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**Nota Técnica**

# Evaluation of chestnut shell and coffee waste with phenol-formaldehyde resin for plywood filler

Avaliação de cascas de castanha e resíduos de café como material de enchimento do adesivo fenol-formaldeído para a produção de compensados

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## ABSTRACT

Chestnut fruit shell and ground coffee waste were evaluated as filler raw materials for plywood adhesive. A phenol-formaldehyde (PF) resin was formulated in the laboratory for plywood manufacture. Specification characteristics were determined on the PF resin, including nonvolatile solids, gel time, viscosity, etc. The laboratory synthesized PF resin was mixed with extender, filler and NaOH. Plywoods were uniformly made with the PF resin mixture and tested for tension shear strength after aging method, modulus of rupture (MOR) and thickness swell according to the procedures of the Korean Standard KS F 3101 and KS F 3114. All plywood made with each filler type showed good physical and mechanical strength properties. The performance test results indicated that the chestnut fruit shell and ground coffee waste would be suitable as filler for plywood adhesive.

**Keywords:** Chestnut fruit shell; Coffee waste; Plywood; Filler; Phenol-formaldehyde resin

## RESUMO

A casca da castanha e resíduos de café moído foram avaliados como material de enchimento para produção de adesivo para compensados. Uma resina de fenol-formaldeído (FF) foi formulada em laboratório para fabricação de compensados. As propriedades da resina FF foram sólidos não voláteis, tempo de gelatinização (gel time),viscosidade, etc. A resina FF sintetizada em laboratório foi misturada com extensor, carga e NaOH. Os compensados foram feitos de maneira uniforme com a mistura de resina FF e testados quanto à resistência ao cisalhamento, módulo de ruptura (MOR) e inchamento em espessura, de acordo com as normas coreanas KS F 3101 e KS F 3114. Todos os compensados fabricados com cada tipo de carga apresentaram boas propriedades de resistência física e mecânica. Os resultados dos testes de desempenho indicaram que a casca da castanha e os resíduos de café moído são adequados como carga para produção de adesivo para fabricação de compensados.


**Palavras-chave:** Casca de castanhas; Resíduos de café; Material de enchimento; Resina fenol-formaldeído

# 1 INTRODUCTION

In worldwide, plywood is the most familiar and predominant wood panel product, together with medium density fiberboard (MDF) and medium density particleboard (MDP). Plywood is traditionally used in everything from furniture to residential construction for such uses as roof sheathing, flooring and concrete forms. All plywood bind wood veneer sheets with resin to form a wood-based panel. In Korea, plywood flooring is used as a covering of an under flooring heating system, known as an Ondol plywood board. Korean Ondol plywood board is well known internationally, and exported widely to the world.

Phenol-formaldehyde (PF) resin, which is ideal for glue of exterior type panel, has been the major binder for the manufacture of wood-based panels such as plywood, laminated veneer lumber (LVL), oriented strandboard (OSB) and fiberboard. Adhesives for structural plywood are composed of PF resin, water, extender, filler, and sodium hydroxide (SELLERS, 1994). Due to the high cost of PF resins, it has become increasingly desirable to reduce the amount of PF resin required in the adhesive mix. Fillers, which are used to describe the nonadhesive ingredients added to the adhesive mix to fill the voids in the cured resin structure, play an important role in achieving this goal.

Fillers can be classified into lignocellulosic and inorganic compounds such as nutshell flours, furfural residue, wood flour, bark and clay (ROBERTSON; ROBERTSON, 1977). Since 1950, the southern pine plywood industry has been using the furfural process residue as a principal filler (SELLERS, 1994). Walnut shell flour was found to be superior and became the more commonly used filler for plywood adhesive (SELLERS, 1985). In 1980, pecan shell flour was introduced with successful results as plywood filler. Its much lower ash content as compared to other fillers was significant quality consideration with respect to the tool wear during panel cutting (SELLERS *et al*., 1990). Acid hydrolysis residues from newsprint waste, and Korean source residues from walnut, pinenut and ginkgonut shells were suitable as alternative fillers for structural plywood adhesives, respectively (OH *et al*., 1997; OH; SELLERS, 1999). In addition, wood barks such as Douglas-fir, alder, hemlock, southern pine, wood particleboard sander dust, and also attapulgite clay have been used as fillers for exterior plywood adhesives (SELLERS, 1994). Quality fillers for adhesives of plywood and LVL have been in high demand, and new filler sources are desired by the wood-based panels industry.

Chestnut trees (*Castanea mollissima* Blume) are of moderate growth rate for Chinese chestnut tree to fast-growing for American and European species. Leading producers of chestnut fruit include China, Italy, Republic of Korea, Turkey, Bolivia and Greece (FAO, 2019). The shell peeling process of the chestnut fruit generates a waste product of shell, which can be used as by-product of resources.

The top 5 producers of green coffee are Brazil, Vietnam, Indonesia, Columbia and Ethiopia, and their global market share of green coffee is above 65% (FAO, 2019). A large amount of ground coffee waste is generated after brewing the ground coffee in every year. The ground coffee waste can be used for alternative renewable sources.

The objective of this research was to evaluate the chestnut fruit shell and ground coffee waste as filler raw materials for plywood adhesive. For this, a phenol-formaldehyde resin was synthesized in the laboratory for bonding plywood.

## 2 MATERIALS AND METHODS

### 2.1 PF resin synthesis

The PF resin formulation procedure synthesized in this study is summarized in Table 1. A mole ratio of 2.15 of formaldehyde to phenol was used in the preparation of the PF resin with a two-stage caustic addition. The general cook procedure was similar to that outlined by Oh and Sellers (1999) and as follows.

Phenol, NaOH (50% solution) and water were charged into a stirred reactor and the reaction mixture was heated 70~75°C. Formaldehyde solution (37%) was then added dropwise over a period of 30 min, while the reaction temperature was held constant by cooling using an ice bath. Then the reaction temperature was gradually increased to 85°C over a period of 30 min. After the reaction temperature was reached at 85°C, the remaining NaOH and water were added to attain the targeted resin solid level. This reaction temperature was maintained until the resin reaching a desired final viscosity between 500 to 1,000 mPa·s (cP) at 25°C. The resin was then cooled to room temperature and stored until use.

### 2.2 PF resin analysis

The viscosity of the PF resin was measured using a Brookfield Viscometer, Model RVF, spindle number 1 and 2 at 2.09 rad/s (20rpm) rotation. The free formaldehyde content was measured using the hydroxylamine hydrochloride method (WALKER, 1964). Gel time was measured using a Sunshine gel timer at 100°C. Resin solid level was determined by heating 1 g of resin on an aluminum pan at 125°C for 2 hours.

### 2.3 Filler

Chestnut (*Castanea crenata* Sieb. et Zucc.) fruit shells were obtained by a commercial food company in Korea. The experimental fillers, used in this study, were chestnut fruit shell and ground coffee (*Coffea arabica* L.) waste prepared by drying them to around 8% moisture content (MC) and grinding them with a laboratory Wiley mill into a 75-μm (200-mesh) powder. Ash content at 600°C during 5 hours and pH of each fillers are shown in Table 2. The control filler was a walnut (*Juglans siensis* Dode) shell flour typically used by plywood industry in Korea.

### 2.4 Adhesive mix procedure

The plywood adhesive mix followed the PF mix procedure described by Oh *et al*. (1997) (Table 3). Mix viscosities (initially and after 24 hr.) were measured with a Brookfield Viscometer Model RVF, spindle number 4 and 5 at 2.09 rad/second (20 rpm).

### 2.5 Plywood manufacture

Rotary-cut Korean pine (*Pinus koraiensis* Sieb. et Zucc.) veneer was supplied by a commercial plywood manufacturing company in Korea. The veneer was 2.1 mm in thickness, 4 to 6% in moisture content and 450 kg/m$^3$ in density. Laboratory plywood panels were similarly manufactured according to the procedure described previously by Oh and Sellers (1999). With each filler types, four 3-ply plywood panels were made. Each panel was 6.3 mm thick and 300 by 300 mm in size. The adhesive spread was applied with a roller coater at a rate of 171 g/m$^2$, single glueline basis. The total assembly time, which includes the time from first adhesive application until hot-pressing, was 30 minutes. A total of twelve panels were hot-pressed at 150°C for 4 minutes. The prepress and hot-press pressures were 1034 kPa and 1379 kPa, respectively.

### 2.6 Plywood performance test

Ten plywood tension shear specimens (85 mm long and 25 mm wide) were cut from each plywood panel. Five specimens were subjected to the two 4-hour boil accelerated-aging method, as defined in the Korean Standard KS F 3101 for Ordinary Plywood (KSA, 2016) tests for exterior plywood bonded with exterior glue.

For comparative purposes, an additional five specimens were subjected to the

3-hour soak in 60°C water accelerated-aging method, according to the Korean Standard KS F 3101 for Ordinary Plywood (KSA, 2016). For both tests, the tension shear strength of each wet specimens was recorded, and the wood failure over the sheared areas was visually estimated after drying the specimen. The panels are considered as meeting the standard if the test specimens average 50% wood failure.

Two test specimens of modulus of rupture (MOR) and thickness swell were cut from each plywood panel, and determined according to procedure KS F 3114 (KSA, 2016). The parallel MOR results were obtained in static bending test. Plywood thickness swell properties were observed after 24-h water soaking.

### 2.7 Statistical analysis

The tension shear strength, wood failure, MOR and 24-h thickness swell were evaluated using the Statistical Analysis System (SAS) programming package (SAS Institute, 1998). Wood failure values (%) are not normally distributed; therefore, the wood failure values were converted to a normal distribution by arcsine transformation (degree) prior to analysis (OH *et al*., 1997).

The analysis of variance (ANOVA) for a completely randomized design was used for the wood failure values, shear strength, MOR and 24-h thickness swell to determine differences within filler types. Significant differences (0.05 level) were further compared by the t-test for Least Significant Differences (LSD) from the SAS program (STEEL; TORRIE, 1980).

## 3 RESULTS AND DISCUSSION

### 3.1 PF adhesive mix properties

The laboratory-synthesized PF resin had 44.5% nonvolatile solids, 1.20 specific gravity, 10.3 pH, 18.6 minutes gel time at 100°C, 1020 mPas viscosity at 25°C, and 0.17% free formaldehyde (Table 1).

Table 1 – Resin synthesis and properties

| **Resin synthesis charge** | | **PF resin properties** | | |
|---|---|---|---|---|
| **Ingredients** | **Amount (moles)** | **Property** | **Basis** | **PF resin** |
| Phenol (90%) | 5.17 | Nonvolatile solids | % | 44.5 |
| HCHO (37%) | 11.15 | pH | -- | 10.3 |
| 1st NaOH (50%) | 1.83 | Specific gravity | -- | 1.20 |
| $H_2O$ | 3.06 | Gel time | min | 18.6 |
| 2nd NaOH (50%) | 1.46 | Viscosity | mPa.s | 1020 |
| $H_2O$ | 5.62 | Free formaldehyde | % | 0.17 |

Source: Author (2019)

The ranges of ash content and pH of the three filler were 0.85 to 1.83% and 4.6 to 5.9, respectively (Table 2). The filler content in the adhesive mixes amounts to 5 to 15% of the total adhesive mix solids. The ash content of the filler affects the extent of tool wear when manufactured plywood is trimmed or cut. The more ash in the filler of the adhesive, the higher the potential of cutting tool wears (SELLERS *et al*., 2005).

The PF adhesive mix contained 69% PF resin solids, 5.4% extender solids, 5.4% filler solids and 1.5% added NaOH solids (Table 3). The ranges of the mix viscosities for filler types, initially and after standing 24 hours at room temperature, were 4560 and 9740 mPa·s, respectively (Table 4). Once fillers are added to adhesive mixes, they should not substantially change the consistency and stability of the adhesive mix over a 24-hour to 72-hour period. Thus, the stability of adhesive mixes is important for the consistency and the mix viscosities were within an acceptable range for roll-coater application of plywood adhesives (SELLERS, 1985).

Table 2 – Ash and pH of filler materials

| **Filler type** | **Ash at 600°C (5h)** | **pH** |
|---|---|---|
| | **(%)** | **--** |
| Walnut shell | 1.83 | 4.6 |
| Chestnut shell | 0.85 | 5.2 |
| Coffee waste | 1.56 | 5.9 |

Source: Author (2019)

Table 3 – Adhesive mix procedure and mix characteristics

| Mix procedure | | | Mix characteristics | |
|---|---|---|---|---|
| **Ingredients** | **Amount (%)** | **Mixing time (min)** | **Characteristics** | **Amount (%)** |
| Water | 17.5 | -- | Resin solids | 30.7 |
| Extender | 6 | 2 | Extender solids | 5.4 |
| Filler | 6 | 2 | Filler solids | 5.4 |
| PF resin | 28 | 1 | NaOH solids | 1.5 |
| NaOH | 1.5 | 19 | | |
| PF resin | 41 | 2 | | |
| Total mix | 100 | | Total mix solids (%) | 43 |
| Total resin | 69 | | Total mix water (%) | 57 |

Source: Author (2019)

Table 4 – Adhesive mix viscosity of filler materials

| Filler type | Viscosity (mPa.s) | |
|---|---|---|
| | **Initial** | **After 24 hours** |
| Walnut shell | 4560 | 9630 |
| Chestnut shell | 5120 | 9740 |
| Coffee waste | 4700 | 7570 |

Source: Author (2019)

## 3.2 Plywood test results

The ranges of wood failure values for all plywood after two 4-hour boil and 3-hour soak in 60°C water were 94 to 97% and 91 to 95%, respectively (Table 5). LSD tests (0.05 level) showed that plywood made with each filler types did not significantly differ from each other in wood failure. The plywood is considered as meeting the standard of average 50% wood failure. Thus, the plywood made with three filler types was comparable in wood failure percentages.

The ranges of shear strength values for all plywood after two 4-hour boil and 3-hour soak in 60°C water were 1320 to 1417 kPa and 1586 to 1684 kPa, respectively (Table 5). LSD test results (0.05 level) showed that there were significant difference in shear strength among the three filler types for both accelerated aging method.

However, the average wood failure and shear strength values exceeded the minimum requirement of 50% wood failure and 600 kPa shear strength for Korean Standard KS F 3101 (KSA, 2016).

The average shear strength after the two 4-hour boil and 3-hour soak in 60°C water aging methods were 1378 and 1633 kPa, respectively (Table 5). The LSD results (0.05 level) showed significant difference in shear strength between the two test methods, because the two 4-hour boil is considered definitely a more severe test regime. The average wood failure after two 4-hour boil and 3-hour soak in 60°C water were 96 and 93%, respectively. The LSD test results (0.05 level) showed that there was significant difference in wood failure between the two test methods. The two 4-hour boil aging method with its lower shear strength (1378 kPa) than the 3-hour soak in 60°C water (1633 kPa), resulted in a higher wood failure percentage (96%) than 3-hour soak method (93%). The results are in agreement with the result previously reported by Oh and Lee (2004). In general, higher shear strength of plywood results in lower wood failure (SELLERS, 1985).

Table 5 – Plywood shear strength and wood failure estimates after testing by two 4-hr. boil and 3-hr. 60°C water soak aging method

| Filler type | Two 4-hr. boil | | 3-hr. 60°C water soak | |
|---|---|---|---|---|
| | **Shear strength** | **Wood failure** | **Shear strength** | **Wood failure** |
| | (kPa) | (%) | (kPa) | (%) |
| Walnut shell | 1417 A1 | 96 A | 1630 AB | 93 A |
| Chestnut shell | 1398 AB | 97 A | 1684 A | 95 A |
| Coffee waste | 1320 B | 94 A | 1586 B | 91 A |
| Average | 1378 | 96 | 1633 | 93 |

Source: Author (2019)

In where: [1] Means with the same letter in the same column are not significantly different (p<0.05).

The MOR range for all plywood produced in this study was 34.8 to 35.6 MPa (Table 6). The LSD test results showed that the MOR did not differ significantly (0.05 level) according to filler type. These MOR values were higher than the minimum requirement (8 MPa) for Korean Standard KS F 3114 (KSA, 2016).

The thickness swell of all plywood ranged from 4.1% to 4.8% after 24-h water soaking (Table 6). The LSD test results showed that the 24-h thickness swell did not differ significantly (0.05 level) according to each filler type.

Table 6 – MOR and 24-h thickness swell of plywood with filler type

| Filler type | MOR (MPa) | 24-h Thickness swell (%) |
|---|---|---|
| Walnut shell | 35.1 (7.3) A | 4.3 (3.9) A |
| Chestnut shell | 35.6 (11.2) A | 4.1 (8.2) A |
| Coffee waste | 34.8 (8.6) A | 4.8 (3.5) A |

Source: Author (2019)

In where: MOR: Modulus of rupture; Values in parentheses are coefficients of variations. Means with the same letter in the column are not significantly different ($p<0.05$).

## 4 CONCLUSIONS

All experimental plywood made from each filler type showed good physical and mechanical strength properties. The results of this research indicate that the chestnut fruit shell and ground coffee waste are suitable raw materials of filler for plywood adhesive. Also, these fillers can be used for adhesive mixes of LVL and Ondol plywood flooring board.

The usage of these resources in wood-based panels industry will enhance the economics of by-product.

## REFERENCES

FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS (FAO). **Statistic of Agriculture and Forest Products**. FAO, Rome, Italy. 2019.

KOREAN STANDARD ASSOCIATION (KSA). **Ordinary plywood**. KS F 3101. KSA. Seoul, Republic of Korea. 2016.

KOREAN STANDARD ASSOCIATION (KSA). **Plywood for flooring**. KS F 3114. KSA. Seoul, Republic of Korea. 2016.

OH, Y-S.; LEE, J-K. Evaluation of Korean softwood plywood bonded with urea-formaldehyde resin adhesive. **Forest Products Journal**, v. 54, n. 2, p. 77-80. 2004.

OH, Y-S.; SELLERS, T. JR. Korean filler raw materials for plywood adhesives. **Forest Products Journal**, [s.l.], v. 49, n. 3, p. 61-64. 1999

OH, Y-S.; SELLERS, T. JR.; KIM, M.G.; STRICKLAND, R.C. Fillers for plywood adhesives made from acid hydrolysis residues of municipal newsprint waste. **Forest Products Journal**, [s.l.], v.47, n. 10, p.107-110. 1997.

ROBERTSON, J.E.; ROBERTSON, R.R. Review of filler and extender quality evaluation. **Forest Products Journal**, [s.l.], v. 27, n. 4, p. 30-38. 1977.

SAS Institute. **SAS system for Microsoft Windows**, Release 6.10. SAS institute, Cary, NC. 1998.

SELLERS, T. Jr. Information processing systems for general access of unpublished wood products research data. **Forest Products Journal**, [s.l.], v. 44, n. 6, p. 33-38. 1994.

SELLERS, T. JR. **Plywood and Adhesives Technology**. Marcel Dekker, N.Y. 1985. 661 p.

SELLERS, T. JR.; MILLER, G.D.; SMITH, W. Tool wear properties of five extender/ fillers in adhesive mixes for plywood. **Forest Products Journal**, [s.l.], v. 55, n. 3, p. 27-31. 2005.

SELLERS, T. JR.; MILLER, G.D.; NIEH, W. L. Evaluation of three fillers in PF adhesives used to bond intermediate moisture content plywood: glueline durability and knife wear. **Forest Products Journal**, [s.l.], v. 40, n. 10, p. 23-28. 1990.

STEEL, R.G.D.; TORRIE, J.H. **Principles and Procedures of Statistics**: A Biometric Approach. 2nd ed. McGraw-Hill Book. Co., New York.1980. 633 p.

WALKER, J.F. **Formaldehyde**. Reinhold, New York. 1964. 96 p.

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Contribution: Conceptualization, Methodology, Project administration, Resources, Supervision, Validation, Writing – original draft, Writing – review & editing

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