

# Reduction of Formaldehyde Emission from Particleboards by Bio-Scavengers<sup>\*1</sup>

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## ABSTRACT

This study was to investigate the effect of adding additive as tannin, rice husk and charcoal, for reducing the formaldehyde emission level, on the adhesion properties of urea-formaldehyde (UF) resin for particleboard. We controlled the hot-pressing time, temperature and pressure to determine the bonding strength and formaldehyde emission. Blends of various UF resin/additives (tannin, rice husk and charcoal) compositions were prepared. To determine and compare the effect of additives (tannin, rice husk and charcoal) content, 0, 5, 10 and 15%, by weight of UF resin, were used.  $\text{NH}_4\text{Cl}$  as hardener added. To determine the level of formaldehyde emission, we used the desiccator, perforator and 20 L-small chamber method. The formaldehyde emission level decreased with increased additions of additive (except rice husk). Also, increased hot-pressing time decreased formaldehyde emission level. At a charcoal replacement ratio of only 15%, the formaldehyde emission level is under F☆☆☆☆ grade (emit  $< 0.3 \text{ mg}/\ell$ ). Curing of the high tannin additive content in this adhesive system indicated that the bonding strength increased. But, in the case of rice husk and charcoal, the bonding strength was much lower due to the inorganic substance. Furthermore, rice husk was poor in bonding strength as well as formaldehyde emission than tannin and charcoal.

*Keywords* : formaldehyde emission, particleboard, tannin, rice husk, charcoal, desiccator method, perforator method, 20  $\ell$ -small chamber method

## 1. INTRODUCTION

Urea-Formaldehyde (UF) resins are based on the manifold reaction of two monomers, urea

and formaldehyde. By using different conditions of reaction and preparation a more of less innumerable variety of condensed structures is possible. UF resins are the most important type

<sup>\*1</sup> Received on January 9, 2006; accepted on May 21, 2006.

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of the so-called aminoplastic resins. Currently, approximately 6 billion tons are produced per annum worldwide, based on a usual solids content of 66% by mass (Dunky 1998). In the past, the important criteria for assessing a wood adhesive for a specified application were its technical properties, its gluing behavior, and its cost. During recent years, a new set of criteria has attained more importance about: the environmental and health aspects of the adhesive itself. Formaldehyde (HCHO) is emitted by many synthetic resins that are used as adhesives for wood and wood products. HCHO is a suspected human carcinogen that is known to be released from pressed-wood products used in home construction, including products made with UF resins (e.g., particleboard, hardwood plywood, medium-density fiberboard (MDF), and panelling) (Thomas 1999). Its official name, given by the International Union of Pure and Applied Chemistry (IUPAC), but it is much better known by its common name, formaldehyde. Formaldehyde is the simplest aldehyde, closely related to methane, methanol, and formic acid. It was first synthesized in 1859 by the Russian chemist Butlerov but not identified with certainty until some years later by the German chemist Hofmann (Butlerov 1859; Hofmann 1868). During the following decades the diverse properties of formaldehyde were investigated in detail, and its high reactivity toward a variety of other substances was determined. At the beginning of this century, the American chemist Baekeland combined formaldehyde and phenol to synthesize the first polymers (Baekeland and Patents). These products later became to be known as the name "Bakelite". During the followings years, more formaldehyde-based polymers were also developed by reaction with other phenols. The 1930s saw the development of another important group of condensation products, the aminoplastics. By the middle of the twentieth century, the aminoplastics started their

successful penetration into of the industrial wood adhesives market. Aminoplastics are condensation products of formaldehyde with amino- and amino-group-containing compounds, especially urea and melamine. The most advantageous properties of both aminoplastics and phenoplastics were their excellent performance as wood adhesives and their high cost effectiveness. UF resins have been being widely used as wood binders, especially in the manufacture of particleboard (Levendis *et al.* 1992). Ease of handling and application, lack of color in the finished product and low cost have led these resins to the most widely used adhesives for bonding wood products. As a consequence, UF and phenol-formaldehyde (PF) resins obtained a dominant position in wood gluing during the 1940s (Pizzi *et al.* 1994).

However, the toxicity of UF resin bonded wood particles due to the emission of formaldehyde and the associated possible health hazards, could act as promotion obstacle under the prevailing environmental concerns (Kavvouras *et al.* 1998). Their low stability of urea-formaldehyde resins results in the liberation of formaldehyde by the glued wood products, a process that can last over years. In 1962 Wittmann published the first studies on the subsequent dissociation of formaldehyde from UF resin-bonded particleboard. Since then, about one hundred publications have reported on the same theme. Although formaldehyde complaints were sporadic during the 1960s, the problem increased dramatically after the oil crisis that began in late 1973 (Sundin 1985).

Recently, governments of many countries have already or are about to impose regulations limiting the emission of formaldehyde from building materials and from the materials used for the manufacture of furniture and fitments. The release of formaldehyde in wood products can be minimized during the manufacturing process, or by posttreatment of the board and

by surface treatment. These include the addition of wax scavengers of urea to the wood finish, treatment of the composite boards with ammonia gas or ammonia salts, and surface treatment of boards with paints, lacquers, veneers, and papers. The surface treatment can be both a physical and a chemical minimization of formaldehyde diffusibility. The most efficient route is the modification of the adhesives chosen. The use of phenolics and isocyanates or of UF formulations with lower molar ratios are powerful concepts used to reduce the formaldehyde emission potential of wood products, in some case to nearly zero (Plath 1994). The molar ratio of formaldehyde to urea (F/U) is the most important factor affecting formaldehyde release in the production of boards. The combination of UF resins with formaldehyde-free adhesive systems may also be a way to reduce formaldehyde emission (Meyer *et al.* 1986; Roffael 1982).

Post-treatment methods to decrease and minimize formaldehyde release are based on compounds like ammonia, ammonium salts, or urea. Another effective way to reduce formaldehyde release is the addition of formaldehyde-binding substances ("scavengers") to the resin or to the wood particles (Ernst 1982; Roffael 1982). A conventional formaldehyde scavenger added to UF resins is urea, which is often used in combination with ammonium chloride (about 20:1). Urea also acts as a buffer controlling the pH and improving the stability of UF resins. Ammonium chloride acts as an acid catalyst of the curing reaction and as a scavenger. The urea/ammonium chloride system apparently has no effect on resin curing and bonding properties if it is added up to 10% of the UF resin dry weight. Other approved formaldehyde scavengers are organic amines. Especially in Sweden, dispersion systems based on formaldehyde-binding paraffins were developed (Sundin 1985; Sundin *et al.* 1997). These wax systems are often added to the wood particles before drying. The am-

ounts necessary to obtain perforator values below 10 mg depend on plant conditions and on the perforator value of the untreated board. The scavenger does not negatively influence the board strength and pressing time. The increase in cost has been reported by the producer as varying between 1 and 5% of the total production cost. In this study, as a eco-environmental scavenger used, make a study of formaldehyde emission reduction. UF resins mixed by three types additive materials measured the physical (thickness swelling, linear expansion and water-absorption) and mechanical properties (bending strength and internal bonding). Also, for measuring of formaldehyde emission the desiccator and perforator methods were used.

## 2. EXPERIMENTAL

### 2.1. Materials

#### 2.1.1. Wood Particle

The wood particles used for manufacturing the particleboard were donated by the Dongwha Enterprise Co., Ltd. in South Korea, and consisted of recycled chips, which had 6% of moisture content. Coarse particle was used for core layer, and fine particle was used for face and back layer.

#### 2.1.2. Urea-formaldehyde (UF) Resin

The urea-formaldehyde resin used for manufacturing the particleboard was donated by the Taeyang Co., Ltd. in South Korea. The solids content of UF resin was 49%.

#### 2.1.3. Scavengers

The three materials, tannin, charcoal and rice husk, were used as scavengers. First, commercial tannin extracts were prepared. These tannins consisted of a fine dark brown powder

with a moisture content of 5%. Second, the charcoal was supplied by the Seoul National University in Korea. Moisture content of the charcoal was 4%. Third, the rice husk with a moisture content of 6% was donated by the Seoul National University in Korea.

## 2.2. Methods

### 2.2.1. Manufacturing Particleboard

The particleboards were manufactured using urea-formaldehyde (UF) resin and three different scavengers, and the boards had dimension of 35 cm × 35 cm × 10 cm (length×width×thickness). The wood particles were placed in a rotary drum mixer and the UF resin was sprayed onto them while rotating the mixer. The quantity of adhesive was calculated as being 10% of the raw material based of the oven-dried weight. The mixture of particles and adhesives was cold pressed at 2 kgf/cm<sup>2</sup> for 2 minutes, in order to ensure the stability of the mat and obtain the proper density gradient of the composites prior to hot pressing. The mixture was then hot pressed in order to form composite boards at a peak pressure of 30 kgf/cm<sup>2</sup> and a temperature of 160°C. The main pressing

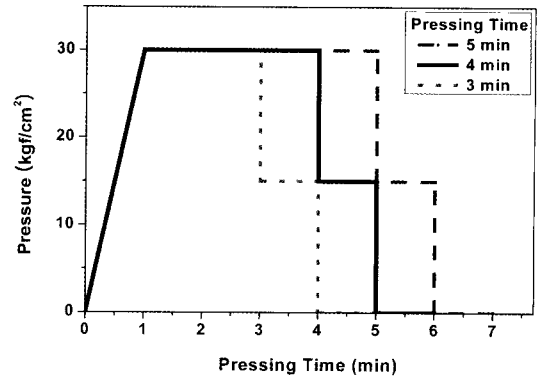


Fig. 1. Multi-hot pressing schedule.

times were 3 to 5 minutes, with the pressure being released in 1 minute of second step. Then, the hot pressed particleboard was cold pressed at 2 kgf/cm<sup>2</sup> for 2 minutes. The main temperature, time and pressure were 160°C, 240 seconds and 30 kgf/cm<sup>2</sup>, respectively. A schematic diagram showing the multi-hot press schedule is shown in Fig. 1.

### 2.2.2. Measuring Formaldehyde Emission

Typical methods for evaluation of formaldehyde emission are desiccator, perforator and 20-liter small chamber. Also, standard of permission for formaldehyde emission are compared

Table 1. Total adhesive systems showing the additive content of the additive

Adhesive	Additive	Content	Hardener
UF	No	100%	NH <sub>4</sub> Cl
		5%	
		10%	
	Tannin	15%	
		5%	
		10%	
	Rice Husk	15%	
		5%	
		10%	
	Charcoal	15%	
		5%	
		10%	

in Tables 2 to 4. And, 20-liter small chamber measured formaldehyde.

#### 2.2.2.1. 24 h-desiccator Method

The method used for the evaluation of formaldehyde emission provided for the JIS (Japanese Industrial Standard) and KS (Korean Standard) is the desiccator method (see Table 2). The manufactured particleboards were pre-conditioned at 25°C and 65% RH for two weeks before testing.

Emission of formaldehyde is determined by placing test samples in a desiccator at a controlled temperature and measuring the quantity of emitted formaldehyde absorbed in a specified volume of water for 24 hours. The interior volume of the desiccator was 11 liters and 50×150 mm specimens were used for each desiccator test. The specimens were cut from sample panels. The sample total surface area was 18,000 mm<sup>2</sup>.

The released formaldehyde was caught in the distilled water and analyzed using a UV spectrophotometer after treatment with acetyl acetone and acetyl acid ammonium.

#### 2.2.2.2. Perforator Method

This method is primarily used in Europe. The perforator value of formaldehyde emission was determined using the DIN EN 120 (European Committee For Standardization, 1991). For this method, a special perforator apparatus is necessary. About 110 g of test pieces are weighed to an accuracy of 0.1 g and put into the round bottom flask. 600 ml of toluene are added. About 1000 ml of distilled water is poured into the perforator attachment. The boiled toluene was passed through the distilled water for two hours. In this process, the distilled water absorbed the formaldehyde and other volatile organic compounds stripped by the boiling

Table 2. Comparison of standard of formaldehyde emission with desiccator method between Korea and Japan

Test Method	Standard	Under 0.3	Under 0.5	Under 1.5	Under 5.0
Desiccator (mg/ℓ)	KS*	S-E <sub>0</sub>	E <sub>0</sub>	E <sub>1</sub>	E <sub>2</sub>
	JIS*	F☆☆☆☆	F☆☆☆	F☆☆	F☆

\* KS : Korean Standard

\* JIS : Japanese Industrial Standard

Table 3. Standard of formaldehyde emission with perforator method

Test Method	Standard	E <sub>0</sub>	E <sub>1</sub>	E <sub>2</sub>
Perforator (mg/100 g panel)	EN 120*	Under 0.8	Under 6.5	Under 15

\* DIN EN 120 : European Committee For Standardization

Table 4. Standard of formaldehyde emission with 20 ℓ-small chamber method

Test Method*	Excellent	Very good	Good	General 1	General 2
20 ℓ - Small Chamber (mg/m <sup>2</sup> h)	Under 0.03	0.03~0.05	0.05~0.12	0.12~0.60	0.60~1.25

\* Korea Air Cleaning Association (KACA)

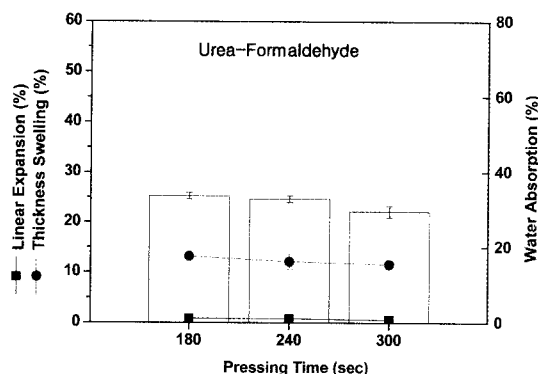


Fig. 2. Physical properties of particleboards bonded with UF adhesive

toluene. The formaldehyde trapped by the water was then quantitatively determined using an UV spectrophotometer after treatment with acetyl acetone and acetyl acid ammonium.

#### 2.2.2.3. 20 ℓ Small Chamber Method

In order to generate formaldehyde emission data from particleboard, the tests were conducted in the JIS A 1901 20-liter small chamber method. The chamber consisted of electro-polished stainless steel chambers. The facilities allowed close control of  $25 \pm 1^\circ\text{C}$ ,  $50 \pm 5\%$  relative humidity, air exchange rate (N) of  $0.5 \pm 0.1/\text{hour}$ , sample loading factor of  $2.0 \text{ m}^2/\text{m}^3$  10%, and an air flow rate on sample surface of 0.1 to 0.3 m/s. Formaldehyde adsorption cartridge 2,4-DNPH (2,4-dinitro- phebylhy draxube) and VOC adsorption tubes, were purchased from Tenax TA. Particleboards were cut into 150 mm×150 mm sections, and the exposed edges sealed with Teflon material to ensure that only the sample surface was measured. The total test duration was 6 days, and air samples were collected on the 1st, 3rd, 5th and 7th day from the chamber outlet. Immediately after sampling, the tubes were tightly sealed and analyzed within a few hours.

#### 2.2.3. Physical Properties

Moisture content, linear expansion, thickness swelling and water absorption were examined using the ASTM D 1037-99 (American Society for Testing and Material, 1999) method. The samples of each type of particleboards were soaked in water for 24 hours. Each value obtained represented the average of five samples.

#### 2.2.4. Mechanical Properties

3-point bending strength and internal bonding strength were determined using a Universal Testing Machine (Hounsfield, 1999) using the ASTM D 1037-99 (American Society for Testing and Materials, 1999). The results were compared with ANSI A208.1-1999 (American National Standard). Each value represents the average of five samples.

## 3. RESULTS and DISCUSSION

### 3.1. Physical Properties

The physical properties (thickness swelling, linear expansion and water absorption) of the particleboards made using the urea-formaldehyde adhesives and with the three different additives are shown in Fig. 2 to 5. In Fig. 2, the thickness swelling of the urea-formaldehyde at press time of 3 min is 13.1%, and then decreases with increasing press time to 11.7% at 5 min. This same trend is shown in case of the additives (tannin, rice husk and charcoal), with the only difference being the degree of thickness swelling, which varies for the amount of additives. The tendency toward decrease thickness swelling both with increasing press time and in the order of tannin, rice husk and charcoal were proved by the results of linear expansion and water absorption.

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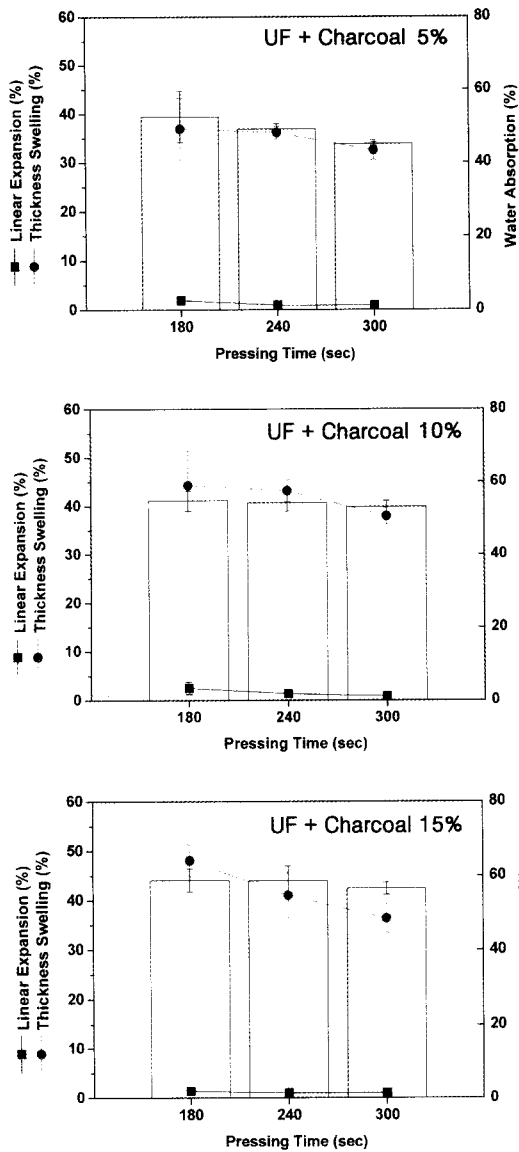


Fig. 3. Physical properties of particleboards made using the UF adhesives and with charcoal.

The overall physical properties of UF adhesives decrease with increasing press time and in the order of rice husk, charcoal and tannin. The thickness swelling, linear expansion and water absorption of the particleboards using the UF adhesives with the three different additives

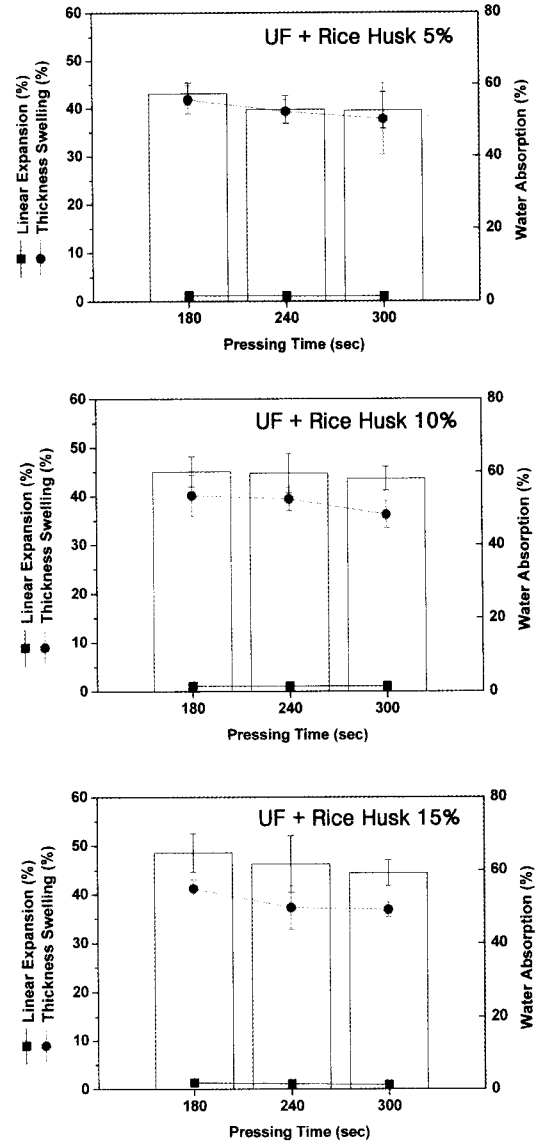


Fig. 4. Physical properties of particleboards made using the UF adhesives and with rice husk.

increased as with decrease press time and increasing amount of additives. Why do these physical properties show the behavior? UF adhesives are thermosetting adhesives, such as phenol-formaldehyde and melamine-formaldehyde adhesives. Accordingly, composites made using

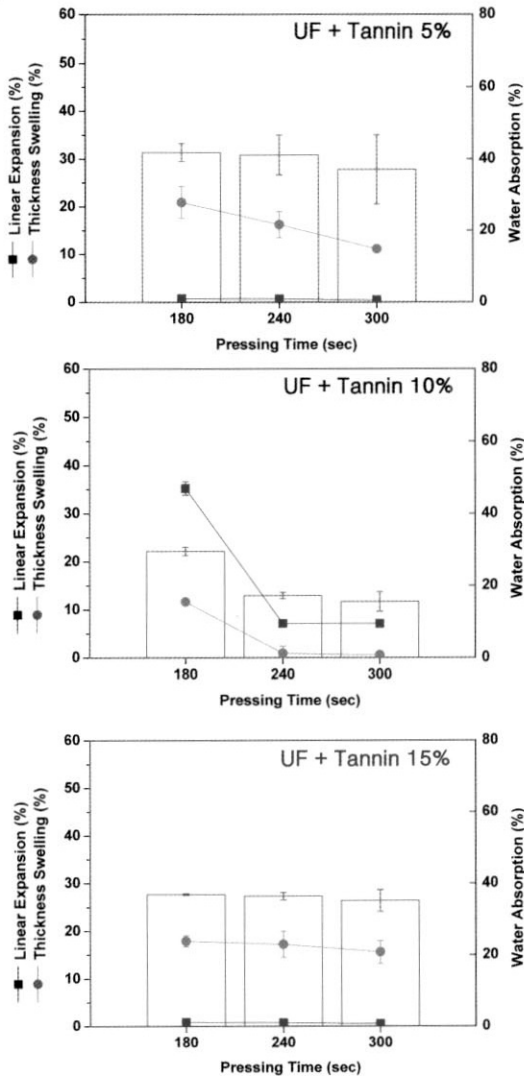


Fig. 5. Physical properties of particleboards made using the UF adhesives and tannin.

UF adhesives show this kind of trend. As the press time increases, molecules of the UF prepolymer and functional group are cross-linked to each other and are hardened. More press time contained more energy to cross-link. That the reason why the physical properties improve with increasing press time. The particleboard using the UF adhesive with tannin

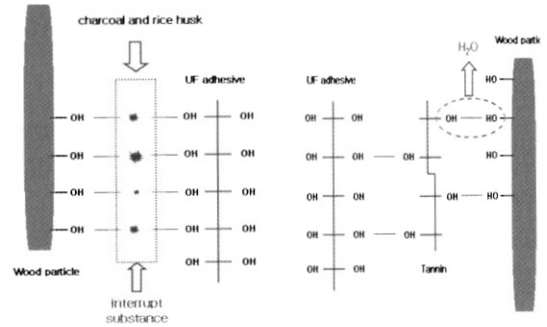


Fig. 6. Schematic diagrams of adhesion between rice husk, charcoal and tannin with UF adhesive (kim *et al.* 2006).

showed the best physical properties, while rice husk and charcoal didn't show enough physical properties. That is why the physical properties, tannin additives are thermosetting adhesives. Accordingly, as the press time increases, tannin functional group are cross-linked to each other and are hardened.

On the other hand, rice husk has the similar components to the wood as well as inorganic substance. Also, charcoal contain inorganic substance. These inorganic substance hampers adhesive strength (see Fig. 6).

## 3.2. Mechanical Properties

### 3.2.1. Bending Strength

The bending strengths of the particleboards using the urea- formaldehyde adhesives with the three scavengers at press times of 180, 240 and 300 sec are shown in Fig. 7. The bending strength of the particleboards made using the UF adhesives with all three additives increased slightly as the press time increased. Compared with ANSI (American National Standard), the bending strengths of all the adhesive systems for all press times are above Grade M-3. As regards the additives, the bending strength of UF adhesive was in the order : tannin > charcoal ≥ rice husk. From these results, tannin



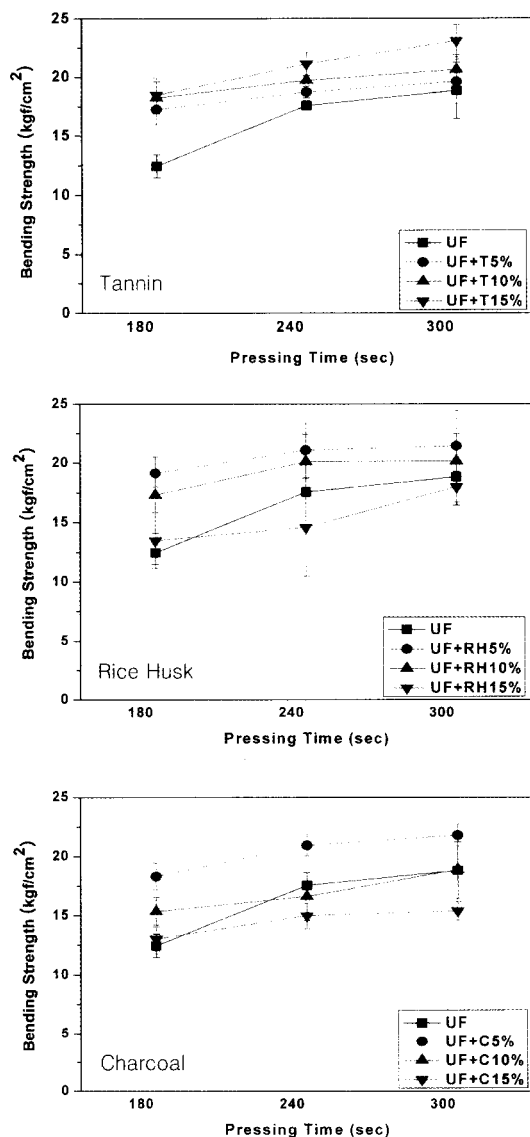


Fig. 7. Bending strength of particleboards with urea-formaldehyde adhesives at three press times.

is best cured, then charcoal and finally rice husk, in that order. The bending strength increases with increasing press time. These results showed the same tendency as the physical properties results. Increasing the amount of additive, except tannin caused the bending strengths of particleboards to decrease,

as shown in Fig. 7. In terms of the physical property results, it is understandable that the board properties were improved by increasing the press time. When the press time was increased, bending strength of the particleboards slightly increased.

### 3.2.2. Internal Bonding Strength

Internal bonding strength provides an overall measure of the boards integrity, which defines how well the core material is bonded together, and this property is influenced directly by the board density, resin content, particle geometry, and raw material type. As shows in Fig. 8. Almost adhesive systems satisfied Grade M-3. Especially, in the case of where tannin used as a scavenger, the adhesive system was distinguished. Increasing the press time from 3 to 5 min caused the internal bonding strength of the particleboards to increase. On the other hand, charcoal or rice husk used as a scavenger little influence on the internal bonding. Fig. 8 shows the difference in internal bonding as a function of press time for all adhesive systems. This tendency was the same as that observed for the bending strength. When the press time was increased from 3 min to 5 min at 160°C, the internal bonding strength of the particleboards using all adhesives system increased slightly.

## 3.3. Measuring Formaldehyde Emission

### 3.3.1. Desiccator Method

Firstly, the results obtained for particleboards are presented in Fig. 9. Formaldehyde emission was decreased with press time. The formaldehyde emission of UF adhesives was 0.944 mg/ℓ at press time 3 min. At 4 min and 5 min, it reached 0.859 and 0.795 mg/ℓ respectively. The results obtained by press time from 3 to 5 min showed that the formaldehyde emission slightly decreased. In UF adhesives, the formal-

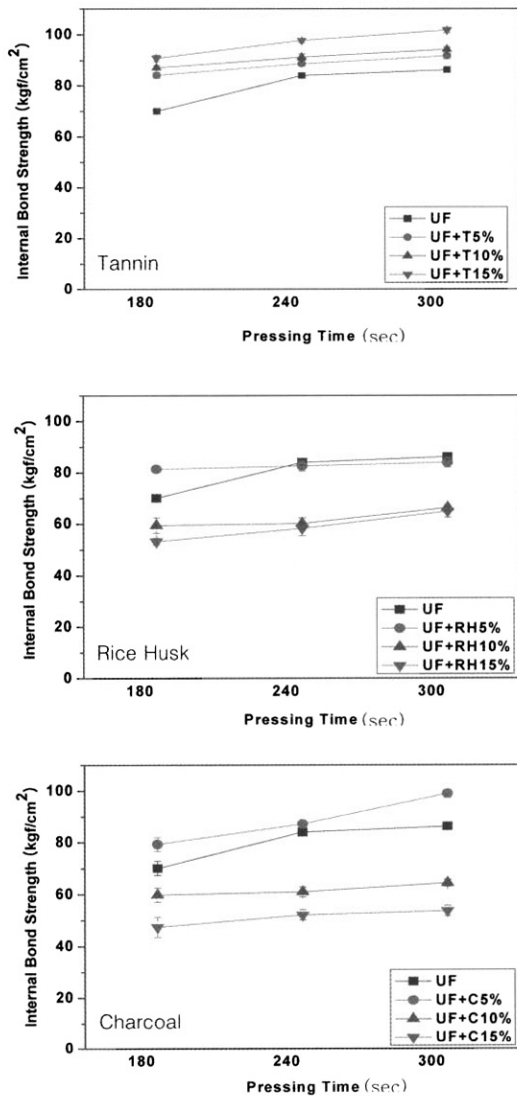


Fig. 8. Internal bonding strength of particleboards with UF-based adhesives at three press time.

dehyde emission was 0.944, 0.859 and 0.795 mg/l at 3 to 5 min, respectively. In UF adhesives, as the press time increased, formaldehyde emission decreased. Fig. 9 shows similar tendencies for tannin additive. A low emission level was observed at 5 min, which increased with an decrease in press time at 4 min. A

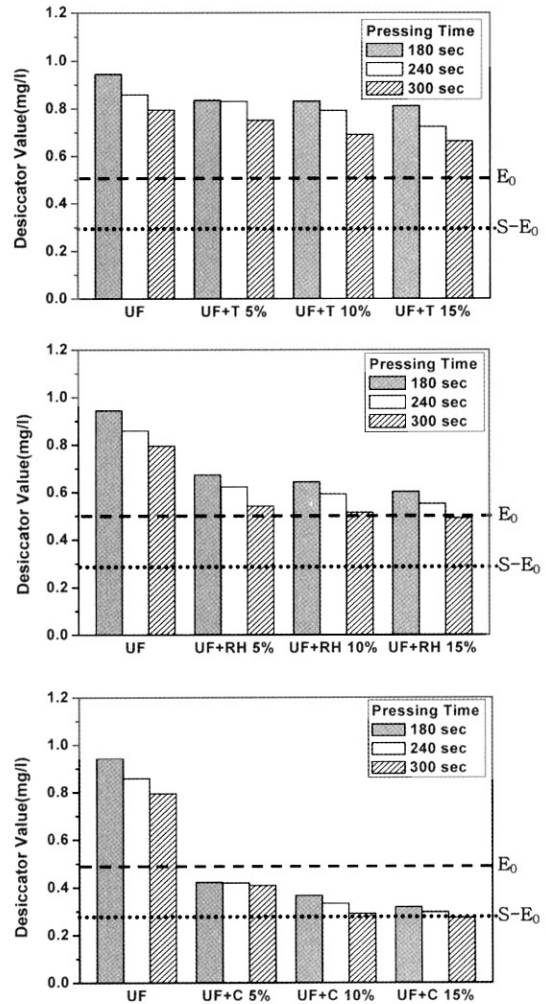


Fig. 9. Desiccator values of particleboards by UF-based adhesives with bio-scavengers.

further press time decrease at 3 min also increased the level of formaldehyde emission. Also, as the amount of additives increasing, formaldehyde emission level slightly decreasing. In particleboard of tannin additive, as the amount of additives increased, the formaldehyde emission level decreased in Fig. 9. Also, the particle board of rice husk additive shows similar tendencies in Fig. 9. Furthermore, as the amount of additives and press time increased, the formal-

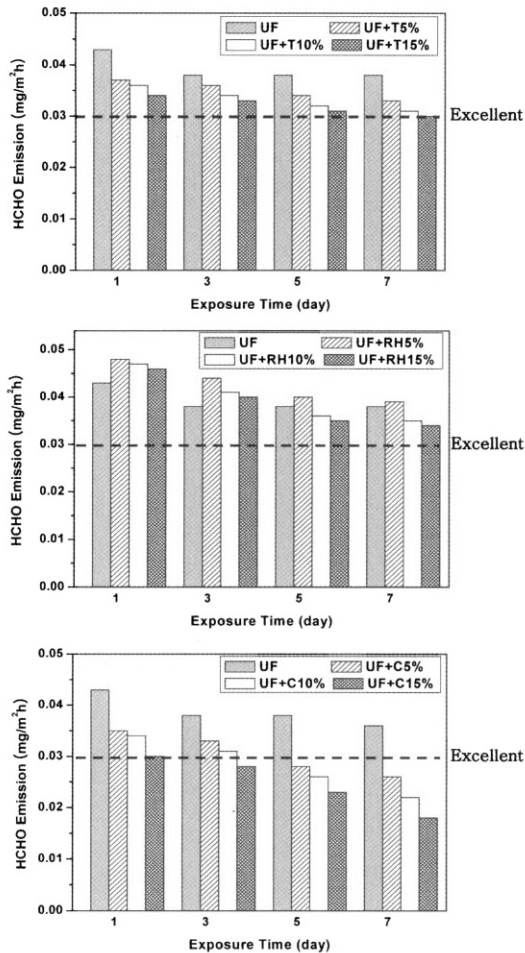


Fig. 10. Formaldehyde emission of particleboards by small chamber method (UF adhesive with bio-scavengers).

dehyde emission rapidly decreased, than UF adhesives and tannin additive. Fig. 9 shows formaldehyde emission level of charcoal additives. According to JIS, the particleboard of charcoal additives satisfied Grade F☆☆☆☆ (emit <0.3 mg/ℓ). In the case of charcoal additives, is extremely effective such as scavengers among additives. According to these results, the increasing amount of additives and press time did effectively reduce formaldehyde emission and in the order: charcoal > rice husk → tannin.

Therefore, press time and amount of additives are important factors to consider for formaldehyde emission. For low formaldehyde emission, the best performance was obtained using charcoal as the scavenger.

### 3.3.2. Small Chamber Method

Fig. 10 shows a representative pattern of formaldehyde emission rate from the particleboards. For UF adhesive, the formaldehyde emission rate started at the maximum of 0.043 mg/m³h and decreased by about 12% after 3 days. The emission rate was reduced to 2.4% of the maximum after 5 days and to 2.4% after 7 days. The cumulative reduction rate of formaldehyde emission was 16.8%, (0.007 mg/m³h) on the 7th day. The chamber experiments showed that UF adhesive with charcoal had the highest emission rate of formaldehyde (see Fig. 10). Compared with KACA (Korea Air Cleaning Association), formaldehyde emission of UF adhesive with charcoal satisfied grade-excellent (emit < 0.03 mg/m³h). While, UF adhesive with tannin and rice husk unsatisfied. From these results, charcoal was a best reducer of formaldehyde emission in that order.

## 4. CONCLUSION

The results of this study are summarized as follows :

UF based adhesives increased the bending strength and internal bonding strength of particleboards with increasing press time, while it was decreased with increasing amount of additives. The physical (thickness swelling, linear expansion and water-absorption) and mechanical properties (bending strength and internal bond), all adhesives system is increased as with decreasing press time and increasing of additives. However, the particleboard made using the UF adhesive with tannin showed the best physical

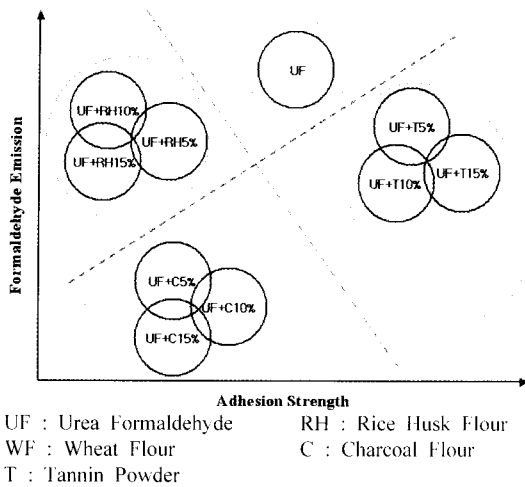


Fig. 11. Representation criterion by bio-scavenger type for UF resin.

and mechanical properties, while rice husk and charcoal didn't show enough physical and mechanical properties. The properties of the particleboards made using the three additives is influenced by the each of additives contained substances. As the tannin additives are thermo-setting adhesives, functional group are cross linked to each other and are hardened. On the other hand, rice husk and charcoal containing inorganic substance such as silica component, hamper adhesive strength of particleboard. These tendencies are increased as with increasing amount of additives.

In desiccator test, formaldehyde emission of all adhesives system decreased, when amount of additive and press time increased. Especially additives, reduced effectively formaldehyde emission (and satisfied Grade F☆☆☆☆). This same trend was shown in the case of the small chamber test. Formaldehyde emission of UF adhesive with charcoal emit  $< 0.03 \text{ mg/m}^2\text{h}$ . While, the emissions of UF adhesive with tannin and rice husk emit on a higher plane.

Finally, The additives of three different materials used such as scavenger distinguish by a

difference of the physical and mechanical properties as well as formaldehyde emission.

## ACKNOWLEDGEMENTS

This work was supported in part by the Research Program 2006 of Kookmin University in Korea. Sumin Kim and Jin-A Kim are grateful for the graduate fellowship provided by the Ministry of Education through the Brain Korea 21 project.

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