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Synthesis and Properties of Melamine Modified Urea Formaldehyde Resin for Impregnation Under New Process

Zhiyong Zheng[1, 2], Huaihao Chen[3], Kaijie Zheng[1], Qi Li[3, *], An Mao[3, *]

[1]Lanling Jiesen Decoration Materials Co., Ltd, Linyi, China

[2]Shandong Yisen Meiju New Material Technology Co., Ltd, Linyi, China

[3]College of Forestry, Shandong Agricultural University, Taian, China

Email address:

wonderfulliqi2364@126.com (Qi Li), dannymaoan@126.com (An Mao)

[*]Corresponding author

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Abstract: In this paper, the basic properties of UF and UMF resins for impregnation under conventional and new processes were compared, and their chemical structures were characterized by ^{13}C NMR. The effects of synthetic process and melamine addition on the bonding strength and formaldehyde emission of UF and UMF resins were studied. The resin synthesized by the new process has higher linear and methylene ether bond content, relatively lower methylene content, longer curing time and storage life and lower free formaldehyde content than the conventional resin; When melamine was added at the initial stage of UF resin synthesis reaction, it did not significantly change the total amount of methylene ether and hydroxymethyl in the resin system, but the content of class I hydroxymethyl increased and the content of class II hydroxymethyl decreased. The reason may be that the added melamine and urea react not only with the remaining formaldehyde in the system, but also with the formaldehyde removed from hydroxymethyl due to dehydroxylation reaction, Hydroxymethylmelamine and monobasic substituted urea are produced. At this time, about 50% of melamine still exists in UMF resin in the form of free or low hydroxymethylate, and does not participate in the formation of the main structure of the resin. Melamine was added in the early stage of the reaction. The modified resin bonded plywood has high bonding strength and low formaldehyde emission.

Keywords: Melamine Modified Urea Formaldehyde Resin, Impregnation, New Process, Synthesis

1. Introduction

The wood-based panel industry has developed rapidly. The adhesive used is mainly UF resin, accounting for more than 90% of the total amount of wood adhesive [1]. It is mainly used for bonding plywood, fiberboard and particleboard. However, UF resin bonded wood-based panel has the problems of formaldehyde emission and poor waterproof. California Act implemented in 2009 (CARB) imposes stricter restrictions on the formaldehyde emission of wood-based panels [2]. Reducing the formaldehyde emission of wood-based panels, improving the wet strength and maintaining the cost advantage has always been an important direction and main goal of UF resin research [3-5]. At present, although reducing the molar ratio of formaldehyde to urea can effectively reduce the formaldehyde emission of its bonded products, the curing speed of low molar ratio resin Slow and poor water resistance, which affects the quality grade and production efficiency of products. Therefore, it is necessary to improve the properties of low molar ratio resin by improving the synthesis process, adding modifier or changing the type of curing agent [6-8].

One of the methods to improve the properties of UF resin is to optimize the resin structure. The basic framework of the classical theory of UF resin synthesis considers that the synthesis of UF resin can be divided into addition reaction of urea and formaldehyde under weak alkaline conditions and polycondensation under weak acidic conditions. Subsequently, other synthetic processes began to be studied. For example, in the UF resin synthesis process under strong acid conditions, urea reacts with formaldehyde to form a large number of cyclic methylene ether bonds (urons), which makes the resin have some special properties [9, 10].

Methylene ether bond is easy to decompose and release formaldehyde during hot pressing curing, so ether bond may be one of the main reasons for formaldehyde release from wood-based panel [11]. But so far, there is no systematic literature on ether bonds (including linear and cyclic methylene ether bonds) on the properties of resin and bonded wood-based panel. Therefore, studying the role of methylene ether bonds in resin structure has important practical significance for a more comprehensive understanding of formaldehyde emission sources, optimizing resin structure and formula, reducing formaldehyde emission from wood-based panel and improving physical and chemical properties of wood-based panel. Another way to improve resin properties is to add modifiers, such as introducing a small amount of melamine into UF system to synthesize UMF polycondensation resin [12, 13].

In the previous study, a new urea formaldehyde resin with low molar ratio was synthesized by alkali strong acid alkali weak acid alkali process. At the same time, melamine modified urea formaldehyde resin was synthesized by combining this process with melamine addition, so that the conventional melamine modified urea formaldehyde resin has unique structure and properties under the new synthesis process. NMR analysis confirmed that the resin under this process contains more methylene ether bonds. In urea formaldehyde resin, these ether bonds mainly exist between urea components, while in melamine modified urea formaldehyde resin, ether bonds exist not only between urea components, but also between cyanuric amine, urea components and melamine components. This makes this kind of resin have some special properties, such as longer curing time and lower free formaldehyde content than conventional resin. In view of the above ether bond characteristics and melamine modification, the author intends to combine the two to explore the structure and performance characteristics of UMF resin under the new synthesis process.

2. Materials and Methods

2.1. Materials

Formaldehyde, 37%, Lanling Jason decoration materials Co., Ltd; Urea, 99%, Tianjin Zhiyuan Chemical Reagent Co., Ltd; Melamine, analytical purity, Tianjin Zhiyuan Chemical Reagent Co., Ltd; Sodium hydroxide, analytical purity, Tianjin Zhiyuan Chemical Reagent Co., Ltd; Sulfuric acid, analytical purity, Tianjin Zhiyuan Chemical Reagent Co., Ltd.

2.2. Instrument

BYK Gardner bubble viscometer, BYK additives & instruments, Germany; Varian inova-400 nuclear magnetic resonance instrument, Varian company of the United States; Hanna hi9124 pH tester, Hanna instruments, Italy; BY302×2/15 universal test press, Suzhou XINXIELI Machine Manufacturing Co., Ltd; CMT4104 microcomputer controlled electronic universal mechanical testing machine, Meister industrial system (China) Co., Ltd.

2.3. Synthesis of UF Resin and Modified Resin

2.3.1. Conventional UF Resin Synthesis

UF resin was synthesized by alkali acid alkali conventional process. First, add all formaldehyde into a three port flask, adjust the pH value to 8.0 with sodium hydroxide solution, and raise the temperature to 70°C, Add the first batch of urea (at this time, the molar ratio is 2.0), continue to raise the temperature to 90°C, keep the temperature for 45 min, add sulfuric acid solution, adjust the pH value to 4.75, start the polycondensation reaction, when the viscosity is O-P, adjust the pH value to 7.8, add the second batch of urea (reaching the final molar ratio of 1.01), stir and lower the temperature and discharge the material. The UF resin synthesized under this process is marked as UF.

2.3.2. New Process UF Resin Synthesis

First, add all formaldehyde into a three port flask, adjust the pH value to 8.0 with sodium hydroxide solution, and raise the temperature to 70°C, Add the first batch of urea (at this time, the molar ratio is 2.65), continue to raise the temperature to 90°C, keep the temperature for 45 min, add sulfuric acid solution, adjust the pH value to 3.0, start the polycondensation reaction, test the viscosity with BYK bubble viscometer, adjust the pH value to 8.0 when the viscosity is H-I, and add the second batch of urea (at this time, the molar ratio is 2.0), continue the reaction at 90°C for 20 min, adjust the pH value to 4.5, start the polycondensation reaction, when the viscosity is o-P, adjust the pH value to 7.8, add the third batch of urea (reaching the final molar ratio of 1.01), stir and cool down the material. The UF resin synthesized under this process is marked as UF-N.

2.3.3. Conventional UMF Resin Synthesis

Melamine and residual urea are added at the initial stage of the second alkaline stage of the above conventional UF resin synthesis, and the material is cooled and discharged after 30 minutes of reaction. The amount of melamine added accounts for 4% of the total urea. After adding melamine, the F/(U + m) molar ratio at the corresponding stage remains unchanged, and the final molar ratio is 1.01. The UMF resin synthesized under this process is marked as UMF.

2.3.4. New Process UMF Resin Synthesis

Melamine and residual urea are added at the initial stage of the third alkaline stage in the UF resin synthesis process with the above new process content, and the material is cooled and discharged after 30 minutes of reaction. The amount of melamine added accounts for 4% of the total urea. After adding melamine, the F/(U + m) molar ratio at the corresponding stage remains unchanged, and the final molar ratio is 1.01. The UMF resin synthesized under this process is marked as UMF-N.

2.4. Performance Test and Characterization

Free formaldehyde content, solid content and curing time shall be determined according to GB/T 14074-2006. Viscosity: measured by BYK bubble viscometer. Storage period: store the resin at 25°C, measure the viscosity with BYK bubble viscometer every day, and characterize the storage period of the resin when the resin viscosity increases to J.

Chemical structure characteristics: it was determined by Varian inova-400 nuclear magnetic resonance instrument. The measurement conditions are: pulse width 12 μs. The delay time is 12s, the measurement frequency is 400MHz, and the cumulative time is 400.

2.5. Plywood Preparation

Poplar veneer: moisture content 10%, thickness 1.6-1.7mm; When mixing, add flour accounting for 25% of the resin weight and 0.5% ammonium chloride curing agent; The gluing amount is 250G/m^3 (double-sided); the hot pressing temperature is 120 OC, the pressure is 1.1 MPa, and the time is 3 min and 4 min.

2.6. Determination of Bonding Strength and Formaldehyde Emission

The bonding strength of plywood shall be determined in accordance with GB 17657-2013; Formaldehyde emission shall be determined according to the dryer method in GB 17657-2013.

3. Results and Discussion

3.1. Basic Properties of Resin Under Different Synthetic Processes

The basic properties of the resin are shown in Table 1. The four resins are transparent liquid in the final stage of their synthesis. After cooling to room temperature, the color of UF and UMF resin immediately changes to white, umf-n resin changes to white after 7 days of storage at room temperature, and uf-n resin remains transparent for 30 days.

Table 1. *Basic properties of resins with different synthetic processes.*

Resin	Appearance	Solid content (%)	Viscosity (mPa·s)	Gel time (s)	Free F content (%)	Storage time (d)
UF	White	50.2	G-H	75	0.17	41
UF-N	Clear	51.5	G-H	103	0.11	49
UMF	White	51.4	G-H	148	0.15	26
UMF-N	Clear	52.4	G-H	187	0.06	33

The final molar ratio of each resin is 1.01, the difference of solid content is small and in the normal range, and the solid content of UMF resin is slightly higher than that of UF resin. Because the bubble viscometer is used to accurately control the viscosity of the resin in the polycondensation stage, the viscosity of UF resin is in the range of g-h, and there is little difference between UMF resin and UF resin. With the addition of melamine, the content of free formaldehyde in the resin decreased. In the new process resin, more formaldehyde participates in the formation of methylene ether bond, so the content of free formaldehyde is lower than that of conventional resin. The effect of new process resin on formaldehyde emission of wood-based panel will be further studied and discussed in the follow-up pressure plate experiment.

The curing time of each resin varies greatly. The curing time of UF resin is the shortest and that of UF-N resin is significantly prolonged. The possible reason is that during the curing process of UF-N resin, due to the thermal fracture of some ether bonds, the molecular weight is reduced, thus delaying the formation of the final macromolecular cross-linking structure of the resin. The addition of melamine reduces the curing speed of UF resin to a certain extent, while the curing speed of UMF resin under the new process is significantly lower than that of conventional UMF resin. Therefore, if this kind of resin is applied to the production practice of wood-based panel, it is recommended to select appropriate curing agent to improve the curing speed, optimize the hot pressing process and ensure a certain production efficiency.

When the viscosity of resin measured by BYK bubble viscometer is less than k, better atomization sizing effect can be obtained and applied to the production of particleboard and fiberboard. The storage period of UF resin is longer than that of UMF resin, and the storage period of new process resin is longer than that of conventional resin. Although a longer storage period can improve the flexibility of resin in wood-based panel production, the curing speed is generally slow. The storage life of the new process resin (UMF-N) added with melamine is close to that of the conventional UF resin.

3.2. ^{13}C NMR Structural Characteristics of Resin Under Different Synthetic Processes

The chemical structures of the above four resins were analyzed by ^{13}C NMR. The chemical shifts are in good agreement with the literature values, with a difference of about 0.9. Based on the absorption of free urea at 162.3 ppm, the corresponding chemical shifts of main functional group components are classified in Table 2, of which the absorption at 48.6 ppm comes from methanol [14].

The chemical shifts of various urea carbonyl compounds, melamine compounds and methylene structures in the spectrum are classified respectively Quantitative analysis and relative content expressed as percentage (Table 2). In general, conventional synthetic processes (UF and UMF) and new synthetic processes The main difference in the structural composition of the resin synthesized by (UF-N and UMF-N) lies in the relative percentage of methylene and methylene ether. The percentage of the total amount of methylene ether of the new process resin is higher than that of the conventional resin, and the content of class I, II and III methylene ether is higher than that of the conventional resin. The content of Quaternary substituted urea in the new process resin is also higher, indicating that the linear methylene ether in the new process resin. The contents of ether bond and cyclic methylene ether bond were higher than those of conventional resin. The percentage of the corresponding total methylene content is lower than that of conventional resin, in which the content of class I and II methylene decreases and the content of class III methylene increases. There was no significant change in the total amount of hydroxymethyl.

Table 2. *The chemical structure of the resin was determined by ^{13}C NMR.*

Component group name	Chemical shift (ppm)	Percentage (%)			
		UF	UF-N	UMF	UMF-N
Free urea	162.4	21.34	22.31	20.06	20.75
Mono substituted urea	160.5	31.85	32.02	35.27	35.17
Binary/ternary substituted urea	159.1	44.04	41.78	41.27	39.69
Tetra substituted urea	153-156	2.81	3.93	3.33	4.28
Total Urea	/	100	100	100	100
Free melamine	166.6	/	/	47.76	48.65
Substituted melamine	165.6	/	/	52.24	51.35
Total melamine	/	/	/	100	100
Formaldehyde	84-90	0.53	0.42	0.47	0.37
Class I hydroxymethyl	63.5	34.51	35.96	38.79	38.37
Class II hydroxymethyl	70.3	8.9	8.1	5.71	5.21
Total hydroxymethyl	/	43.42	44.04	44.48	43.61
Class I methylene	45.7	15.12	12.23	16.12	13.61
Class II methylene	52.4	20.15	17.61	17.93	16.57
Class III methylene	58.6	3.19	3.93	3.42	3.92
Total methylene	/	38.38	33.74	37.34	34.11
Class I methylene ether	67.7	11.63	13.71	12.26	14.01
Class II methylene ether	74.2	4.42	4.89	3.64	4.33
Class III methylene ether	77.5	1.57	3.24	1.77	3.62
Total methylene ether	/	17.59	21.78	17.54	21.93
Total CH_2	/	100	100	100	100

Conventional UF resins generally contain about 18% ether bonds. A large number of ether bonds generally occur under the condition of high formaldehyde/urea molar ratio. Generally, more linear methylene ether bonds can be generated by addition polycondensation under alkaline conditions, while more cyclic methylene ether bonds can be generated under strong acid conditions. In this experiment, the initial molar ratio of 2.65 and the polycondensation reaction under the acidic condition of pH 3.0 can increase the content of linear and cyclic methylene ether bonds in the resin at the same time. The ether bond has good water solubility, so the resin with higher ether bond content has a longer storage period, which corresponds to the results of storage period determination.

Before and after adding melamine, the total amount of methylene ether in UF and UMF and UF-N and UMF-N resins has not changed significantly, indicating that when melamine was added at the initial stage of the second alkaline stage in the synthesis process, most of the ether bonds have been formed in the UF system. After melamine was added, the total amount of ether bonds in the system has not been changed, but it is not clear whether melamine and melamine and urine have been formed Ether bonds between elements.

After adding melamine, there was no significant change in the total amount of hydroxymethyl, but the percentage of class I hydroxymethyl increased and the percentage of class II hydroxymethyl decreased. The reason may be that the added melamine reacted not only with the remaining formaldehyde in the reaction system, but also with the formaldehyde removed from class I and II hydroxymethyl by dehydroxylation reaction to form a new class I hydroxymethyl (hydroxymethylmelamine and monobasic substituted urea), so as to reduce the relative percentage of class II hydroxymethyl in the system. This inference can be further confirmed by the increase of monobasic substituted urea and the decrease of binary/ternary substituted urea in the system.

Melamine was added in the middle and late stage of the resin synthesis process and did not participate in the polycondensation reaction. At this time, the formaldehyde concentration in the reaction system is low, resulting in a certain limit on the hydroxymethylation degree of melamine. Therefore, there is still a considerable part of melamine after the synthesis reaction (about 50%) exists in UMF and umf-n resins in the form of free or low hydroxymethylate. Due to the poor water solubility of melamine, it is difficult to stably exist in the resin solution, making the color of UMF resin turbid and the storage life shortened. These incompletely hydroxymethylated cyanuric amines did not participate in the formation of the main resin structure in the resin synthesis process, but the resin was cured Whether it can be used as formaldehyde capture agent or participate in the formation of the final network cross-linking structure of the resin needs to be further studied [15].

Considering that the formaldehyde concentration, molar ratio and pH value of the system are different when melamine is added in different stages of the synthesis process, it is of certain practical significance to study the effective utilization of melamine in the modified resin so as to design a reasonable formula and process. The new process resin has different chemical structure and properties than the conventional resin. The effects of this kind of resin on the physical and mechanical properties and formaldehyde emission of bonded man-made panels need to be further studied. Through reasonable selection of curing agent, optimization of hot pressing process, making full use of the characteristics of this kind of resin and giving full play to its advantages, it will have practical significance and practical value for the production practice of wood-based panel.

3.3. Bonding Strength of Plywood

The test results of bonding strength are shown in Table 3. The dry bonding strength is high, and the wet bonding strength decreases in varying degrees. Although some specimens are not fully opened after soaking in 63°C hot water for 3h, However, the strength cannot meet the requirements of class II plywood in the national standard GB/T 9846.3-2004 (greater than 0.7MPa), the main reason should be the low molar ratio. For the new process resin, appropriately prolonging the hot pressing time is conducive to promote the resin curing and improve the bonding strength, which corresponds to the determination results of curing time. These measures can improve the curing process of the resin; for the conventional resin, it may lead to excessive curing of the resin and reduce the bonding strength.

Table 3. *Bonding strength and formaldehyde emission of plywood.*

Resin	F content (mg/L)		Dry bonding strength		Wet bonding strength	
	3min	4min	3min	4min	3min	4min
UF	1.56	1.46	1.31	1.33	0.51	0.47
UF-N	1.81	1.68	1.12	1.19	0.43	0.64
UMF	1.50	1.46	1.39	1.45	0.71	0.77
UMF-N	1.36	1.28	1.22	1.14	0.72	0.78

The wet bonding strength of the modified resin with melamine is higher than that of the corresponding unmodified resin. The addition of melamine can effectively improve the water resistance of the resin; The bonding strength of the new process modified resin (UMF-N) catalyzed by ammonium chloride is higher than that of the conventional modified resin (UMF) low; melamine is added at the initial stage and copolymerized with other components to enhance the resin network cross-linking structure and effectively improve the water-resistant bonding strength of wood-based panels; melamine is added at the final stage and does not participate in the polycondensation reaction, and the formaldehyde concentration in the reaction system is relatively low. A considerable part of melamine exists in the resin in free state and has been cured Whether this part of melamine can participate in the formation of the final network cross-linking structure of the resin needs to be further studied.

3.4. Formaldehyde Emission of Plywood

The formaldehyde emission of plywood bonded with resin in the new process is high, but the formaldehyde emission of plywood bonded with UMF resin is low. The reason may be that a more stable methylene ether bond is formed between melamine in UMF resin, while the ether bond of urea formaldehyde resin will decompose to produce formaldehyde under the same hot pressing conditions; the formaldehyde emission of the modified resin bonded plywood with melamine is low. From the test results, the aldehyde reduction effect is more obvious when melamine is added at the end of the reaction. At this time, the free melamine in the resin may play a role in capturing formaldehyde.

It can be seen from the results that the content of free formaldehyde in the resin is not absolutely positively correlated with the formaldehyde emission of wood-based panel. The chemical structure of resin, the selection of curing agent and hot pressing process all affect the formaldehyde emission of wood-based panel. Some free formaldehyde in the resin will escape during hot pressing. At the same time, the possible fracture of ether bond will also release some formaldehyde and change the resin structure. Therefore, it should be comprehensively considered to solve the problem of formaldehyde release from wood-based panel.

4. Conclusions

The resin synthesized by the new process has higher linear and cyclic methylene ether content and lower methylene content than the conventional resin. The curing time and storage period of the new process resin are long, and the content of free formaldehyde is low; When melamine was added in the initial stage of UF resin synthesis reaction, it did not significantly change the total amount of methylene ether bonds and hydroxymethyl groups in the system. However, the content of class I hydroxymethyl increases and the content of class II hydroxymethyl decreases. The reason may be that the added melamine and urea react not only with the residual formaldehyde in the system, but also with the formaldehyde removed from class I and II hydroxymethyl by dehydroxylation reaction, A new class I hydroxymethyl group (hydroxymethyl melamine and monobasic substituted urea) was formed. When melamine was added at the initial stage of UF resin synthesis reaction, a considerable part of melamine was added (about 50%) exists in the liquid resin in the form of free or low hydroxymethylate, which does not participate in the formation of the main structure of the resin.

Melamine can improve the water resistance of the resin and reduce the formaldehyde emission of plywood. Melamine is added to the new process of synthesis at the initial stage of the reaction. The modified resin is bonded to plywood with high bonding strength and low formaldehyde emission.

Author Contributions

The Manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript. Zhiyong Zheng and Huaihao Chen contributed equally and should be considered as co-first authors.

Conflicts of Interest

The authors declare that they have no competing interests.

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