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Effect of addition of polyvinyl acetate to melamine-formaldehyde resin on the adhesion and formaldehyde emission in engineered flooring

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Abstract

The objective of this research was to investigate the effect of adding polyvinyl acetate (PVAc), for reducing the formaldehyde emission level, on the adhesion properties of melamine-formaldehyde (MF) resin for fancy veneer and plywood in engineered flooring. We controlled the hot-press temperature, time and pressure to determine the bonding strength and formaldehyde emission. Blends of various MF resin/PVAc compositions were prepared. To determine and compare the effect of PVAc content, 0, 30, 50, 70 and 100% PVAc floorings, by weight of MF resin, were used. Wheat flour, 25% by weight of adhesive, was added as material to increase the quantity. To determine the level of formaldehyde emission, we used the desiccator method. The formaldehyde emission level decreased with increased additions of PVAc. At a PVAc replacement ratio of only 30%, the formaldehyde emission level of the coated sample by UV-curable coat was under E_1 grade. Curing of the high MF resin content in this adhesive system (MF resin with PVAc) was well processed indicating that the bonding strength was increased. In the case of PVAc only, the bonding strength was much lower due to the already high temperature of 120 °C. The adhesion layer was broken by high temperature and pressure. The sample with 30% PVAc added to MF resin (MF resin: PVAc = 70:30) showed good bonding strength compared with MF resin only in all cases, hot-press temperature, time, pressure and boiling test.

Keywords: PVAc; Bonding strength; Formaldehyde emission; Flooring; Fancy veneer

1. Introduction

With rising economic standards in Korea, there has been increasing demand for a wide range of flooring products. Recently, there have been many concerns about human health and the environment. PVC flooring, laminated paper lacquered with bean oil, which was used in most Korean houses has been replaced by wood flooring materials, especially in new apartments. There are three types of wood flooring: laminate flooring, engineered flooring and solid wood flooring. The laminate flooring consists of HDF (high-density fiberboard) as the core material, while the engineered flooring consists of plywood with a thin fancy veneer bonded onto the face of the plywood using urea-formaldehyde and melamine-formaldehyde (MF) resins as hot-press adhesives. Many consumer products containing formaldehyde-based resins release formaldehyde vapor, leading to consumer dissatisfaction and health-related complaints. These emissions have resulted in various symptoms, the most common of which is irritation in the eyes and the upper respiratory tract. Formaldehyde has also been found to produce nasal carcinomas in mice and rats after exposure to 14.1 and 5.6 ppm of formaldehyde, respectively, over a long period of time. These findings have led to an intensified interest in the indoor environment. Consumer products, specifically construction materials, are a major source of formaldehyde in the indoor environment [1–3].

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The Korean government has started controlling indoor air quality since 2004. The law from the Ministry of Environment regulates the use of pollutant emission building materials and prohibits the use of materials that emit formaldehyde more than 1.25 mg/m^2 h (JIS A 1901, small chamber method). This is equivalent to E_2 grade (>5.0 ppm) when changed to the desiccator method (JIS A 1460). Most suppliers and people are concerned about how to reduce pollutants from building materials and how to control indoor air quality [4].

Polyvinyl acetate (PVAc) adhesive is commonly known as resin emulsifier or simply as ''white glue''. It is manufactured by polymerizing vinyl acetate monomer and stabilizers with other polymers to copolymers. It comes in liquid form with viscosity ranging from 2000 to 3000 centipoises at 21 °C. PVAc is an odorless, non-flammable adhesive. It can be used in cold temperatures and solidifies quickly. Its application is very easy and it does not damage the tools during the cutting process. However, PVAc adhesive's mechanical resistance decreases with increasing temperature. It loses bonding resistance capacity at over 70 °C [5,6].

Huang et al. studied the miscibility of thermo-setting resins and PVAc blends. It was found that the phenolic/PVAc blends are miscible in the amorphous phase over the entire composition range. Results obtained by DSC, FT-IR and high-resolution solid-state 13C NMR revealed strong inter-association hydrogen bonds between the hydroxyl group of phenolic and the carbonyl group of PVAc [7].

In this study, we focused on the effect of adding PVAc, for low-formaldehyde emission level, on the adhesion properties of MF resin for fancy veneer and plywood in engineered flooring. We controlled the hot-press temperature, time and pressure.

2. Experimental

2.1. Materials

Resin was prepared at an F/M molar ratios of 1.75, with a solid content of 60%. After addition of water to formalin, i.e. 38.5% by weight of formaldehyde in water, the pH was adjusted to 9.0 by the addition of a 1 M sodium hydroxide solution (because the methylolated intermediates of the reaction rapidly condense under acidic conditions) and melamine was added. As hardener, 10% of ammonium chloride solution was used. The viscosity as measured using a Brookfield Viscometer Model DV-II+ was 75 cPs at 21 °C. PVAc in liquid form with the following characteristics was used: density, 1.1 g/cm^3; viscosity, 2000 cPs at 21 °C; pH value and ash ratio, 5% and 3%, respectively. PVAc adhesive was supplied from Tae Yang Chemical Co. Ltd (Incheon, Korea). The molecular structure of MF resin and PVAc are shown in Fig. 1.

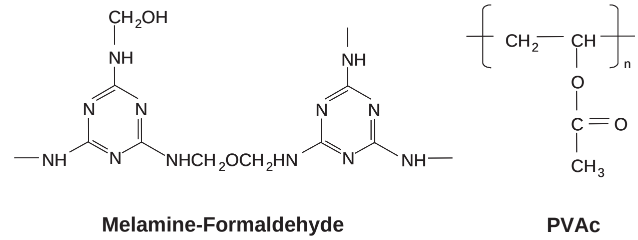

Fig. 1. Molecular structures of MF resin and PVAc.

The fancy veneers were of 0.5 mm thickness, the species was birch, and the plywoods manufactured in Indonesia were supplied from Dongwha Enterprise Co. Ltd.

2.2. Blend preparations

Blends of various MF resin/PVAc compositions were prepared as content ratios. To determine and compare the effect of PVAc content, 0, 30, 50, 70 and 100% PVAc floorings, by weight of MF resin, were used. Each composition stirred for 20 min and five blending systems were prepared. Wheat flour, 25% by weight of adhesive, was added as material to increase the quantity. To determine effect of wheat flour content, it was added at 15, 20, 25 and 30%.

2.3. Sample preparations

We set various hot-press temperatures (60, 80, 100, 120, 140 and 160 °C), times (110, 160, 200 and 240 s) and pressures (3, 5, 8 and 12 kgf/cm^2). The main temperature, time and pressure were 120 °C, 160 s and 5 kgf/cm^2, respectively.

2.4. Formaldehyde emission

The Japanese standard method of determining the formaldehyde emission from samples, before coating and after coating, was used. The 24 h desiccator method uses a common glass desiccator with a volume of 10 l. Ten test specimens, with dimensions of 5 cm × 15 cm, are positioned in the desiccator. The emission test lasts 24 h in the covered desiccator at a temperature of 20 °C. The emitted formaldehyde is absorbed in a water-filled petri dish and is analyzed by JIS A 1460, Building boards determination of formaldehyde emission–Desiccator method.

2.5. Bonding strength

To determine the bonding strength between the fancy veneer and plywood, we tested with a Universal Testing

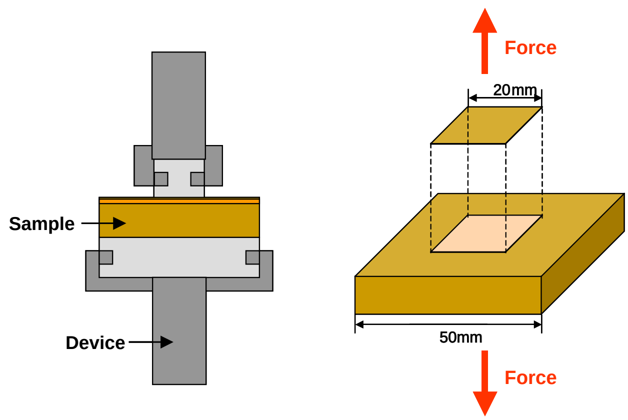

Fig. 2. Test of surface bonding strength.

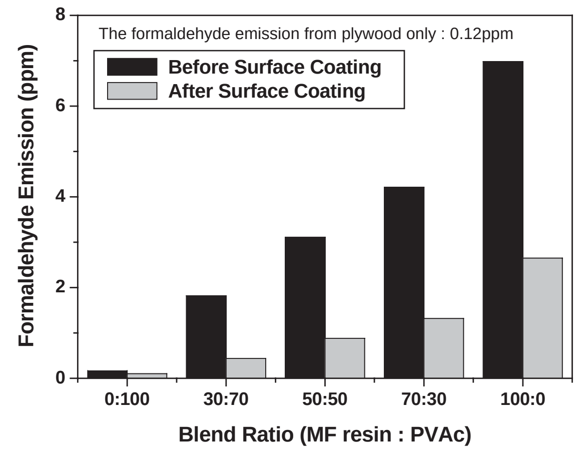

Fig. 3. Formaldehyde emission from various adhesive blend ratios.

Machine (Hounds Co.) in the tensile mode. Samples were cut at 5 cm × 5 cm (length × width) and a knife mark of 2 cm × 2 cm (length × width) was made on the surface (fancy veneer face) at a depth of 0.5 mm. We bonded the upper device of UTM and the knife mark of 2 cm × 2 cm on the surface with hot-melt adhesive. Another face was bonded to under the device like the internal bonding strength test specimen. It is shown in Fig. 2. The tests were performed at crosshead speeds of 2 mm/min.

3. Results and discussion

3.1. Formaldehyde emission

Today, MF resin is mainly used as thermosetting wood adhesives for wood panels. These thermosets of the family of amino resins are widely applied in the coating industry as resin crosslinkers such as hexakis-(methoxymethyl)-melamine (HMMM), protective coatings, decorative laminates and wood composite particleboard for household products, and also as molding compounds in dinnerware [8]. MF resin gives excellent adhesive performance, good moisture resistance and tends to give lower formaldehyde emission than UF resin. However, in this study, the formaldehyde emissions from the products glued with MF resin were much higher than we expected before surface coating. They were more than 7 ppm and exceeded the E_2 grade of formaldehyde emission level in the Korean Standard as shown in Fig. 3. When we coated the surface with UV-curable urethane acrylate coating for flooring, the emission level was reduced more than third.

In this resin system, of MF resin and PVAc, the role of PVAc was to reduce formaldehyde emission by replacement of the formaldehyde system resin and to increase the initial bonding strength through the high viscosity and room-temperature curable property of PVAc. The formaldehyde emission level was decreased with increasing additions of PVAc as replacement for MF resin. At a PVAc replacement ratio of only 30%, the formaldehyde emission level of the coated sample was under E_1 grade.

The formaldehyde emission from these samples was only caused by the surface adhesion with MF resin. The emission from samples bonded with PVAc only was just 0.12 ppm, which is similar to the level of non-bonded plywood. The formaldehyde level of plywood only was 0.12 ppm. When we coated engineered flooring with UV-curable urethane acrylate coating, the surface of the fancy veneer area was covered and this interrupted the emission of formaldehyde from the flooring surface.

3.2. Bonding strength

The bonding strengths of the engineered flooring samples bonded with various adhesive systems at a press temperatures of 120 °C for 160 s are shown in Fig. 4.

The bonding strength of the engineered flooring samples, non-treated (before boiling), made using the MF resin and PVAc increased as the MF resin content was increased. Because the hot-press temperature of 120 °C was high, MF resin was cured as a thermosetting adhesive. On the basis of KS F3110 method, we boiled the samples at 60 ± 3 °C for 4 h, dried them for 20 h, boiled them again for 4 h and finally dried them for 3 h to determine the waterproof property of each adhesive system. This result also showed the tendency of the bonding strength to increase as the MF resin content was increased. When only PVAc was used, there was no waterproof property. We found that PVAc was weak in water.

Fig. 5 shows the relationship between bonding strength and formaldehyde emission of the engineered floorings bonded by MF resin with various contents of PVAc (before surface coating). As bonding strength

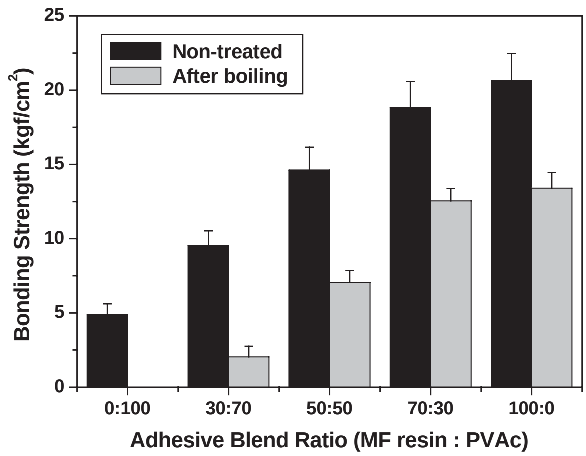

Fig. 4. Bonding strength of various adhesive systems; non-treated and after boiling.

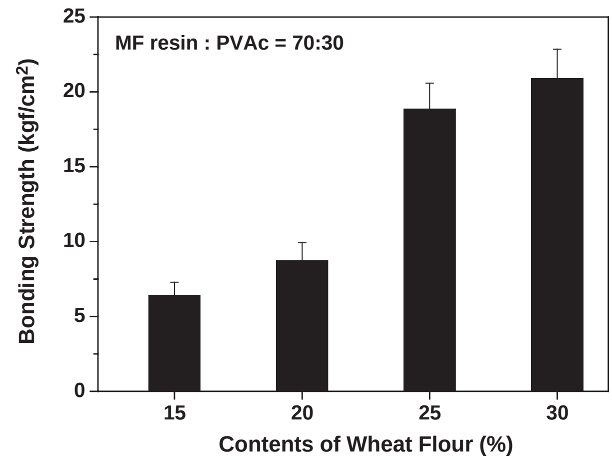

Fig. 6. Bonding strength according to wheat flour content.

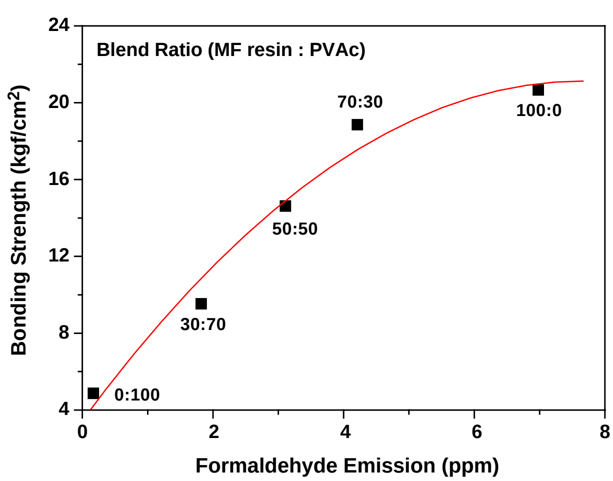

Fig. 5. Relation between bonding strength and formaldehyde emission of the engineered floorings boded by MF resin with as content of PVAc, (before surface coating).

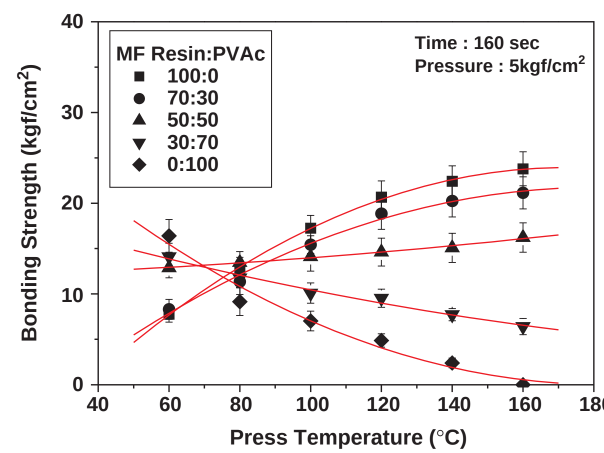

Fig. 7. Bonding strength of various adhesive systems according to hot-press temperature.

increased, formaldehyde emission also increased. The reduction ratio of formaldehyde emission was much higher than the reduction ratio of bonding strength, when 30% of PVAc was added. It is possible to reduce formaldehyde emission with just small decreasing bonding strength by adding just 30% of PVAc.

When we blended MF resin and PVAc together, wheat flour was used as material to increase the quantity and viscosity. Fig. 6 shows the effect of wheat flour contents on bonding strength. It was sharply increased with the increase of all resin weight from 20 to 25% in MF resin: PVAc = 70:30 system. We need to fill up the grain holes with wheat flour in the adhesive blend. As the species of fancy veneer, we can control contents of the wheat flour content because of different veneer surfaces. Especially, in the case of oak and teak, there are many grain holes and rugged surface.

For the hot-press temperature, the bonding strength of each adhesive system was investigated for 160 s of press time at a pressure of 5 kgf/cm^2. As shown in Fig. 7, the samples of high MF resin content above 50% showed increasing bonding strength with increasing temperature. However, PVAc adhesive's mechanical resistance decreased with increasing hot-press temperature. In the case of PVAc only, the bonding strength was sharply decreased. The adhesion layer of PVAc was broken at high-temperature conditions over 60 °C, in agreement with the reports by some other researchers [6]. They reported that PVAc loses its bonding resistance capacity over 70 °C. The addition of 30% MF resin did not raise the bonding strength with increasing hot-press temperature.

Fig. 8 showed the effect of hot-press time on bonding strength at 120 °C by a pressure of 5 kgf/cm^2. The basic property of thermosetting resins, whereby the bonding strength increases with increase in hot-press temperature [9], was shown in the case of MF resin component ratios

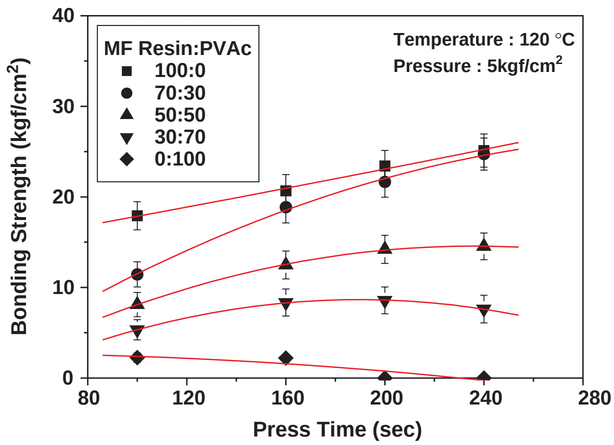

Fig. 8. Bonding strength of various adhesive systems according to hot-press time.

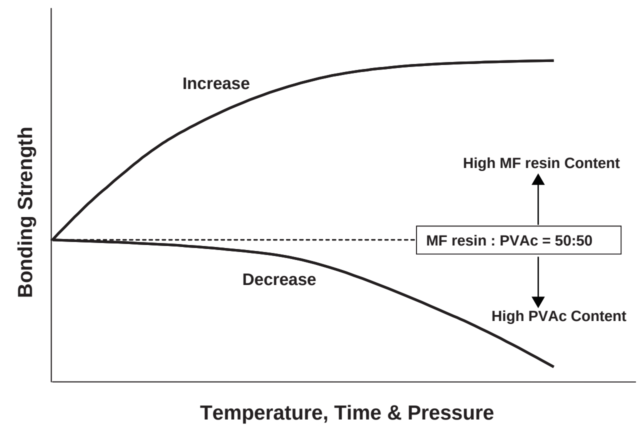

Fig. 10. Relation between bonding strength and hot-press temperature, time and pressure of the engineered floorings bonded by MF resin with, as content of PVAc.

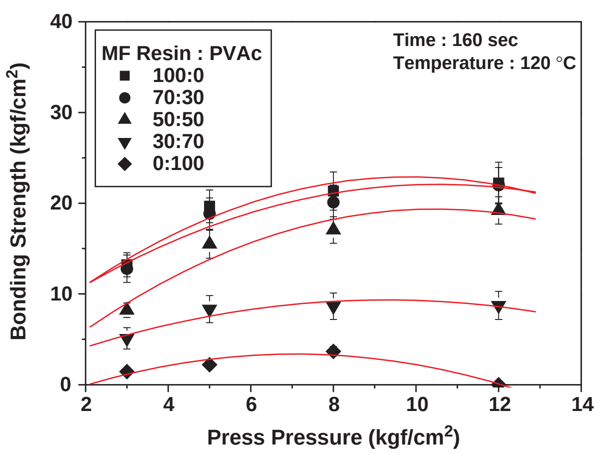

Fig. 9. Bonding strength of various adhesive systems according to hot-press pressure.

of more than 50%. Although the adhesion of PVAc was broken again after 200 s, the effect of time on bonding strength was weaker than that of temperature.

Finally, the effect of hot-press pressure is shown in Fig. 9, which shows that bonding strength was increased till 12 kgf/cm^2 in the case of the 70:30 and 50:50 (MF resin: PVAc) resins. This tendency was the same as that in the time and temperature cases. Thermosetting resin is sensitive to curing time, temperature and pressure [10]. With high MF resin contents in this adhesive system (MF resin with PVAc) curing was well processed, indicating that the bonding strength was increased. In the case of PVAc, the bonding strength was much lower due to the already high temperature of 120 °C. The sample with 30% PVAc added to MF resin (MF resin: PVAc = 70:30) showed good bonding strength compared with MF resin only in all cases, hot-press temperature, time, pressure and boiling test.

We put various results of bonding strength together in Fig. 10. In case of high MF resin content (more than 50%), bonding strength is increased with increase in hot-press temperature, time, pressure. It shows the tendency of curing of typical thermosetting adhesive because of high MF resin content. On the other hand, bonding strength of high PVAc content was decreased over 80 °C of press condition. PVAc loses bonding resistance capacity. The adhesion layers of PVAc were broken by high temperature.

4. Conclusion

PVAc was added to reduce formaldehyde emission in the adhesion between plywoods and fancy veneers by replacing the formaldehyde system resin. The formaldehyde emission level was decreased with increased additions of PVAc as replacement for MF resin. At a PVAc replacement ratio of only 30%, the formaldehyde emission level of the coated sample was under E1 grade. The formaldehyde emission from these samples was only caused by the presence of MF resin. With high MF resin contents in this adhesive system (MF resin with PVAc) was well cured, indicating that the bonding strength was increased. In the case of PVAc only, the bonding strength was much lower due to the already high temperature of 120 °C.The adhesion layer was broken by high temperature and pressure. The sample with 30% PVAc added to MF resin (MF resin: PVAc = 70:30) showed good bonding strength compared with MF resin only in all cases, with hot-press temperature, time, pressure and boiling test. According to the results of formaldehyde emission and bonding strength, the adhesive system with an 'MF resin: PVAc ratio of = 70:30' was the best formula for adhesion between plywood and veneer. Although the bonding strength was increased with increase in MF resin content in this adhesion, the formaldehyde emission level was also increased.

Acknowledgements

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