

Effect of Microwave Pretreatment on the Properties of Particleboard Made from Para Rubber Wood Sawdust with the Addition of Polyhydroxyalkanoates

(Kesan Prarawatan Ketuhar Gelombang Mikro ke atas Sifat Papan Partikel yang Diperbuat daripada Habuk Gergaji Kayu Getah Para dengan Polihidroksialkanoat)

JUTARUT IEWKITTAYAKORN*, PIYAPORN KHUNTHONGKAEW, WILAIWAN CHOTIGEAT & KUMAR SUDESH

ABSTRACT

The aim of this study was to investigate the effect of microwave pretreatment on the properties of particleboard made from para rubber wood sawdust with the addition of polyhydroxyalkanoates (PHAs). Para wood sawdust was pretreated with microwaves at different wattages (400, 600 and 800 W) and pretreatment times (5, 10 and 15 min) before being mixed with 10 wt. % of pure PHAs and 10 wt. % freeze-dried cells which contained 46.90% wt. % PHA, in order to make particleboard. The physical and mechanical properties of the particleboard panels obtained were also tested based on the Japanese Industrial Standards A 5098 for particleboard (JIS A5908) including modulus of rupture (MOR), internal bonding (IB), water absorption (WA) and thickness swelling (TS). The results showed that only panel made from 10 wt. % freeze-dried cells and treated with 400 watts for 5 min passed the minimum requirement of IB (1.62 ± 0.06 MPa) and nearly passed that of MOR (7.86 ± 0.67 MPa) but not passed that of WA ($40.09 \pm 6.53\%$) and TS ($34.69 \pm 5.43\%$). The panels without the addition of PHA cannot test WA and TS values because the panels dispersed immediately when submerged in water. The MOR of panels decreased with increasing wattages and pretreatment time. The wattage and pretreatment time of microwave had little effect on the WA and TS. Based on this study, microwave treatment is an appropriate method for the pretreatment of materials and PHAs can be promoted as potential additives for making particleboard.

Keywords: Microwave; para rubber wood sawdust; particleboard; PHA; polyhydroxyalkanoates

ABSTRAK

Tujuan penyelidikan ini adalah untuk mengkaji kesan prarawatan ketuhar gelombang mikro pada sifat papan partikel yang diperbuat daripada habuk gergaji kayu getah para dengan penambahan polihidroksialkanoat (PHA). Habuk gergaji kayu getah para telah diprарawat dengan ketuhar gelombang mikro pada watt yang berbeza (400, 600 dan 800 W) dan masa prarawatan (5, 10 dan 15 min) sebelum dicampur dengan 10 % bt. PHA tulen dan 10 % bt. sel kering sejuk beku yang mengandungi 46.90% % bt. PHA untuk membuat papan partikel. Sifat fizikal dan mekanik panel papan partikel yang diperolehi juga diuji berdasarkan Piawaian Industri Jepun - A 5098 bagi papan partikel (JIS A5908) termasuk modulus rekahan (MOR), ikatan dalaman (IB), penyerapan air (WA) dan ketebalan pembengkakan (TS). Keputusan kajian menunjukkan bahawa hanya panel yang diperbuat daripada 10 % bt. sel kering sejuk beku dan dirawat dengan 400 watt untuk 5 min melepasi keperluan minimum IB (1.62 ± 0.06 MPa) dan hampir melepasi MOR (7.86 ± 0.67 MPa) tetapi tidak melepasi WA ($40.09 \pm 6.53\%$) dan TS ($34.69 \pm 5.43\%$). Panel tanpa penambahan PHA tidak boleh menguji nilai WA dan TS kerana panel terserak serta-merta apabila ditenggelami air. Panel MOR menyusut dengan peningkatan watt dan masa prarawatan. Kuasa dan masa prarawatan ketuhar gelombang mikro mempunyai sedikit kesan ke atas WA dan TS. Berdasarkan kajian ini, rawatan ketuhar gelombang mikro adalah satu kaedah yang sesuai untuk prarawatan bahan dan PHA boleh dipromosikan sebagai aditif yang berpotensi untuk membuat papan partikel.

Kata kunci: Habuk gergaji kayu getah para; ketuhar gelombang mikro; papan partikel; PHA; polihidroksialkanoat

INTRODUCTION

The rubber tree (*Hevea brasiliensis*) is a precious economic crop in Thailand and Malaysia. It is the primary source not only of natural rubber but also of rubber wood. The life cycle of the rubber tree in producing milky latex (the primary source of natural rubber) is 25-30 years; after this age, the tree is usually harvested and used in wood industries to make furniture and wood products. In addition, the branches and the trunks are also used as raw materials to make particleboard panels. Para rubber wood

sawdust or rubber wood dust which is a major by-product of cutting, grinding, drilling or otherwise pulverizing wood with a saw or other mechanical device can also be considered as a new source of raw materials that can replace rubber wood in manufacturing panels. Moreover, the utilization of sawdust could help to reduce the amount of wood from forest which is wasted, by using the wasted products as a raw material in the production of valuable products such as particleboard. Particleboard is widely used for making furniture, interior decoration and appliances.

Generally, the particleboard production process uses urea-formaldehyde (UF) as an adhesive (Dunky 1997). However UF is classified as a hazardous chemical with carcinogenic properties. Therefore, the investigation of other types of adhesive, which can be used as bonding materials for para rubber wood sawdust is potentially interesting.

Polyhydroxyalkanoates (PHAs) are biopolymers, which can be synthesized by various types of microorganisms such as, *Cupriavidus necator*, *Bacillus megaterium* and *Pseudomonas putida* under certain nutrient limited conditions but in the presence of excess carbon sources (Merugu et al. 2012). The properties of PHAs are similar to those of some thermoplastics with melting temperatures ranging from 50°C to 180°C and elastomeric materials (Sudesh et al. 2000). However, unlike most petrochemical based plastics, PHAs are biodegradable and biocompatible. PHAs are attractive materials for various applications in many industries such as medicine, agriculture and tissue engineering, the production of nano composites and polymer blends and processes such as chiral synthesis (Philip et al. 2007). Recently, Baskaran et al. (2012) studied the application of PHAs as an adhesive in the particleboard production process. The results showed that the properties of particleboard made from oil palm trunks with the addition of only PHAs were superior to those of particleboard without the addition of PHAs. Thus, certain types of PHAs have the potential to be used as an adhesive to make particleboards.

The type of adhesive used is one of the main factors in the manufacture of particleboard and the pretreatment system is also important. Steam treatment is the conventional method used to hydrolyze starch granules and promote the proper distribution of PHAs to fill up the spaces between the particles (Baskaran et al. 2013). However, it has been noted that this treatment requires high consumption of power and takes a long time. Therefore other treatments requiring lower power consumption and taking a shorter time such as microwave treatment, would be of interest to manufacturers of particleboard. Microwave has been widely applied for improving wood properties (Torgovnikov & Vinden 2009).

Therefore, the objective of this research was to investigate the effect of microwave pretreatment on the properties of particleboard made from para rubber wood sawdust using PHAs in the form of freeze-dried cells and pure PHA as an adhesive. The freeze-dried cells are fermented cells which consist of PHA polymers, while the pure samples are mainly PHA granules obtained through an extraction process described below.

MATERIALS AND METHODS

FREEZE-DRIED CELL AND PURE PHA

The PHA used in this study were bacterial polyesters synthesized from *Cupriavidus necator* Re2058 (Paramasivam et al. 2016). The PHA was added into the panels in the form of freeze-dried cells and pure PHA.

The pure form of PHA used in this study consisted mainly of PHA granules obtained from extraction which was carried out by adding 1 L chloroform into 10 g of freeze-dried cells and stirring for 5 days at room temperature, then filter it using Whatman filter paper No. 1, leaving the cell debris and the supernatant was then concentrated through rotary evaporation and the polymer precipitated by dropping it in cold methanol. The excess methanol was then removed and the polymer allowed to dry in a petri dish under a fume hood until pure PHA (white powder) was observed (Chia et al. 2010).

MICROWAVE PRETREATMENT OF PARA RUBBER WOOD SAWDUST

Para rubber wood sawdust was obtained directly from Rataphum Parawood Industry, Songkhla, Thailand. It was then dried in an oven at 103±2°C for 24 h or until the moisture content reached 2-3% which was considered as a dry basis (Rahman et al. 2013), before being milled and sieved to a size of 1 mm. Then, the samples were mixed with water in a ratio of 1:2 before being treated with microwaves in a microwave oven (LG model MS2323) with various levels of microwave power (400, 600 and 800 watt) and pretreatment times (5, 10 and 15 min).

The pretreated samples were then dried at a temperature of 65°C for 24 h in a hot air oven until a constant weight was reached.

PRODUCTION OF PARTICLEBOARD PANELS

PHAs in the form of freeze-dried cell and pure PHA were mixed with 10% wt. of the pretreated para wood sawdust which had been subjected to different pretreatments of microwave power and time and agitated in a blender for 5 min until a homogenous composition was achieved. There were thus 20 experiments as shown in Table 1. Each experiment was conducted in three replicates; hence 60 panels were made. The panels were manufactured in a compression molding machine (VDU-100 model LCC-140). The mixture was placed in a mold in order to manually form the panels, then pressed using the compression molding machine at a temperature of 180°C and a pressure of 200 kg/cm² for 20 min. The dimensions of the panels used in the experiments were 20.5 × 20.5 × 0.5³ cm. All the panels were then conditioned in a refrigerator at a temperature of 20°C and a relative humidity of 65% until an equilibrium moisture content of 12% was reached corresponding to that used in Baskaran et al. (2013). Then, the panels were cut based on the Japanese Industrial Standards (JIS 2003) and their mechanical and physical properties were analyzed.

TESTING OF PARTICLEBOARD PROPERTIES

Mechanical and physical testing of the particleboard panels were carried out based on the Japanese Industrial Standards A 5098 (JIS. 2003) in terms of the modulus of rupture (MOR), internal bond strength (IB), water absorption (WA) and thickness swelling (TS).

TABLE 1. Experiments of particleboard production

Experiment	Type of PHA	Level of microwave power (watts)	Pretreatment time (min)
1			5
2	Freeze-dried cells	400	10
3			15
4			5
5	Freeze-dried cells	600	10
6			15
7			5
8	Freeze-dried cells	800	10
9			15
10			5
11	Pure PHA	400	10
12			15
13			5
14	Pure PHA	600	10
15			15
16			5
17	Pure PHA	800	10
18			15
19			Pure PHA
20*	No PHA added	No treatment	No

*control

MODULUS OF RUPTURE AND INTERNAL BOND STRENGTH (JIS. 2003)

Specimens of the particleboard with dimensions of $5 \times 13 \text{ cm}^2$ and $5 \times 5 \text{ cm}^2$ were prepared from each panel in order to investigate the MOR and IB, respectively. Both the tests were conducted on an Instron Testing System, Model UTM-3365 with a load cell capacity of 100 N.

WATER ABSORPTION AND THICKNESS SWELLING (JIS. 2003)

Square particleboard specimens with dimensions of $5 \times 5 \text{ cm}^2$ were prepared from each panel in order to test the WA and TS. The square samples were soaked in water at room temperature (25°C) for 24 h. The weight and thickness of the square sample were measured before and immediately after soaking and used to calculate the WA and TS. The results were reported in terms of the swelling as a percentage of the value before soaking.

STUDY OF THE MORPHOLOGICAL PROPERTIES OF THE PARTICLEBOARDS

A scanning electron microscope (JSM-5800LV) was used to study the morphological properties of the para rubber wood sawdust and the bonding quality between the sawdust particles with the addition of freeze-dried cells and pure

PHA in the panels. The samples studied were cut with a cross section of $0.5 \times 0.5 \text{ cm}^2$ before being coated with gold by an ion sputter coater.

RESULTS AND DISCUSSION

EFFECT OF MICROWAVE PRETREATMENT ON PARA RUBBER WOOD SAWDUST

The morphological analysis of the para rubber wood sawdust after microwave pretreatment is shown in Figure 1. It can be seen that the microstructure of the pretreated sawdust particles was more obviously porous and denatured than the sawdust particles which were not subjected to the pretreatment process. Moreover, the pretreated sawdust particles after treatment with a microwave power of 800 watts showed the highest degree of porosity compared to the sawdust particles pretreated with microwave power of 600 watts and 400 watts, with the same pretreatment time. Further, the structure of the pretreated sawdust particles pretreated for 15 min showed a greater degree of denaturing than those pretreated for 10 and 5 min at the same microwave power. These results probably contributed to the bonding ability between the PHA and the pretreated sawdust particles.

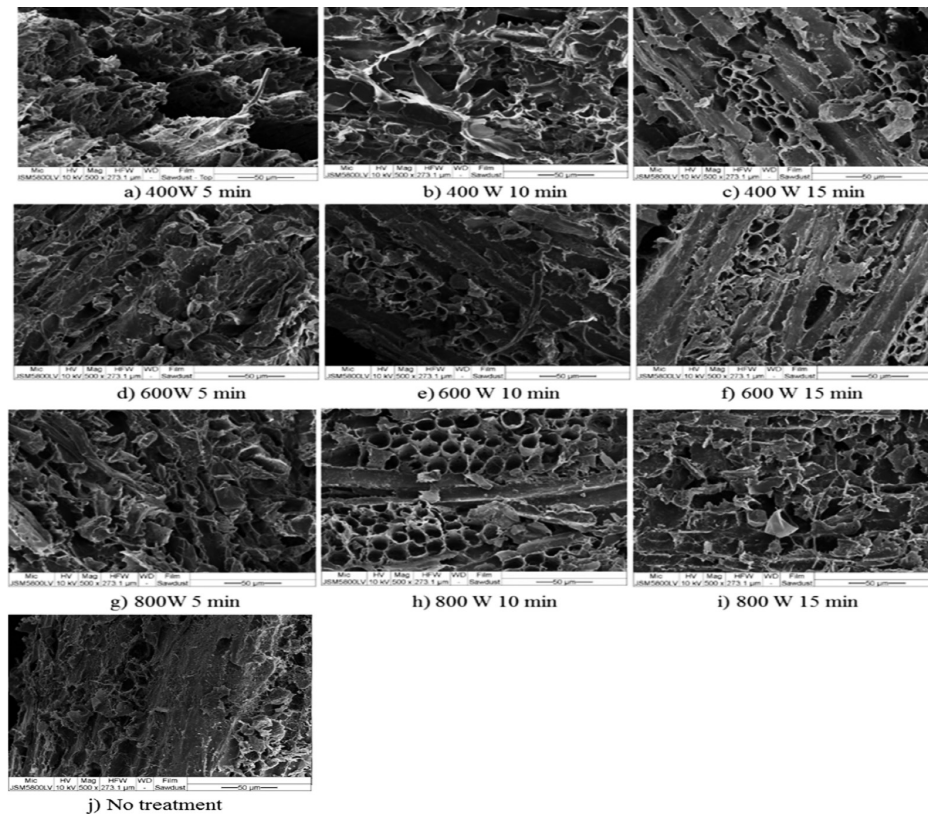


FIGURE 1. Scanning electron micrographs of cross sections of the para rubber wood sawdust particles after microwave pretreatment

EVALUATION OF THE MECHANICAL AND PHYSICAL PROPERTIES OF THE PARTICLEBOARD PANELS

MODULUS OF RUPTURE AND INTERNAL BOND STRENGTH

The MOR results of the specimens are shown in Figure 2. The samples treated with microwaves and freeze dried cells as well as pure PHA had lower MOR values than those of the control samples. It was also clear that the panels made with pure PHA showed a decreasing trend of MOR values as the pretreatment time increased above 10 min whereas the panels made using freeze-dried cell PHA showed a decreased MOR with increasing pretreatment time at 400 watts treatment. These results correspond to Hermoso and Vega (2016) that microwave has effect on the MOR of wood. MOR will decrease as a function of the MW energy and time consumption applied. It was however observed that only two experiments including experiment 2 (Treatment with 400 watts for 5 min) and exp. 20 (no PHA added and no treatment) of the panels nearly passed and the minimum requirement of the Japanese Industrial Standard A 5098 (JIS 2003) which specifies a minimum MOR value of 8.0 MPa, giving the MOR value of 7.86 ± 0.67 and 11.29 ± 1.41 MPa, respectively.

The IB values of the specimens are shown in Figure 3. It was noted that all the panels passed the minimum requirement of the Japanese Industrial Standard A 5098 (JIS 2003) of a minimum IB strength of 0.15 MPa.

The panels made with freeze-dried cell PHA gave higher IB values than the panels made with pure PHA. This might be due to the non PHA cellular material in the freeze-dried cells improving the bonding ability of binderless particleboard. It was also noted that the panel obtained from experiment 2 (Treatment with 400 watts for 5 min) gave the highest IB values (1.62 ± 0.06 MPa).

EVALUATION OF THE WATER ABSORPTION AND THICKNESS SWELLING OF THE PANELS

The dimensional stability of the specimens in terms of water absorption and thickness of swelling are shown in Figures 4 and 5, respectively. The results showed that panels made with freeze-dried cell produced lower values for WA and TS than the panels made with pure PHA. This result disagrees with the previous study of Baskaran et al. (2012) in which the TS and WA of the panels made from palm trunk with the addition of freeze-dried cell were much higher than those made with pure PHA. However, none of the panels passed the requirement of the Japanese Industrial Standards which was in agreement with the findings of Baskaran et al. (2012). A typical type 8 particleboard should not have a TS exceeding 12%. The lowest TS value obtained in this study was 21% from the panel pretreated with 600 watts for 10 min. This might be due to PHAs being insoluble in water whereas UF is water soluble. Thus, the particle bonds resulting from the PHAs were not as strong as the bonds from UF when the particleboard was submerged in water.

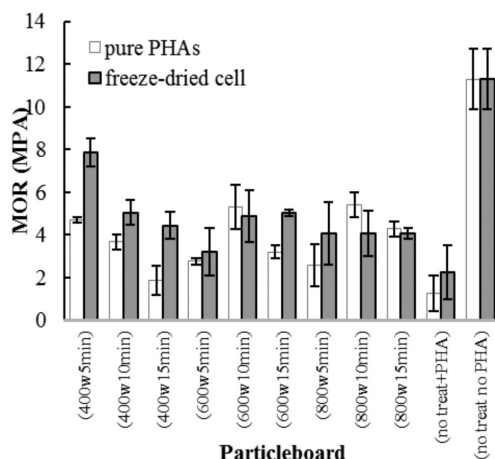


FIGURE 2. Modulus of rupture of the experimental panels

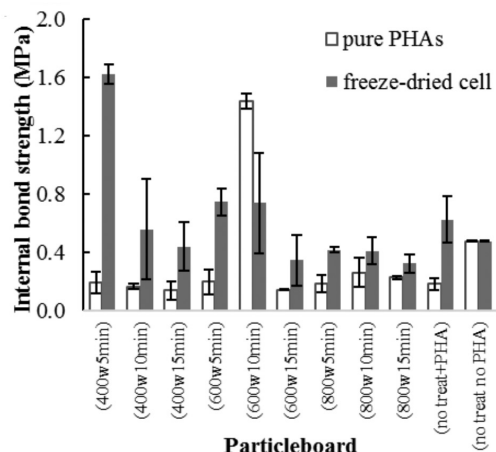


FIGURE 3. Internal bond strength of the experimental panels

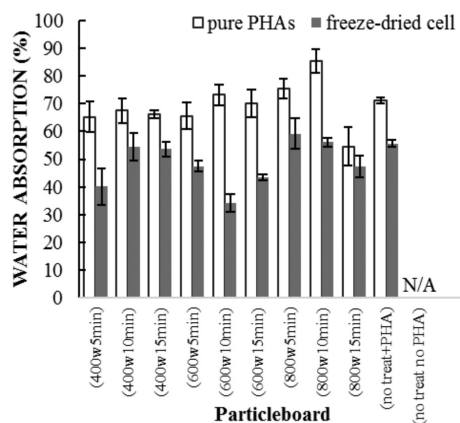


FIGURE 4. Water absorption of the experimental panels

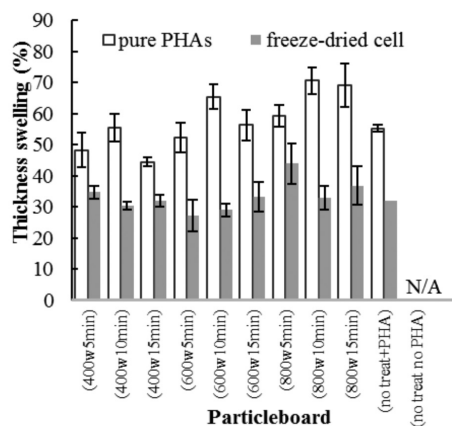


FIGURE 5. Thickness swelling of the experimental panels

More water was able to penetrate into the particleboard produced using PHAs resulting in the particle adhesion breaking down, leading to higher swelling. In addition, it was noted that the WA and TS values of the panels pretreated with various wattages and pretreatment times gave similar values for the two types of PHAs used in their manufacture, i.e. the values for the freeze dried PHA were all broadly similar as were all those for the pure cell PHA. Thus, the wattage and pretreatment time had little effect on the WA and TS values of the panels.

Moreover, it was observed that the panels without the addition of PHA produced no WA and TS values because the panels dispersed immediately when submerged in water. This result confirms that PHAs act as a binder.

EVALUATION ON THE MICROSTRUCTURE OF THE PANELS

The scanning electron microscope (SEM) of the sawdust particles pretreated with microwaves had a rougher structure than that of untreated particles (Figure 6). This

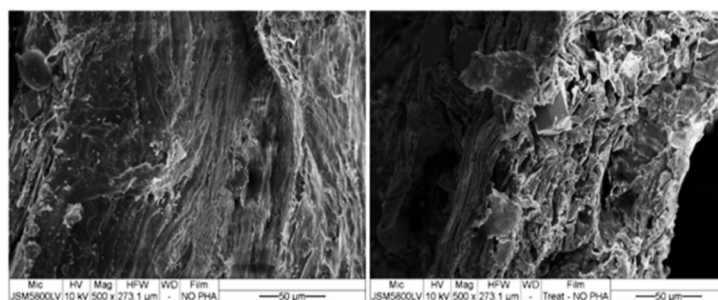


FIGURE 6. Scanning electron micrographs of cross sections of binderless panels from particleboard without the addition of PHAs, (a) no PHA added and no treatment, (b) treatment with 500 watts no PHA added

result can be concluded that the microwave treatment was able to increase the surface area which in turn supported the interaction between the PHAs and the particles. In the SEM of the pretreated particleboard the PHAs in the form

of both freeze-dried cells and pure PHA can clearly be seen dispersed within the cells either encrusted on them or filling up the spaces between the sawdust particles (Figures 7 and 8).

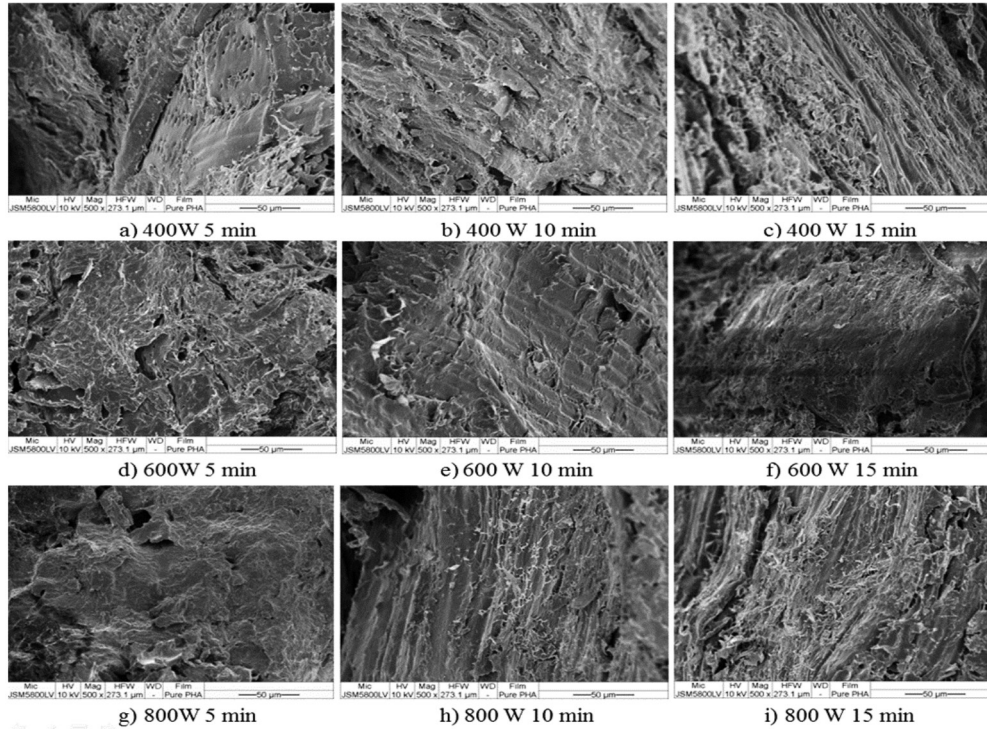


FIGURE 7. Scanning electron micrographs of cross sections of binderless panels from particleboard with the addition of pure PHAs at different microwave pretreatment times and wattages

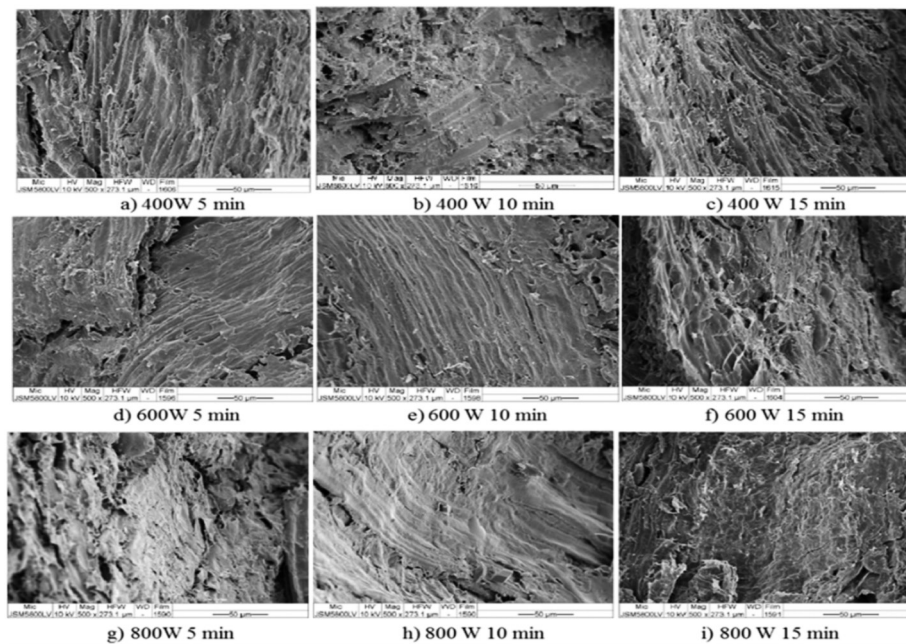


FIGURE 8. Scanning electron micrographs of cross sections of binderless panels from particleboard with the addition of freeze-dried cell PHA at different microwave pretreatment times and wattages

CONCLUSION

The pretreated sawdust particles treated with microwaves showed obvious porosity and were more denatured than untreated sawdust particles. The microwave has also effect on the mechanical properties particleboard panels. The wattage and pretreatment time had an influence on the hardness of the panels, but little effect on their WA and TS values. The mechanical properties of panels made from 10 wt. % freeze-dried cells and treated with 400 watts for 5 min pass requirement of the Japanese Industrial Standards A 5098 for particleboard (JIS A5908) which specifies a minimum MOR and IB, except the WA and TS values.

ACKNOWLEDGEMENTS

This research was supported by the Department of Molecular Biotechnology and Bioinformatics, Faculty of Science, Prince of Songkla University and the Long Term Research Grant Scheme (LRGS), Ministry of Education, Malaysia.

REFERENCES

- Baskaran, M., Hashim, R., Sudesh, K., Sulaiman, O., Hiziroglu, S., Arai, T. & Kosugi, A. 2013. Influence of steam treatment on the properties of particleboard made from oil palm trunk with addition of polyhydroxyalkanoates. *Ind. Crop. Prod.* 51: 334-341.
- Baskaran, M., Hashim, R., Said, N., Raffi, S.M., Balakrishnan, K., Sudesh, K., Sulaiman, O., Arai, T., Kosugi, A., Mori, Y., Sugimoto, T. & Sato, M. 2012. Properties of binderless particleboard from oil palm trunk with addition of polyhydroxyalkanoates. *Composites Part B* 43(3): 1109-1116.
- Chia, H.K., Ooi, F.T., Saika, A., Tsuge, T. & Sudesh, K. 2010. Biosynthesis and characterization of novel polyhydroxyalkanoate polymers with high elastic property by *Cupriavidus necator* PHB-4 transformant. *Polym. Degrad. Stab.* 95: 2226-2232.
- Dunky, M. 1997. Urea-formaldehyde (UF) adhesive resins for wood. *International Journal of Adhesion & Adhesives* 18: 95-107.
- Hermoso, E. & Vega, A. 2016. Effect of microwave treatment on the impregnability and mechanical properties of *Eucalyptus globulus* wood. *Maderas, Cienc. tecnol.* 18(1): doi.org/10.4067/S0718-221X2016005000006.
- JIS-A 5908, 2003. Particleboards. *Japanese Standard Association*, Tokyo, Japan. p.1-24.
- Merugu, R., Rudra, M.P.P., Girisham, S. & Reddy, S.M. 2012. PHB (Polyhydroxybutyrate) production under nitrogen limitation by *Rhodobacter capsulatus* KU002 isolated from tannery effluent isolated from tannery effluent. *Int. J. Chem. Tech. Res.* 4(3): 1099-1102.
- Paramasivam, M., Chhajer, P., Kosugi, A., Arai, T., Brigham, C.J. & Sudesh, K. 2016. Production of P(3HB-co-3HHx) with controlled compositions by recombinant *Cupriavidus necator* Re2058/pCB113 from renewable resources. *Clean Soil Air Water*. DOI: 10.1002/clea.201500714.
- Philip, S., Keshavarz, T. & Roy, I. 2007. Review polyhydroxyalkanoates: Biodegradable polymers with a range of applications. *J. Chem. Technol. Biotechnol.* 82: 233-247.
- Rahman, Kh-S., Islam, Md. N., Rahman, Md. M., Hannan, Md. O., Dungani, R. & Khalil, HPS. Ab. 2013. Flat-pressed wood plastic composites from sawdust and recycled polyethylene terephthalate (PET): Physical and mechanical properties. *SpringerPlus.* 2: 629.
- Sudesh, K., Abe, H. & Doi, Y. 2000. Synthesis, structure and properties of polyhydroxy-alkanoates: biological polyesters. *Prog. Polym. Sci.* 25(10): 1503-1555.
- Torgovnikov, G. & Vinden, P. 2009. High-intensity microwave wood modification for increasing permeability. *Forest Products Journal* 59(4): 84-92.
- Jutarutlew kittayakorn*, Piyaporn Khunthongkaew & Wilaiwan Chotigeat
Department of Molecular Biotechnology and Bioinformatics
Faculty of Science, Prince of Songkla University
Hat Yai, Songkhla, 90112
Thailand
- Wilaiwan Chotigeat
Center for Genomics and Bioinformatics Research
Department of Molecular Biotechnology and Bioinformatics
Faculty of Science, Prince of Songkla University
Hat Yai, Songkhla, 90112
Thailand
- Kumar Sudesh
Ecobiomaterial Research Laboratory
School of Biological Sciences
Universiti Sains Malaysia
11800 Penang, Pulau Pinang
Malaysia

*Corresponding author; email: jutarut.p@psu.ac.th

Received: 31 August 2016

Accepted: 17 January 2017