

Hydrogen Peroxide and Ferrous Sulphate Activated Wood Particles for Binderless Particleboard

Suhasman¹⁾, Muh. Y Massijaya²⁾, Yusuf S Hadi²⁾, Adi Santoso³⁾

1) Post Graduate Student at Bogor Agricultural University, Bogor/
Faculty of Forestry, Hasanuddin University, Makassar

2) Forest Product Department, Faculty of Forestry,
Bogor Agricultural University, Bogor

3) Forest Products Research and Development Center, Ministry of Forestry, Bogor

Corresponding author: email suhasman@yahoo.com (Suhasman)

Abstract

A series of study have been conducted to analyze the effect of pretreatments and particle sizes on physical and mechanical properties of binderless particleboard (BP). The BP made of particles from sengon wood (*Paraseranthes falcataria* L Nielsen) and pine wood (*Pinus merkusii* Jungh et. de. Vriese) which passed through 10 mesh sieve, were used to analyze the effect of particle pretreatments. The pretreatments were: immersion in boiling water for 30 minutes; the immersion in boiling water followed by oxidation using hydrogen peroxide and ferrous sulphate; and oxidation using hydrogen peroxide and ferrous sulphate without immersion in boiling water. The results showed that the best phisycal and mechanical properties was found in board made from sengon wood particle with oxidation using hydrogen peroxide without immersion in boiling water; It had the internal bond (IB) of 6.95 kgf cm⁻² and the thickness swelling (TS) of 5.74%. In the next step, the sengon wood with several particles sizes namely; passed through 20 mesh, 10 mesh, 5 mesh, 2.5 mesh, and shaving were used to produced BP. The results indicated that the MOR decreases when the particle size increases, while the IB, MOE, and TS were relatively similar for all particle sizes.

Key words: binderless particleboard, hydrogen peroxide, oxydation, *Paraserianthes falcataria*, *Pinus merkusii*

Introduction

The development of manufacturing technology of binderless particleboard has been conducted by several researchers. The methods used such as steam injection pressing (Kawai *et al.* 2002, Xu *et al.* 2003, Widyorini *et al.* 2005a, 2005b, 2005c, Xu *et al.* 2005, 2006), the enzymatic activation (Kharazipour *et al.* 1998, Hüttermann *et al.* 2001, Widsten *et al.* 2004, Müller *et al.* 2007) or by oxidation using hydrogen peroxide and catalyst (Karlsson & Westermarck, 2002, Widsten *et al.* 2003). These efforts have been conducted to overcome the weakness

of the conventional particleboard technology using adhesive. According to Maloney (1993), about 90% of the wood panel products were made by using urea formaldehyde adhesive. Whereas formaldehyde compounds were potential causes of cancer, eye and throat irritation and respiratory problems (Marutzky 1989; Meyer *et al.* 1986; Henderson 1979; Baumann *et al.* 2000; in Li 2002).

The oxidation method used in manufacturing the binderless particleboard was one of the most interesting methods because of its simplicity in the process and the characteristics of its product. The

result of Karlsson & Westermark (2002) research by using norway spruce and scot pine woods indicated that particleboard which was made with this method had the internal bond of 14.5 kgf cm^{-2} , far above the standard which was determined in Japanese Industrial Standard (JIS) A 5908 2003. While its thickness swelling was 11% or under the JIS (Max. 12%). On the other hand, Widsten *et al.* (2004) reported that the particle which was oxidized using radiation method showed that hardwood group was more reactive than softwood, though there was no significant difference for oxidation using chemical material.

Although somewhat potential, so far there was no report about tropical wood usage in manufacturing binderless particleboard with oxidation method. Whereas it is possible that characteristic of the product will be strongly determined by the type of material used. Therefore the development of particleboard technology using tropical woods needs to be done. In Indonesia, the potential of wood utilization seem shifted towards usage of woods from community forest. Therefore this research was more focused on the utilization of woods which was more likely found in community forest, such as sengon. Furthermore, to evaluate the comparison between hardwood and softwood, pine wood was also used. Pine wood was preferred because it was much more developed as a community forest; whilst there was very limited kind of softwood found in Indonesia, mostly were agathis and pine wood.

In the oxidation method, accessibility of the wood chemical component was presumably considered as one of the key factor in determining the success of oxidation process. As presumed, the smaller the particle size, the accessibility of chemical component by the oxidator will be higher. This is proven in Karlsson

& Westermark (2002) research which indicated that omission of 24% of fine particle component (less than 0.25 mm in size) would reduce internal bond value about 1/3 in comparison with the board made from combination of coarse and fine particle. However, if the size of the particle was too small, it was potential to reduce mechanical strength of the product because of the lack of overlay area between particles. The objective of this research was to study the characteristics of binderless particleboard which was made with oxidation method, and to analyze the influence of particle size on the characteristic of its products.

Materials and Methods

The influence of particle pretreatments

Materials preparation

The samples used to analyze the effect of particle pretreatment on the properties of binderless particleboard, were two wood species namely sengon wood and pine wood. The woods in the air dry condition were then converted into shavings particles. The particles henceforth converted into fine particle using a hammer mill machine. The particles used in this research were particles which passed through 10 mesh sieve.

Pretreatments

Air dried particles were treated in the following way: 30 minutes immersion in boiling water named treatment (R-30); treatment (F-H) where the air dried particles directly oxidized with 20% hydrogen peroxide based on weight of dry particle and 5% ferrous sulphate based on weight of hydrogen peroxide, and the last one is (R30-F-H) consecutively following this: immersion in boiling water, air drying, oxidizing with 20% hydrogen peroxide and 5% ferrous sulphate.

Board manufacturing and testing

The oxidized particles were conditioned in room temperature for 90-120 minutes. Each type of particles had been made into a mat and hot pressed at temperature of 180°C for 20 minutes with a pressure of 25 kgf cm⁻². The sizes of the board were (30 x 30 x 0.7) cm³ with a target density of 0.75 g cm⁻³. The boards were then conditioned for 10 days before being cut into test samples. Parameters tested including water absorption (WA), thickness swelling (TS), modulus of rupture (MOR), modulus of elasticity (MOE), and internal bond (IB). The tests were carried out according to JIS A 5908 2003.

The influence of particle size

The material used in this research were sengon wood particles with different sizes, i.e. particles which passed through 20 mesh, 10 mesh, 5 mesh, 2.5 mesh, and shavings. Each type of particles was oxidized by hydrogen peroxide and ferrous sulphate as a catalyst at the same levels of previous research. Oxidized particles were then conditioned for 90-120 minutes and hot pressed at a temperature of 180°C for 15 minutes with a pressure of 25 kgf cm⁻². The size, density targets, and test procedure were similar to the previous stage.

Results and Discussion

The influence of particle pretreatments

The results showed that the dimensional stability of binderless particleboards using oxidation pretreatment were much better than the binderless particleboard using only immersion in boiling water pretreated without oxidation pretreatment. The TS of binderless particleboard using oxidation pretreatment were only 5.74-5.97% for sengon wood and 5.08-10.58% for pine wood in 24 hours water immersion. The TS was even better than the particleboard

made of sengon wood using 6% waterbased polymer isocyanate (Suhasman *et al.* 2007) and 10% melamine formaldehyde adhesives (Suhasman *et al.* 2008). This was an excellent indicator, since dimensional stability was considered a serious problem in commercial particleboard products.

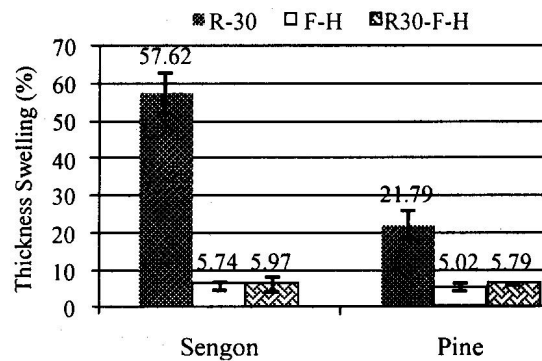


Figure 1 TS of binderless particleboard. (R-30: 30 minutes immersion in boiling water, F-H: Oxidation using hydrogen peroxyde and catalyst, R30-F-H: Combination of 30 minutes immersion in boiling water and oxidation).

The value of WA of binderless particleboard had the same pattern with dimensional stability. The WA of binderless particleboard with immersion in boiling water pretreatment without oxidation was 40% higher than the binderless particleboard using oxidation pretreatment for sengon wood, while for the pine wood was 17% higher.

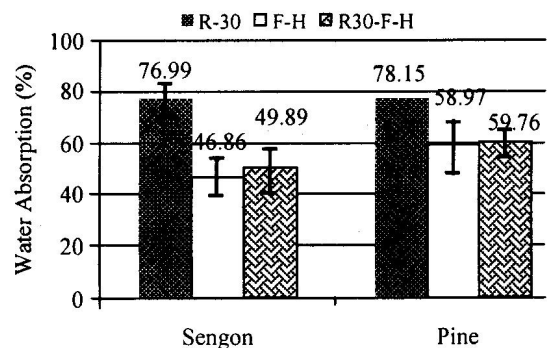


Figure 2 WA of binderless particleboard

Mechanical strength of board made from oxidation particle pretreatment were much higher than the boards made from particles treated with just immersion in boiling water. The MOR and MOE value of particleboard made of sengon particle were two times higher than the particle board using immersion in boiling water particle pretreatment, while the value of internal bond was 15 times higher. MOE and IB value of the board were even higher than the boards of the same type of wood that used water-based polymer isocyanate (Suhasman *et al.* 2007) which only reached 16,000 kgf cm⁻² for the value of MOE and 0.7 kgf cm⁻² for IB, as well as the particleboard using melamine formaldehyde (Suhasman *et al.* 2008) the MOE had only reached 12,060 kgf cm⁻² and IB 1.71 kgf cm⁻². However, for pine species, the values of mechanical properties were extremely low, leading to no prospect of further development.

In comparison to JIS A 5908 2003, it appeared that the particleboard made of sengon wood almost fulfilled all the parameters standard of the 8 types of particle board in terms of its TS (max 12%), MOR (min. 80 kgf cm⁻²), MOE (min 20,000 kgf cm⁻²), and IB (min 1.5 kgf cm⁻²). Only the particle board made with method of oxidation without immersion in boiling water pretreatment that had a lower MOR values from standard.

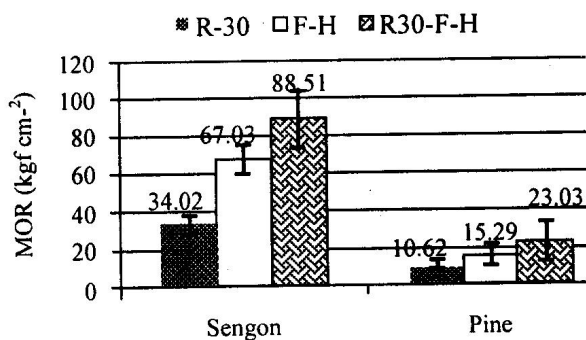


Figure 3 MOR of binderless particleboard

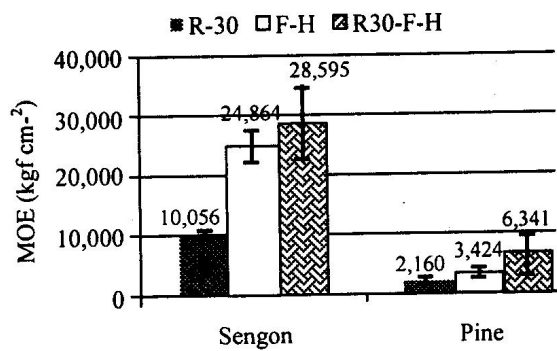


Figure 4 MOE of binderless particleboard

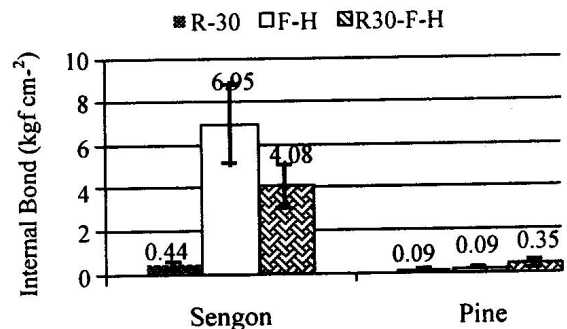


Figure 5 IB of binderless particleboard

However, for particleboard made of pine wood, its mechanical properties were very low. The distinction was caused by differences of wood density, where the pine wood had a density of 0.55 g cm⁻³ and sengon 0.33 g cm⁻³. Having a lower density, sengon wood would possibly achieve high compression ratios, so that the contacts between particles will also be more intensive. In addition, another important factor was the difference of lignin structure. Pine wood is grouped as soft wood i.e. the overall lignin was composed by guaiacyl units, while sengon is in the group of hard wood which lignin was composed by guaiacyl and siringyl units. Widsten *et al.* (2003) indicated that the reactivity of the two types of lignin units were different, in which siringyl lignin tend to be more reactive compared to the guaiacyl lignin.

The physical and mechanical properties were presented in Fig. 1-5, it also showed that the oxidation pretreatment without

immersion in boiling water to produce particleboards, had the best properties in terms of dimensional stability and IB; but on the other hand, the combination of immersion in boiling water and oxidation pretreatment produces binderless particleboard with a better MOR and MOE parameters. The facts in this study indicated that the oxidation pretreatment using hydrogen peroxide and the catalyst had successfully activated the chemical components of wood, especially lignin, causing it to bond directly when hot pressed. The bonding possibly occurred because the hydroxyl radical that was produced from hydrogen peroxide and ferrous sulphate, the resulted electrophilic would attack the group electron-rich lignin. The results of this reaction were lignin phenoxy radical, hydroxylation, and demethoxylation (Nguyen 1982). Therefore, when the oxidized particles were hot pressed, the covalent bonds would be formed (Widsten *et al.* 2003, Pantze *et al.* 2008).

The influence of particle size

Material used in this step were particles in several sizes, i.e. passed through 20 mesh, 10 mesh, 5 mesh, 2.5 mesh, and shaving. Distribution of particle size for each group could be seen in Table 1.

Particle size groups were obtained from a number of particles taken of certain sizes. For example, particles that passed through

the 10 mesh sieve consisted of several particle sizes as shown in Table 1. The proportion of particle size distribution indicated that the majority of the particles will be restrained in the most coarse sieve. However, each group had a small particle size powder. In the Karlsson & Westermarck (2002) experiment, when small particles (less than 0.25 mm) were removed, it would decrease the IB of about 1/3 and were observed at similar density of the boards.

Fig. 6-9 showed the effect of wood particle size on the physical and mechanical properties of particleboard. Dimensional stability as well as MOR and MOE tend to decrease when particle size increases. However, the IB values tend to increased.

The effectiveness of the material surface oxidation process would be determined by the level of lignin components accessibility that existed on wood particles. In the context of this study, variations in the size of the particles that passed through 20 mesh, 10 mesh, 5 mesh, 2.5 mesh and shaving particle showed high dimensional stability and mechanical properties. The result showed that in the range of particle size, lignin was still quite easy to reach by the oxidant, although this approach in laboratory experiments which using wafer (larger size), showed that the bonding failed.

Table 1 Particle size distribution for each particle size group

No.	Particle size group	Proportion of particle distribution (%)						
		Pass 2.5 mesh / of 5 mesh	Pass 5 mesh / of 10 mesh	Pass 10 mesh / of 20 mesh	Pass 20 mesh / of 40 mesh	Pass 40 mesh / of 60 mesh	Pass 60 mesh / of 80 mesh	Pass 80 mesh
1	Pass 20 mesh	-	-	-	58.32	14.50	17.52	9.66
2	Pass 10 mesh	-	-	66.31	23.46	4.12	3.81	2.31
3	Pass 5 mesh	-	55.62	11.47	21.64	3.50	5.11	2.66
4	Pass 2.5 mesh	47.60	24.97	4.43	14.33	2.82	3.74	2.1
5	Shaving				no data			

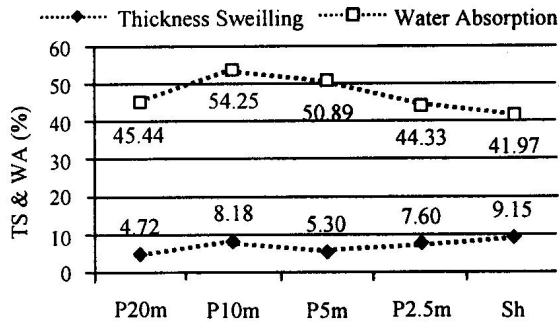


Figure 6 TS and WA of binderless particleboard, (P20m: pass 20 mesh, P10m: pass 10 mesh, P50m: pass 5 mesh, P2.5m: pass 2.5 mesh, Sh: Shavings)

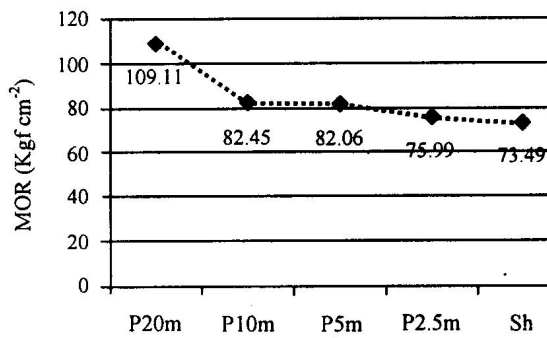


Figure 7 MOR of binderless particleboard

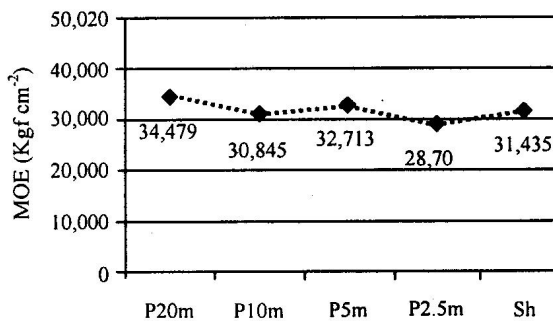


Figure 8 MOE of binderless particleboard

Decomposition of hydrogen peroxide to produce hydroxyl radicals in the presence of a ferrous sulphate was exothermic reaction that generated a high temperature (>100°C). This condition might cause the burning of particles as observed in several experiments by using any small particles that passed through 20 mesh sieve. Therefore, despite the fact that the tendency of particles that passed 20 mesh

would produce a board with high MOR and MOE values, the use of this size was not feasible. Therefore, the use of particle size that passed through 10 mesh or coarser were more feasible to be developed, but this method was only effective until particles had passed through shaving.

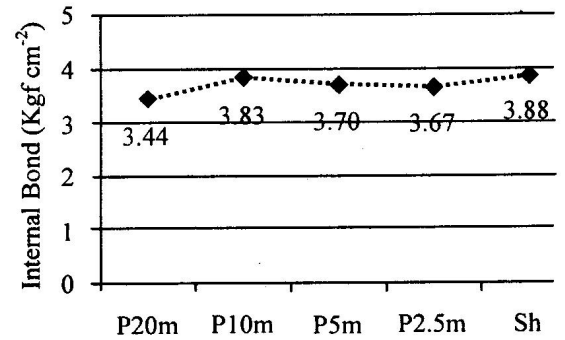


Figure 9 IB of binderless particleboard

Conclusion

This research had successfully developed a binderless particleboard manufacturing methods through oxidation pretreatment using hydrogen peroxide and ferrous sulphate. The best results obtained on particle board using sengon wood species. Combination of pre-treatment of particle board by immersed in boiling water for 30 minutes and followed by oxidation produced a board with the best MOR and MOE values. But in terms of internal bond, oxidation pretreatment without immersion in boiling water produced better particle board. The dimensional stability values and the IB of the board were far above the minimum standard as specified in JIS A 5908 2003.

In terms of board characteristics relationship with particle size, the results of this study showed a tendency to increase dimensional stability, MOR and MOE board when the particle size decreased, but the opposite occurred for the IB. However, given the exothermic

reaction that could cause burning of wood particles, the use of very fine particles (20 mesh pass) was not recommended.

Acknowledgement

The authors are grateful to the Directorate General of Higher Education, Ministry of National Education for funding this work by Competitive Research Grants Under the National Priorities Batch IV for Budget Year 2009; 523.3/13.11/PL/2009.

References

- Hüttermann A, Mai C, Kharazipour A, 2001. Modification of lignin for the production of new compounded materials. *Appl. Microbiol. Biotechnol.* 55:387-394.
- Japanese Standards Association [JSA], 2003. *Particleboards*. Japanese Industrial Standard (JIS) A 5908-2003. Japan.
- Kawai S, Zhang M, Okudaira Y, Xu J, Widyorini R, Han G. 2002. Development of kenaf bast fiberboard and core binderless particleboard. 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. pp 129-134.
- Karlsson O, Westermark U. 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. Pp 149-153.
- Kharazipour A, Hüttermann A. 1998. Biotechnological Production of Wood Composites. In: Bruce A, J W Palfreyman, editors. *Forest Product Biotechnology*. UK, Taylor & Francis Ltd. Pp 141-150.
- 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. Pp 58-67.
- Maloney T M. 1993. *Modern Particleboard and Dry-Process Fiberboard Manufacturing*. USA: Miller Freeman Inc San Francisco.
- Müller C, Kharazipour A. 2007. Enzymatic Modification of Wood Fibres to Activate their Ability of Self Bonding. In: Kharazipour A R, C Müller, C Schöpfer, editors. *A Review of Forests, Wood Products and Wood Biotechnology of Iran and Germany*. Germany.
- 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.
- Pantze A, Karlsson O, Westermark U. 2008. Esterification of carboxylic acids on cellulosic material: Solid state reactions. *Holzforschung*. 62: 136-141
- Suhasman, Massijaya MY, Hadi YS. 2007. Pengaruh Penambahan Lapisan Karton Daur Ulang Terhadap Kualitas Papan Komposit. *Prosiding Seminar Mapeki X*, Fakultas Kehutanan Universitas Tanjung Pura, Pontianak, 9-11 Agustus 2007.
- Suhasman, Yuniarti AD, Massijaya MY, Hadi YS. 2008. Performance of composite board manufactured from stem and branch of tree. In: Li J, chairman. *Proceeding of International*

- Symposium on Wood Science and Technology* IAWPS; Harbin, 27-29 Sep 2008. China. pp 463-464.
- Widsten P, Qvintus-Leino P, Tuominen S, Laine JE. 2003. Manufacture of fiberboard from wood fibers activated with fenton's reagent ($H_2O_2/FeSO_4$). Germany. *Holzforschung* 57: 447-452.
- Widsten P, Tuominen S, Qvintus-Leino P, Laine JE. 2004. The influence of high defibration temperature on the properties of medium-density fiberboard (MDF) made from laccase-treated softwood fibers. *Wood Sci Technol* 38: 521-528.
- Widyorini R, Xu J, Umemura K, Kawai S. 2005a. Manufacture and properties of binderless particleboard from bagasse I: effects of raw material type, storage methods, and manufacturing process. *Japan. J. of Wood Sci.* 51 : 648-654.
- Widyorini R, Xu J, Watanabe T, Kawai S. 2005b. Chemical changes in steam-pressed kenaf core binderless particleboard. *Japan. J. of Wood Sci.* 51 : 26-32
- Widyorini R, Higashihara T, Xu J, Watanabe T, Kawai S. 2005c. Self-bonding characteristics of binderless kenaf core composites. *Japan. J. of Wood Sci.* 39: 651-662.
- Xu J, Han G, Wong E D, Kawai S. 2003. Development of binderless particleboard from kenaf core using steam-injection pressing. *Japan. J. of Wood Sci.* 49:327-332.
- Xu J, Widyorini R, Kawai S. 2005. Properties of kenaf core binderless particleboard reinforced with kenaf bast fiber-woven sheets. *Japan. J. of Wood Sci.* 51 : 415-420.
- Xu J, Widyorini R, Yamauchi H, Kawai S. 2006. Development of binderless fiberboard from kenaf core. *Japan. J. of Wood Sci.* 51 : 415-420.

Riwayat naskah (*article history*)

Naskah masuk (*received*) : 10 October 2009
 Diterima (*accepted*) : 20 December 2009