Characteristics of Particleboards Using Tannin Resin as Novel Environment-Friendly Adhesion System

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Characteristics of Particleboards Using Tannin Resin as Novel Environment-Friendly Adhesion System

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Key Words
Green adhesive · Tannin · eMDI · Particleboard · Formaldehyde

Abstract
The objective of this study was to develop an environment-friendly adhesion system for particleboard using tannin-based resin. Tannin-based resin was used in the surface layer of particleboard to replace urea-formaldehyde resins. In addition, the additive with various content ratios in particleboard was evaluated to confirm the effect of formaldehyde reduction performance by the perforator and desiccator methods. As the amount of additive was increased, formaldehyde reduction performance would significantly be increased. Manufactured boards containing additive above 3 wt% in the surface layer satisfied grade SE0 in the results of the desiccator method. The mechanical properties of manufactured boards satisfied the minimum requisite quality standard. This study discussed the effects and possibilities of tannin-based resin to reduce formaldehyde emission in indoor environments.

Introduction

Sick building syndrome (SBS) symptoms that are experienced by a building's occupants may be caused by volatile organic compounds (VOCs) and formaldehyde, which are known to be emitted from building materials and furnishings. In addition, proliferation of moulds in indoor environment is also a factor of SBS [1–4]. In particular, there has been an increasing focus on formaldehyde because of its potential for causing harmful diseases such as cancer and asthma. Therefore, formaldehyde was reclassified from "probably carcinogenic to humans (Group 2 A)" to "carcinogenic to humans (Group 1)" in June 2004 by the International Agency for Research on Cancer (IARC), part of the World Health Organization (WHO) [5].

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Particleboard is widely used in the manufacture of furniture, floor underlayment, cabinets, stair treads, home constructions, tabletops, vanities, speakers, sliding doors, lock blocks, interior signs, displays, table tennis and pool tables, electronic game consoles, kitchen worktops and work surfaces in offices, educational establishments, laboratories and other industrial products [6]. Many consumer products of particleboard have been made with urea-formaldehyde (UF) and are known to be emission sources of formaldehyde, which have a tendency to emit formaldehyde permanently once installed in an indoor environment [6,7].

With growing interest in the residential environment, indoor air quality has become a major factor of comfortable indoor environment. In the interest of indoor air quality, there is a need to reduce formaldehyde contents in residential environment, porous material are used widely as an absorbent, such as charcoal, activated carbon and graphite. In addition, ventilation and bake-out method are efficient techniques for improving indoor air quality [8–11]. However, these methods have disadvantages in terms of frequent replacement of absorbent and energy consumption. Therefore, the use of low emission materials or materials with no added formaldehyde is considered a fundamental method to reduce formaldehyde emission in indoor environments. The possibility of using replacement materials for UF and phenol-formaldehyde (PF) adhesives has also been studied for some time. Especially, emulsifiable methylene diphenyldiisocyanate (eMDI) is a non-formaldehyde resin of an isocyanate type, and it is used to manufacture zero-emission board, which emits formaldehyde at under 1/10 of SE0 (super emission zero: under 0.3 mg/L) board according to Korean standard, KS F 3104 [12]. This resin has many advantages, such as not requiring water in the manufacture of resins because of the use of the moisture in the wood for curing. Also, because adhesion is possible at strands of high moisture content, production in the drying process can be increased. While eMDI is easy to manufacture as stated above, it also has a weakness. The surface of the particleboard adhered to the surface of the press machine when eMDI was sprayed as the surface layer of particleboard in manufacturing [13,14].

In addition, tannin, soybean and cashew nut shell liquid (CNSL) are considered appropriate natural materials to replace formaldehyde-based resin [15–18]. Amongst the possible alternatives, tannin is an excellent renewable resource that can be used for replacing petroleum-derived phenolic compounds. Tannins are naturally occurring phenolic compounds that have been a subject of extensive research leading to the development of a wide range of industrial applications. Tannin is mainly concentrated in the inner layer of the bark and has been used in the adhesive industry in Africa, South America and Oceania to obtain the low formaldehyde emission levels required for environment-friendly adhesives. In the last decade, several approaches to the problem of producing low formaldehyde emission wood panels using these wood adhesives have been developed. Moreover, hardeners cause formaldehyde emission even when tannin adhesive is used [19–24].

In this study, to make environment-friendly furniture materials, tannin resin and eMDI resin were used in the manufacturing of particleboard. Formaldehyde contents and emissions were measured by the desiccator and perforator methods. In addition, mechanical properties were also measured in order to confirm that the manufactured particleboards meet the Korean standard for particleboard (KS F 3104). The characterization of the particleboard produced would therefore present an informative guide to building and furniture designers for the appropriate use of these wood-based products for interior building applications.

Methods

Materials

The wood particles used for manufacturing the particleboard were donated by the Dongwha Enterprise Co., Ltd. in South Korea. The moisture content of wood particles was 1.5% and the ratios of wood particle used for the surface layer and core layer were 41.0% and 59.0%, respectively. Resins for manufacturing of particleboard and the additive named Dongwha Chemical material (DCM) that were used as a scavenger for formaldehyde were also donated by the Dongwha Greenchem., Ltd, in South Korea.

Preparation of Adhesion Systems

Condensed tannin resin of 50% solids was used with 35 wt% hexamine solution and 35 wt% formaldehyde solution as a hardener for application in particleboard production. In general, tannin adhesives use hexamine as a hardener, but in this case hexamine tends to degrade the mechanical properties of particleboard. Thus, formaldehyde and hexamine were used in this study as a hardener with the same content to prevent the degradation of mechanical properties. In addition, the method of using eMDI resin in the core layer of particleboard was considered. However, considering the cost and the

Table 1. Preparation of resin and manufacturing conditions of particleboard

Sample name		Experimental step 1				Experimental step 2			
		#1	#2	#3	#4	#5	#6	#7	#8
Surface	Resin type	Tannin				Tannin			
	Chip for blending (g)	590.0	590.0	590.0	590.0	590.0	590.0	590.0	590.0
	Resin (g)	177.0	177.0	177.0	177.0	177.0	177.0	177.0	177.0
	Wax emulsion (g)	25.1	25.1	25.1	25.1	25.1	25.1	25.1	25.1
	35% Hexamine (g)	12.6	12.6	12.6	12.6	12.6	12.6	12.6	12.6
	35% HCHO (g)	12.6	12.6	12.6	12.6	12.6	12.6	12.6	12.6
	37% DCM (g)	–	–	–	–	15.9	31.9	47.8	63.8
Core	Resin type	Tannin	eMDI	eMDI	eMDI	eMDI			
	Chip for blending (g)	639.0	639.0	639.0	639.0	639.0	639.0	639.0	639.0
	Resin (g)	102.2	22.4	22.4	22.4	22.4	22.4	22.4	22.4
	Wax emulsion (g)	11.2	11.2	11.2	11.2	11.2	11.2	11.2	11.2
	35% Hexamine (g)	7.3	–	–					
	35% HCHO (g)	7.3	–	–					
	37% DCM (g)	–	–	8.7	17.3	17.3	17.3	17.3	17.3

manufacturing process, the method using eMDI resin in all layers or in the surface layer of particleboard was excluded.

Particleboards were prepared in two separate steps to confirm the optimum adhesion system. First, three types of adhesion systems were used in step 1 as a preliminary step: Tannin-Tannin, Tannin-MDI and Tannin-MDI containing additive resource were measured. The DCM was composed of 37% water-urea solution. The effect of DCM on the variation of ratio in the surface layer was evaluated in step 2. The preparation of resin and manufacturing conditions of particleboards are listed in Table 1.

Formaldehyde Content by Perforator Method

The perforator value of the formaldehyde content was determined using the European standard - EN 120:1992 method [25]. A sample of the particleboard (110 g) and 600 mL of toluene were placed into a flask, and then the perforator was filled with 1,000 mL of distilled water. Formaldehyde released from the particleboard in the boiled toluene was passed through the distilled water for 2 h. In this process, the distilled water absorbed the formaldehyde stripped by the boiling toluene. The formaldehyde trapped by the water was then quantitatively determined by using UV photometers, after treatment with acetyl acetone and acetyl acid ammonium.

Formaldehyde Emission by Desiccator Method

The formaldehyde emissions from particleboards were determined using a desiccator according to the Korean standard, KS M 1998 method [26]. The desiccator test was used to determine the quantity of formaldehyde emitted from the building boards and was carried out using a 10-L volume glass desiccator. The quantity of formaldehyde emitted was determined from the concentration of formaldehyde absorbed in the distilled or deionized water when the test pieces of the specified surface area were placed in a desiccator filled with a specified amount of distilled or deionized water after 24 h had elapsed. The sample surface areas were 1,800 mm^2, as specified by the Japanese JIS A 1460 [27] and Korean standards, KS M 1998. Throughout the 24 h, the temperature of the dry oven containing the desiccators was set to 20°C.

Mechanical Strength Test

Particleboards were estimated on three-point bending strength (MOR) and internal bond strength (IB) by using a universal testing machine (Hounds Co.). Each value represents the average of eight tested particleboard samples.

The bending strength test measured the maximum load (P) at the surface of the specimen by applying a load of about 10 mm/min, and it is classified by equation (1) as follows.

$$\text{Bending strength } (\text{N/mm}^2) = \frac{3}{2} \times \frac{PL}{bt^2} \qquad (1)$$

P is the maximum load (N), L is span (mm) and b and t are the width (mm) and thickness (mm) of a specimen, respectively.

The internal bond strength test measured the bonding of the specimen at the steel or aluminium block, and the

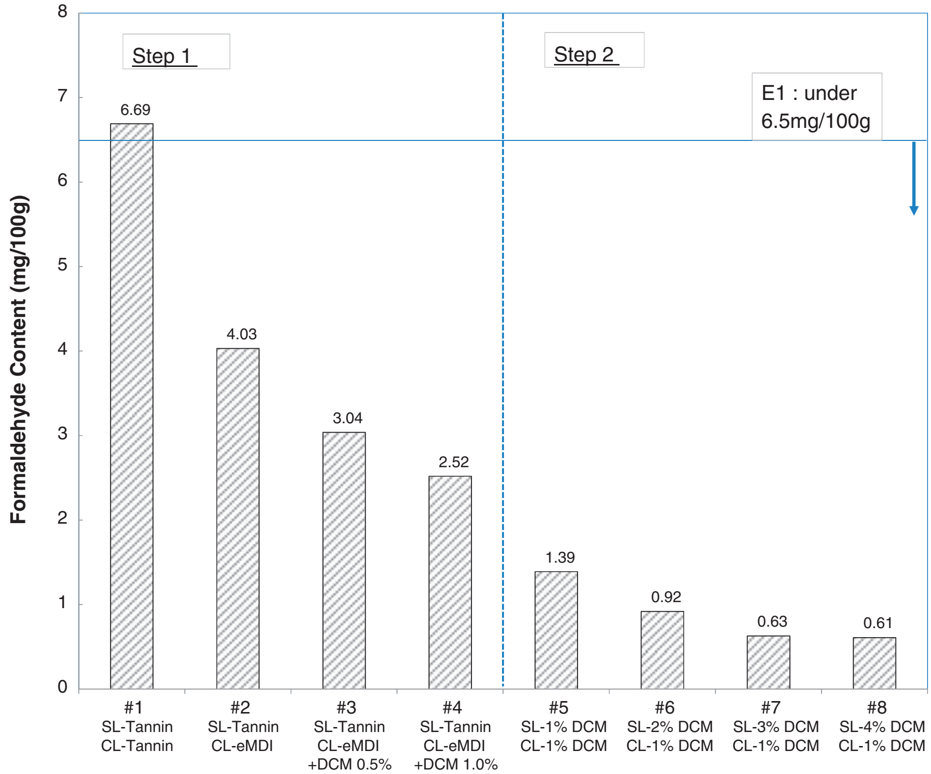

Fig. 1. Formaldehyde content from particleboards by perforator method.

tensile load was applied to the surface of the specimen perpendicularly. Maximum load (P) was measured when the pull out type fracture occurs, and it was calculated by equation (2). The tensile loading rate was 2 mm/min.

$$\text{Internal bond strength } (\text{N/mm}^2) = \frac{P}{bL} \qquad (2)$$

P is the maximum load (N) when the pull out type fracture occurs, and b and L are the width (mm) and length (mm) of a specimen, respectively.

The above criteria and qualifications that were complied with the KS F 3104 in Korea were established based on ISO 768:1972: Fibre building boards – Determination of bending strength, ISO/DIS 3931: Fibreboards – Transversal internal bond, ISO/NP 16893: Wood-based panels – Particleboard, ISO 16893-1:2008: Wood-based panels – Particleboard [12].

Results and Discussion

Formaldehyde Content of Manufactured Particleboards

To confirm an optimum adhesion system, three types of adhesion systems were measured for formaldehyde content by perforator method as a preliminary step. The results of the perforator method for the formaldehyde content in the experiments are shown in Figure 1.

Although using a non-formaldehyde-based resin system, all specimens that were measured contained formaldehyde at a high ratio. These results can be explained by the following two factors: formaldehyde was used as a hardener and is contained in the wood particles used in the manufacturing particleboard. Most of the wood particles obtained from furnishing and building materials waste as well as the re-used wood particles would contain formaldehyde. The formaldehyde content of wood particles used for the manufacturing of particleboards was also measured by the perforator method. The average value was 1.13 mg/100 g. Specimen #2 showed a lower content value of 4.03 mg/100 g than specimen #1. In comparison with specimen #1 using tannin resin for all layers, the use of eMDI resin in the core layer was effective to reduce formaldehyde content. When compared with specimens #2 and #3, the effect of DCM was identified. By using DCM at a 0.5 wt% of wood particles mass ratio in the core layer, the formaldehyde content was reduced by approximately 25%. When increasing the amount of DCM until 1 wt%, the formaldehyde content was reduced by 37% compared with specimens #2 and #4. Accordingly,

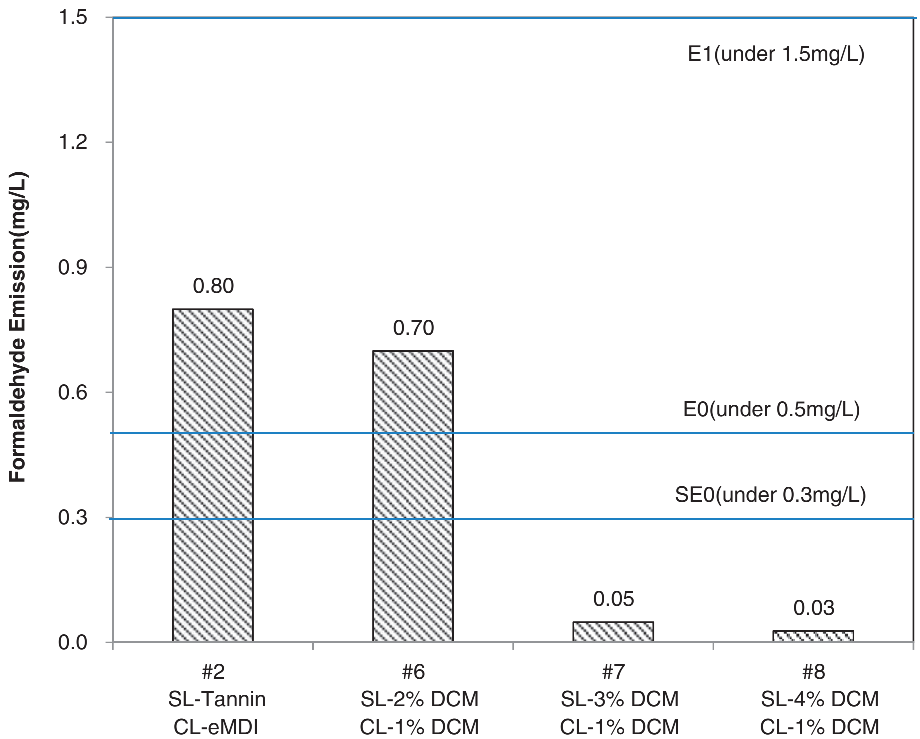

Fig. 2. Formaldehyde emission from particleboard by desiccator method.

the formaldehyde content should decrease with resin containing DCM, which plays an important role as a scavenger in particleboard.

In step 2, particleboards were prepared with various contents of DCM in the surface layer to maximize the formaldehyde reduction effect. In the core layer, DCM was added at an amount of 1 wt% of the wood particle mass ratio used in the core layer, whereas in the surface layer, DCM was added with various ratios from 1 wt% to 4 wt% of the wood particle mass ratio used in the surface layer. The formaldehyde content was lowered as the content of DCM was increased. In particular, the result of specimen #7 showed a lower content value of 0.61 mg/100 g. The value showed a reduction of about 85% in comparison with specimen #2, which did not contain DCM. DCM showed a relatively linear reduction performance and stabilized above the content ratio of 3 wt%. Thus, it was estimated that the addition amount of DCM of 3–4 wt% was adequate.

Formaldehyde Emission of Manufactured Particleboards

In contrast to Europe which uses the perforator value for classification of wood-based products, in Korea the formaldehyde emissions of wood-based panels are usually evaluated by the desiccators test. In the desiccator method, the formaldehyde emission of four specimens was confirmed to verify the reduction performance of DCM for reducing formaldehyde after 7 days from the date of manufacture. The results of formaldehyde emission by desiccator tests are shown in Figure 2.

The formaldehyde emission of the particleboards was lowered by an increase in the content of DCM, as indicated by the perforator test results. Specimens #2 and #6 showed lower levels than the E1 grade (under 1.5 mg L^{-1}) of formaldehyde emission level in the Korean Standard, KS M 3104, and the values were 0.80 mg/L and 0.70 mg/L, respectively. In particular, for the specimens of #7 and #8, which contained more than 3 wt% of DCM in the surface layer, the results of formaldehyde emissions were sharply reduced and the emission values were 0.05 mg/L and 0.03 mg/L, respectively. The values were significantly lower than the SE0 grade (under 0.3 mg/L) of formaldehyde emission level in the Korean Standard. From the results of specimens #7 and #8, using an additive with more than the specific mass ratio in the surface layer of the particleboard was effective to reduce formaldehyde in the desiccator method. In the desiccator method, the formaldehyde emitted from the exposed surface of the specimen is absorbed into the distilled water within 24 h in the 10-L desiccator. Because of the characteristics of the desiccator test method, the formaldehyde reduction was considerably more efficient when applying resin on the

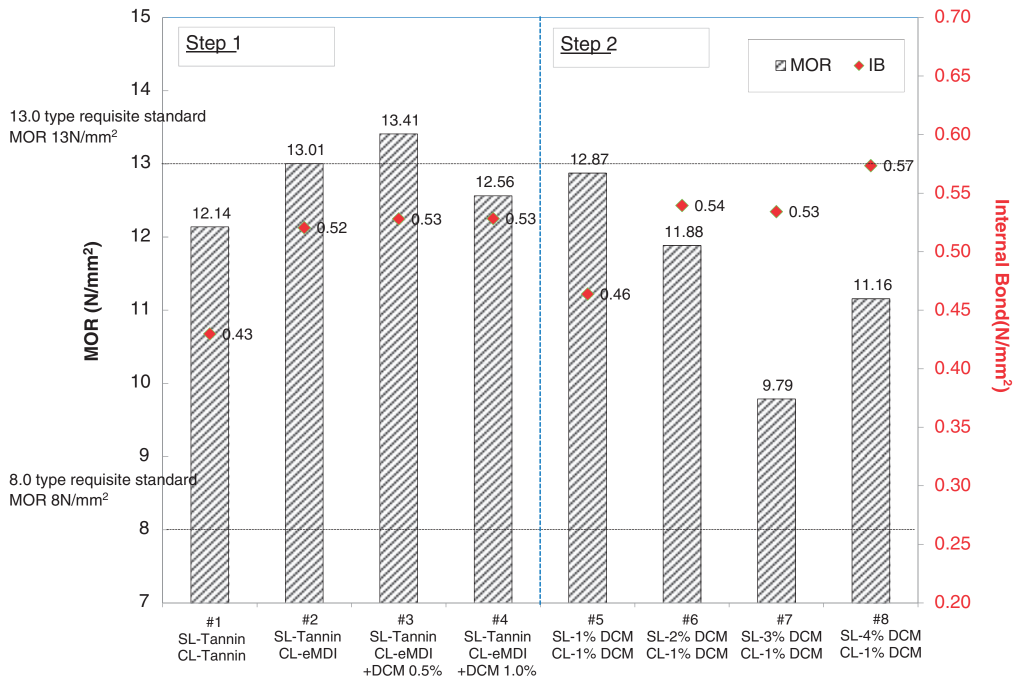

Fig. 3. Mechanical properties of particleboard with various adhesion systems by using a universal testing machine.

surface layer of the particleboard, using an additive as a scavenger, than in the core layer because the exposed surface of the specimen would consist most of the surface layer of the particleboard

Mechanical properties

The mechanical properties of all manufactured particleboards are shown in Figure 3.

In the cases of specimens of step 1, the internal bond strength of the reference board made with Tannin resin in all layers showed a value of 0.43 N/mm^2. When MDI resin and resin containing the additive were used on the core layer, each value of the manufactured board was higher than that of the reference board and the values were 0.52 N/mm^2 and 0.53 N/mm^2, respectively. In addition, the bending strength of the reference board showed a value of 12.14 N/mm^2. When MDI resin and resin containing the additive were used on the core layer, each value of manufactured board was higher than that of the reference board and the values were 13.01 N/mm^2 and 13.41 N/mm^2, respectively. According to the results, it was also confirmed that the mechanical properties were affected by resin used in the core layer and by resin with the additive; however, the values showed narrow variations in comparison with the formaldehyde content results. In the case of the specimens of step 2, the effect of DCM in the surface layer according to various content ratios was evaluated to confirm the impact on the mechanical properties. The internal bond strength of specimen #4 without DCM added on the surface layer showed a value of 0.53 N/mm^2. When resin containing DCM was used on the surface layer with various content ratios, each value of manufactured board was similar or higher than the result of specimen #4; the values were 0.56 N/mm^2, 0.54 N/mm^2, 0.53 N/mm^2 and 0.57 N/mm^2, respectively.

Unlike the results of internal bond strength, the results of bending strength of the specimens in step 2 showed different aspects. The result of specimen #4 with DCM not added on the surface layer was 12.56 N/mm^2. In the cases of resin containing additive on the surface layer, the results of bending strength were similar or lower than the result of specimen #4; the values were 12.87 N/mm^2, 11.88 N/mm^2, 9.79 N/mm^2 and 11.16 N/mm^2, respectively.

Based on KS F 3104: particleboard criteria, type 8.0 is the minimum requisite quality standard for general uses. The type 8.0 requirements are as follows:

1. Bonding strength should be over 8.0 N/mm^2 for both the length and width direction.
2. Internal bond strength should be over 0.15 N/mm^2.

The results showed that the mechanical properties of all the manufactured particleboards met the type 8.0 requirement for the particleboard. Therefore, internal bond strengths showed outstanding performance above the criteria of type 18.0 (above 0.30 N/mm^2). However, the bending strengths values showed a range from 9.79 to

13.41 N/mm^2 excluding specimens #2 and #3; the other specimens could not meet the higher criteria of type 13 (above 13.0 N/mm^2).

Conclusions

The adequacy of using tannin resin to make environment-friendly furnishing materials has been confirmed as suitable through the experimental research of this study. The adhesion system method of dividing the core and surface layer showed better performance than using tannin resin in all layers. The emission of formaldehyde as used in the hardener was sharply reduced by containing an additive that acted as a scavenger. In particular, the effect of the scavenger maximizes the application to 1 wt% in the core layer and 4 wt% in the surface layer and the value was 0.03 mg/L by the desiccator method. In the cases of mechanical properties, all specimens satisfied the minimum requirements of the Korean Standard. However, further studies on the performances of bending strength are considered necessary.

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