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Effects of an Inorganic Compound Added to Amino Resin Binders for Manufacture of Plywood

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Abstract : Curing behavior and structural property of an inorganic compound added urea-formaldehyde(UF) and urea-melamine-formaldehyde(UMF) were studied. In addition, tensile strength and formaldehyde emission of plywoods made of those resin binders were studied. Curing temperature and structure were not changed, but tensile strengths of plywoods manufactured both with a UF resin and a UMF resin were decreased slightly as increased amount of inorganic compound. Formaldehyde emissions from plywoods were reduced as increased amount of inorganic compound. Wheat flour as an extender was helped to reduce of formaldehyde emission. From the result of this study it might be estimated that using appropriate amount of inorganic compound and proper resin system can be strengthened bond strength and reduced formaldehyde emission.

Keywords : formaldehyde emission, inorganic compound, plywood, urea-formaldehyde resin

1. Introduction

Cheap and colorless amino resin binders are widely used for furniture and for interior wood composites. However, amino resin binder used wooden materials considered as formaldehyde emitting materials due to its fatal weakness against moisture. The formaldehyde emission from the wood based panels for interior applications is considered as a major factor affecting sick house syndrome in newly built houses. Therefore, the issue of formaldehyde emission has one of the most important topics of amino resin binder in last few decades [1-3]. Both the reversible curing mechanism of UF resins and the low resistance against hydrolysis of cured UF resins are major factors affecting formaldehyde emission of UF resin bonded wood-based boards. The formaldehyde emission during curing on board manufacturing or during using boards has been controlled and reduced greatly by technical advancement. The hydrolysis mechanisms of cured UF resins were investigated and the reversible reaction schemes of UF resins were revealed. Numerous researches focused on the formaldehyde emission and the water resistant were done on UF resins but few of them have been reported. Most attention has been focused on control the formaldehyde emission from amino resin binders through resin preparation methods. Even though many

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researchers investigated resin preparation methods to improve formaldehyde emission of amino resins, there is limited approach to use secondary formaldehyde reducing or capturing materials [4-8]. Therefore, this study was conducted to investigate proper formaldehyde reducing material for amino resin binders. A silicon-rich inorganic compound was chosen as possible formaldehyde reducer and its performance as an additive to amino resin binder was examined. Tensile strength and formaldehyde emission of plywood as a function of an inorganic additive to urea-formaldehyde resin and urea-melamine-formaldehyde resin were studied.

2. Materials and methods

2.1. Materials

Inorganic rock powder was obtained from Davistone Co. The chemical composition of inorganic rock powder, as determined using an X-ray fluorescence spectrometer (Shimadzu XRF-1700, Japan), is shown in Table 1 which present the high concentration of SiO_2 and Al_2O_3 elements. A urea-formaldehyde (UF) resin (formaldehyde/urea (F/U) mole ratio of 1.10) was obtained from a plywood producing company and a laboratory synthesized urea-melamine-formaldehyde (UMF) resin (F/U ratio of 1.15 and M content of 5%) was used.

2.2. Manufacturing plywood

Radiata pine veneers (400 ㎜ (l) ×400 ㎜ (w) ×2 ㎜ (t)) with 1% of moisture content were used for manufacturing 3-plies of plywood. A 150 g of resin was mixed with (or without) 30 g of flour as filler and at the same time 0, 1, 3% of a silicon-rich inorganic compound were added to resin mix. Then a 130 g/㎡ of the resin mix was spread on each veneer. After assembled 3-plies of plywood, cold pressed for 30 minutes and hot pressed for 4 minutes at 130℃ and 10kgf/㎠.

2.3. Physical and mechanical test

Solids content of UF resin was 52.8% and free formaldehyde amount of 1.04% was determined by sodium sulfite method. Solids content and free formaldehyde amount of UMF resin were 52.8% and 1.06%, respectively. Viscosity of resins was measured using Brookfield Viscometer at 25℃ and gel times were measured using the common laboratory methods. The pH of the resins was measured with a pH meter at 25℃. Gel times at 100℃ were measured with a Sunshine Gel Timer. DSC curves of resins were obtained using a TA DSC Q10. Approximately 5 mg of resin was sealed in an aluminum pan and run was carried out by increasing temperature of chamber from 30℃ at a heating rate of 10℃/min to 200℃. FT-IR spectra were recorded on a. Nicolet NEXUS spectrophotometer. Dry and wet tensile strengths (TS) of plywood samples were measured by the Korean standard of KS F 3101 (Ordinary plywood).

2.4. Formaldehyde emission test

Formaldehyde emission(FE) amounts were measured. Prepared plywoods were cut in 50 ㎜×150 ㎜ and then those were put into a conditioning room(20℃ and 65% of relative humidity). After 1 week, formaldehyde emission amount of those samples were measured by the Korean standard of KS M 1998-4(Determination of the formaldehyde emission of building interior products-desiccator method).

3. Results and discussion

3.1. Physical and mechanical properties

A silicone-rich compound did not affect physical properties such as viscosity, pH, gel time and solids content of resin mixes and those were summarized in Table 2. Cure temperatures of a UF and a UMF were increased slightly with addition of a

Table 1. Chemical composition of silicon-rich powder by weight percent.

Element	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	LOI[a)]
Content (%)	73.2	15.8	0.6	1.37	0.16	3.97	3.86	0.88

a) Loss of ignition

Table 2. Properties of resin mixes

Resin Mix	Viscosity (cP)	pH	Gel Time (min)	Solids Content (%)
UF	198	8.35	225	52.8
UF+1%[a)]	196	8.38	226	52.8
UF+3%	195	8.37	228	52.9
UMF	201	8.50	248	52.8
UMF+1%	200	8.48	250	52.9
UMF+3%	198	8.49	250	53.0

a) Amount of silicon-rich powder

silicone-rich inorganic compound (Fig. 1 and 2). Cure temperatures of UF mixes were far lower than that of UMF mixes. Curing behavior of these resin mixes tended to depend on the intrinsic curing behavior of resin itself; normally pure UF cure faster than UMF under the same curing condition. Cure temperature of a UF resin itself was 91.72℃, and 92.48℃ with 1% of a silicone-rich compound added mix and increased up to 92.79℃ with 3% of a silicone-rich compound added mix. Cure temperature of a UMF resin itself was 102.79℃, 1% of a silicone-rich compound added mix was 103.22℃ and increased up to 103.98℃ with 3% of a silicone-rich compound added mix. A slight increase of cure temperature of these resin mixes might be simply caused by the formation of hydrogen bonds between a UF or a UMF and a silicone-rich inorganic compound. Even though the cure temperature of resin binders raised slightly with addition of a silicone-rich inorganic compound, no new band occurred

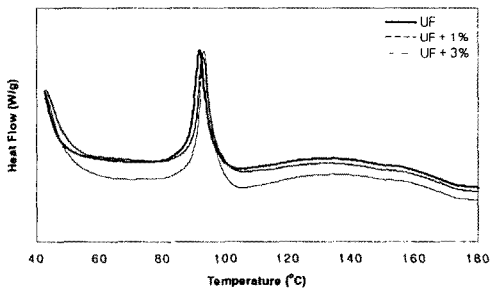

Fig. 1. DSC thermograms of UF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

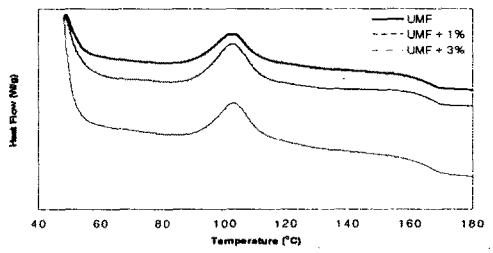

Fig. 2. DSC thermograms of UMF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

by chemical bonding between these amino resins and a silicone-rich inorganic compound observed from FT-IR spectra (Fig. 3 and 4). Only a simple spectral difference between a UF and a UMF existed wavenumber around 820 cm^{-1} at which small -NH wagging bands

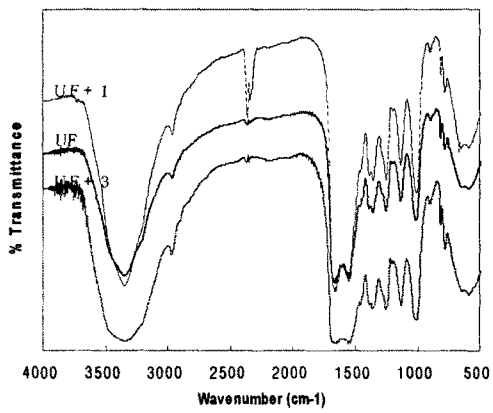

Fig. 3. FT-IR spectra of UF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

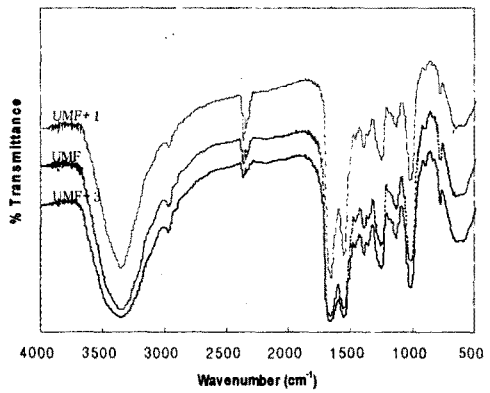

Fig. 4. FT-IR spectra of UMF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

of UMF mixes appeared. Tensile strengths of plywood prepared with UF resin without filler were shown in Fig. 5 and tensile strengths of plywood prepared with UF resin with filler were shown in Fig. 6. As shown in Fig. 5 and 6, tensile strengths of plywoods were weakened as inorganic additive added;

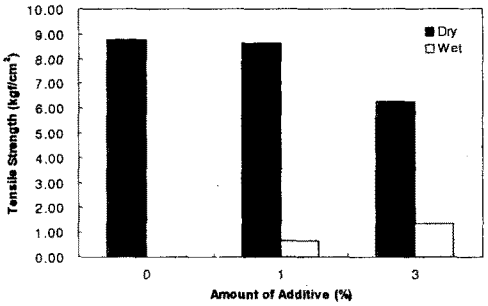

Fig. 5. Tensile strength of plywoods prepared with UF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

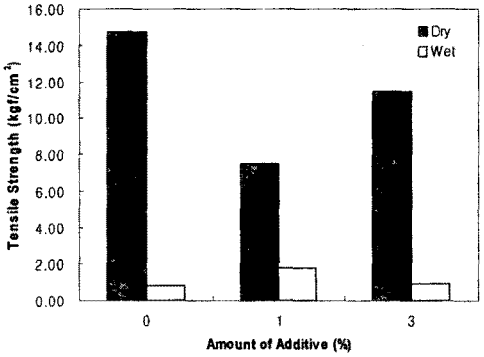

Fig. 6. Tensile strength of plywoods prepared with UF resin contained 20% of flour and added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

the added inorganic compound tended to disturb curing of resin binders. Tensile strengths of plywood prepared with UMF resin with or without filler were shown in Fig. 7 and 8, respectively. Tensile strengths of both cases were weakened by inorganic additive and tensile strength showed the lowest values at 1% of silicone-rich compound addition and it increased at 3% of silicone-rich compound addition, but still not stronger than that of 0% of control. Added

silicone-rich compound presumably acted as a curing catalyst for used UMF resin system. From this presume it might be deduced that 1% of silicone-rich compound was not enough to help curing of UMF resin binder but 3% of it tended to help curing of UMF resin binder a little. It is generally known that UMF resin is more stronger and more water-resistant than UF resin. For that reason, when compared UF resin and UMF resin, UMF resin showed much higher tensile strengths for both dry and wet conditions.

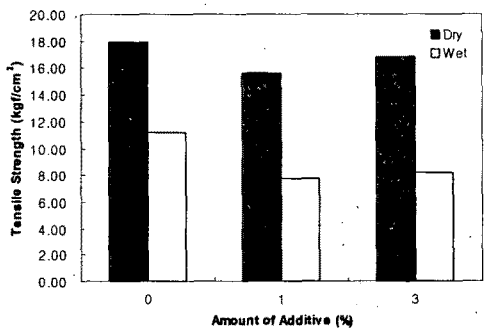

Fig. 7. Tensile strength of plywoods prepared with UMF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

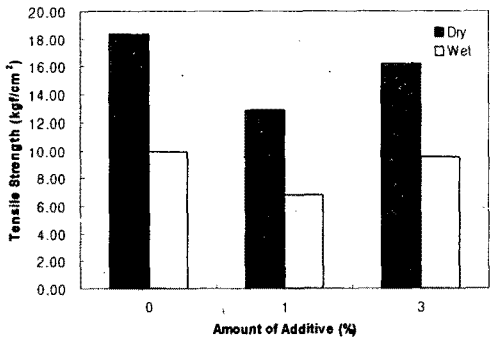

Fig. 8. Tensile strength of plywoods prepared with UMF resin contained 20% of flour and added silicone-rich inorganic compound at 0, 1 and 3 wt.%.

3.2. Formaldehyde emission

Formaldehyde emission amount of plywoods prepared with UF resin without filler was reduced as inorganic compound addition amount increased. Formaldehyde emission amount was reduced with addition of filler, but formaldehyde emission amount was not much affected by the amount of inorganic compound (Fig. 9). On the contrary, formaldehyde emission of plywoods prepared with UMF resin without filler was not much

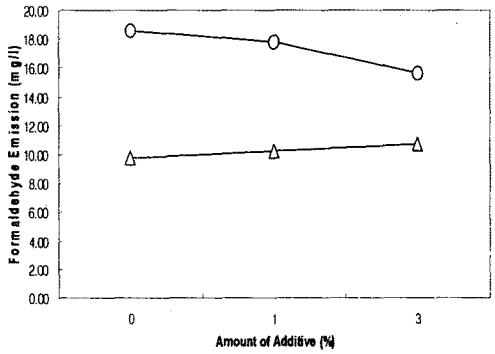

Fig. 9. Formaldehyde emission of plywoods prepared with UF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%: (O) without filler and (△) with 20% of flour filler.

affected by addition amount of inorganic compound, but when filler was added formaldehyde emission amount was reduced about 50% at 1% of inorganic compound and a little more formaldehyde emission amount was reduced at 3% of inorganic compound (Fig. 10). From the results of tensile strength and formaldehyde emission amount, it may be deduced that the resin type used was not a critical factor to affect both bond strength and formaldehyde emission, but the use of filler tended to a major factor to influence them. Hydroxyl group rich starch and amino group rich gluten components of filler may be reacted with some of formaldehyde existed in both UF resin and UMF resin, and then

hydroxyl group rich inorganic compound may be captured some of remainder formaldehyde. With these results, both maximizing tensile strength and reducing formaldehyde emission of plywood can be achieved from the optimum combination of filler, resin, and additives.

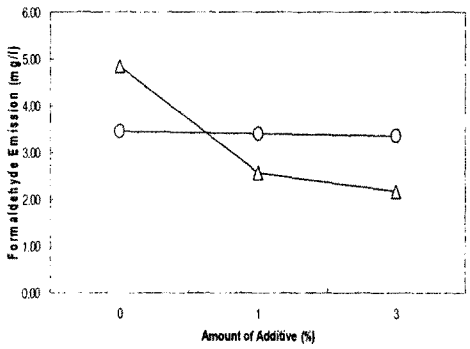

Fig. 10. Formaldehyde emission of plywoods prepared with UMF resin added silicone-rich inorganic compound at 0, 1 and 3 wt.%: (O) without filler and (△) with 20% of flour filler.

4. Conclusion

An adequate amount of inorganic compound did not affect physical properties of two amino resin binders. Therefore it could be used as an additive of amino resin binders for plywood. Cooperative interaction of filler and inorganic compound helped to reduce formaldehyde emission of plywood. Both improvement of tensile strength and reduction of formaldehyde emission of amino resin binders bonded plywood are possibly achievable at once by using adequately formulated adhesive mix.

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