Studies on Particle Board. (IX)

Suitability of species as raw material for particle board production. (No. 2)

Manufacture of particle board using larch wood extracts for particle bonding. (2)

By

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I. Introduction

The previous paper²⁾ suggested a new manufacturing method of particle board using the tannin-like constituents of SUDAJII particle or larch wood extracts, especially the arabogalactan, and paraformaldehyde as a means of replacing synthetic resin bonding agent. To increase the adhesive quality of these extracts, however, since it was necessary to apply high pressure during hot-pressing, the resultant board inevitably increased its density. Therefore, the authors tried to apply this process to the surface layer of a three-layer board, using urea resin only in inner layer.

In the case of SUDAJII particle, however, since the extracts required for adhesion were not sufficient, the resultant board did not show appropriate properties. In the case of larch wood particle, on the other hand, although the extracts required for adhesion were sufficient (the extracts of larch in Nagano prefecture was 8.87%), the water-proofing property was inferior as compared with other dry-strength properties.

The wood particle used in the experiment is 0.2 mm in thickness which means that the surface area of the particle is less than that of a granule or fiber. It seems in utilizing the extracts of such particles, that since the surface area of particle is so small, the possibility of reaction is little indeed, and therefore the water-proofing adhesive quality of the board becomes worse as mentioned above. According to P. NASHAN⁴⁾, resin can be formed by condensation between carbohydrate and formaldehyde. It is presumed, therefore, that if a chance of reaction is given, the water-proofing adhesive quality may also improve.

From this viewpoint, this study deals with the effects of zinc chloride as an active agent to provide a chance for a reaction, or as a catalyzer to accelerate the reaction between larch wood extracts and formaldehyde and to improve the water-proof adhesive quality of the resultant board. In addition, the effects of three factors, paraformaldehyde, zinc chloride and moisture content of particle on the adhesive quality of larch particle, or the qualities of the resultant board were studied. Morever, this study again deals with the possibility of using larch wood particle containing reactive extractions as a reactive agent for the surface layers of a three-layer board.

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II. Preliminary test on utilization of catalyzer

The previous paper²⁾ showed that the water-proofing quality of the board was somewhat improved by addition of 2.25% of ammonium (28% solid) as a catalyzer when adding formaldehyde to larch wood particle. This work was undertaken to determine the effects of zinc chloride as a catalyzer on acceleration of a reaction between larch wood extracts and formaldehyde, and on the water-proof adhesive quality of the resultant board.

1. Experimental procedure

(1) Preparation of specimens

Species of wood particles was heartwood of larch (*Larix Kaempferi* SARG., wood extracts 8.29%). The size of wood particle was $0.2mm \times 1 \sim 3 mm \times 20mm$.

(2) Curing condition

Zinc chloride contents used were 0%, 0.1%, 0.2%, 0.3% and 0.4% of absolute dry-wood weight of particles. Paraformaldehyde content used was 4% of absolute dry-wood weight of particles. Wood particle moisture content before pressing was 25%. As a control, urea resin (TD 5 II) content 11%. The hardener, catalyst 376, was used at a rate of 3% of urea resin. The temperature of hot press plates 140° C, and the pressing time 15 min. The pressure was applied in a step-down system of $35 \text{ kg/cm}^2 \rightarrow 20 \text{ kg/cm}^2 \rightarrow 10 \text{ kg/cm}^2$ every 5 min. The size of board was $30 \text{ cm} \times 30 \text{ cm} \times 5 \text{ mm}$. The specific gravity of board about 0.8. Three replications were used for each condition. In the case of the board using urea resin, wood particle moisture content before pressing was 15%, and the pressing time 5 min. The pressure was applied in a step-down system of 35 kg/cm^2 (2 min) $\rightarrow 20 \text{ kg/cm}^2$ (2 min) $\rightarrow 10 \text{ kg/cm}^2$ (1 min).

(3) Testing procedure

Sampling of specimens is shown in Fig. 1. All board specimens were tested after conditioning for three weeks at 20°C temperature and 65% relative humidity. Bending strength was tested by JIS A 5908 (1961) and YouNG'S modulus was measured simultaneously. Water absorption was tested by JIS A 5907 (1961) (water soaking at 25°C for 24 hours), and thickness swelling was measured at the center of specimen (size: 5×5 cm).



Fig. 1 Sampling of specimens.

2. Results

The properties of the board when using zinc chloride as a catalyzer for a reaction between larch wood extracts and formaldehyde during hot-pressing, are as shown in Fig. $2\sim$



Fig. 2 The effect of zinc chloride contents on bending properties.

Fig. 3 The effect of zinc chloride contents on water-proofing properties.

Table 1. The properties of mono-layer board using resin adhesive.

Specific gravity	Bending strength (kg/cm ²)	Young's modulus $\times 10^4 (\text{kg/cm}^2)$	Water absorption (%)	Thickness swelling (%)
0.72 (0.02)	462 (51)	5.9 (0.7)	38.5 (4.2)	15.1 (4.8)

Note: () Standard deviation.

3. The analysis of variance showed that bending strength, Younc's modulus, water absorption and thickness swelling in soaking between 0% and other various quantities of zinc chloride were significant at the 1% level of probability, but between 0.1 to 0.4% of zinc chloride were not significant among them at the 5% level of probability. Between them, however, all board properties showed an increasing tendency with increase of zinc chloride content. Table 1 shows the properties of mono-layer board using urea resin as a control. The board adding even 0.1% of zinc chloride had virtually the same properties as the board using urea resin. This means that the reaction between larch wood extracts and formaldehyde was accelerated by using zinc chloride catalyzer.

These results suggest that the water-proofing quality is exceedingly improved by adding a small amount of zinc chloride when hot-pressing the larch board adding formaldeyde.

III. The effects of paraformaldehyde and zinc chloride contents

Since the effect of zinc chloride as a catalyzer was recognized at the hot-pressing of larch heartwood particles adding formaldehyde in the preliminary test, this experiment was undertaken to determine quantitatively the effects of paraformaldehyde and zinc chloride in order to investigate optimum curing condition.

1. Experimental procedure

(1) Preparation of specimens

Species of wood particles was heartwood of larch from Hokkaido (wood extracts 5.66%). The size of wood particle was $0.2mm \times 1 \sim 3 mm \times 20 mm$.







Fig. 4 The effects of paraformaldehyde and zinc chloride contents on bending strength.



(2) Curing condition and testing procedure

Zinc chloride contents used were 0.1%, 0.2%, 0.3% and 0.4% of absolute dry-wood weight of particles and paraformaldehyde contents 0%, 1%, 2% and 4%. Wood moisture content before pressing was 25%. Other curing conditions, and testing procedure were the same as in preliminary test.

2. Results

(1) The effect of paraformaldeyde contents

The previous paper²⁾ showed that in the case of adding paraformaldehyde the effects of difference in paraformaldehyde contents among 1%, 2% and 4% on the board properties were not significant. In the present experiment, although zinc chloride as a catalyzer was added together, bending properties did not show any obvious difference between 0% and other various contents of paraformaldehyde (Fig. 4~ 6, Table 2), whereas water absorption and thickness swelling in soaking were reduced



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	Bending strength		Water a	bsorption	Thickness swelling	
	Sig.	ρ(%)	Sig.	ρ(%)	Sig.	ρ(%)
A. Zinc chloride content	**	50.4	**	42.7	**	48.4
B. Paraformaldehyde content	**	6.3	*	13.5	**	21.4
A×B		43.3		43.8	**	30.2
E						

Table 2. Analysis of variance (1).

Note: ** 1% confidence level, * 5% confidence level.

Young's modulus between paraformal dehyde contents or zinc chloride contents is not significant at the 5% level of probability.

at the 1% level of probability with increase of paraformaldehyde content, and water-proofing properties were improved. It seems to be considerably influenced by presence of zinc chloride, in other words, this means that even if paraformaldehyde is not added to the particles, water-proofing properties are somewhat increased by presence of zinc chloride. Besides, the value of bending properties of these boards was less than of larch from Nagano prefecture in the preliminary test. This seems to be evidently influenced by shortage of wood extracts of larch from Hokkaido.

(2) The effect of zinc chloride contents

Bending strength, water absorption and thickness swelling in soaking between zinc chloride contents were significant at the 1% level of probability, and each of the properties were increased with increase of zinc chloride contents. This was contrary to phenomenon in the preliminary test in that these properties between 0.1% and 0.4% of zinc chloride contents were not significant at the 5% level of probability. As mentioned above, it was quite characteristic that the effect of adding zinc chloride on the water-proofing properties was proved even in the case of 0% of paraformaldehyde, because the board disintegrated during water soaking when both paraformaldehyde and zinc chloride were not added to the particle²⁹. This means that some water unsoluble substance is formed in larch wood extracts during hot-pressing, by presence of zinc chloride instead of paraformaldehyde. But, since the present experiment shows that most stable properties are produced in the case of 2 to 4% of paraformaldehyde and 0.3 to 0.4% of zinc chloride, the effect of paraformaldehyde can not be overlooked.

IV. Manufacture of three-layer particle board

As mentioned above, to increase the self-bonding ability of larch wood extracts in wood particles and the chance of reaction between larch wood extracts and paraformaldehyde, it was necessary to apply high pressure during hot-pressing and to use thinner wood particles. Therefore, since the resultant board increased its density inevitably, the previous paper²³ suggested the new manufacturing method of three-layer board applying this process to surface layer and using urea resin only in inner layer. This work was undertaken to apply the effects of zinc chloride as a catalyzer, confirmed by above chapter, on the surface layer of three-layer board, and to investigated again the relationship between the curing conditions which are moisture content of particle during hot-pressing and board specific gravity, and the properties of three-layer board.

The reasonable surface moisture content in manufacturing three-layer board is about 25% generally for 10% of inner layer moisture content in the case of 1:2 in weighing ratio

of surface to core particle³⁾. When this process utilizing the wood extracts is applied to the surface of three-layer board, however, since the bonding reaction between larch wood extracts, especially arabogalactan, and formaldehyde is affected by the surface moisture content, the optimum moisture content in the surface layer could be changeable.

If the surface particles have excessive high moisture, the wood extracts at the surface layer would transfer to the inner layer with steam flow during hot-pressing of the particle mattress, and consequently the bonding ability of surface layer might be decreased, while the low moisture content of the surface layer would cause the reduction of the bonding reaction. Therefore, the effects of the moisture content in surface layer could be more severe for the self-bonding board than that for the board using urea resin.

As for the board specific gravity, in the case of three-layer board pressing, the inner layer of particles shows low compressibility and therefore the density of the surface layer increases¹⁰. If this process utilizing the wood extracts is applied to the surface layer, it seems possible to produce a three-layer board with lower density than that of a mono-layer board.

1. Experimental procedure

(1) Preparation of specimens

Species of wood particles were larch heartwood from Hokkaido for surface and sapwood of the same larch for core. The size of them were $0.2 \times 1 \sim 3 \times 20$ mm for the surface layer, $0.5 \times 3 \sim 6 \times 40$ mm for the core.

(2) Curing condition

Based on the result of the above chapter, paraformaldehyde and zinc chloride contents used for surface particles were 4% and 0.3% absolute dry-wood weight respectively. Wood particle moisture contents before pressing were 25% and 30% for surface particles. Urea



Code letter

- C: Bending properties
- D: Tensile strength perpendicular to surface
- G: Wood screw holding power
- EF: Water absorption & Thichness swelling

Fig. 7 Sampling of specimens.

resin (Plyamine TD 511.45 solids) content for core particles was 7 %. Hardener, catalyst 376, was used at rate of 3% of urea resin. Core particle moisture content before pressing was 10%. Weighing ratio of surface to core particles 1:2 in dry state. The size of board was $45 \times 45 \times 2$ cm. The specific gravities of board were about 0.65 and 0.7. The temperature of hot-press plates 140 °C, and the pressing time 15min. The pressures were applied in a step-down system such as $25 \text{ kg/cm}^2 \rightarrow 15 \text{ kg/cm}^2 \rightarrow$ 5kg/cm² every 5 min for 0.65 board specific gravity and $30 \text{kg/cm}^2 \rightarrow 20 \text{ kg/cm}^2 \rightarrow$ 10kg/cm² every 5min for 0.7 board specific gravity. Two replications were used for each condition.

(3) Testing procedure

Sampling of specimens is shown in Fig. 7. All board specimens were tested

after conditioning for three weeks at 20°C temperature and 65% relative humidity. Bending properties, tensile strength perpendicular to surface and wood screw holding power were tested by JIS A 5908 (1961), and water resistant properties were tested by JIS A 5907 (1961).

2. Results

(1) The effect of moisture content in surface layer

From the view point of plasticization of surface layer, high moisture content in the surface layer is effective in manufacture of a three-layer board, as described in the author's previous paper¹⁾³⁾. In this experiment surface smoothness of board was improved at 30% of surface particle moisture content as compared with 20%. As shown in the result of the analysis of variance (Table 3), however, all the properties, except the bending strength, between 25% and 30% of surface moisture content were not significant at the 5% level of probability. Especially, the bending strength was significantly decreased at the 1% level of probability with increase of the surface moisture content from 20% to 30% (Fig. 8). The

Table 3. Analysis of variance (2).	e (variance	of	Analysis	3.	Table
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	Bending strength		Young's modulus		Tensile strength		Wood screw holding power	
	Sig.	ρ(%)	Sig.	ρ(%)	Sig.	ρ(%)	Sig.	ρ(%)
A. Specific gravity	**	43.0	**	58.9	**	56.5]]
B. Moisture content	**	25.6		41.1		44.5	*	16.5
$A \times B$		31.4)	*)		
E	l							

Note: ** 1% confidence level, * 5% confidence level

Water-proofing properties between specific gravities or moisture contents are not significant at the 5% level of probability.







Fig. 9 The effects of surface particle moisture contents and board specific gravity on YOUNG'S modulus in bending.



Fig. 10 The effects of surface particle moisture contents and board specific gravity on tensile strength perpendicular to surface.











Fig. 13. The effects of surface particle moisture contents and board specific graviy on thickness swelling.

other properties also somewhat tended to reduce with increase of surface moisture content (Fig. $9\sim13$). It seams that the effect of excessive moisture in surface layer during hotpressing on squeezing of the reactive materials from the surface to inner layer is more significant than that on the plasticization of surface layer.

- (2) The effect of board specific gravity
- The results of the analysis of variance showed that bending strength, Yound's modulus

and tensile strength perpendicular to surface were increased at the 1% level of probability with increase of board specific gravity from 0.65 to 0.7. Other properties, except water absorption, also tended to increase with increase of board specific gravity. Therefore, the plasticization of surface which is characteristic of three-layer board and also self-bonding of the wood extracts seemed to be accelerated by high pressure during hot-pressing. However, since the tensile strength perpendicular to surface was increased with increase of board specific gravity, it might be considered that the general improvement of the properties with increase of board specific gravity is also included.

In any case, it was recognized that rather high specific gravity should be required even in three-layer board in order to improve the condition of self-bonding.

(3) Comparison with the three-layer board using urea resin adhesive

Comparing the three-layer board having self-bonding surface layer with that of only urea resin adhesive (Table 4), the board properties, especially water-proofing properties, in this experiment are apparently inferior to those of urea resin bonded board. This seems to be evidently influenced by shortage of wood extracts of larch from Hokkaido.

	Surface moisture content				
Items	25%	30%			
Specific gravity	0.65(0.02)*	0.66(0.02)			
Moisture content (%)	9.1 (0.2)	9.1 (0.2)			
Bending strength (kg/cm ²)	368 (21)	298 (22)			
Young's modulus $\times 10^4$ (kg/cm ²)	4.6 (0.1)	4.2 (0.1)			
Tensile strength** (kg/cm ²)	3.3 (0.6)	3.7 (0.5)			
Wood screw holding power (kg)	58.9 (8.0)	42.8 (7.9)			
Water absorption (%)	50.0 (1.8)	50.4 (0.8)			
Thickness swelling (%)	13.2 (0.2)	12.9 (1.7)			

Table 4. The properties of three-layer board using urea resin adhesive³⁾.

Note : * standard deviation, ** perpendicular to surface.

Manufacturing variables are the same as those in this study, except adhesive for surface particles. Urea resin (Plyamine TD 511, 45% solids) content for surface is 11%.

V. Conclusion

By using larch heartwood from Honshu island (containing 8 to 10% of wood extracts) for surface particles (mixing 4% of paraformaldehyde, 0.3 to 0.4% of zinc chloride) and urea resin adhesive for inner layer, a new manufacturing method of three-layer board utilizing larch wood extracts could be established. However, it is inevitably recongnized as a fault in this type of board that the quantity of larch wood extracts is changed by growing district, and this makes the board properties unstable.

VI. Summary

The cost of particle board is strongly influenced by the cost of adhesive, which frequently is the biggest single item in the total production cost. To reduce this production cost, although wood species is restricted, the manufacturing method of particle board using larch wood extracts as a means of replacing synthetic resin bonding agent has been investigated in an earlier paper. However, since the optimum condition of reaction between larch wood extracts and formaldehyde has not been found, water-proofing adhesive quality was extremely inferior. Therefore, the present study deals with improvement of reactive condition using zinc chloride as a catalyzer in the above-mentioned manufacturing method, and establishment of new manufacturing method of three-layer particle board applying this method only to surface layer.

The results obtained in this study are summarized as follows:

(1) The measurement of the cold water wood extracts showed the quantity of larch wood extracts from Hokkaido was lower than that from Honshu (Ex. Akita and Nagano prefecture). This means that the quantity of wood extracts was changeable in accordance with habitat.

(2) The preliminary test on the effect of adding zinc chloride as a catalyzer showed that bending properties as well as water-proofing adhesive qualities were so improved by small amount of zinc chloride that they almost corresponded to that of the board using urea resin.

(3) The effects of paraformaldehyde and zinc chloride on the board properties using larch heartwood from Hokkaido were studied and consequently it was found that the addition of 4% of paraformaldehyde and $0.3 \sim 0.4\%$ of zinc chloride gave appropriate properties to the resultant board. Compared with the result of the preliminary test using larch from Honshu island mentioned above, however, the properties of the board using larch from Hokkaido were somewhat inferior. The effect of difference in wood extracts was definitely recognized.

(4) Based on the optimum curing condition suggested above, the properties of threelayer board utilizing the wood extracts in surface particles were studied and consequently the following curing conditions required for this method were recognized: paraformaldehyde content 4%, zinc chloride content 0.3%, board specific gravity 0.7 and moisture content of surface particles 25%. Compared with the three-layer board using urea resin, however, the board properties were evidently inferior. This seems to be apparently influenced by shortage of wood extracts. Therefore, it is necessary for the raw material of this method to restrict the habitat of larch wood.

(5) Consequently, by using larch heartwood from Honshu island to surface layer and adding appropriate quantities of paraformaldehyde and zinc chloride, it was found that a new method of three-layer board using wood extracts could be established.

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パーティクルボードに関する研究(IX) 原料樹種に関する研究(第2報)

カラマツの成分利用によるパーティクル

ボードの製造(2)

岩下 睦"

概 要

パーティクルボード製造における原価のうち,接着剤の占める比率は比較的高い。この接着剤コストを さげるために特定樹種ではあるが,カラマツ水溶性成分を利用し,パラホルムアルデヒドを添加する成板 方法を前報において報告したが,反応最適条件が得られなかったため,耐水接着性がきわめて低かった。 そこで,この研究は上記の成分利用に際して,触媒として塩化亜鉛を用い,接着反応条件の改善方法の検 討を行ない,あわせて,表層にのみこの考え方を適用した新しいタイプの3層ボードの製造法を確立しよ うとしたものである。得られた結果を要約すると次のごとくである。

(1) 冷水抽出法によって水溶性成分量を検討した結果,北海道産カラマツは成分量において,内地産 カラマツ(秋田県,長野県)に比較して,低い値が得られ,水溶性成分量は立地条件により差異があるこ とが認められた。

(2) 長野県産カラマツを用い,予備的に触媒としての塩化亜鉛添加の効果を検討した結果,ごく少量 の添加で,成板されたボードの曲げ性能はもちろん,耐水接着性はユリア樹脂使用のボードの材質と比較 して大差なく,むしろそれ以上に向上したほど,きわめて顕著に改善された。

(3) 次に北海道産カラマツ心材部を用い,パラホルムアルデヒドおよび塩化亜鉛添加量を検討した結果,パラホルムアルデヒド4%,塩化亜鉛 0.3~0.4%程度添加した場合に良好な材質が得られることが 認められた。しかしながら,上記内地産カラマツを使用した予備試験の結果に比較して,いくぶんボード 材質がおとっており,水溶性成分量の差が明らかに認められた。

(4) 以上得られた最適成板条件をもとにして,表層成分利用による3層ボードの材質を検討した結果,パラホルムアルデヒド添加量4%,塩化亜鉛添加量0.3%,ボード比重を高く(0.7),表層小片含水 率をある程度低く(25%)する必要が認められた。しかしながら,ユリア樹脂使用の3層ボードと比較す ると,材質は明らかに低下しており,水溶性成分量の不足に原因されるものと思われ,カラマツの立地条 件を限定する必要性が認められた。

(5) 以上の結果,表層小片に内地産のカラマツ心材を利用し,適当量のパラホルムアルデヒドと塩化 亜鉛を添加することにより,新しいタイプの成分利用による3層ボード製造の可能性が得られた。

(1) 木材部材質改良科材質改良研究室長·農学博士