

Particle Oxidation Time for the Manufacture of Binderless Particleboard

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Abstract

The oxidation treatment using hydrogen peroxide and ferrous sulphate of wood particles can form free radicals of the wood chemical components essentially required in manufacturing binderless particleboard. The oxidation process is expected to have a certain optimal time. Therefore, the purpose of this study was to analyze relationship between the oxidation time and the characteristic of produced binderless particleboard. Three wood species from community forest, namely, sengon (*Paraserianthes falcata*), gmelina (*Gmelina arborea*), and mindi (*Melia azedarach*) were used for treatment. The air-dried wood particles of 10~20 mesh in size were oxidized using 20% hydrogen peroxide based on particle dry-weight and 5% ferrous sulphate based on hydrogen peroxide weight. The oxidized particles were conditioned in room at different periods of time (15, 30, 45, 60, 75, and 90 min) prior to the board production. The oxidized and conditioned particles were then hot-pressed at 180°C for 15 min with a specific pressure of 25 kgf cm⁻². Results showed that oxidation treatment for 15 min was sufficient to produce a binderless particleboard with good physical and mechanical properties. The characteristics of the particleboard, such as dimensional stability, modulus of rupture, modulus of elasticity, and internal bond were equivalent to that of particleboard made of particles with a longer oxidation time. The particle board made of sengon showed excellent thickness swelling (only 5.04%) and modulus of elasticity (37.184 kgf cm⁻²). This research result indicated that sengon was the most suitable raw material for binderless particleboard production compared to other observed wood species.

Key words: binderless particleboard, sengon, gmelina, mindi, oxidation, hydrogen peroxide.

Introduction

Nowadays, particleboard technology still faces some environmentally related problems. The major adhesives (96.6 %) (Li 2002) are made of formaldehyde compound; therefore they may cause formaldehyde emissions when used for particleboard production. Formaldehyde compound can cause cancer, irritation of the eyes and throat, and many respiratory problems. In addition, the popular adhesives today, such as urea formaldehyde, melamine formaldehyde, phenol formaldehyde, and isocyanate are resins that use raw materials of petroleum-derived compounds being not renewed (Roffael 1993). Thus, viewed from the aspects of raw materials, the adhesives are also not environmentally friendly.

Efforts have been made to develop environmentally friendly products, including the use of lignin as a renewable material for adhesive production. Utilization of lignin as an adhesive has actually been done for a long time. The first patent on the use of lignin from the waste of pulp industry as an adhesive existed in the 1900's (Nimz 1983). However, the patented adhesive still needs to be copolymerized with formaldehyde compounds (Santoso 2003).

The potential use of lignin as adhesive has inspired some researchers to develop methods that can directly activate the lignin compound in wood particle. The activation of wood lignin compounds is expected to bind the wood fibers or particles without adhesive addition (Karlsson and Westermarck 2002; Widsten *et al.* 2003; Widsten and Kandelbauer 2008).

One of the methods to activate the lignin for producing binderless particleboard is the oxidation of wood surface

using chemical reagent (Karlsson and Westermarck 2002; Widsten *et al.* 2003). Oxidizing wood particles with hydrogen peroxide and ferrous sulphate would produce hydroxyl radicals that can produce covalent bonds of particles through the hot-pressed application. Further investigation had been carried out to better understand the bonding mechanisms in manufacturing binderless particle board (Pantze *et al.* 2008) and it was concluded that direct ester formation was one of the important mechanisms that had a role in self bonding of particles. However, radicals produced in oxidation process were not very stable (Pantze 2006) as found in irradiated beech where only 10% of radicals survived at room temperature. However, in our previous study (Suhasman *et al.* 2010a; 2010b), the oxidation pretreatment of particles for 90~120 min were used for binderless particleboard production showed that the manufactured board had good properties especially in terms of dimensional stability and modulus of elasticity. The aim of this research was to investigate the effect of oxidation time in manufacturing binderless particleboard made of three wood species from community forest.

Materials and Methods

Material Preparation

The material used were obtained from three wood species of community forest namely; sengon (*Paraserianthes falcata* L Nielsen), gmelina (*Gmelina arborea*) and mindi (*Melia azedarach*). The wood in air-dried condition was converted into shavings by using shaving machine. The shavings were then converted to particle and sorted out. The particles used for the board production were

particles which passed through 10 mesh sieve and restrained at 20 mesh sieve.

Oxidation Temperature

Exothermic reaction during the oxidation process was measured using a thermocouple that was connected to cynorecorder. Wood particles that had been prepared were sprayed with hydrogen peroxide and ferrous sulphate in the blender. Levels of hydrogen peroxide used were 20% based on dry weight of the particles, while the levels of ferrous sulphate were 5% based on hydrogen peroxide weight. After spraying, the oxidized particles were immediately placed in plastic containers and the thermocouple plugged in the particle. Temperature changes were automatically recorded in 45 second intervals by the cynorecorder. This observation was held for 25 min.

Binderless Particleboard Production and Characteristics

Particles from each wood species were oxidized using hydrogen peroxide with level 20% based on particle dry-weight and ferrous sulphate in level 5% based on hydrogen peroxide weight. The oxidized particles were then conditioned at room condition with mean temperature of $\pm 29^{\circ}\text{C}$ and relative humidity of $\pm 80\%$ in various time, i.e. 15, 30, 45, 60, 75, and 90 min. These oxidized particles were then hot-pressed at the temperature of 180°C for 15 min with the specific pressure of 25 kgf cm^{-2} . The sizes of particleboards were $(30 \times 30 \times 0.7) \text{ cm}^3$ with target density of 0.75 g cm^{-3} . The produced particleboards were then conditioned for 14 days before being cut for subsequent test. The test was carried out according to Japanese Industrial Standard (JIS) A 5908 2003.

Results and Discussion

Oxidation Temperature

Exothermic reaction during oxidation process produced high temperature. The data of temperature changes as noted in the cynorecorder showed that the temperature in the early stage of oxidation process occurred very rapidly as shown in Figure 1. Sengon and mindi species reached peak temperature only within 1.5 min, whereas gmelina attained the peak temperature after 3.8 min. This phenomenon indicated that substitution of certain group of the wood chemical component, such as metoxyl group with radical group as mentioned by Widsten (2002) would have a different reaction speed. In other words, the reactivity of wood chemical compound in the oxidation process varied with different wood species.

Peak temperature attained within a short time (maximum 3.8 min) indicated that the oxidation process actually occurred very rapidly. However, in our experiment, hot pressing process which was applied immediately after exceeding the peak temperature generated an explosion in the compressed mat. This result indicated that despite the

fact that the decreasing temperature had occurred at a later stage, oxidation process was not fully completed. As we know, the acceleration of chemical reactions could be performed through the addition of a catalyst or increasing the temperature. In the case of this research, presumably the very rapid increase of temperature was due to the hot compression (180°C) which had caused the oxidation process in certain parts of the wood particles took place too quickly and simultaneously causing the explosion. Based on this fact, in manufacturing binderless particleboard, the shortest oxidation time taken before the hot-pressed application should be at least 15 min.

Binderless Particleboard Characteristics

In general, particleboard density was not influenced by the length of oxidation time. This fact could be seen from the distribution density of each board which had no particular tendency of being influenced by the differences in oxidation time. The trend of differences was actually more to the influence of the wood species. All three types of boards were designed to achieve the target density of 0.75 g cm^{-3} . However as shown in Figure 2, the highest density was shown by particleboard from sengon wood, while the lowest density was shown by the particleboard made of mindi wood. This phenomenon indicated that the three types of raw materials each had a different tendency in achieving their target density.

Visual observation during oxidation process showed that mindi wood reacted very rapidly at times when the previous particle that had been sprayed by hydrogen peroxide, it reacted directly when the particle then sprayed with ferrous sulphate. Since the occurred reaction was exothermic reaction, this process was easy to observe by increasing the particle temperature and releasing water vapor due to heating. This was different with both sengon and gmelina woods which still had a lag time of 2~3 min before the water vapor formed by the exothermic reaction. This quick process was an indication that mindi had the more reactive chemical compound during oxidation process in comparison with the other two species. Because of that reactivity, it was expected that the number of degraded chemical components also tend to be higher, resulting in lower board density.

Apart from its density, the moisture content of produced board had an opposite tendency. Particleboard made of sengon wood had the lowest moisture content than the other two. Nevertheless, the equilibrium moisture content of the three types of boards tend to be much lower than that of solid wood which is generally above 10%. Pressure at high temperatures (180°C) for 15 min would cause the reduction of amorphous regions in the cell wall of wood particles, so the hydrophobic of the board would decrease. At the temperature of 180°C , it had also exceeded the glass transition point of lignin (140°C) (Hill 2006), allowing lignin to move easily. Some part of the lignin could be exposed to the surface of particle as visually

observed with darker colour on the produced particleboard. Since lignin is a hydrophobic material, its presence may inhibit the absorption of water during the conditioning process. This is the reason the equilibrium moisture content of particleboard tends to be low.

Based on variation of moisture content parameter, the oxidation time appeared to have no specific correlation with

the variation of board moisture content. In other words, difference of oxidation time did not affect the moisture content of the board. This means that in the oxidation time range of 15~90 min, there was no significant effect on the equilibrium moisture content of produced board.

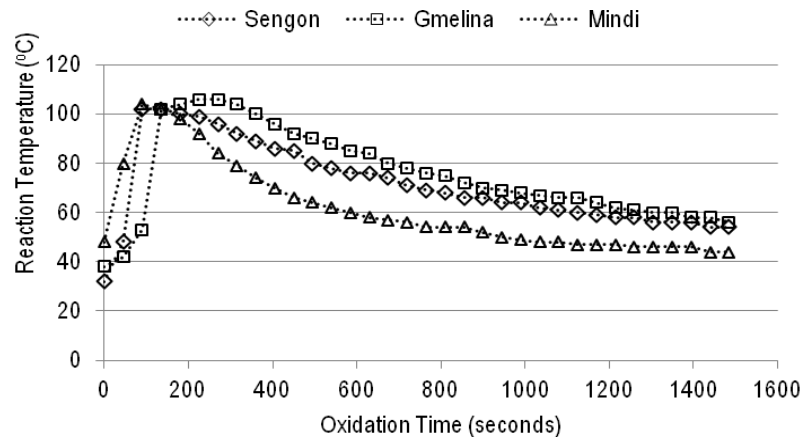


Figure 1. Oxidation reaction temperature.

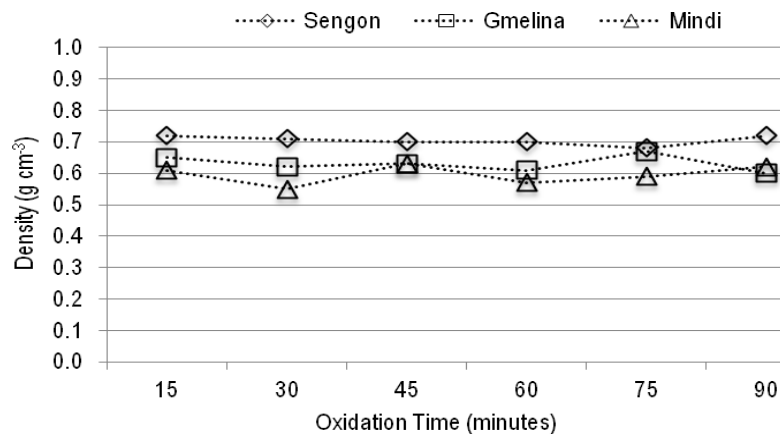


Figure 2. Density of particleboards in various oxidation time.

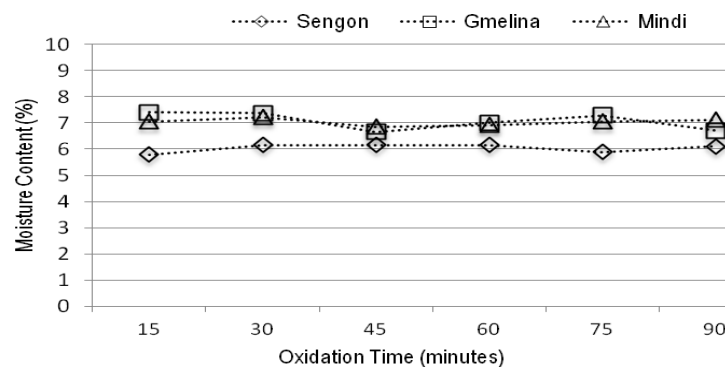


Figure 3. Moisture content of particleboards in various oxidation time.

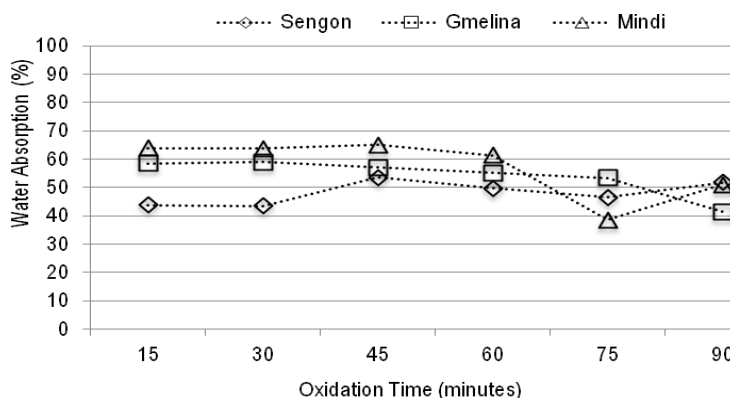


Figure 4. Water absorption of particleboards in various oxidation time.

Just like to moisture content, the lowest water absorption of particleboard also occurred in particleboard made of sengon wood, while if viewed from oxidation time, it showed that there was no significant difference in the various oxidation time. Figure 4 showed that water absorption of particleboard made of gmelina and mindi decreased slightly simultaneously with the increase of oxidation time, but sengon wood was quite the contrary as it slightly increased. Both tendency of decrease or increase were relatively small, indicating that it was only because of an intrinsic variation of particleboard and not due to the differences of oxidation time. This means that oxidation time treatment did not affect the water absorption properties of the board.

Results also showed that the thickness swelling of produced particle boards varied with wood species used as raw material. Figure 5 showed that the board made of sengon wood had the lowest thickness swelling. In other words, the best dimensional stability was found on the board made of this wood species. Board obtained from gmelina wood had the lowest dimensional stability. The results indicated that the high dimensional stability of binderless particleboard could be produced from sengon and mindi woods as their thickness swelling were below 12%, the maximum requirement of JIS A 5908 2003.

The fact that binderless particleboard has a low thickness swelling is one of the attractive characteristic, because the thickness swelling of particleboard has always been a problem with the conventional particleboard which uses adhesive. Even attempts to overcome these problems such as addition of paraffin or chemical modification of wood particles had been carried out. However, at the produced binderless particleboard, the highest dimensional stability was achieved without any extra effort. Apparently, changes in lignin structure due to the oxidation treatment had an important role in improving the dimensional stability of boards.

Considering the effect of oxidation time on the board thickness swelling characteristic, it appeared that oxidation time had no direct correlation with board thickness swelling parameter. This mean that the oxidation period from 15 to

90 min could be used in particleboard manufacturing process without any doubt of differences in dimensional stability.

The values of modulus of rupture (MOR) and modulus of elasticity (MOE) of each type of boards were presented in Figure 6 and 7. The data in the figure showed that the correlation between oxidation time to the MOR and MOE of board characteristics have the same tendency with the other parameters, i.e. there was no direct relationship. MOR values were relatively similar with different oxidation time applied. It seemed that hydroxyl radicals formed during the oxidation process remained stable at those intervals of time just as when the boards were hot-pressed, it could still produce a board with relatively similar MOR and MOE values.

Based on raw material types, it appeared that the board made of sengon wood had much higher MOR and MOE values than the other two boards. Since binderless particleboard manufacturing process solely rely on interactions between the chemical components in wood particles, so it could be concluded that the type of sengon wood experienced a more balanced oxidation process. However, lignin component had an adequate process of radical formation, whereas the cellulose component was not susceptible to interference. This needed to be considered because in the process of oxidation, although the hydroxyl radical that was formed tend to attack the electron-rich lignin (Nguyen 1982), cellulose and hemicellulose components could also be affected. Whereas, cellulose was a major component that played a role in the strength of wood.

In contrast to the other parameters of physical and mechanical properties, the highest value of internal bond held by the particleboard from mindi wood. As discussed earlier, the exothermic reaction temperature in mindi wood was higher than the sengon wood and was quicker than the gmelina wood. This indicated that mindi wood was more reactive in the oxidation process. This reactivity seemed to correlate directly with the formation of radical group during the oxidation process, as such when the board was hot-pressed it was able to form a stronger bond.

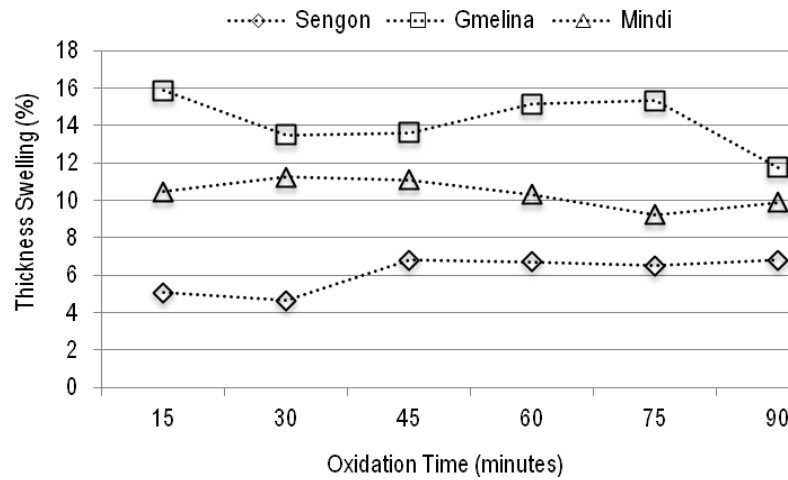


Figure 5. Thickness swelling of particleboards in various oxidation time.

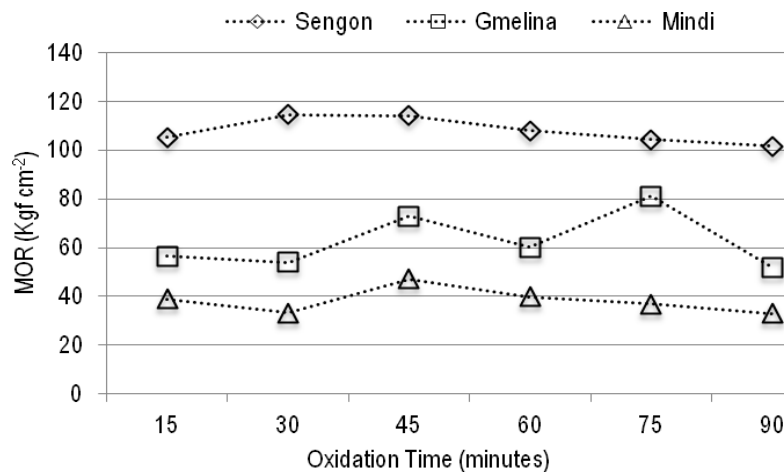


Figure 6. Modulus of rupture of particleboards in various oxidation time.

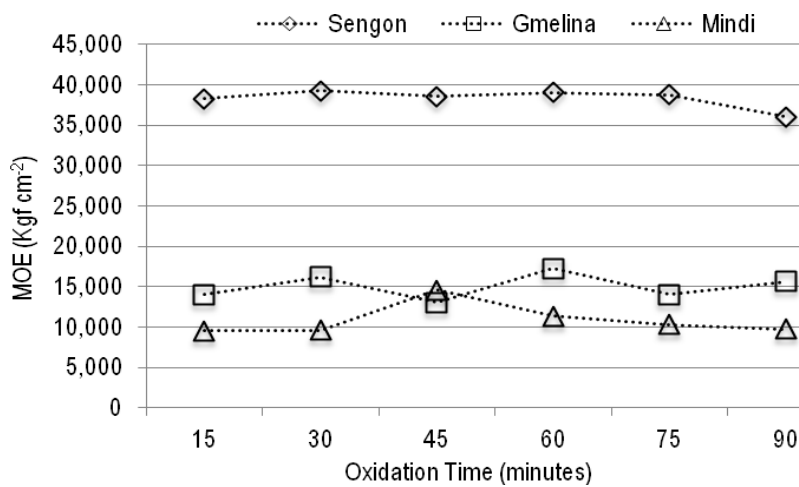


Figure 7. Modulus of elasticity of particleboards in various oxidation time.

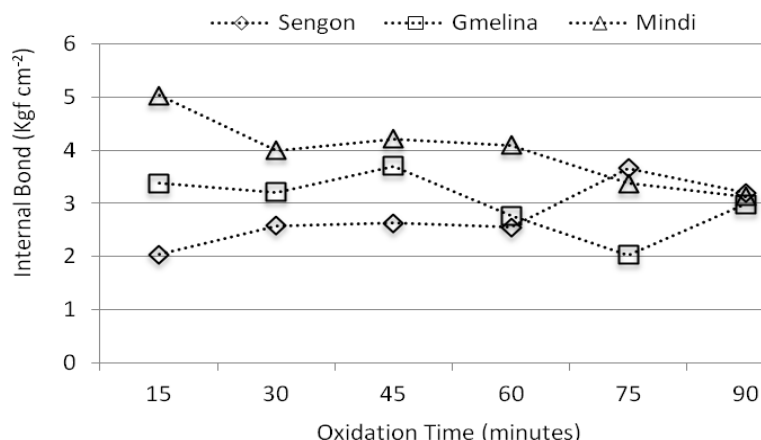


Figure 8. Internal bond of particleboards In various oxidation time.

Furthermore, when viewed from the relationship between the values of internal bond and the oxidation time as shown in Figure 8, it appeared that the tendency of internal bond values relatively similar to the other physical and mechanical properties of boards, such that there are no correlation with oxidation time. Thus, in terms of bonding formation, the time difference of 15~90 min did not give a specific effect in terms of bonding strength of the board.

Conclusions

Based on this study, it was concluded that peak temperature of oxidation process took place very rapidly (below 4 min), but complete oxidation had not been attained until several min. In terms of binderless particleboard manufacturing, oxidation for at least 15 min would be sufficient to produce binderless particleboard with good characteristics. Furthermore, due on relationship between oxidation time and binderless particleboard characteristics it could be concluded that there were no significant differences as well as certain trends in physical and mechanical properties of particleboard which were produced from particles oxidized with various time in the range of 15~90 min. This phenomenon is very attractive, because if this technology were to be developed on a industrial scale, it would be more flexible to adapt it to other production operations, without any fear that the quality of the product will decrease. Furthermore, when viewed from the characteristics of the board for each species, it appeared that the sengon wood is the most suitable species as raw material in manufacturing binderless particleboard. Its MOE is very excellent. Nevertheless, two other species (gmelina and mindi) remain useful as raw materials even though its products are not as good as the board from sengon wood species.

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References

- Hill, C.A.S. 2006. Wood Modification, Chemical, Thermal and Other Processes. John Wiley & Sons, Ltd.
- Japanese Industrial Standar (JIS) A 5908-2003. [JSA] Japanese Standards Association 2003. Particleboards. Japan.
- Karlsson, O; U. Westermark 2002. Resin-free Particleboard by Oxidation of Wood. In: Humphrey, P.E., Compiler. Proceedings of the 6th Pacific Rim Bio-Based Composites Symposium & Workshop on the Chemical Modification of Cellulosics. Portland, Oregon, USA. 149-153.
- Li, K. 2002. Use of Marine Adhesive Protein as A Model to Develop Formaldehyde-free Wood Adhesives. In: Humphrey, P.E., Compiler. Proceedings of the 6th Pacific Rim Bio-Based Composites Symposium & Workshop on the Chemical Modification of Cellulosics. Portland, Oregon, USA. 58-67.
- Nguyen, T. 1982. The Role of Lignin in Surface-Activated Bonding of Lignosellulose-Characterized by Differential Scanning Calorimetry. J. Adhesive 14(3-4): 283-294.
- Nimz, H. 1983. Lignin Based Wood Adhesive. In: Pizzi, A., Editor. Wood Adhesives, Chemistry and Technology. Marcel Dekker Inc. New York. 247-288.
- Pantze, A. 2006. Studies of Ester Formation on a Cellulose Matrix. Luleå University of Technology [Thesis]. Sweden.

- Pantze, A.; O. Karlsson; U. Westermark. 2008. Esterification of Carboxylic Acids on Cellulosic Material: Solid State Reactions. *Holzforschung* 62: 136-141
- Roffael, E. 1993. Formaldehyde Release from Particleboard and Other Wood Based Panels. Forest Institute Malaysia (FRIM).
- Santoso, A. 2003. Sintesis dan Pencirian Resin Lignin Resorsinol Formaldehida untuk Perekat Kayu Lamina . Institut Pertanian Bogor [Disertasi].
- Suhasman; M.Y. Massijaya; Y.S. Hadi; A. Santoso. 2010a. Binderless Particleboard Made from Wood Particles Activated using Hydrogen Peroxyde and Ferrous Sulphate. *Jurnal Ilmu dan Teknologi Kayu Tropis*. 8 (1).
- Suhasman; M.Y. Massijaya; Y.S. Hadi; A. Santoso. 2010b. Karakteristik Papan Partikel dari Bambu tanpa Menggunakan Perekat. *Jurnal Ilmu dan Teknologi Hasil Hutan*, Departemen Hasil Hutan IPB. 3 (1): 38-43
- Widsten, P. 2002. Oxidative Activation of Wood Fibers for the Manufacture of Medium-Density Fiberboard (MDF). Helsinki University of Technology [Dissertation].
- Widsten, P.; P. Qvintus-Leino; S. Tuominen; J.E. Laine. 2003. Manufacture of Fiberboard from Wood Fibers Activated with Fenton's Reagent ($H_2O_2/FeSO_4$). Germany. *Holzforschung* 57: 447-452.
- Widsten, P.; A. Kandelbauer. 2008. Laccase Applications in the Forest Products Industry: A Review. Elsevier. *Journal of Enzyme and Microbial Technology* 42 : 293-307.
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