MANUFACTURE OF UREA - MELAMINE - FORMALDEHYDE GLUE USED FOR LAMINATED VENEER LUMBER PRODUCTION

Tran Van Chu[1], Trinh Hien Mai[2], Ly Tuan Truong[3]
[1,2,3]*Vietnam National University of Forestry*

SUMMARY

In this study, glue Urea - Melamine- Formaldehyde (UMF) was created to produce laminated veneer lumber (LVL), then the quality of LVL produced from Urea-Formaldehyde glue (UF) of Gia Han Company was compared to that of LVL produced from UMF made in Laboratory of Wood Industry College, Vietnam National University of Forestry (VNUF). The research results show that: The technology of producing UMF glue and LVL is suitable with current production condition and can be well applied in Vietnam. This technological process has been tested and applied in production at some factories and enterprises in Vietnam. The UMF glue produced in this study was a good adhesive and met the requirements of a glue used for high-quality LVL production. The quality criteria of the UMF glue were much better than those of UF glue from Gia Han Company. Some quality criteria of LVL produced from the UMF reached the requirements of the standards CAN3 - 0437 - M85 and JAS S-11-15.2. The LVL produced from the UMF glue can be used for furniture and construction.

Keywords: Fast growing plantation wood, Laminated Veneer Lumber, Urea - Formaldehyde glue, Urea - Melamine - Formaldehyde glue.

I. INTRODUCTION

In recent years, the forest products processing industry in Vietnam has been growing very well, confirming very important position and role of the sector in the development of the national economy. One of the reasons for this success is the right policy in science and technology to promote the processing of forest products, including the technology of wood-based panel production.

Among the kinds of wood-based panels, LVL possesses a lot of valuable properties and is widely used to replace timber in construction, such as girders, beams, door frames, window or bending details. LVL can improve some disadvantages of natural wood such as more uniform in properties, better weather resistance, large-sized details overcoming the limitations of tree's diameter and height (especially for plantation timber).

LVL's structure is similar as that of plywood, however LVL is thicker than plywood and the veneer grains are arranged parallel. There are 3 methods to create LVL: step by step pressing, once step pressing and high frequency pressing. All of those methods to produce LVL are very strict in pressing parameters and especially adhesives and additives.

This happens as LVL has great thickness and includes many parallel veneers. Heat is difficult to transmit into the core layer (veneer), this affects the bonding quality of LVL product. The glue should have high solid content, high viscosity, permeability into veneers, good spreading, high elasticity, etc. Most of the UF glues used for plywood and particle board in Vietnam market is not suitable to produce high quality LVL.

Therefore, the study to create an UF glue with appropriate quality criteria for LVL production from plantation wood, in technological condition of Vietnam is a very essential and meaningful task. There have been many solutions to enhance the quality of the glue. One of these solutions is modification of UF glue by different chemicals, such as phenol, melamine, epoxy, etc.

This article presents the modification of UF glue by melamine to create UMF glue used for

LVL production in technological condition of Vietnam.

II. MATERIALS AND METHODS

2.1. Raw materials used in the experiments

2.1.1. Round wood

Wood used in the experiments was *Styrax tonkinensis*. Pierre at age of 7-8 years and harvested from Doan Hung District, Phu Tho Province. *Styrax tonkinensis*. Pierre is a very flammable wood, pinkish white, heart wood and sap wood are differentiable, timber vessels are scattered single or dual, diameter of timber vessels varies from 100-200 µm, parenchyma cells arrange along the trunk, uniform arranged rays are accounted for 10-30% of the timber volume, there is no texture layer and glue tube, cellulose content is 47-49%, lignin content is 22.3%. The wood basic density is 0.38-0.41g /cm^3, shrinkage coefficient: 0.29, parallel compression strength: 192.105N /m^2, static bending strength: 505.105N /m^2, low natural durability, easy processing and easy cracking.

The above mechanical, physical and chemical properties of *Styrax tonkinensis*. Pierre show that the wood can fully meet the requirements of raw material used for LVL production technology.

2.1.2. Chemicals

a. **Urea (H_2N-CO-NH_2):** Urea was produced in Duc Giang Chemical Factory. Some basic properties: crystal, colorless, soluble in water and oil, hygroscopic, molecular mass: 60; density: 1.335 g/cm^3, water soluble at 20^oC: 104.7g/100g H_2O, melting temperature: 132^oC. Content of substances in Urea: Cl 0.0003%, SO_4 0.001%, NH_3 0.005%, Fe 0.0002%, Pb 0.0002%.

b. **Melanine ($C_3H_6N_6$):** Melamine is an organic base, low water soluble, chemical formula is $C_3H_6N_6$, white powder. Other names: 2,4,6-Triamino-s-triazine, Cyanurotriamide, Cyanuramide. Melamine used in this study was produced by Chengdu Yulong Chemical Co. Ltd, China.

c. **Formaldehyde (H-CHO).** Formaldehyde was produced in Duc Giang Chemical Factory. Some basic properties: Liquid, colorless, molecular mass: 60; density: 1:05 g/cm^3. Content of other substances in Formaldehyde: Cl 0.0002%, SO_4 0.001%, Fe 0.0002%, Pb 0.0002%.

d. **Caustic soda (NaOH):** NaOH was produced in Duc Giang Chemical Factory. Some basic properties: crystalline, white, soluble in water, molecular weight: 40; density: 2.13 g/cm^3 melting point: 321^oC. Content of other substances in Caustic soda: Na_2CO_3<0.005%, SO_4 0.005%, Cl 0.005%, N 0.001%, PO_4 0.001%, SiO_3 0.01%, Fe 0.003%, Ca 0.01%, K 0.05%.

d. **Amoniclorua (NH_4Cl):** NH_4Cl was produced in Duc Giang Chemical Factory. Some basic properties as follows: crystalline, white, soluble in water.

g. **Glue used for control LVL:** The glue used in the experiments for control LVL production was Urea-formaldehyde (UF) of Gia Han Company, Taiwan. This is one of the glues used widely in many Southeast Asian countries such as Malaysia, Indonesia, Thailand. In Vietnam, glues of Gia Han Company have been used extensively in factories of plywood, LVL and particleboard production.

2.2. Experimental methods

2.2.1. Prescription and process of making UMF glue

a. Prescription of making UMF glue

Urea 98%: 350 pbw; Melamine 99%: 10 pbw; Formaldehyde 37%: 700 pbw; NaOH 10%: a sufficient amount; NaCl 20%: a sufficient amount.

b. Process of making UMF glue

Use a technical balance with precision of 0.01g to weight the chemicals: urea, melamine and formaldehyde. Pour the amount of formaldehyde in the glue pot. Use NaOH 10%

to adjust the pH of formaldehyde to 7.5-8.5. Pour 2/3 amount of urea (first time) and entire amount of melamine into the glue pot. Stir carefully until urea and melamine were well dissolved. Increase the temperature of the solution up to 90+/-1^{o}C in 40-50 minutes, simultaneously maintain this temperature for about 30 minutes. NH_4Cl solution was used to adjust pH of the solution to 4.8-5.0 until the end of the reaction.

When the reaction ended, immediately use NaOH to neutralize, adjust the pH to 7.0-7.5, then pour 1/3 amount of urea (second time) and stir well, then cool the solution. When the temperature in the glue pot was about 40^{0}C, finish the cooking process.

2.2.2. Quality of the experiment boards

LVL thickness: 35mm; number of veneers: 15; veneer thickness: 3.6mm; veneer moisture content after drying: 10%; veneer moisture content after glue spreading: 20%. Quality of LVL were tested as the standards in table 01.

Table 01. Testing standards of LVL

N_0	Criteria	Sign	Unit	Testing standard
1	Thickness swelling	TS	%	CAN3 – 0437 – M85
2	Shear strength	σ_T	MPa	JAS S-11-15.2
3	Modulus of rupture	MOE	MPa	CAN3 – 0437 – M85

2.2.3. Process of producing LVL

In this study, the process of producing LVL is presented in figure 01:

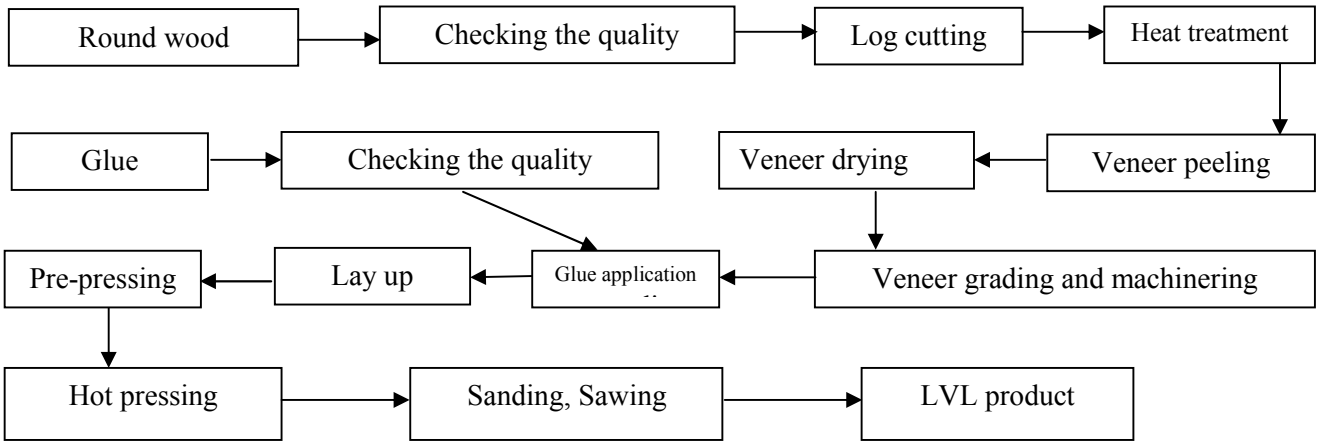

Figure 01. Technical process of producing LVL

After harvesting, *Styrax tonkinensis*. Pierre wood with diameter of 20-25cm were debarked and cut into logs with a length of 1.35m. Then the wood was boiled in water (heat treatment) before peeling. The purpose of heat treatment was to soften the wood before peeling and remove some of the wood extractives. Parameters of the heat treatment: Temperature: 60-70^{o}C; Time: 9-10 hours; cooling to the environment temperature: 2 hours.

After the heat treatment, the logs were peeled to create veneers with thickness of 3.6 mm. Here were the parameters of peeling lathe: sharpness angle (β): 20^{o}; clearance angle (α): 3^{o}; vertical distance (h) between knife edge and spindle axis: +1mm; speed of chuck spindle: 150 r/min; speed of knife movement (peeling speed): 5.8 mm/s; vertical distance (h_o) between nose bar and knife edge: 0.3 mm; compression rate (Δ): 20%.

After peeling, the wet veneers were dried by a roller kiln (with air circulation along the axis of the kiln. Required moisture content of the veneers after drying was 10%, moisture content of the veneers after glue spreading was 20%. The parameters of veneer drying: inlet temperature: T = 60 - 70^{o}C; outlet temperature: T = 110 - 120^{o}C; roller speed: 0.75 m/s; air circulation speed: V = 3m/s. Veneer quality after drying as follows: depth of checks: 49.52-60.11%; frequency of checks: 4.02 – 5.13 marks/cm; thickness variation: 1.77 - 2.46%; recovery rate of veneer: 55 - 62%. Color of the veneers after drying was similar as that of the natural timber. The veneers with such quality criteria met entirely the requirements of materials used for LVL and plywood.

The UMF glue was stabilized and tested quality before using. Then the veneers were spreaded by UMF glue and layed up. UMF spreading amount: 180 – 220 g/m^2. Number of veneer was 15 for each product. The veneers was layed up with parallel grain direction and the principle: right side-right side. Then the veneer pack was hot pressed by following parameters: temperature 140^{o}C, pressing pressure: 1.5 MPa; pressing time: 65 minutes (1,2 min/mm).

2.2.3. Methods of testing

The quality of glue was tested according to the standard GB/T4897-77. The contact angle of glue was measured by measurement device JJC-L of Polymer Chemistry Laboratory in Hanoi University of Technology. The pH was measured by pH meter (HI 9224 Microprocesser printing pH meter). The viscosity of the glue was measured by a viscometer (Rion Viscoteter VT-04).

Veneer's pH was measured as place 3g of wood power in a 50 ml glass cup (including 30ml distilled water) at room temperature. Use a glass rod to stir wood powder for 5 minutes. Let stand for 10 minutes and continue stirring for 5 minutes and let stand for 5 minutes again. Then use a pH meter to measure the pH value of the veneer.

Resistance to acid of veneer was measured as place 25 g wood power in a triangle glass bottle at room temperature. Pour 250 ml of distilled water in the bottle and shake well for 20 minutes. After cooling to room temperature, pour the solution into the filtering device. Add 2 cups of glass (350 ml) each contained 50ml purification water. Calibrate with a solution of 0.025 mol/l sulfuric acid to pH = 3. Number of sulfuric acid mol used is called resistance to acids of the veneer.

Some quality criteria of LVL were tested on a physical and mechanical testing machine, at the Laboratory of Wood Industry College, Vietnam National University of Forestry (VNUF).

III. RESULTS AND DISCUSSION

3.1. Technical parameters of glue

- Glue UF used for control LVL: The technical parameters of the UF glue through the tests as follows: liquid; milky; solid content: 47%; density 1.25-1.27 g/ml; viscosity: 100-180 mPa.s (at 30^{o}C) (Rion Viscoteter VT-04) as standard GB/T 14074.7-93 [12]; pH: 7.0-7.2 (at 20^{0}C); gel time: 67s (at 100^{o}C); free formaldehyde: less than 1.0%; storage time at 30^{0}C: 2 months.

- Glue UMF: The technical parameters of the UMF glue through the tests as follows: liquid, light yellow; solid content: 50+/-1%; density: 1.26-1.28g/ml; viscosity: 132-195 Pa.s (at 20^{o}C); pH 7.0-8.2; gel time: 65s; curing time: 90s (when use 1% hardener NH_4Cl); lifetime: 4-6h; storage time: 2.5 months; free formaldehyde: less than 1.0%.

The above results showed that: the UF glue in milky or cream liquid, the UMF in light yellow liquid. Therefor color of the LVL would be very nice. Dry content of UMF glue (50+/-1%) was higher than the UF glue (47%),

hence the polymersation ability of the UMF glue was higher than that of the UF glue. These glues had almost equal viscosity, however, the contact angle of the UMF glue was higher.

The lifetime of the UF glue was lower than that of UMF glue. Curing time of the UMF glue was higher than the UF glue with the same amount of hardener. Meanwhile, gel time of the UMF glue was lower than that of the UF glue. With the gel time of the UF glue was 67s, and the curing time was 80s, the heat transfer rate of *Styrax tonkinensis*. Pierre wood was low, and LVL thickness was thick (35 mm), it was very difficult to cure the glue UF thoroughly. Meanwhile, the gel time of the UMF glue was 65s, curing time was 90s, the glue UF could be cured more thoroughly.

Formaldehyde free content of both glues was less than 1.0%, so the toxic levels of both glues reach the requirement.

Test results of the contact angle between two glues and *Styrax tonkinensis*. Pierre wood according time is presented in table 02.

Table 02. The contact angle between two glues and Styrax tonkinensis. Pierre wood

Contact angle (^{o})	Time (min)					
	1	**2**	**4**	**6**	**8**	**10**
UF glue	35	33	30	26	24	22
UMF glue	42	40	37	34	31	27

The UMF glue caused higher contact angle, therefore the spreading ability of the glue was lower, permeability and bonding quality were reduced. Hence, one step hot pressing method is often applied to produce LVL with these kinds of glue. If many step hot pressing method is used, the LVL can be bloomed incase glue has high contact angle.

3.2. Moisture content, pH and acid resistance of *Styrax tonkinensis*. Pierre veneer

Moisture content, pH and acid resistance of *Styrax tonkinensis*. Pierre veneer are presented in table 03.

Table 03. Moisture content, pH and acid resistance of Styrax tonkinensis. Pierre veneer

Veneer color	Moisture content (%)	pH	Acid resistance
Green brown	11.23	7.2-7.6	1.09
Red brown	8.91	7.1-7.3	1.01
Yellow white	11.64	5.6 - 6.1	0.29

The results in table 03 shows that: There were significant differences in moisture contents, pH, and acid resistance of veneers in different colors. The yellow white veneers were mostly from sapwood, hence their acid resistance was less than that of the green brown and red brown veneers mostly from heartwood. The bonding quality between glue and heartwood was low due to common UF glue is hardened in an acid environment while heartwood was largely neutral and slightly alkaline. With 1% NH_4Cl, pH value of the UF glue remains in the neutral limits, so if we do not use high pressing temperature, the hardening of the UF glue is difficult. Meanwhile, the UMF glue with 1% NH_4Cl was acidic, so the glue could be hardened at low pressing temperatures.

There are many solutions to treat the veneers before gluing spreading, however, the price of LVL product could be increased. Therefore, the use of modified UMF glue

would bring high efficiency for LVL.

3.3. LVL properties

Some properties of LVL are presented in table 04.

Table 04. Some properties of LVL

Type of product	TS, %			Tensile strength σ_T, MPa			MOR, MPa		
	Min	Max	TB	Min	Max	TB	Min	Max	TB
Control LVL – UF glue	2.68	3.95	3.31	0.126	0.137	0.131	97.62	99.71	98.66
LVL – UMF glue	2.09	3.24	2.66	0.143	0.187	0.165	99.54	101.45	100.49

The results in table 04 shows that: The value of MOR, tensile strength of the control LVL with the UF glue (98.66 MPa and 0.131 MPa) were lower than those of the LVL produced from the UMF glue (100.49 MPa and 0.165 MPa). In particular, thickness swelling of the control LVL was much higher than that of the LVL with the UMF glue (3.31% and 2.66%).

This reflects that, with the same wood and technological parameters to produce LVL, quality of LVL made from the UMF glue was better than that of LVL made from the UF glue and met the requirements of LVL type I used for construction. Meanwhile, LVL made from the UF glue only met the requirements of LVL type II used for construction.

This is can be explained as follows: UF glue is a thermosetting resin with network structure molecular. The UF molecules contain many OH groups, affinity for water. In the hardened glue still exist some original - free CH_2OH which do not react, so the glue still is hygroscopic and can absorb water. When air humidity changes, the hardened glue layer absorbs water in the air or emits water into the air, hence the volume of the hardened glue layer changes, creates residual stresses and cracks in the hardened glue layer.

When melamine molecular was combined into molecular of UF glue, a new kind of glue was formed and has wider network structure and more closely molecular structure. Therefore, the LVL produced from the UMF glue displayed better quality than the LVL produced from the UF glue.

IV. CONCLUSIONS

The UMF glue possesses valuable properties, namely: simply process of creating the UMF glue, good bonding quality, nice color, plastic glue membrane, low cost, meet the requirements of a glue used for LVL. This UMF glue has overcome disadvantages of common UF glues used for resin wood, and is suitable for producing LVL from plantation wood and other kinds of wood-based panels. The technical parameters of the UMF glue were better than those of UF glue from Gia Han company. Technology of making UMF glue and producing LVL is suitable with practical condition and can be applied well in Vietnam. This technological process has been tested and applied in production at some factories in Vietnam. The created UMF glue is a good adhesive, meets the requirements of an adhesive used for producing high-quality LVL. Some properties of LVL produced from the UMF reached the requirements of the standards CAN3 - 0437 - M85 and JAS S-11-15.2. The LVL produced from the UMF glue can be used for furniture and construction.

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NGHIÊN CỨU TẠO KEO UREA – MELAMINE – FORMALDEHYDE DÙNG TRONG SẢN XUẤT VÁN LAMINATED VENER LUMBER (LVL)

Trần Văn Chứ[1], Trịnh Hiền Mai[2], Lý Tuấn Trường[3]

[1,2,3]Trường Đại học Lâm nghiệp

TÓM TẮT

Mục đích của bài viết này là nghiên cứu, tạo ra keo Urea - Melamine- Formaldehyde (UMF) dùng trong công nghệ sản xuất ván LVL (Laminated vevneer lumber) và so sánh về chất lượng của ván LVL tạo ra từ keo Urea – Formaldehyde (U-F) thông dụng của Hãng Gia Hân và keo Urea – Melamine - Formaldehyde (UMF) được tạo ra tại Phòng Thí nghiệm Viện Công nghiệp gỗ - Trường Đại học Lâm nghiệp. Kết quả nghiên cứu cho thấy: Công nghệ tạo keo dán UMF và công nghệ tạo ván LVL hoàn toàn phù hợp với điều kiện về công nghệ và áp dụng tốt vào thực tế sản xuất của Việt Nam. Quy trình công nghệ này đã được thử nghiệm và áp dụng vào sản xuất tại một số nhà máy, xí nghiệp của Việt Nam; Keo UMF được tạo ra là chất kết dính tốt, đáp ứng tốt yêu cầu và rất phù hợp của keo dán dùng cho công nghệ sản xuất ván LVL chất lượng cao. Các chỉ tiêu chất lượng của keo UMF tốt hơn hẳn so với keo U-F của Hãng Gia Hân dùng trong ván LVL đối chứng; Một số chỉ tiêu chất lượng của ván LVL từ keo UMF đáp ứng tốt yêu cầu của tiêu chuẩn CAN3 – 0437 – M85 và JAS S-11-15.2. Ván LVL từ keo UMF có thể làm đồ mộc và xây dựng.

Từ khóa: Gỗ mọc nhanh rừng trồng, keo Urea - Formaldehyde, keo Urea - Melamine - Formaldehyde, ván Laminated Veneer Lumber.

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