their high durability and fast curing adhesive [4]. With increasing crude oil prices and environmental awareness, the bio-based adhesive was started to promote as an alternative bonding agent [5]. Due to this problem, starch as a polymer of D-glucose which is stored in granules as a food source in most plants was used as a supplementary for synthetic polymers [6]. Starch is also considered as a cheap material and easier to adapt for any chemical modification thus makes it as an interesting material. Starch was used as an adhesive in variable products, as binders, sizing material, glues and pastes [7].

Recently, starch-based wood adhesive was widely used in interior applications [8]. The bonding results produced from starch based adhesive generally was formed from both mechanical interlocking and van der Waals forces. It wet a polar surface of cellulose and penetrate into pores, thus formed a strong adhesive bond [8]. While the modified starch is normally used as stabiliser or emulsifier and recently was precisely develop to be used as a binder in particleboards manufacturing [9]. Particleboards when exposed to the places with high moisture conditions, are easier to be attacked by the decay [3].

Several trials have been made to blend starch with biodegradable synthetic polymers, such as polyvinyl alcohol (PVOH) due to its biodegradable behaviour and possess excellent mechanical properties [10]. In this study, the 30/70 PVOH/CS sample showed the highest weight loss compared to pure PVOH. This finding was associated with higher corn starch content in the packaging film. The PVOH, which is biodegradable due to its high hydrolysability, exhibited a higher resistance to soil burial degradation [10]. Boron / boric acid as reported in the various study was successfully accommodated as antibacterial and antifungal agents when they are mixed with polymers such as starch and PVOH [11]. The significant weight loss was reported in enzymatic degradation test on thermoplastic starch (TPS) when it is added with nano-SiO_2 content up to 6 % wt [12].

Therefore, the main objective of this study was to investigate the decay resistance of particleboard bonded with starch/PVOH/SiO_2. Approximately 70 % of starch ratio with 30 % of PVOH and 3 % of Nano silicon dioxide (SiO_2) will be compared with their native starch and commercial binder.

MATERIALS AND METHODS

Oil Palm Starch Extraction Process

The oil palm starch extraction process followed procedure from previous studies [12 - 13] with a slight modification and has been described details in the previous publication [15].

Oil Palm Starch Modification Process

In this study, the oil palm starch was modified with polyvinyl alcohol (PVOH) and crosslinked with 2 % of boric acid. The modification method was carried out based on previous studies [14 - 15] with a slight modification in term of amount of chemical and additives used in the modification process. Polyvinyl alcohol (PVOH) with molar mass = 99.000 g/mol (Sigma-Aldrich), glycerol (20 %) and Tween 80 (1 %) were used in this study. Boric acid with purity 98 % with a melting point at 171 °C and Tween 80 were purchased from Sigma-Aldrich. The starch ratio used in this modification process approximately 70 % mixed together with 30 % of PVOH was prepared. All the mixtures used in this modification activity were weighed based on 15 % of the adhesive formulation. Firstly the 30 % PVOH was weighed and added with distilled water until it is marked up to 400 ml and placed in a 1000 ml beaker before being heated at 90 °C for 1 h. Then, the mixtures underwent constant stirring until all the PVOH crystals were completely dissolved. Next, the 70 % of oil palm starch samples and 20 % wt. of glycerol was added to the mixture and stirred for 1 h until dissolved. After that, 2 % of boric acid and 1 % of tween 80 were mixed together with starch and glycerol and stirred again for another 1 hr. The physical blending of 3 % of Nano silicon dioxide (SiO_2) was done into the mixture in the last phase of 3 h of overall modification process. Finally, all the mixtures were then poured into a container and oven-dried at 60 °C before being used as a binder for particleboard manufacturing.

Particleboard Manufacturing

The particleboard was made from 70 % *Acacia mangium* and 30 % mixed hardwoods supplied by a local particleboard company in Negeri Sembilan, Malaysia. The density of particleboard was produced at 0.80 g/cm^3 and the dimension for each board is 20.1 X 20.1 X 0.5 cm. The moisture content of particles was 8 %. Oil palm starch was extracted from oil palm trunk as described previously in other research work [13- 14]. About 15 % of starch based adhesive made from native oil palm starch and modified oil palm starch were prepared in this formulation and were weighed based on dry weight (w/w). A commercial binder, urea formaldehyde (UF) was used and using similar resin level. The starch based adhesive was prepared by mixing the starch powder with 150 ml of hot distilled water (80 °C) and manually mixed with particles until all the particles were completely covered with the starch based adhesives within 5 min. Meanwhile, the board bonded with commercial binder (UF) was mixed following standard commercial particleboard manufacturing. Then, the mixture was poured into the mould to form a mat with a dimension size of 20.1 cm x 20.1 cm x 0.5 cm, followed by pre-pressing with a cold press for about 2 min. Later, the mat was pressed by hot pressing at a temperature of 165 °C with a pressure of 5 MPa for 15 min. Finally, all the panels were cooled and kept in a conditioning room at a temperature of 25 °C ± 2 °C with relative humidity of 65 °C ± 2 °C for 7 days before further testing were conducted.

Testing and evaluation

Soil burial decay test was performed with a slight modification in sample size and it was conducted in laboratory room in a close container according to BS 1982-2 (1990) [18]. All the test specimens in the soil burial and borer test evaluation were cut into dimension 5 cm x 1 cm x 0.5 cm and 3 replicates were prepared from each type of boards. Oven dried weight of all test specimens was recorded before testing. Each sample was weighed at an accuracy of 0.01 g before being placed in the container. The soil burial test was performed in a polyethylene container filled with soil which is had passed through the mesh sieve size of 450 µm. All the specimens were then buried 3 cm deep in the container then were left in the incubator set up at a temperature of 27 °C for four different period times (2, 4, 6 and 8 weeks). All the specimens were removed, cleaned and oven dried in an oven overnight before final weight was recorded. Meanwhile, for borer test, it was carried out based on BS EN 47 (2005) [19]. The test specimens were tested in test arena in a close container that has a solid rubberwood planks attacked by powder post beetles (*M. rugicollis)*. Then the samples were buried in between the two solid rubberwood planks that previously exposed to the insects. Next, the container was sealed and stored in the room without direct sunlight. After 10 weeks exposure, each sample was lightly brushed and dried for the overnight before the final weight was taken. The fungus test on the test specimens was conducted according to the ASTM D 2017 (1995) [19] with slight modification. In this experiment, the samples were exposed to the fungal strains such as *G.trabeum Pers.ex.Fr* (ATCC No.11539) and *Trametes Versicolor (L.ex.Fr.) Pilat*. (ATCC No. 42462). Before the testing was conducted, the fungal strains approximately 10 mm was inoculated on petri dish MEA and grown in sterilized glass culture vessels for about two weeks before being exposed to the test samples. Ten replicates for each type of board samples were cut into dimension 25 mm x 25 mm x 9 mm. The incubation of fungi in culture bottles was carried out for twelve weeks in the culture room of controlled environment of 25 °C and 85 % relative humidity. After incubation of 12 weeks, all the test specimens were removed from the culture bottle and lightly brushed before oven dried for overnight in the oven at a temperature of 100 °C. The final weight at nearest 0.01 g was taken after oven dried process. All the samples in the evaluations were calculated as weight loss and expressed as a percentage as in equation (1).

The percentage of weight loss was calculated by using an equation (1):

$$\text{Weight loss, \%} = \frac{W_1 - W_2}{W_1} X\ 100 \qquad (1)$$

where;

W_1 = initial weight before exposure (g)

W_2 = weight after exposure (g)

Statistical Analysis

All samples were analysed by using SPSS software for the window (version 20) and reported as the mean values. The comparison of mean values was tested by using Duncan Multiple Range Test at $p < 0.05$.

RESULT AND DISCUSSIONS

Soil Burial Trend Analysis

The weight loss analysis results for the boards bonded with native oil palm (OPS_100), modified oil palm starch (MOPS_70:30) and the control board (UF_100) were illustrated in Fig.1. Overall results showed increasing values from week 2 to week 6 except for the board bonded with native oil palm starch (OPS_100) was reduced a value approximately 5.6 %.

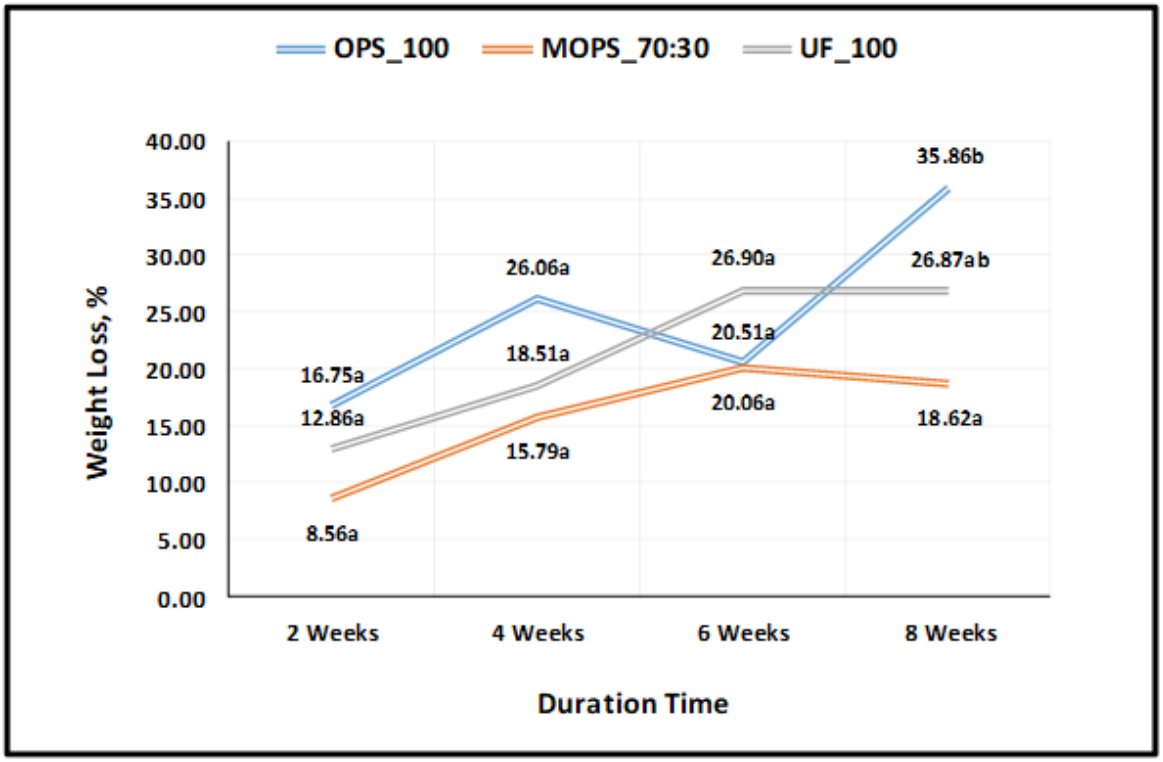

Figure 1. Soil burial trend analysis for boards bonded with native Oil palm starch (OPS_100); modified Oil palm starch (MOPS_70:30) and Urea formaldehyde (UF_100). Data is expressed as means; values in parentheses show standard deviations. Values with the same letter are not significantly different ($p < 0.05$)

However, this value was increased again to 35.86 % after 8 weeks exposure. Higher starch content may consume the food to microorganisms activity thus encouraged the higher degradation process. The lowest value was found in the samples bonded with modified oil palm starch (MOPS_100) which only obtained a value of 18.62 %. The addition of boric acid and Nano silicon dioxide (SiO_2) in the modified oil palm samples may attribute to the findings. Higher PVOH content up to 30 % in the starch modification may also affect the results. This statement was in agreement with previous research in soil burial test that used PVOH in the packaging film [10]. Statistical analysis also proved that boards bonded with native oil palm starch (OPS_100) and modified oil palm starch (MOPS_70:30) were significantly different to each other.

Borer Test Analysis

The borer test results had shown no significant different for all samples in weight loss percentage as could be seen in Table 1. The higher value was found in the sample bonded with native oil palm starch (OPS_100) obtained a value of 75.57 %. Meanwhile, the board bonded with modified oil palm starch (MOPS_70:30) achieved the lowest value (36.55 %) amongst other samples. MOPS_70:30 also proved more resistant approximately 4.38 % from the control sample (UF_100). This may be due to the hydrophilic starch characteristics was covered by using a 3 % of Nano silicon dioxide that was protected the samples from absorbed the moisture which is attributed from the soil.

Table 1. Borer test and fungal strains analysis

Samples	Weight Loss (%)		
	Borer Test (10 weeks)	Fungal strains Test (12 weeks)	
		Trametes Versicolor (CV)	*G.trabeum Pers.ex.Fr.* (GTRA)
OPS_100	73.57 (31.77)a	40.57 (13.36)b	38.59 (26.38)b
MOPS_70:30	36.55 (2.36)a	14.33 (8.22)a	16.30 (7.41)a
UF_100	38.15 (0.97)a	17.87 (3.21)a	17.75 (7.10)a

Note: Data is expressed as means; values in parentheses show standard deviations. Values with the same letter are not significantly different ($p < 0.05$). Native Oil palm starch (OPS_100); modified Oil palm starch (MOPS_70:30) and Urea formaldehyde (UF_100).

Fungal Strains Test Analysis

The mean percentage of weight loss due to decay by *C.versicolor* (CV) and *G.trabeum* (GTRA*)* was shown in Table 1. The high weight losses were observed in the samples bonded with native oil palm samples (OPS_100) when exposed to both CV and GTRA fungal strains. The weight loss values obtained were about 40.57 % and 38.59 %, respectively. These findings were supported by statistical analysis that shown a significant different at $p < 0.05$. Higher starch content and hydrophilic characteristics of oil palm starch samples [20] may also contribute to the findings. The board bonded with modified oil palm starch (MOPS_70:30) was shown the lowest values of 14.33 % and 16.30 % when exposed to both CV and GTRA, respectively. The control board (UF_100) was shown less resistance to fungi approximately 24.7 % and 8.90 % as compared to modified oil palm starch (MOPS_70:30) samples when it was exposed to both fungal strains.

CONCLUSIONS

The presence of Nano silicon dioxide (SiO_2) plus lower starch ratio in the starch modification process successfully influenced the findings. SiO_2 and boric acid were roles as a water repellent and antifungal agents thus prevented the microorganism's activity in the final particleboard. The starch-based adhesives made from modified oil palm starch can be suggested as a potential binder in the particleboard manufacturing in the future.

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REFERENCES

[1] Monteiro, S., Martins, J., Magalhães, F., and Carvalho, L. (2016). Low Density Wood-Based Particleboards Bonded with Foamable Sour Cassava Starch: Preliminary Studies. *Polymers (Basel)*, *8*(354), 1-11.
[2] Müller, C., Kües, U., and Schöpper, C. (2007). 16. Natural Binders, 347–381.
[3] de Melo, R. R., Stangerlin, D. M., Campomanes Santana, R. R., and Pedrosa, T. D. (2015). Decay and termite resistance of particleboard manufactured from wood, bamboo and rice husk. *Maderas. Ciencia y Tecnologia*, *17*(1), 55-62.
[4] Moubarik, A., Pizzi, A., Allal, A., Charrier, F., and Charrier, B. (2009). Fungal decay resistance and durability of wood products made from borax-impregnated wood and bonded with corn starch and tannin adhesive. *Annals of Forest Science*, *66*, 1-9.
[5] Pizzi, A. (2016). Wood products and green chemistry. *Annals of Forest Science*, *73*(1), 185–203.
[6] Imam, S. H., Gordon, S. H., Mao, L., and Chen, L. (2001). Environmentally friendly wood adhesive from a renewable plant polymer: characteristics and optimization. *Polymer Degradation Stability*, *73*(3), 529–533.
[7] Imam, S. H., Mao, L., Chen, L., and Greene, R. V. (1999). Wood adhesive from crosslinked poly (vinyl alcohol) and partially gelatinized starch: preparation and properties. *Starch - Stärke*, *51*(6), 225–229.
[8] Imam, S. H., Mao, L., Chen, L., and Greene, R. V. (1999). Wood Adhesive from Crosslinked Poly (Vinyl Alcohol) and Partially Gelatinized Starch: Preparation and Properties. *Starch - Stärke*, *51*(6), 225–229.
[9] Jhonson, A. C., and Yunus, N. (2009). Particleboards from Rice Husk: a brief introduction to Renewable materials of Construction. *Jurutera*, 12–15.
[10] Azahari, N. A., Othman, N., and Ismail, H. (2011). Biodegradation Studies of Polyvinyl Alcohol / Corn Starch Blend Films in Solid and Solution Media. *Journal of Physical Science*, *22*(2), 15–31.
[11] Bursali, E. A., Coskun, S., Kizil, M., and Yurdakoc, M. (2011). Synthesis, characterization and in vitro antimicrobial activities of boron/starch/polyvinyl alcohol hydrogels. *Carbohydrate Polymers*, *83*(3), 1377–1383.
[12] Dimens, N., and Issue, S. (2014). Enzymatic degradation of Poly (ε -Caprolactone) and Starch blends bontaining

SiO_2 nanoparticle by Amyloglucosidase and α-Amylase. *International Journal of Nano Dimension*, *5*(6), 549–555.
[13] Sulaiman, N. S., Hashim, R., Amini, M. H. M., Sulaiman, O., and Hiziroglu, S. (2013). Evaluation of the properties of particleboard made using oil palm starch modified with epichlorohydrin. *BioResources*, *8*(1), 283–301.
[14] Noor, M. A. M., Dos Mohd, A. M., Islam, M. N., Mymensingh, Mehat, and N. A. (1999). Physico-chemical Properties of Oil Palm Trunk Starch. *Starch-Stärke*, *51*(8–9), 293–301.
[15] Norani Abd Karim, Rokiah Hashim, Othman Sulaiman, and Salim Hiziroglu (2015). Extraction of Oil Palm Starch: A Comparative Study. *International Conference Environmental Research and Technology (ICERT 2015)* 121–125.
[16] Yin, Y., Li, J., Liu, Y., and Li, Z. (2005). Starch crosslinked with poly (vinyl alcohol) by boric acid. *Journal of Applied Polymer Science*, *96*(4), 1394–1397.
[17] Tang, S., Zou, P., Xiong, H., and Tang, H. (2008). Effect of nano-SiO2 on the performance of starch/polyvinyl alcohol blend films. *Carbohydrate Polymers*, *72*(3), 521–526.
[18] British Standard BS 1982-2, (1990) Fungal resistance of panel products made of or containing materials of organic origin. Method for determination of resistance to cellulose-decomposing microfungi.
[19] British Standard BS EN 47, (2005) Wood preservatives - Determination of the toxic value against larvea of Hylotropes bajulus (Linnaeus) - Laboratory method. *British Standard Institution*, London, 1–24.
[20] Maran, J. P., Sivakumar, V., Thirugnanasambandham, K., and Sridhar, R. (2014). Degradation behavior of biocomposites based on cassava starch buried under indoor soil conditions. *Carbohydrate Polymers*, *101*, 20–8.