

Review Article

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Effects of chemical modification and nanotechnology on wood properties

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Abstract: As a green material, wood is widely used in building decoration, railway construction, and other fields. However, the wood itself has inherent defects of being easy to absorb water and deform, rot, and decrease in strength. The physical and mechanical strengths and stability of artificial fast-growing forest wood are even worse. As wood modification can improve the dimensional stability, durability, strength, and other properties of wood, it has been widely used. Chemical modification is the main method of wood modification. The development of nanotechnology has brought more possibilities for wood modification. Owing to the extensive literature available, this article summarizes the representative achievements of wood chemical modification and nanotechnology. The principle, production process, advantages, and disadvantages of various wood chemical modification methods were analyzed, compared, and evaluated. Finally, according to the application status of wood-modified materials, the problems existing in the current wood chemical modification methods and the

application of nanotechnology, and the development trend in the future are analyzed.

Keywords: wood, chemical modification, principle, process, characteristics

1 Introduction

In recent years, research on wood modification technology has been progressing well, and many achievements in the related fields are of great significance for the improvement of existing technologies. However, there is currently a lack of relevant reviews to comprehensively sort out, compare, and summarize the existing studies. In today's industrialization, with the decrease of resource availability and the increase of resource demand, it is more and more necessary to explore and popularize practical, green, and sustainable building materials such as wood [1–3]; Moreover, some suitable, cost-effective wood modification technologies will greatly promote this process. This article compares and summarizes the principles, production processes, advantages, and disadvantages of various modification methods. By analyzing the existing problems in the current technology and the difficulties that may be encountered in actual use, the improvement ideas and development directions are proposed to provide follow-up research.

Nowadays, although the use of green building materials such as wood and bamboo has attracted more attention [4–11], it has not been widely used due to their insufficient stability and overall strength [12–14]. The existing modification technologies can also bring various side effects while improving the ability of some aspects. For example, heat treatment modification will affect the strength of wood [15], and furfuryl alcohol modification treatment will make wood brittle and darker in color. Therefore, in recent years, the research of wood modification technology mainly focuses on the further improvement of existing wood technology. By improving the production process, changing or adding chemical reagents, the original defects are made up,

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and the properties of modified wood are more diversified. In the overall trend, researchers have tried to improve the properties of modified wood while taking into account economic and environmental factors [16]. As a new type of modifier, nanomaterials have broad application prospects because of their good impregnation effect and strong functionality, which fit the development trend of wood modification technology at present.

There is no doubt that the emergence of nanomaterials and the development of nanotechnology are extremely important for the improvement of wood properties. The existing traditional chemical modification methods mainly improve the properties of wood by impregnation. Therefore, inevitably there will be problems of insufficient impregnation and uneven dipping. In contrast, due to its high surface area to volume ratio and small size, nanomaterials can easily penetrate into the wood matrix, impregnate more fully, distribute more evenly, and obtain better modification effects than the traditional modification methods. In addition, nanomaterials can also be used as a carrier for biocides. The use of fungicides to protect wood has always been a difficult idea. One of the problems is that it is difficult to maintain the activity of the substances in the fungicides for a long time. Nanocarriers can introduce fungicides into the wood while maintaining the high activity of the active substance. Although the technology is still in the research stage, the prospects are promising. In addition, nanotechnology can be combined with traditional modification technology, which can not only further improve the properties of wood but also reduce the side effects of traditional modification methods. For example, heat treatment improves dimensional stability and durability while making the wood brittle. However, if the wood is pretreated with nanomaterials, the transformation degree of amorphous cellulose to crystalline cellulose can be improved, and the crosslinked part of lignin can be improved so as to reduce the damage of heat treatment on the wood strength. Therefore, the development and application of nanotechnology are very important for wood modification.

So far, researchers have achieved certain results. Breakthroughs have been made in nanotechnology, acetylation, furfuryl alcoholization, thermosetting resin modification, polymer monomer modification, and other technologies, and the properties of modified wood have been further improved. However, these studies are complicated, with different treatment processes and modification effects. Without systematic sorting, comparison, and evaluation, it is difficult to choose and apply in actual use. To evaluate a modification method, a variety of factors such as the performance, production cost, and pollution degree of the modified wood should be considered.

Most of the existing literature reviews focus on the field of wood chemical modification technology and nanotechnology application and summarize their development history [17]. However, there has not been a comprehensive review and evaluation of the research progress in wood modification. This article is not just a simple list of research results, but in-depth analysis and evaluation of the pros and cons of different modification methods, the difference in the properties of modified wood caused by the same modification method using different production processes, and its impact factor. On this basis, the shortcomings and improvement directions of the existing methods are summarized. This is missing from the existing research.

A comprehensive review and evaluation of research in the field of wood modification has not been completed. This work is very important and inseparable from modern engineering. As a renewable resource, wood has the characteristics of low energy consumption, low greenhouse effect, and low air pollution. In addition, the wood is light in weight, high in strength, and has good seismic performance [18,19,21]. The casualties and property losses caused by previous strong earthquakes at home and abroad are far lower than other buildings. In addition, wood has excellent thermal insulation and mechanical properties. Under the same thickness conditions, the thermal insulation value of wood is about 16 times that of concrete, 400 times that of steel, and 1,600 times that of aluminum. Therefore, more and more engineering structures choose to use wood, which is more friendly to the environment and the health of users while ensuring that safety requirements are met [18–20]. However, the defects of the existing fast-growing forest in terms of stability and strength severely limit its application. Therefore, there is an urgent need for a modification method to enhance its related properties and expand its scope of application [1,3,13,22]. A comprehensive review of the existing research can provide references and ideas for the continuous improvement of technology.

This article reviews the research milestones in the field of wood chemical modification. Wood chemical modification technology is divided into nonreactive cell wall modification and reactive cell wall modification [18], as shown in Figure 1. On the basis of this classification, this article analyzes the factors and solutions that hinder its further development through a summary of the principles, processes, and characteristics of the existing modification technology. In addition, this article summarizes the current application of nanotechnology as a green innovation method in the field of wood modification and the existing problems. It provides ideas and references for the research and development of more economical and effective modification technologies.

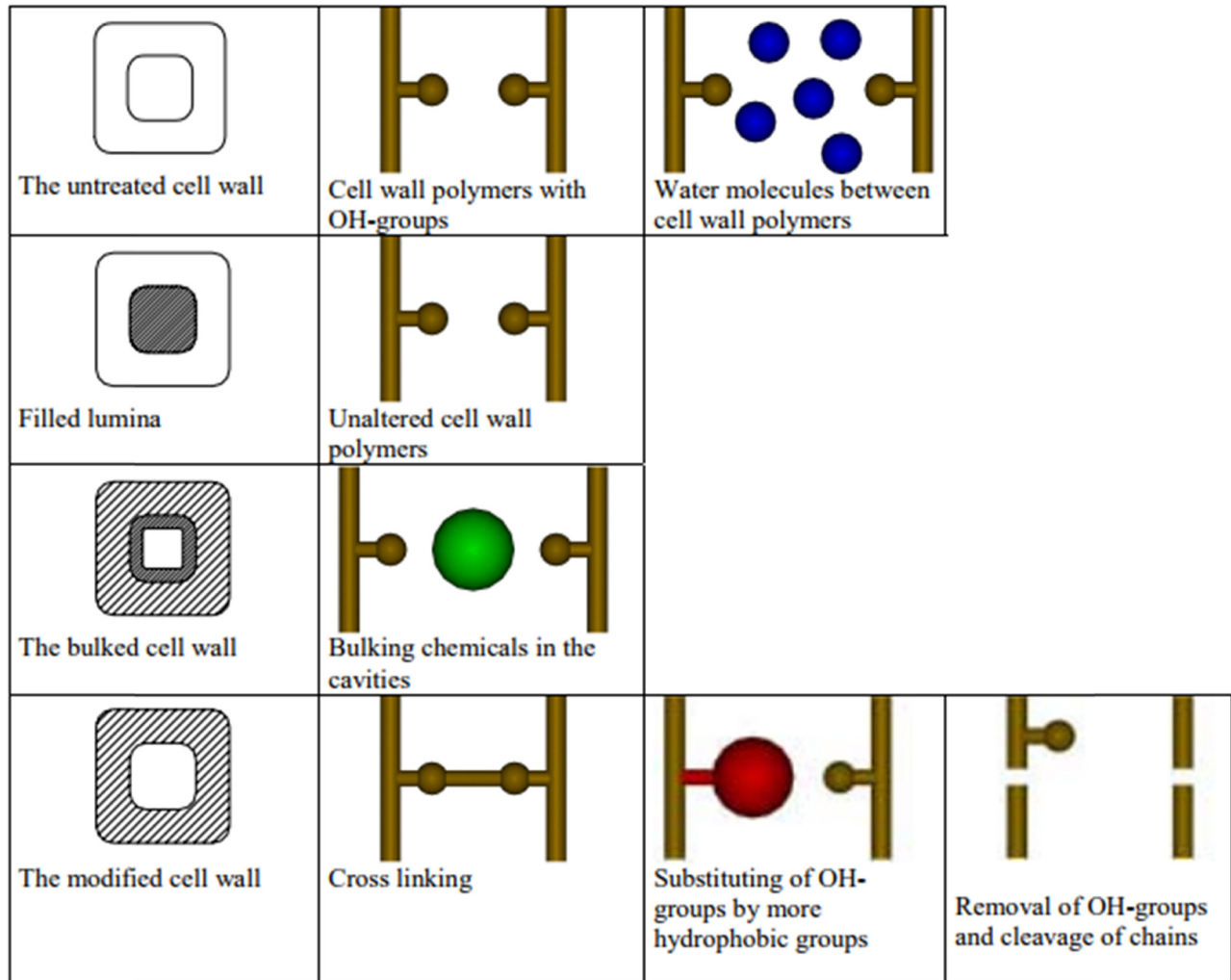


Figure 1: Cell wall reaction diagram of different chemical modifications [18].

2 Acetylation

2.1 The principle

Acetylation is a process in which acetyl groups (CH_3CO) are used to replace the available hydroxyl groups (OH) on wood polymers to produce acetic acid as a byproduct. As early as 1928, Fuchs in Germany began to use chemical reagents such as acetic acid as the main reagent, and sulfuric acid as a catalyst to react with pine wood powder. This was the first wood acetylation experiment [18]. Since the 1940s, many laboratories in the world have conducted acetylation experiments on different species of wood in various ways [23]. Early wood acetylation was mainly based on the liquid-phase reaction to produce acetic acid. The entire reaction process requires the catalysis of reagents such as zinc chloride. Later, several

other catalysts including sodium acetate [24], dimethylformamide, sodium acetate, magnesium persulfate, trifluoroacetic acid, boron trifluoride, *etc.*, were also used [1]. However, nowadays, with the development of chemical technology, the acetylation process has been continuously simplified, and catalysts are no longer necessary for many acetylation reactions [25]. At present, the purpose of using catalysts is more to shorten the reaction time and enhance the stability of wood in the reaction process. At the same time, in order to further improve the stability of the acetylated wood, coatings can be added to the wood surface to shield ultraviolet light to reduce the degradation of the wood polymer. In general, the cost of acetylation treatment of wood is getting lower and lower, and the performance of the product continues to improve. Wood processing is gradually becoming industrialized.

Acetylation can significantly reduce the water absorption of wood and improve the stability and durability of

wood. This is due to the esterification of hemicellulose in the cell wall, which reduces hydrogen bonding with water [1]. Due to the larger size of the acetyl group, the accessibility of the unmodified hydroxyl group is limited [26–28]. This results in very low moisture content in the cell wall, which is not conducive to bacterial invasion and thus improves durability. Therefore, the increase in weight percent gain (WPG) indirectly reflects the degree of acetylation of wood and is highly correlated with the improvement of wood properties [29–31]. Some scholars also agree with another mechanism: by modifying wood with acetic anhydride (AA), the relationship between the observed dimensional stability and the weight gain rate may be related to the expansion of the cell wall [32]. Under this theory, the degree of hydroxyl substitution is not the main reason that affects the efficiency of wood acetylation [33]. In addition, the specific mechanism of the high resistance of acetylated wood to fungal corrosion is still controversial. One of the most widely accepted theories is that acetylation inhibits the consumption of wall polymers and blocks the cell wall micropores, thereby preventing the action of fungal catalysts [34].

2.2 Production process

The process of acetylation usually consists of three steps [23,24]. The first step is drying and pretreatment, drying the water in the wood cell walls to facilitate the penetration of chemical agents. The second step is acetylation, adding a certain amount of AA and other reagents to the reaction tank to diffuse the wood, which is the main modification process. The last step is to recover the chemicals and byproducts left in the reaction tank. This can be done by vacuum heating, followed by drying the wood.

There are four main treatment methods for wood acetylation, chloroacetyl method, ketene method, thioacetic acid method, and AA method [23,27,30]. The chloroacetyl method uses chloroacetyl and wood to react, and the acetylation effect is good, but the byproducts of the reaction will cause the degradation of wood components and reduce the strength of the product. Pretreatment of wood with lead acetate, *etc.*, can reduce side effects, but the cost is relatively high, so it is rarely used at present. The ethylene method dissolves gaseous ethylene in acetone, toluene, and other solvents. Then, it reacts with the wood at a low temperature. The method is simple in process and does not produce harmful byproducts. However, the degree of acetylation of the modified wood is lower than the other methods, and the reaction

time is longer, which is difficult to apply to industrial production [33–35]. The thioacetic acid method uses a catalyst or a swelling agent to pretreat the wood to increase the degree of acetylation of wood. The AA method is currently the most used method in actual production, with low cost, simple process, recyclable treatment liquid, and nontoxic byproducts.

There are two main AA treatment processes: the liquid-phase method and the gas-phase method. Early acetylation treatments are all liquid-phase methods, which are usually suitable for wood with larger dimensions. The acetylation reaction of the liquid-phase method is completed in a liquid environment, and the wood is placed in a vacuum treatment tank. The mixture of AA and catalyst is then introduced and heated according to the thickness of the wood. After the reaction, the remaining AA and the byproduct acetic acid are recovered. The disadvantage of this method is that it is difficult to recover the treatment liquid. The acetylation reaction of the gas-phase method is completed in a gaseous environment. The AA vapor or the mixed vapor of AA and the catalyst is passed into a closed processing tank. When the reaction is complete, the excess gas is recovered to complete the acetylation process. When the gas-phase treatment is used, the diffusion rate of the drug is inversely proportional to the square of the thickness of the specimen and so gas phase treatment is not suitable for large size wood processing. The common difficulty of the two methods is that when traditional catalysts such as pyridine are used, the generation of by-product acetic acid, *etc.*, leads to the degradation of wood components. Therefore, more stable and reliable catalysts are a hot topic in their research. Some scholars use N-bromosuccinimide (NBS) as a catalyst to acetylate wood with AA. The degree of acetylation is determined by the weight gain rate (WPG). Figure 2 shows that in the concentration range of 1.0–3.0%, the presence of NBS greatly improves the acetylation rate [35].

2.3 Characteristics of acetylated wood

The most fatal inherent defects of wood are biodegradability over time and dimensional instability under alternating environmental conditions [36]. Acetylation can significantly improve this problem. In addition, acetylation significantly reduces the moisture absorption of wood; improves the ability to resist fungal corrosion, and moth-eaten; significantly improves the strength compared to untreated wood; the carbon footprint is much lower than those of steel and concrete; and the fire performance is significantly improved. However, the residual

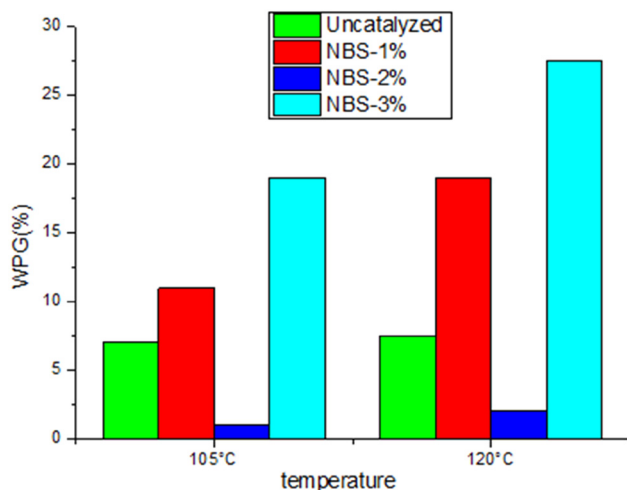


Figure 2: Comparison of the weight gain rate between untreated wood and wood treated with different concentrations of NBS solution [35].

acetic acid byproduct in acetylated wood has greater acidity and has a certain impact on the strength of the wood. It is easy to remove the acid in the product in the case of wood particles and fibers but it is somewhat difficult in solid wood [1]. Additionally, it is also difficult to recover the catalyst from the waste liquid, which is also a problem that cannot be ignored in production [37].

The effect of acetylation treatment is generally evaluated by the weight gain rate of the treated wood. The main factors affecting the weight gain rate of acetylated wood are tree species, wood size, heartwood, wood moisture content, reaction liquid type or ratio, pretreatment conditions, reaction conditions, *etc.* [38–40]. Acetic anhydride (AA) and vinyl acetate (VA) have different modification effects on different tree species under the action of six different catalysts. The catalyst using potassium acetate, potassium carbonate, and sodium carbonate can significantly improve the acetylation of Scots pine wood powder. However, the effect of sodium carbonate on the acetylation of VA is much worse than that of potassium acetate [37]. Potassium acetate can effectively catalyze the grafting reaction of AA on wood, and WPG can usually reach more than 30%. If sodium carbonate is used for catalysis, the WPG of modified wood is usually not more than 20%. In addition, studies have shown that the modified effect of young wood and mature wood is different. Young wood has a significant adverse effect on acetylation and the reduction of young wood WPG is about 50% of that of mature sapwood. The chemical difference between catalytic acetylation and noncatalytic acetylation is believed to be the cause of this behavior [40].

Larsson Brelid's research shows that when the acetyl content is 15.1%, the wood decay rate is significantly reduced. When the content of acetyl was 18.5%, the control effect on all kinds of rotten bacteria was the best. The effect of wood acetylation on the protection of Marine borers was not obvious, but the degree of wood acetylation decreased with the increase of the acetyl content [39,41,42]. Behbood Mohebbi [43] studied the ability of acetylated wood to resist microbial infestation in field soils and observed that soft rot fungi dominate in field soils, and they are one of the important causes of degradation of acetylated and nonacetylated wood. However, the acetylation modification can enhance the resistance of wood to fungal corrosion, and the corrosion of moderately acetylated wood is much slower than that of untreated wood, as shown in Figures 3 and 4. But after 7 years of acetylation treatment, compared with nonacetylation treatment, the defense effect of acetylation treatment on soil microorganisms is weaker. Table 1 shows the influence of various factors on the weight gain rate during the acetylation process, which indirectly reflects the influence on the acetylation effect.

The cost of acetylated wood products needs to be reduced and the physical and mechanical properties need to be further improved. In the future, the development trend of acetylated modification treatment methods should be high performance, harmless, and green. By improving the treatment methods, reducing the use of chemical reagents or other measures, the pollution can be reduced, the process can be simplified, and the product quality can be improved. Of course, this may require new breakthroughs in the study of the mechanism of acetylation.

3 The chaff alcoholize

3.1 The principle

Furfuryl alcohol is also called furan methanol or oxy-methylene methanol. Adding a catalyst under normal temperature or heating conditions can cause polycondensation to generate resin. The principle of furfuryl alcohol modification is to impregnate furfuryl alcohol molecules and acidic catalysts into the wood. After heating and with the action of the catalyst, the furfuryl alcohol molecules undergo *in situ* polycondensation to form a polymer resin, which is filled in the wood voids to inflate the cell wall, thereby improving part of the properties of

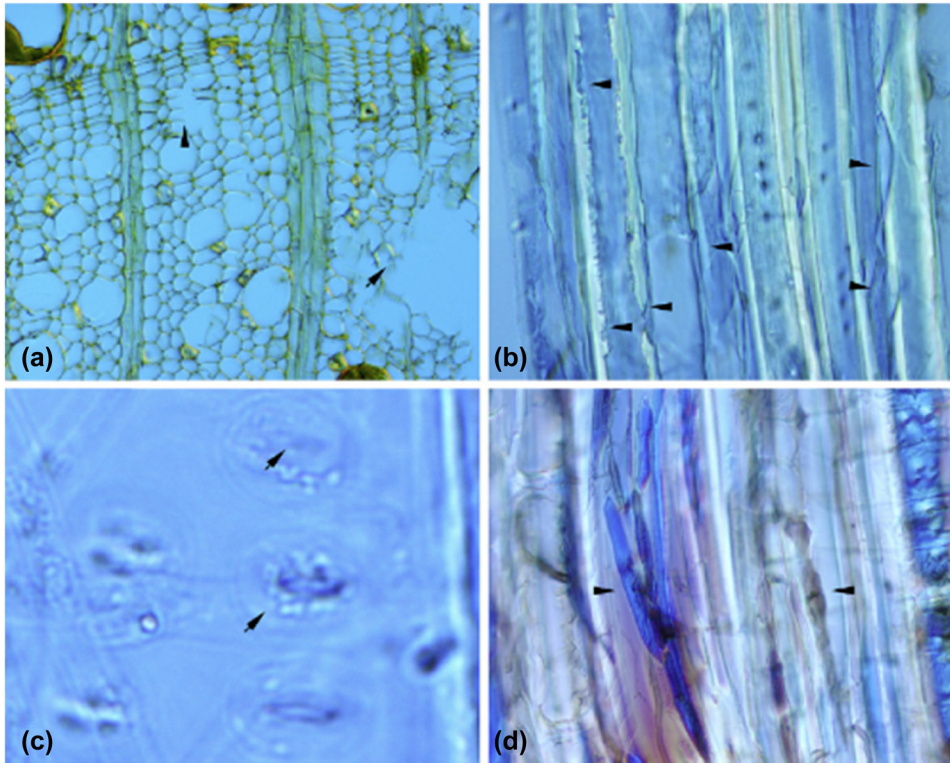


Figure 3: Severe biodegradation in nonacetylated beech: (a) and (b) White rot decay (arrow heads); Nomarsky. (a) cross section; Nomarsky. (b) Radial section; Nomarsky. (c) Bacterial degradation in pits (arrow heads); radial section, Nomarsky. (d) Soft rot decay (arrow heads); radial section, Nomarsky [43].

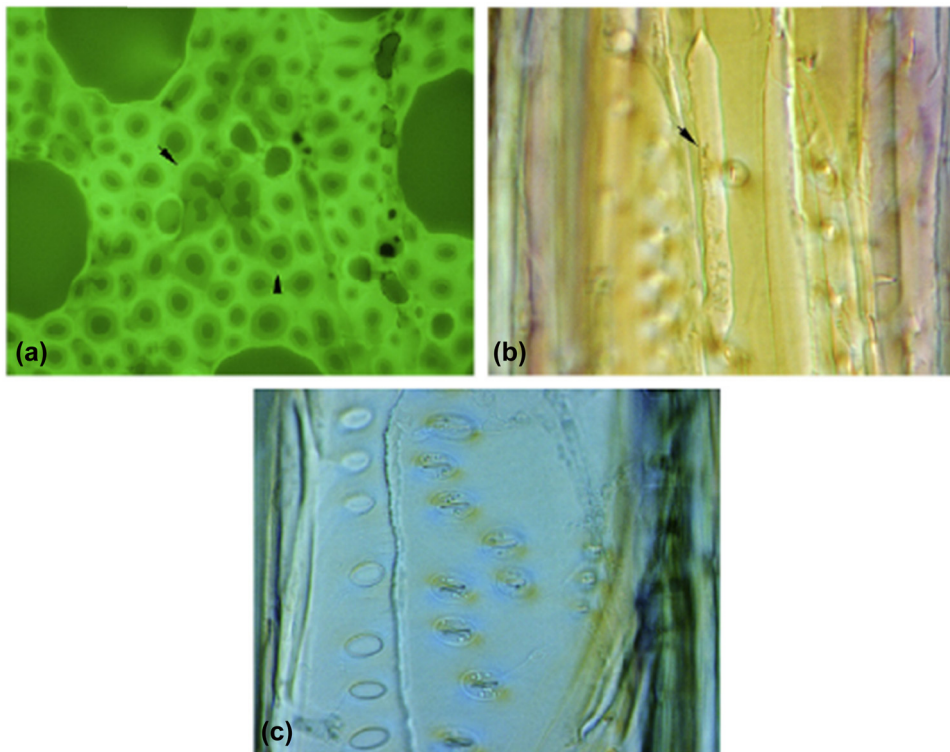


Figure 4: Microbial degradation in acetylated beech wood: WPG 9.6%. (a) Darker area indicates white rot decay developing (arrow heads); cross section, fluorescent light. (b) Soft rot decay (arrow heads); radial section, Nomarsky. (c) Bacteria in vessel pits; radial section, Nomarsky [43].

Table 1: The influence of the change of different factors on WPG [2,36,38,39]

Tree species	Modifier	Catalyst	Conditions	WPG (%)
Scots pine	AA	—	AA concentration	
			0 mmol/g	-3
			14 mmol/g	2.4
			28 mmol/g	3.6
			42 mmol/g	6.5
			56 mmol/g	7.6
Scots pine	CA	K ₂ CO ₃	CA concentration	
			0 mmol/g	-3.8
			14 mmol/g	10.8
			28 mmol/g	11.6
			42 mmol/g	13.8
			56 mmol/g	15.1
Scots pine	AA	K ₂ CO ₃	AA Amount	
			0.4 mmol/g	17.5
			0.7 mmol/g	20.5
			1.2 mmol/g	19
			1.5 mmol/g	18
			1.9 mmol/g	18.2
Scots pine	AA	Na ₂ CO ₃	0.5 mmol/g	16
			0.9 mmol/g	17.5
			1.5 mmol/g	18
			1.9 mmol/g	18.2
Scots pine	AA	CH ₃ COOK	0.6 mmol/g	19
			1.1 mmol/g	21
			1.5 mmol/g	22
			1.9 mmol/g	21
Scots pine	CA	K ₂ CO ₃	0.4 mmol/g	15.5
			0.7 mmol/g	15.3
			1.1 mmol/g	15
			1.5 mmol/g	14.7
Scots pine	CA	Na ₂ CO ₃	0.5 mmol/g	3
			1.0 mmol/g	6
			1.5 mmol/g	7
			2.0 mmol/g	8
Scots pine	CA	CH ₃ COOK	0.5 mmol/g	6
			1.0 mmol/g	12
			1.5 mmol/g	13
			2.0 mmol/g	14
Scots pine	AA	—	80°C, 3 h	8.3
Scots pine	AA	—	120°C, 3 h	9.6
Scots pine	AA	—	120°C, 6 h	18.8
Beech wood	AA	—	80°C, 3 h	8.0
Beech wood	AA	—	120°C, 3 h	
Beech wood	AA	—	120°C, 6 h	20.4
Poplar	AA	—	80°C, 3 h	16.8
Poplar	AA	—	120°C, 6 h	18.8
Scots pine	AA	pyridine	Pine wood with false rings	19.8
			Normal pine wood	21.3
			Pine wood with discontinuous rings	19.2
			Compression pine wood	14.1
			Tension beech wood	12.9
Beech wood	AA	pyridine	Normal beech wood	15.2

the wood. At present, furfural is prepared from biomass containing hemicellulose or xylan by acid dehydration in industrial production, and furfural is further hydrogenated to furfuryl alcohol. The antiseptic properties of furfuryl alcohol are related to xylan, which is one of its raw materials. Xylan is a natural biological preservative with antibacterial properties, mainly found in the hemicellulose of natural plant cells. The nano-xylan prepared by it has a smaller particle size and is easier to penetrate into the pores of the wood. Soaking the wood under normal pressure can obtain a better anti-corrosion effect.

The research on the modification of wood with furfuryl alcohol was initiated by Dr. Alfred Stamm, the pioneer of wood modification in the early 1950s [41]. Irving Goldstein [42,44] conducted an early study on the furfurylation of wood. However, these methods used zinc chloride as a catalyst, which led to the decomposition of cellulose, thereby reducing the strength of the modified wood and the result was not commercialized. In the early 1990s, Professor Marc Schneider and Dr. Mats Westin developed a method using cyclic carboxylic anhydride (maleic anhydride) as a key catalyst [45–47]. The solution prepared by this method has good stability at room temperature and an excellent modification effect. The polymerization reaction of furfuryl alcohol in wood is a complex chemical reaction [48]. There are still some [49] discussions as to whether the process of furfuryl alcohol only swells the wood cell wall without chemical reaction. Some scientists believe that this is a chemical modification process because the furfuryl alcohol polymer can react with itself and may react with the lignin in the cell wall to form a covalent bond [50]. Another view is that wood furfuryl alcoholization is a process of impregnation and filling modification, and furfuryl alcohol polymer fills wood cell walls more instead of chemically reacting with cell walls [51–53].

Figure 5 shows the distribution of maleic anhydride (F2) in cells using confocal laser scanning microscopy (CLSM). It can be observed that the polymerization effect of resins distributed in different positions is different. The resins directly adhered to the surface of the cell wall have the best polymerization effect, and the resins distributed in the center of the cell cavity and the cell wall [53] are the second. Thygesen *et al.* [54] believe that cell wall aggregation constitutes a restricted environment for FA polymerization, resulting in a shorter coupling length in the FA resin.

3.2 Production process

The general process of wood bran alcoholization is very similar to acetylation, which is divided into three steps.

The first step is to dry the wood; the second step is to configure furfuryl alcohol solution to treat the wood and impregnate it under normal pressure; and the third step is to cure the impregnated wood and then dry it.

The modification process of furfuryl alcohol is to make furfuryl alcohol enter wood cells through pressure impregnation under the action of an acidic catalyst. After curing furfuryl alcohol at high temperatures, the modified wood is dried. Wood impregnation is usually done by the full cell method. The main factors affecting the impregnation effect include the impregnation process parameters, wood properties, modifier properties, and the choice of catalyst. Impregnation process parameters mainly include pressure and time, and the results of impregnation will differ with different wood properties. When the molecular diameter of the modifier is smaller than the cell wall pore, the modifier can enter the wood cell wall. The lower the specific gravity of wood, the higher the weight gain rate after impregnation [53]. The viscosity, molecular weight, and polarity of the modifier will affect the impregnation effect. The lower the viscosity of the modifier, the better the permeability in the wood and the faster the flow rate [54]. The molecular weight and the polarity of the modifier determine whether the modifier can impregnate the wood cell wall. The modifier with a molecular weight between 290 and 470 can enter the wood cell wall, while the modifier with a molecular weight of 820 can only enter the wood cell cavity [55]. Wood's own cellulose, hemicellulose, and lignin have polar groups such as hydroxyl, carboxyl, and carbonyl. Therefore, it is difficult for nonpolar modifiers to enter the wood cell wall. Only polar modifiers can swell the wood cell wall and enter [56].

During the immersion curing process, the choice of the catalyst, curing time, and curing temperature are the main factors that need to be considered. The molecular weight of the catalyst needs to be small enough and has a certain affinity for the wood material and the modifier to meet the requirement of penetrating deeply into the wood pore wall without being separated from the modifier. Under the action of a strong acid catalyst (such as p-toluenesulfonic acid), the main reaction is free radical polymerization, which leads to a high degree of ring-opening reaction. The furfuryl alcohol polycondensation reaction is violent and it is difficult to stabilize the reaction system. However, the wood impregnation solution needs to be stable during storage and impregnation and so strong acid catalysts are not suitable for wood impregnation [57]. Schneider *et al.* [58] showed that maleic anhydride has stronger acid sites than mixed organic acids, resulting in a higher resin WPG. In addition, at a shorter time and at a lower temperature, maleic anhydride is

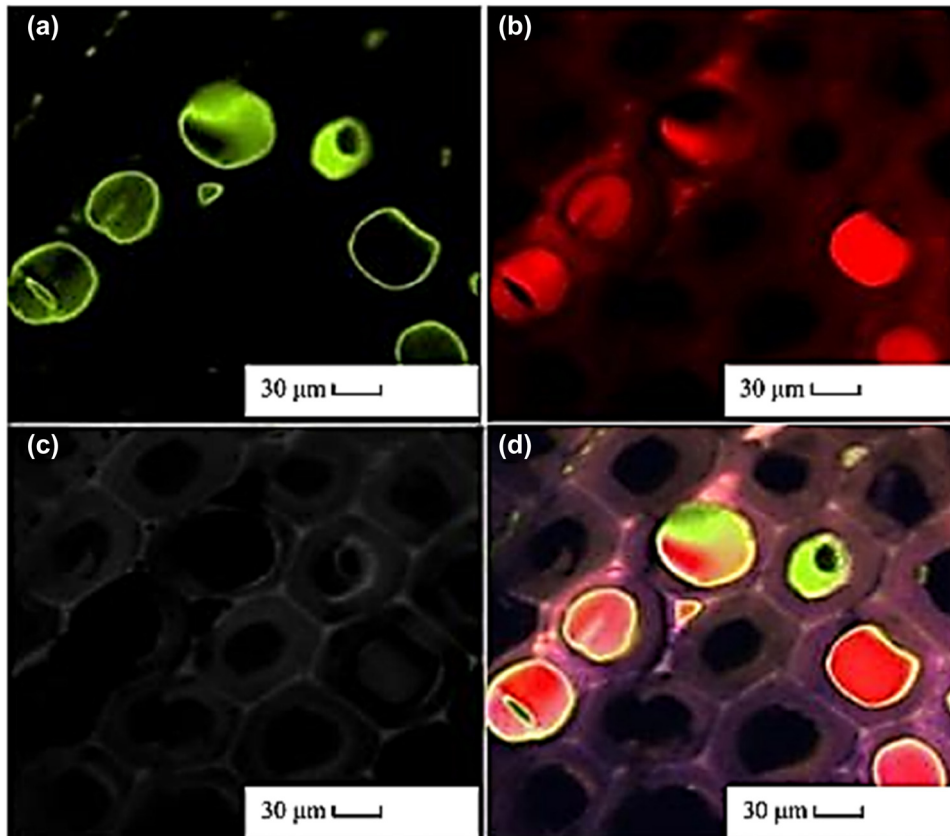


Figure 5: The scanning microscope image of wood treated with maleic anhydride: (a) resin I with a better degree of polymerization in the first channel (650–700 nm); (b) resin II with a lower degree of polymerization in the second channel (700–750 nm); (c) anatomical structure of wood; and (d) reconstruction images obtained from all channels [53].

beneficial for the polymerization of furfuryl alcohol, but at a higher temperature and a longer curing time, it will cause more serious degradation of the cell wall. When the temperature is constant, the mass fraction of the catalyst is in the range of 5–20%, and the curing speed of the resin increases as the mass fraction of the catalyst increases, and the curing time is shortened [59]. The higher the curing temperature of the furfuryl alcohol resin-modified wood, the faster the resin curing speed, but when the curing temperature exceeds 100°C, the evaporation rate of the moisture in the wood must be strictly controlled to avoid the wood cracking due to drying stress [55].

3.3 Modification effects

The advantage of furfuryl alcohol treatment is that furfuryl alcohol is mainly made from crop wastes, and the cost is low [60]; it can reduce the equilibrium moisture content of wood; significantly improve the compressive strength, dimensional stability, and biological durability

of wood along the grain; delay the aging process degradation of wood components and deformation of wood cell walls; decreases the water expansion and shrinkage of wood more than 50% lower than that of the untreated wood [61]; improve the wood resistance to insects [62]; burning does not release any volatilization organic compounds or polyaromatic hydrocarbons; similar to the volatile levels of untreated wood burning [63]. The reason for the corrosion resistance of the modified wood may be that the internal voids of the modified wood are reduced, the moisture absorption is reduced, and the moisture content is lower than the moisture content required for microbial reproduction [64].

However, the disadvantages of the furfuryl alcohol modification treatment are that the process is complicated, time-consuming, has low curing efficiency, and the treated wood becomes slightly brittle, and the color becomes brown. The furfuryl alcohol modification treatment has no significant effect on the static bending strength and elastic modulus of wood, but it will reduce the impact strength of wood and increase its brittleness [65]. The decrease in the impact strength is related to the weight

gain rate [66], and furfuryl alcohol modification will also reduce the water absorption of wood [65], as shown in Figure 6. In the process of furfuryl alcohol modification, some additives can be added to improve its performance in certain aspects. For example, some scholars added ammonium dihydrogen phosphate (ADP) as a flame-retardant additive, which ultimately improved the flame retardancy of the modified wood and reduced the rate of heat release and the promotion of char formation effectively inhibit the production of smoke during the combustion process [69]. Table 2 shows the differences in properties of different concentrations of furfuryl alcohol modification, additive modification, and dihydroxymethyl dihydroxyvinyl urea resin (DMDHEU) modified wood [60,65,69].

The effect of furfuryl alcohol modification depends on the retention of furfuryl alcohol in wood. With a

high retention rate, the wood is fully impregnated, the modification effect is good, and the hardness and resistance to biological erosion and chemical degradation are significantly improved, with high-temperature stability. At lower retention, many attribute enhancements will also appear but the enhancement is slightly lower. Therefore, the modification of furfuryl alcohol should take into account the modification effect and cost, and control the resin retention. Some scholars have established mathematical models to determine resin retention, but there are defects such as high requirements for samples, large quantities, and poor model applicability [67,68]. This technology still needs further exploration. Simultaneously simplifying the process of furfuryl alcohol modification and improving the strength of wood while ensuring the modification effect is the future development direction of the furfuryl alcoholized wood modification method.

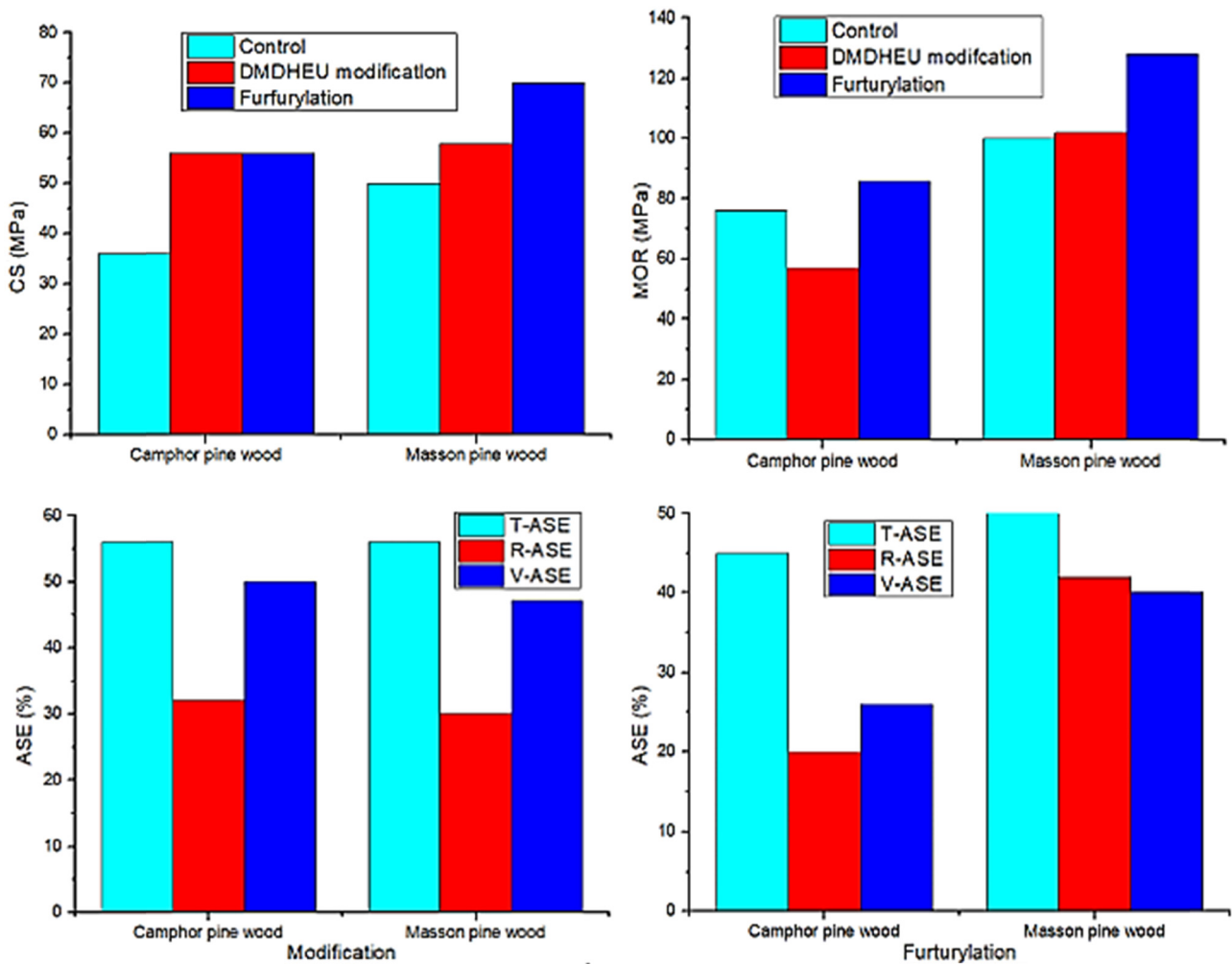


Figure 6: Comparison of properties of modified wood and unmodified wood [65].

Table 2: Properties of wood treated with furfuryl alcohol (FA) and other modified methods

Sample	MOR (MPa)	MOE (GPa)	CS (MPa)	Wood density (g/cm ³)	WPG (%)	ASE (%)
Unmodified	—	305	—	0.734	—	—
25% FA	—	154	—	0.932	10.1	23.4
50% FA	—	163	—	0.973	21.3	36.5
100% FA	—	142	—	1.062	33.9	42
Control	88.42	11.7	49.84	0.37	—	—
Poly furfuryl alcohol (PFA)	78.77	14.1	78.14	0.63	67.75	—
Wood/PFA/ADP	83.26	15.52	71.26	0.58	68.34	—
Control	75	—	50	—	15	48.2
DMDHEU	55	—	61	—	27	40.4
Frufurylation	87	—	76	—	46	39.3

Note: WPG, weight gain rate; EMC, equilibrium moisture content; ASE, anti-expansion; CS, grain compressive strength; MOR, modulus of rupture; WL, weight loss rate.

4 Thermosetting resin modification

Thermosetting resin refers to a type of synthetic resin that is cross-linked and cured into insoluble and insoluble substances through a chemical reaction under the action of heat, pressure, or a curing agent. The thermosetting resin is a kind of high molecular weight polymer material, in which molecular chains are cross-linked together to form a rigid three-dimensional network structure. It has excellent characteristics such as high strength, good heat resistance, and good dimensional stability. In lignocellulose, free hydroxyl groups absorb moisture from the atmosphere, causing chemical reactions, forming covalent bonds, and causing wood deformation. This process is reversible. Modifying wood with thermosetting resin is one of the ways to reduce wood expansion and contraction and improve wood dimensional stability. Currently, there are three main types of thermosetting resins used for wood modification: urea–formaldehyde resin (UF), phenolic resin (PF), and melamine–formaldehyde resin (MF) [106].

4.1 The urea–formaldehyde resin modification

The curing process of urea–formaldehyde resin is to transform the linear soluble resin into the body structure resin with hinge chlorine as a curing agent. The curing mechanism of the urea–formaldehyde resin is the reaction of formaldehyde and ammonium chloride to produce hexamethylenetetramine and hydrochloric acid, and the acid production reactive functional groups of the resin help the resin to form a three-dimensional network structure [67]. The preparation process is to add the curing

agent (ammonium chloride, *etc.*) to the resin solution first, then immerse the sample, immerse after vacuum treatment, and finally, dry and solidify.

The modification of the urea–formaldehyde resin can improve the stability, durability, and strength of the wood. Yinluan *et al.* [68] used different molar ratios of melamine urea–formaldehyde resin (MUF) to treat hardwood and coniferous wood. The study found that the effect of MUF-modified wood is related to the structure and species of the wood itself. The MUF modification effect is good when the Wood has a loose structure and large pores. After the modification, the microstructure of the wood is damaged, causing the volume shrinkage of poplar and fir (the ASE value is negative), as shown in Figure 7. Furuno *et al.* [70] and Behr *et al.* [71] also found this phenomenon in their studies. Furuno *et al.* [70] believe that the collapse of the cell wall is the cause of the negative ASE value, while Behr *et al.* [71] believe that the high temperature and alkaline impregnation are the cause for the negative swelling. After modification, the compressive strengths along the grain of pine, fir, eucalyptus, and vertebral wood increased by 66.33, 43.18, 32.67, and 13.28%, respectively [68]. In addition, the urea–formaldehyde resin can also be composite-treated with polyammonium phosphate treated with polyethylenimine as a modifier, and the two can form a good synergistic effect, which ensures the bonding strength and improves the flame-retardant performance of the material [72]. It is worth mentioning that with the research of nanomaterials and nanotechnology, some scholars have tried to modify the wood with the urea–formaldehyde resin and nano-SiO₂ composite material. The hardness of wood after modification is significantly improved, but the improvement of dimensional stability is not as good as the traditional modification method [73]. The

urea–formaldehyde resin has the advantage of low price, but the urea–formaldehyde-modified liquid is prone to polymerization during storage, has a short storage period, and is not easy to store.

4.2 The phenolic resin modification

Phenolic resins are also widely used in the wood industry. The phenolic resin modification method includes five steps: drying, resin impregnation, resin semi-curing, heating, and hot pressing. First, the wood is dried and then impregnated with the phenolic resin in the impregnation cylinder. After impregnation, it is left for a few hours to drain the excess resin and then partially dried and heated again. The purpose of this re-drying or heating is to make the impregnated resin into a semicured structure. Finally, at a high temperature, the sample is densified to a predetermined thickness by hot pressing for 45 min [74].

Shams *et al.* [75] systematically studied the penetration and distribution of resin in wood and the effect on wood crystallinity and surface wettability during the process of preparing dense wood by impregnating low-molecular-weight water-soluble phenolic resin with hot pressing. The changes in the mechanical properties of cured wood and untreated wood were studied. Studies have shown

that the young's modulus and flexural strength of the phenolic-resin-impregnated compressed wood are higher than the untreated compressed wood, especially low-density wood. Low-molecular-weight phenolic resin can enter the cell wall and so the modification effect is better than that of large-molecular-weight phenolic resin [76]. In the wood impregnation process, the degree of resin entering the cell wall depends on the molecular weight of the resin. When the average molecular weight of the phenolic resin is between 290 and 480 g/mol, the resin mainly exists in the wood cell wall. When the average molecular weight of the phenolic resin exceeds 800 g/mol, most of the resin is mainly filled in the cell cavity. During the heating and curing process of the resin distributed in the cell cavity, the evaporation of water will cause the shrinkage and deformation of the wood cell cavity, causing defects such as shrinkage during the drying process of the wood and affecting the drying level of the wood and dimensional stability. The study of Bakar *et al.* [77] showed that phenolic resin modification can effectively improve the resistance of oil palm wood (OPW) to white rot fungi and termites, as shown in Figures 8 and 9. To further improve the addition reaction and condensation reaction of the phenolic resin, nanoparticles and surfactants can be added. When the alkane surfactant and nano-cuo work together, the apparent activation energy of the phenolic resin decreases, and the reaction enthalpy increases, making the addition and condensation reaction of the phenolic resin

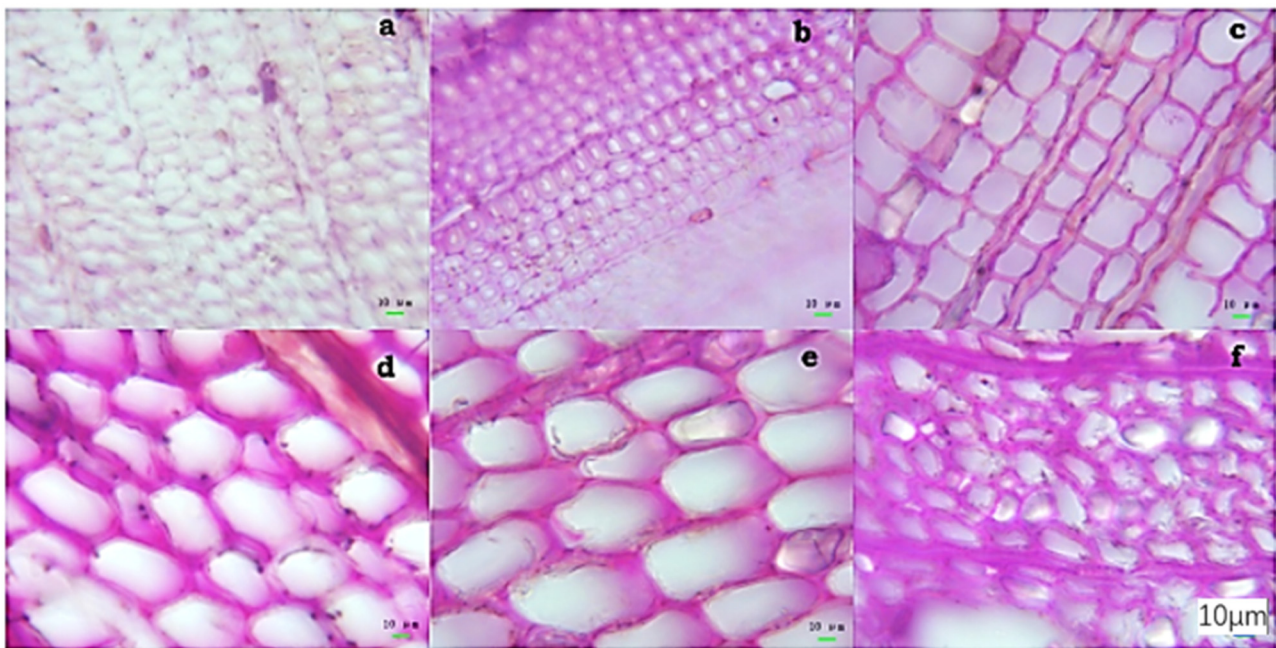


Figure 7: Light micrograph section of (a) untreated eucalyptus; (b) treated eucalyptus; (c) treated Chinese fir; (d) untreated pine; (e) treated pine; and (f) poplar after burial for 377 days [68].



Figure 8: OPW after cleaning at the end of the termite test. Top left: untreated OPW; top right: treated OPWD-0%; bottom left: treated OPWD-25%; and bottom right: treated OPWD-50% [76].

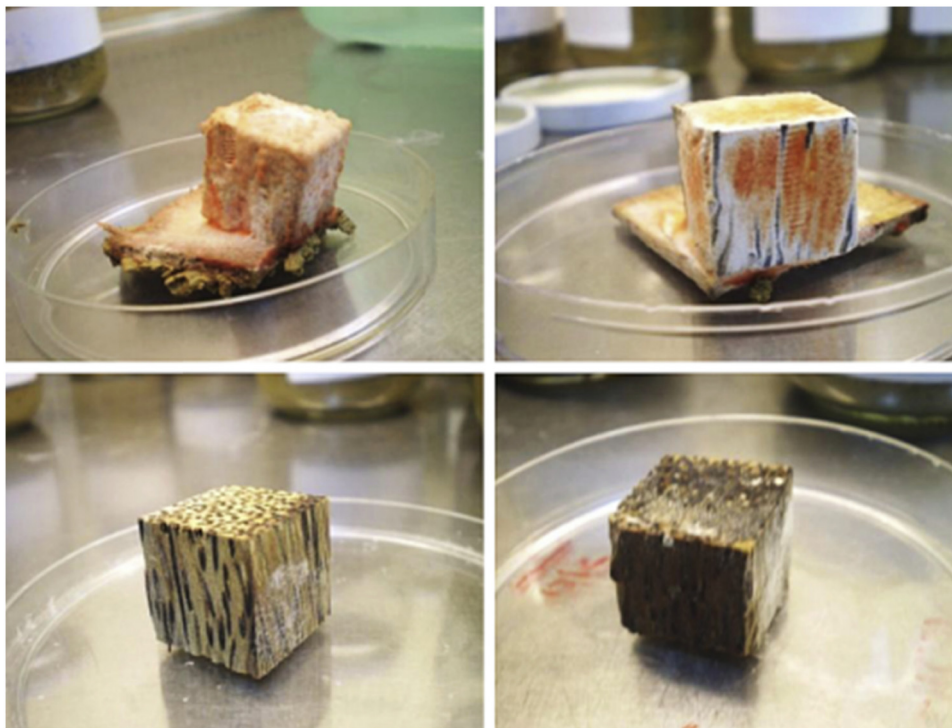


Figure 9: OPW after the end of the decay test. Top left: untreated OPW; top right: treated OPWD-0%; bottom left: treated OPWD-25%; and bottom right: treated OPWD-50% [76].

easier to complete. This makes up for the effect of nano cuo alone on diffusion control [78].

The shortcomings of phenolic resin in the application are that its curing temperature is relatively high, the drying process is difficult to control, and during the modification of phenolic resin, the treated OPW releases a large amount of free formaldehyde, which is harmful to the human body. In order to make it suitable for indoor materials, the hot-pressing compression program in the process is changed and the drying time after the process is extended; hence the curing state of the resin can be improved, which can effectively reduce the release of formaldehyde. Compared with the change of the hot-pressing process, extending the compression time is more beneficial to reduce the emission of formaldehyde [79]. In addition, phenolic resin is the product of polycondensation of phenol and formaldehyde in an alkaline medium containing excess formaldehyde, and phenol is produced on a large scale from raw materials extracted from petroleum. Considering the increasing demand for phenol and the depletion of fossil resources, the replacement of phenol by renewable raw materials has become a hot issue. At present, researchers have studied different types of thermal pyrolysis liquefaction and its components as substitutes for phenol: total thermal pyrolysis liquefaction [80], separated lignin [81], and phenol reaching components [82], Separation and cleaning of phenol [83,84], methylated bio-oil [85], palm kernel shell bio-oil [86] and cowhide lignin bio-oil [87,88], *etc.* However, this resin has more or fewer shortcomings in terms of strength, odor, or color, and there is still room for improvement. Zabelkin [89] found that some of the shortcomings come from the neutral compounds in PL, which reduces the quality of the resin. After removing the neutral compounds in the modified phenolic resin with organic solvents such as *n*-hexane and benzene, the water resistance coefficient of the resin was increased by more than 60% [76].

4.3 The formaldehyde resin modification

In 1940, Stamm and Seborg [90] and his colleagues conducted the first experiment of impregnating wood with formaldehyde-based resin. The formaldehyde resin impregnation treatment improves the dimensional stability of the wood and improves the biological resistance to fungi, termites, and marine borers. In recent years, there has been more and more research on the use of melamine-formaldehyde (MF) resin to impregnate the wood, especially in Europe [91,92]. The modification process of the formaldehyde resin is to impregnate the wood in resin, and all impregnation is

carried out in the decompression process with 10% resin. After 20 min of vacuum, the solution was soaked in a desiccator. Then, the vacuum was released and the liquid was stored in liquid for 2 h, and then stored at 20°C/65% RH. After immersing for 7 days, it was dried at 60°C for 24 h, and then at 140°C for 24 h. This curing scheme proved to be the best curing scheme for resins [93].

The principle of modifying wood with the melamine-formaldehyde (MF) resin is as follows: the amino groups in melamine react with formaldehyde to generate methylol melamine [94]. Methyl melamine improves the miscibility with water and limits self-coagulation during storage [95]. This allows low-molecular-weight aqueous solutions of methylol melamine monomers and oligomers to be polymerized *in situ* inside the wood. Many small molecules can enter the nanopores of the cell wall, making the modification more thorough [96,97]. Cured MF resin contains polar groups that can react with water molecules [98]. Altgen *et al.* studied the relationship between water absorption rate of the MF resin, curing method, and temperature, as shown in Figure 10. Figure 10a–c [99] are, respectively, the adsorption isotherm, desorption isotherm, and hygroscopic hysteresis isotherm of the cured MF resin; Figure 10d [99] represents the amount of hydrogen exchange and D2O absorption under different curing conditions; Figure 10e represents the FTIR spectrum of the cured MF resin after drying and H-D exchange. When the relative humidity is about 80%, the moisture content of the resin under different curing conditions is basically the same, but when the relative humidity is more than 80%, the moisture content of the MF resin under the curing condition of 130°C increases sharply. The hysteresis effect of the water content of the mildly cured MF resin was significantly reduced, while the mildly cured or highly cured MF resin almost had no hysteresis effect.

Considering the wood of the *Picea asperata* tree as the research object, Gindl and Gupta [97] tested the hardness and Young's modulus of the wood cell wall treated with the MF resin by using a nano notch. The results showed that the treatment increased Young's modulus of the second layer of the secondary cell wall by 33%, and the hardness and compressive strength were also enhanced. The dimensional stability, water resistance, and swelling property of wood modified with melamine resin were improved [100,101]. However, the main disadvantage of this treatment is the high production cost, and the wood product is prone to cracking under dry and wet cycle conditions.

In summary, the current thermosetting resin modification has been proven to significantly improve the durability of wood, but the production cost, process

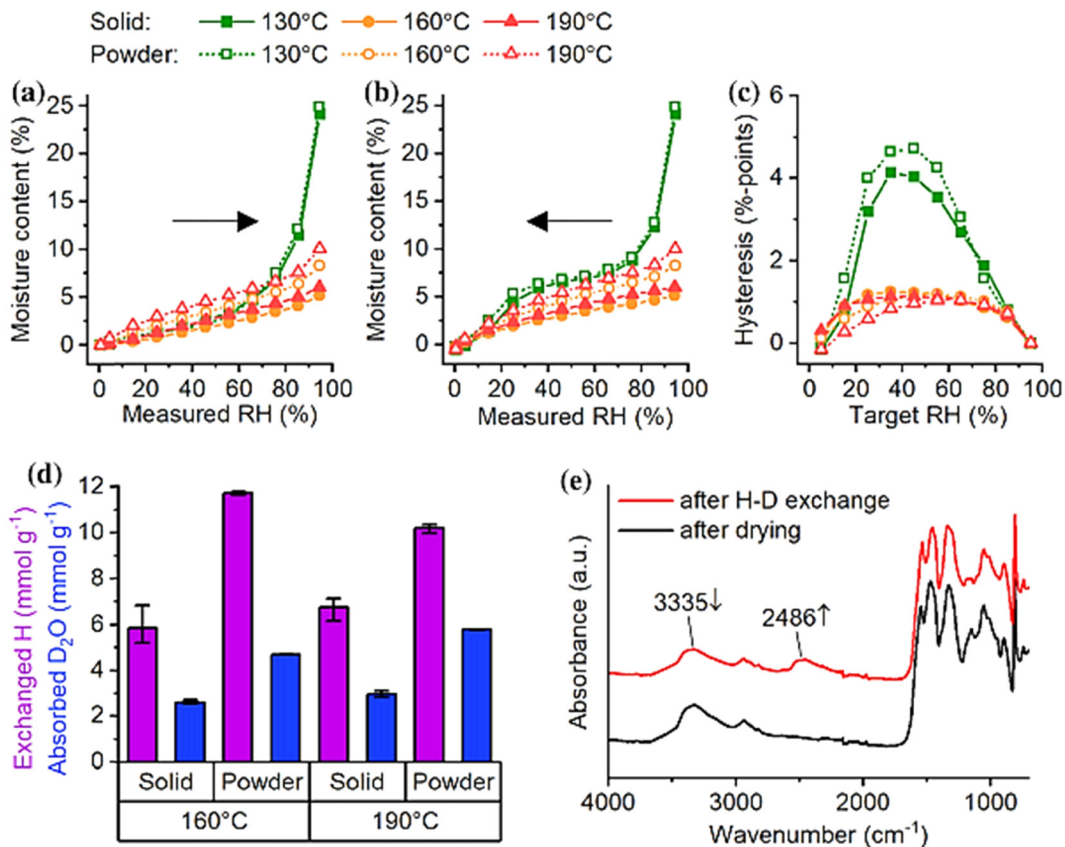


Figure 10: Sorption behavior of cured MF resin: absorption isotherms (a), desorption isotherms (b) and hysteresis (c) measured in the hygroscopic range (0–95% RH), exchanged hydrogen and absorbed D₂O measured by H–D exchange (d), as well as FTIR spectra of cured MF resin (190°C) after drying and H–D exchange (e). Each column in (d) represents the average of three replicates, and the error bars show minimum and maximum [99].

difficulty, environmental pollution, and impact on the wood strength restrict its application.

The future research of urea–formaldehyde modification technology should focus on the optimization and storage of treatment reagents to improve their stability and reduce pollution. The optimization of phenolic resin modification process should pay attention to the protection of

wood under a high-temperature environment during the modification process and improve the drying process. How to reduce production costs and protect the wood from cracking under dry and wet cycle conditions is the future development direction of the melamine-formaldehyde resin modification. The modified characteristics of the three resins are summarized, as shown in Table 3.

Table 3: Various modification characteristics of resins

Types	Advantages	Disadvantages	Improvement method
Urea resin	Improve the stability, durability, and strength of wood; the price is low	Difficult to store; the improvement of dimensional stability is not as good as that of traditional modification methods	Combined with other modifiers (polyethylene imine, <i>etc.</i>) to form a synergistic effect
Phenolic resin	Modulus and bending strength increase; improves resistance to white rot fungi and termites	High curing temperature, the drying process is difficult to control; the byproduct formaldehyde is harmful to human health	Change the hot compression process; extend the drying time after the process
Formaldehyde resin	The dimensional stability, durability, hardness, and compressive strength of wood are improved	High production cost; the cracking condition of wood products under drying and wetting cycles	Control the temperature and humidity during modification

5 Monomer modification of the polymer

5.1 The principle

Lumber itself has inherent shortcomings of large porosity, easy moisture absorption bibulous, poor dimensional stability, and so on; these defects make the wood easy to expand and contract, leading to cracking and deformation. To date, only a few modification methods, such as acetylation or furfuryl alcoholization, have been introduced to the market due to the limitations of scale and product cost. One approach to wood treatment relies on hydrophilic reactants penetrating the hygroscopic wood cell walls to form covalent bonds with the functional hydroxyl groups of natural polymers (lignin, cellulose, and hemicellulose).

Another method is to impregnate the organic monomer or oligomer into the wood under vacuum pressure and then undergo free radical polymerization of the organic monomer impregnated into the wood through free radical initiation or radiation. This is a monomer modification of the polymer. The organic monomer modification fills the cavities and pores on the wood. Therefore, the performance of wood is improved to a certain extent. This is the modification of polymer monomers. Introducing low-molecular-weight monomers into the wood and converting them into polymer chains that fill the micro/nano gaps in the cell wall provides permanent dimensional stability and water resistance.

5.2 Production process

The process of modifying wood with polymer monomers mainly includes the following steps. First, the sample is dried and then placed in a desiccator to equilibrate to room temperature. Second, the wood is loaded into a vacuum pressure impregnation tank to extract the air in the wood. Then, the reagent and catalyst are injected into the impregnation tank. The impregnation of wood is done at normal pressure. After the impregnation is completed, the pressure will be released and the remaining chemical reagents will be recovered [102,103]. The curing time, curing temperature, and the catalyst type are important factors that affect the modified wood. The curing time and temperature are important factors that affect whether the agent can be fixed in the modified wood. The curing time is within 24 h, and the curing temperature is within 120°C. The weight gain rate of wood increases with the

increase of time and temperature. When the curing time exceeds 24 h, as the curing temperature increases, due to thermal degradation, the wood component materials are damaged and the wood weight gain rate will decrease. A suitable catalyst can improve the fixing efficiency of the copolymer in wood. However, if the concentration of the added catalyst exceeds a certain limit, the weight gain rate of the treated material will not continue to increase [106].

5.3 Characteristics of the modification of polymer monomers

Organic monomers are usually formed by free radical polymerization, and free radical initiators are generally nonacidic or alkaline and so the degradation of wood components, especially hemicellulose, can be avoided during the modification process. The physical and mechanical strengths of the organic-monomer-modified wood can be improved to a certain extent. As early as the 1960s, Karpov *et al.* [102] used a mixture of polyethylene and polystyrene to impregnate the wood, which was cured by radiation to prepare plastic composite wood. The wood treated with polymethyl methacrylate (PMMA) or PVC/PMMA mixed reagents has anti-expansion coefficients as high as 87 and 83%, respectively [103]. Because the organic monomers polymerize in the wood cell channels and the cell wall micropores, thereby blocking the water outlet channels, the moisture absorption of the processed wood is significantly reduced [104]. In addition, with the development of nanotechnology, related products such as polymer nanocarriers can protect the hydrophilic active ingredients from excessive leaching, thus further improving the impregnation effect of wood preservatives [73].

Under normal circumstances, the polarity of the organic monomer is low due to the fact that the free radical polymerization reaction occurs mainly in the wood cell cavity and most of the monomer cannot react with the cell walls of the wood. So the combination with organic polymer chemical bonds between woods is weak and the cell walls of the wood and peeling off occur between polymers and the modified material durability is poorer [105].

Therefore, in the modification process, some unsaturated monomers with functional groups are usually added to increase the interface between organic monomers and wood, such as glycidyl methacrylate, acrylic acid, trimethylpropane trimethacrylate, polymaleic acid–maleic acid–maleic anhydride, *etc.* These monomers can improve the interaction between the polymer and wood cell wall components

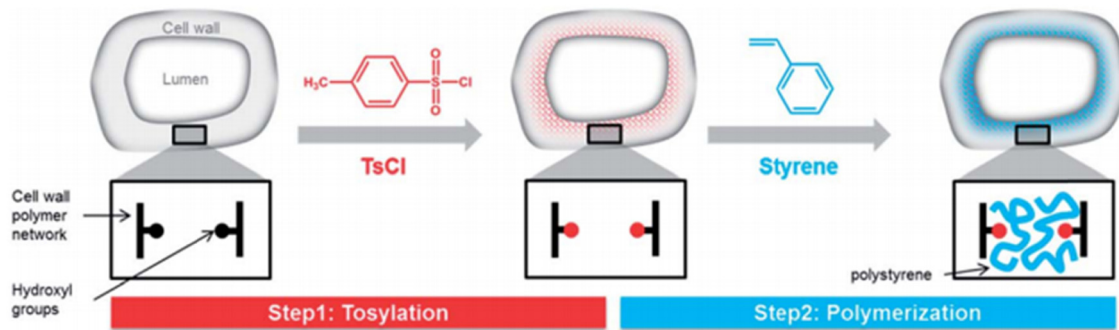


Figure 11: Schematic representation of the modification route: functionalization of cell wall –OH groups with tosyl groups, followed by *in situ* polymerization of the styrene monomer [107].

to a certain extent, thereby improving the properties of wood. Coupling agents are also commonly used to improve the interface compatibility between the polymer and wood cell wall components, thereby improving the overall performance of the monomer-treated wood. At present, the commonly used coupling agents mainly include isohydrate coupling agent, acid anhydride coupling agent, epoxy coupling agent, silane coupling agent, *etc.* [106,108]. Ermeidan *et al.* [107] invented a simple method to insert styrene monomer into the tosylated cell wall and carry out free radical polymerization under relatively mild conditions, as shown in Figure 11. This produces a low wood weight increase, reduces the water absorption rate of the wood cell walls, and significantly improves dimensional stability, as shown in Figure 12.

Another problem the modification of organic monomers faces is that traditional vinyl monomers are toxic and volatile, and sealing measures are required. Therefore, the cost is relatively high and the process is complex and difficult to promote and apply. Therefore, looking for a nontoxic, nonirritating odor, and nonvolatile monomer to replace the traditional vinyl monomer is of great significance

to the further development of vinyl monomer modification technology [109]. At present, scholars have developed water-soluble organic monomers such as 2-hydroxyethyl methacrylate (HEMA) [110] as an alternative.

6 Paraffin wax modification

6.1 The principle

Wax treatment is strictly not classified as a chemical modification because there is no chemical reaction in the whole treatment process. However, compared with chemical modification methods such as heat treatment, the process flow, and effect of wax treatment are similar to the above-mentioned chemical modification. Therefore, this method is also summarized here. Paraffin wax is a cheap byproduct of petroleum refining, mainly composed of normal alkanes with carbon numbers C18–C50 [111]. As shown in Figures 13 and 14 [112], during the paraffin impregnation modification process, the impregnated paraffin

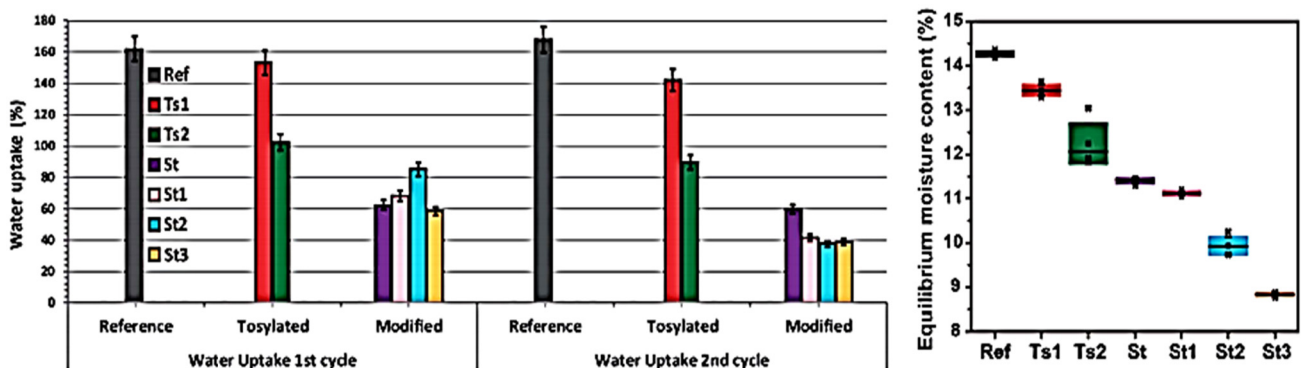


Figure 12: Left: water uptake values for unmodified wood (Ref), tosylated wood (Ts1, Ts2), and modified wood (St, St1, St2, and St3). Right: equilibrium moisture content (EMC) values of reference, tosylated, and modified samples [107].

is widely distributed in the wood ducts, tracheids, and fibers, reducing the pore size and porosity of the wood [112], and draining a large amount of water from the wood cells. The wood water absorption channel is reduced and the growth and reproduction of wood-rot fungi are inhibited. At the same time, the wax forms a protective layer of wax film on the surface of the wood to isolate the moisture in the soil from contacting the wood, blocking the contact of termites with the cellulose in the wood and inhibits decay and prevent termites. This is of great significance to the promotion and application of wood [113–116].

6.2 Production process

Wax treatment is strictly not a chemical modification because there is no chemical reaction throughout the treatment. However, compared with physical modification methods such as heat treatment, the process flow and effect of wax treatment are more similar to the above-mentioned chemical modification methods. The surface coating film is convenient and quick but there is a disadvantage that once the surface layer is destroyed, the modification effect will disappear. The method of impregnation is to impregnate the wax into the wood by means of normal pressure or high pressure. Some scholars at home and abroad have explored the method of normal pressure and high-pressure wax impregnation of modified wood to isolate the contact of moisture and wood, which improves the moisture absorption and dimensional stability of the modified wood to a certain extent [117]. Nowadays, the “wax injection method” is usually used to modify wood with paraffin wax. Wax injection

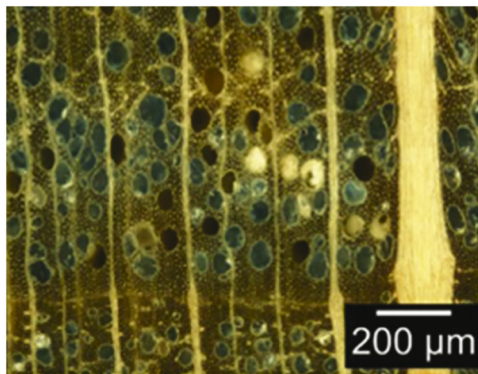


Figure 13: The cross section of wax-treated material under a light microscope [112].

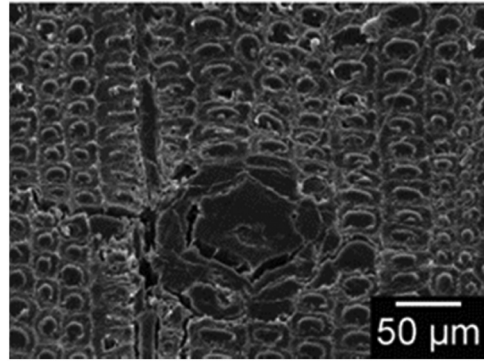


Figure 14: The cross section of the wax-treated material under an electron microscope [112].

technology is to inject wax into the wood to drain excess water and fill the gaps between the pipe holes and cells of the closed wood to improve the dimensional stability of the wood without affecting the mechanical properties of the wood. The wax injection process consists of three stages. The first stage is vacuuming, putting the wood into a pressure test tank, vacuuming, and maintaining the pressure for a period of time before entering the second stage of high-pressure immersion, and injecting molten wax into the instrument. Soak the solution, apply pressure and hold for a while. The third stage is pressure relief and cooling. After the wax treatment is completed under normal pressure for a period of time, the wood can be taken out.

The wax injection time, treatment temperature, wax selection, and wood pretreatment methods will all affect the effect of the wood modification. If the wax injection time is too short, the impregnation effect is poor; if the wax injection time is too long, too much wax will enter the interior, which will damage the cell fibers and will easily lead to residual moisture, which makes it difficult to stabilize the wood. The processing temperature needs to consider the boiling point of the wax and the temperature that the cell cavity wall of the wood itself can withstand. Wax used in the wax treatment previously mostly used low melting point paraffin, which can improve the waterproof performance of wood and increase the density and strength of the treated material, but the surface of the treated material is not resistant to high temperatures. Therefore, some scholars have used high melting point wax to improve the high-temperature stability of the modified wood, and the effect is good [117].

Wood pretreatment can reduce wood hydroxyl groups, increase wood interface compatibility, increase the permeability of paraffin wax in wood, and obtain modified materials with better properties [112]. Scholz *et al.* [118] found that the pretreatment of wood with heat treatment,

acetylation, DMDHEU, and then impregnation modification with montan wax can significantly improve the wood modification effect. After heat treatment before paraffin modification, the bending strength and Brinell hardness of the wood were increased by nearly half. The compression rate and bending modulus of acetylated wood treated with wax were increased by 47 and 66%, respectively, compared with acetylated wood. The influence of paraffin modification on the moisture content of the material is the solid content of paraffin emulsion, drying temperature, and drying time in descending order [119].

6.3 Modification characteristics of paraffin wax

Paraffin treatment can significantly reduce the water absorption of wood, thereby improving the dimensional stability of wood, and at the same time, it can also improve the mechanical properties of wood to a certain extent. Scholz *et al.* [118] found that the moisture absorption of wood modified by paraffin wax was reduced and the dimensional stability was improved. In addition, the choice of wax affects the result of the impregnation. Non-polar or low-polar wax penetrates the wood deeper than high-polar wax, despite the similar molecular size and viscosity. Paraffin impregnation can also improve wood light aging resistance, weather resistance, and resistance to decay fungi. Scholz *et al.* [120] studied the termite resistance of the modified wood, and the results showed that the quality loss of wax-treated beech was reduced and the amide wax showed excellent termite resistance. The antitermite performance is related to the type of wax and the feeding preference of specific termites. However, wax treatment can only reduce the attack of termites and cannot be completely avoided. Brischke and Melcher [121] studied the long-term outdoor durability effect of wax-treated wood, and the results showed that paraffin treatment can significantly improve the outdoor corrosion resistance of wood but the modification effect becomes worse with time. Compared with wood treated with biological insecticides, the water capture and internal decay of wax-treated wood require more attention. Paraffin wax is also widely used in wood preservative treatment, forming a composite system with preservatives to work together on wood. Liu [122] and Jiamin Wang *et al.* [123] studied the treatment effect of the copper azole–paraffin emulsion composite system on wood, and the results showed that the durability and dimensional stability of wood treated by

the composite system was significantly improved. In addition, paraffin wax can also be used as a water-repellent agent and preservative to treat wood, which can effectively enhance the loss resistance of preservatives without affecting the impregnation effect. The synergy between the two can play an effective role in wood modification. The study of Qirui *et al.* [124] provides evidence for this conclusion.

Although paraffin treatment of wood can improve some properties of wood, there are still some problems that restrict its wide application. First of all, paraffin wax is mainly used to modify wood by means of physical filling, so there are common problems such as incomplete penetration and uneven penetration, which need to be improved by pretreatment of wood. Second, the temperature of paraffin wax modification is not easy to control, and the melting point of paraffin wax must be taken into account with the high-temperature stability of the wood itself. Moreover, the wood treated with low-melting paraffin wax is not resistant to high temperatures and it is difficult to maintain its hydrophobicity. The treatment of high-melting wax is a solution. The process elements, characteristics, and improvement directions of paraffin modification are summarized, as shown in Figure 15.

7 Nanotechnology

With the in-depth study of various properties of wood [125–128], researchers found that wood itself has defects such as poor dimensional stability. These problems limit its use. Traditional chemical modification technologies often have shortcomings such as insufficient impregnation and large side effects. Fortunately, with the development of nanomodification technology, it is possible to solve these problems. At present, the main applications of nanomaterials include three aspects: nanometal impregnated wood, nanocarrier, and nanocoating. The use of nanomaterials can not only be used as a supplement to traditional modification methods and reduce the side effects of traditional modification but can also be used alone as a new modification method to give the wood more excellent properties. For example, nanomaterials can be made as carriers of fungicides. This technology can solve the problem that the activity of active substances is difficult to maintain for a long time, thereby improving the anticorrosion performance of wood. There is no doubt that nanochemical modification technology will become an important part of wood modification methods.

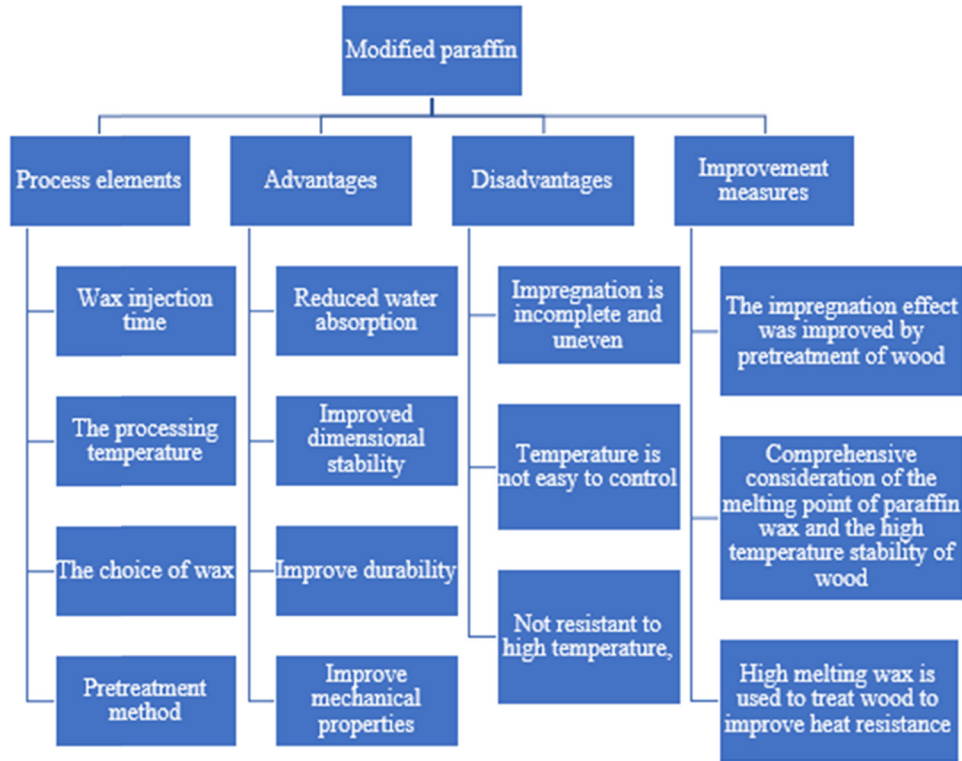


Figure 15: Paraffin modification process elements, advantages, and disadvantages, and improvement methods.

7.1 The principle

Nanotechnology is used for manipulating materials with structure sizes between 1 and 100 nm. Compared with traditional materials, nanomaterials have a higher surface-to-volume ratio, and thus, exhibit greater activity in surface-related phenomena [129]. Therefore, various innovative applications can be explored in many disciplines through nanotechnology. The application of nanotechnology in wood science is mainly to enhance the durability and stability of wood through impregnation, coating, and other processes. This is due to the fact that the size of the nanoparticles is much smaller than the voids in the wood cell wall, and the nanoparticles can penetrate deeply into the wood base [130,131], changing the surface properties and various properties of the wood. In addition, if the size of the nanoparticles is smaller than the diameter of the pits in the wood cell wall, complete penetration and uniform distribution can be achieved [132], making the modification effect more sufficient and lasting. This is difficult to achieve with traditional physical and chemical modification methods. At present, there are three main ways to use nanotechnology to modify wood: nanometal impregnation, polymer nanocarrier-assisted modification, and coating treatment.

7.2 Application in wood modification technology

7.2.1 Nanometal-impregnated wood

Decades ago, metal nanoparticles were used for wood impregnation to slow down its degradation rate [133]. Due to their small size, nanoparticles can penetrate deeply and evenly into the pores of the wood, thereby protecting the wood [134]. Wood impregnation can be divided into diffusion, capillary, and pressure treatments. In industry, the impregnation of wood is mainly carried out by high pressure. Vacuum and pressure cycles are used to impregnate the material into the wood. In the laboratory, one or a combination of techniques is usually used to impregnate wood. Common methods include pressure, ultrasonic impregnation, and microwave-assisted impregnation.

Nanometals for impregnation can usually be synthesized using two chemical methods, one is the gas-phase method and the other is the liquid-phase method [135,136]. The synthesis method will affect the stability, hydrophilicity, and other physical and chemical properties of nanometals [137]. The produced nanometal usually has the characteristics of high surface-to-volume ratio, uniform particle size distribution, and good stability.

The use of nanometal impregnated wood can significantly improve the durability of wood and other properties and are probably the following reasons. First, in the process of dispersing nanoparticles in wood, nanoscale metals may interact with bacteria, causing bacteria to die or deactivate the enzymes required for the degradation reactions [137]. In this process, the degree of interaction between the metal and the wood matrix should be controlled. A low degree of interaction will cause the impregnated substance to separate from the substrate, and the material loses its function [138]. Second, when using nano-copper compounds to treat wood, in some cases, the fungus may not be able to recognize nano-copper. After the nanoparticles enter the fungal cell wall, they will form reactive oxygen species with the fungal cell, which affects the fungus's erosion of wood. Third, nanoparticles will dissolve after entering fungal cells, interfere with the steady-state process of fungal cells, and produce toxic reactions, thereby reducing the fungal erosion of wood [139].

Metal nanoparticles such as copper, gold, and silver are widely used due to their good and stable impregnation effect. Nano-copper can protect wood without chromium and arsenic [140]. Akhtari and Nicholas research shows that the use of nano-copper-impregnated wood can greatly improve its resistance to termites [141]. In the presence of polystyrene, nano-copper oxide treatment of wood can further improve dimensional stability [142]. The treatment of wood with nano-silver-based compounds can not only improve the resistance of wood to fungi but also the water absorption capacity of wood

and improve the dimensional stability of wood [143]. Zinc-based nanocompounds can also be used to impregnate wood. Studies have shown that compared with copper, zinc-based compounds are more effective in resisting termite and white-rot fungal rot (Figure 16). After leaching, only zinc oxide can play an effective protective role. Bak and Nemeth [144] compared the effects of zinc oxide, zinc borate, copper borate, silver, and copper on fungi and found that zinc oxide can provide long-lasting and effective protection to the wood, even after leaching. This is difficult to achieve with other materials. Francés Bueno *et al.* [145], while improving the durability of wood, found that after the wood was impregnated with SiO_2 , TiO_2 , and ZnO_2 , its fire resistance also improved. Among them, 3% silica treatment has the best effect.

Nano-silica is also a commonly used nanomaterial for impregnation. Because of its low price, easy availability of raw materials, and good modification effects, it has been widely used. Traditional modification methods usually use modifiers such as sodium silicate. The modifier is introduced into the wood through a double-diffusion method to improve the performance of the wood. This modification method greatly improves the wear resistance and flame retardancy of the wood [146] but has poor water resistance. This is because the various salt components in the water glass and the unreacted salt in the wood cell cavity have strong hygroscopicity. In order to improve this, researchers began to use nano-silica to modify wood. There are usually two ways to use, one is to directly use nano-silica sol as a modifier and use the sol-gel method to prepare wood-silica composites [147]. Nanosol is a

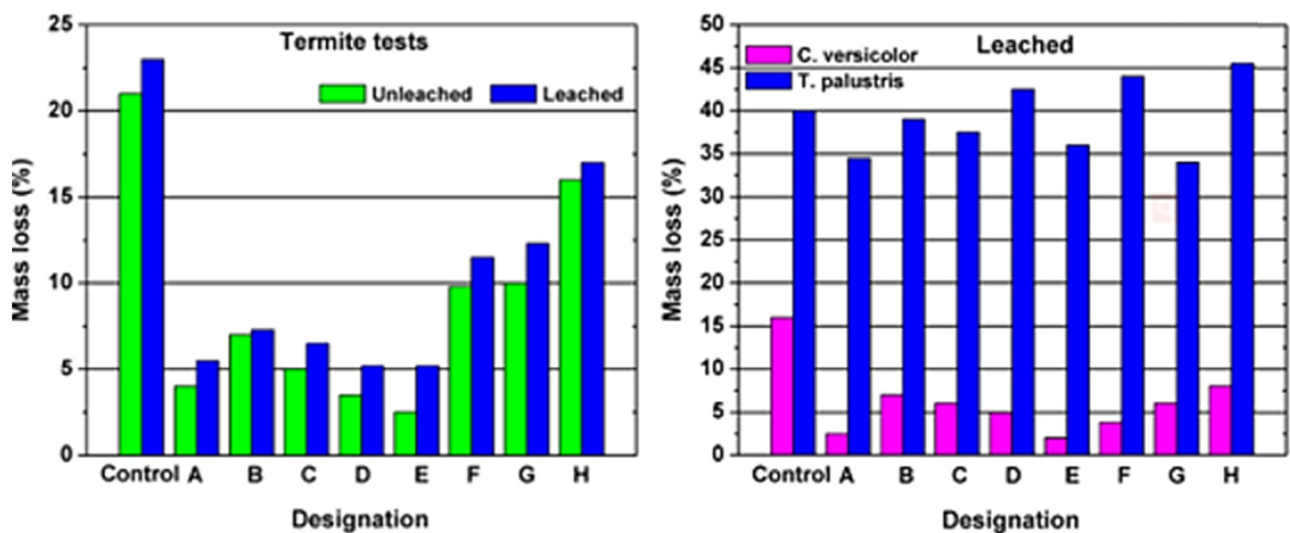


Figure 16: The mass loss (%) following termite resistance for leached and unleached pine wood: (a) nano-zinc oxide; (b) nano-zinc oxide plus binder A; (c) nano-zinc oxide plus binder B; (d) nano-zinc borate; (e) nano-zinc borate plus binder A; (f) nano-copper oxide; (g) nano-copper oxide plus binder A; and (h) nano-copper oxide plus binder B [134].

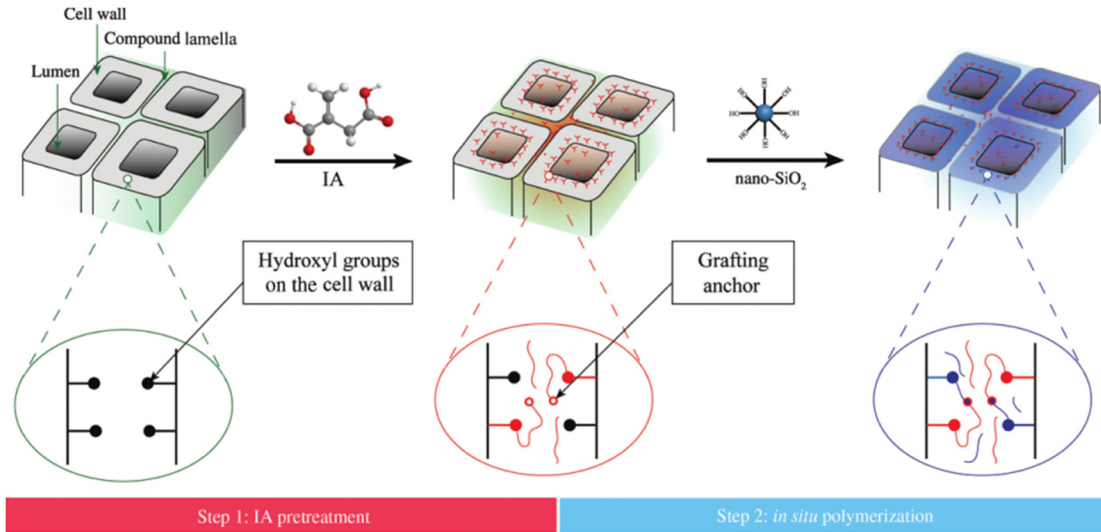


Figure 17: Schematic of the modification process: functionalization of the wood cell wall $-OH$ groups with IA, followed by *in situ* polymerization of nano- SiO_2 [149].

nanoscale transparent dispersion made by mixing inorganic particles with water or organic liquid. The high surface-to-weight ratio and low size allow it to easily penetrate the wood and distribute it evenly [148]. The impregnation effect of silica sol depends on the impregnation time. As the immersion time increases, the quality and quantity of the SiO_2 gel in the composite material increase. The processing technology stimulates the hydroxyl group in the wood to bond with the cell wall and the wood OH group combines with the silica sol to form a covalent bond. The microstructure of wood can be transformed into a composite material and the thermal and mechanical properties are significantly improved. Another method is to graft nano-silica as a reinforcing material onto the modified wood to further improve its various properties. For example, Han *et al.* [149] grafted nano- SiO_2 onto itaconic acid (IA)-modified wood, which significantly increased the thickness of the cell wall, reduced the moisture absorption of the wood and improved the dimensional stability of the wood. Figure 17 shows its technological process.

In addition to using a single metal nanoparticle to modify wood, multiple metal nanoparticles can also be mixed. For example, Cristea *et al.* [150] studied the effect of the mixed use of zinc oxide nanoparticles and silver nanoparticles on wood and the results showed that the mechanical properties of wood, such as hardness, adhesion, abrasion resistance, and water vapor diffusion resistance, were slightly improved. Mixed nanoparticles can protect the wood from weathering problems such as ultraviolet rays [151].

It is worth mentioning that nanochemical modification methods can be combined with traditional modification

methods to obtain better modification effects. For example, the traditional thermal modification method will cause the degradation of low cell wall polymers, which will lead to the decline of wood mechanical strength and other properties. If the wood is impregnated with nano-silver suspension, the degree of conversion of amorphous cellulose to crystalline cellulose can be increased, and the cross-linked part of lignin can be increased. This can improve the adverse effects of thermal degradation at high temperatures, improve thermal conductivity, and modification effects [166]. The modification process is shown in Figure 18.

Current studies have shown that metal nanoparticles mainly composed of copper, silver, boron, and zinc have a better control effect on white rot and brown rot. But they have a poor control effect on mold [152,153]. Some metal nanoparticles are still in the preliminary research stage but they have great potential for development. For example, titanium dioxide (TiO_2) has great potential in wood preservative treatment. Preliminary studies have shown that the use of TiO_2 treated wood has significantly improved antibacterial, antifungal, and anti-ultraviolet properties [154]. It is foreseeable that the application range of wood [155,156] will be expanded due to the popularization of these technologies.

7.2.2 Polymer nanocarrier

The polymer nanocarrier is a submicron carrier delivery system in the nanoscale and microscopic category, that

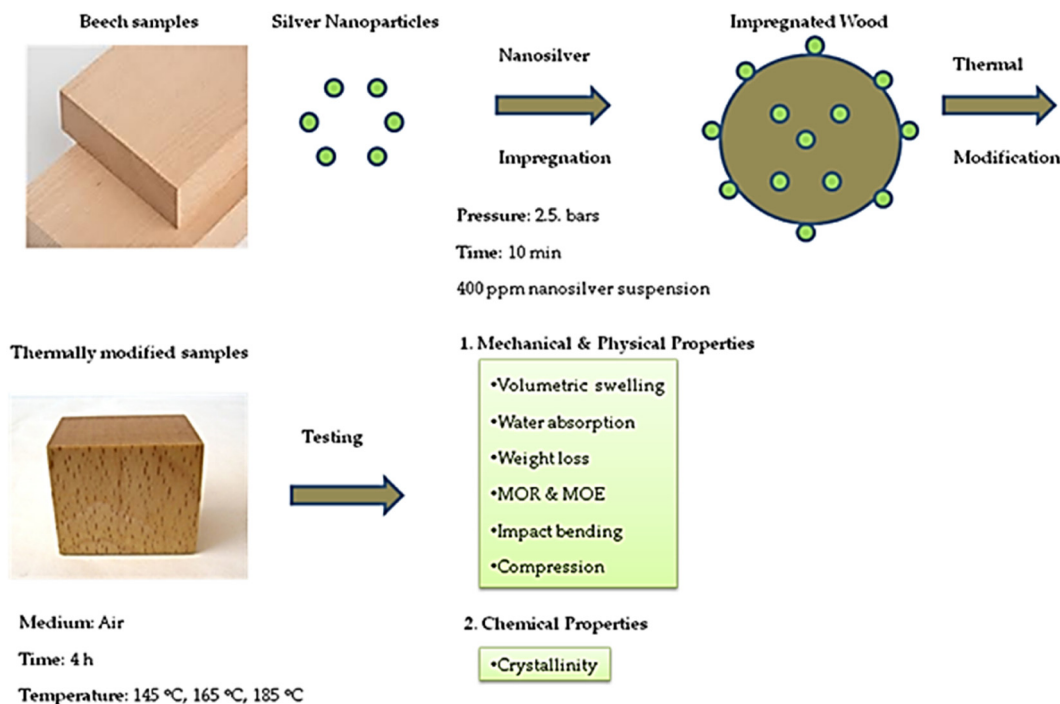


Figure 18: Technological process of impregnating nanometer silver suspension [166].

is, nanoparticles are used as carriers to conduct or transport certain substances through physical encapsulation or chemical bond connection. At present, nanocarrier technology is widely used in the field of medicine [157]. In the wood preservative industry, compared with nanometal impregnation and coating treatment, there are fewer related research literature studies on this technology but can be expected in the future.

The application of polymer nanocarriers in wood preservation is mainly as a carrier of biological fungicides. Nanocarriers can be introduced into the wood while maintaining the high activity of active substances. The usual operation is to combine the biological fungicides with polymer nanoparticles, suspend the nanoparticles in water, and use this suspension to perform conventional pressure treatment on the wood, thereby introducing the biological fungicides into the solid wood [158]. When the nanocarrier is injected into the tree trunk, it needs to be transported through the xylem or phloem with a diameter of a few microns. The small size of the nanomaterial makes this step possible. Once it enters the wood, the polymer matrix acts as a storage for the biocide and controls its release rate in the wood, while protecting the unreleased biocide from exposure to the environment. The colloidal stability of nanocarriers determines their transport characteristics through plant vascular tissues. Its colloidal stability is determined by chemical

design, surfactants, or surface charges that interact with the complex under plant conditions [159]. The stability of the final product is closely related to the properties of the polymer matrix. The polarity of the polymer matrix can effectively slow down the release rate of the bactericide but it also affects the particle size and suspension stability of the nanoparticles. As the size of nanoparticles increases, their suspension stability decreases. The release rate of the bactericide is negatively related to the hydrophobicity of the polymer matrix. Encapsulation of active ingredients in polymer nanocarriers can be carried out by a variety of techniques, such as nanoprecipitation, emulsification, emulsification, and coagulation.

At present, some fungicides, such as chlorothalonil, have been successfully added to polymer nanoparticles [160]. Polyvinylpyridine and polyvinylpyridine-styrene can be used as the polymer matrix. However, how to maintain control of the size and suspension stability of the nanoparticles during the whole process is still a problem, and the surfactant system of the nanoparticles needs to be improved.

7.2.3 The coating process

Although traditional coatings can improve the mechanical strength of wood to a certain extent, they will have a

negative impact on the flexibility of wood and the transparency of the coating system. In addition, the adhesion of the coating and the substrate is weak, poor wear resistance, poor durability, and other problems cannot be ignored. But the emergence of nanocoating makes it possible to solve these problems [161]. Due to its large surface-to-volume ratio, nanocoating can be used for large-volume materials that require extremely thin coatings. While improving wood durability, mechanical properties, fire resistance, UV absorption, and reducing water absorption, their nanosize ensures the maintenance of transparency.

The common method to enhance the function of the coating is to add nanoparticles [162]. There are usually two methods for adding nanoparticles to coatings: solution mixing and *in situ* mixing [163]. The solution mixing method is usually to add the polymer in a suitable solvent and make it uniformly dispersed. Then the nanocoating is applied to the wood surface by brushing or dipping [164]. The *in situ* mixing method is a chemical method in which the monomers are directly combined and then polymerized. Nanoparticles are synthesized *in situ* by chemical methods such as hydrothermal reaction or sol-gel deposition on the wood surface. The coating made by *in situ* polymerization has better adhesion and is not easy to separate.

Nanocoatings can significantly improve the durability, water repellency, dimensional stability, and UV absorption properties of wood. Nanometal oxides such as zinc oxide have strong antibacterial properties [165]. Therefore, the nanocoating made by adding nanoparticles can prevent the growth of bacteria, fungi, and other microorganisms. Because of its own characteristics, wood easily interacts with water, which can cause problems such as moisture and cracking. Conventional chemical modification methods can improve the hydrophobicity of wood but they have not completely isolated the contact between wood and water [166], and the chemical reagents used in the treatment process can easily pollute the environment. The use of nanochemical modification technology does not have these problems. Wu *et al.* [167] used zinc oxide nanoparticles and stearic acid-modified products to make a superhydrophobic coating on the surface of poplar wood, which not only has stronger hydrophobicity but also further improves the dimensional stability of the modified wood and is easy to prepare. The production process is green and environmentally friendly. In addition, many researchers have used other materials to make multifunctional coatings [168], which are widely used in various fields and have great development prospects.

Ultraviolet radiation is an important factor in the degradation of wood. The polymer structural components of wood will be decomposed during the photodegradation process [169], which will reduce the water resistance of wood and cause further biodegradation. Nanoparticles can be used to improve the UV resistance of the coating and provide a high level of UV protection without affecting the surface transparency. Compared with natural materials, the larger surface-to-volume ratio of nanoparticles greatly improves the efficiency of blocking ultraviolet rays. However, the absorption efficiency of different nanoparticles is also different. Åkerlund *et al.* [170] have shown that nano-zinc oxide has a very wide absorption band in the ultraviolet region, so the performance is very good. However, the absorption band of copper oxide is relatively narrow, and the color change is observed to be large after the test, which is not suitable as a coating that blocks ultraviolet rays. In addition, Cataldi *et al.* [171] found that if the material has the largest absorption rate in different parts of the ultraviolet spectrum, a synergistic effect can occur. Based on this view, complex chemical bonds can be formed between inorganic and organic nanomodified materials to enhance the modification.

7.3 Problems

Although nanochemical modification has many advantages, there are still some problems that need to be solved. First of all, in the process of impregnating wood with nanomaterials, due to the higher surface energy of nanomaterials, accumulation may occur, resulting in the formation of microcomposite materials in the wood substrate. This area will become the weak point of the wood, which is easy to damage under the action of a larger load. Therefore, it is necessary to modify the surface of nanomaterials to improve their stability and make them easy to disperse. Second, although nanotechnology has been widely used in wood processing, there have been more and more criticisms and discussions about the potential health and environmental risks of nanomaterials recently. Studies have shown that some nanoparticles may cause inflammation of cytokines and have certain secondary genetic toxicity [172]. Fortunately, as far as observations are concerned, the toxicity of nanomaterials is not severe and does not cause bioaccumulation. The toxicity of nanomaterials needs further research to understand their causes. Based on this, we need to find ways to reduce or even eliminate toxicity.

8 Conclusions

In this paper, the principle, production process, advantages and disadvantages, and improvement direction of various wood modification methods were combined and summarized. This article provides ideas and references for the research and development of more economical and effective modification technologies. Various wood chemical modification methods have their own characteristics. At present, only acetylation and furfuryl alcoholization modification methods have been put into use on a large scale for commercial production because of lower cost, simple manufacturing process, and excellent modification effect. Thermosetting resin modification, polymer monomer modification, nanotechnology, and paraffin wax modification are still in the research stage due to high cost or excessive influence on wood strength. The advantages and disadvantages of various modification methods and the development direction are summarized as follows.

- (1) Acetylation significantly reduces the moisture absorption of wood, improves wood strength, resistance to fungal corrosion, and fire resistance. The effect of acetylation treatment is generally evaluated by the weight gain rate of treated wood. The main factors that affect the weight gain rate of acetylated wood are tree, wood size, heartwood, wood moisture content, reaction solution type or ratio, pretreatment conditions, reaction conditions, and so on. The difficulty that acetylation modification needs to overcome in the future is to reduce the influence of byproducts such as acetic acid on the strength of wood and the recovery of chemical reagents, which can be achieved through optimization of catalysts and modification processes.
- (2) The advantages of furfuryl alcohol treatment are that the materials are easily available, the cost is low, and the effect is good. Furfuryl alcohol modification can significantly improve the compressive strength, dimensional stability, aging resistance, and biological durability of wood along the grain. Moreover, the combustion of modified wood does not emit volatile organic compounds. However, the modification of furfuryl alcohol will affect the toughness, impact strength, and color of the wood. The functional mechanism of furfuryl alcohol modification, especially the polymerization of furfuryl alcohol in wood, needs further study, which is of great significance for the directional improvement of the modification process and modification effect.
- (3) The performance of polymer organic monomers improves the physical and mechanical strengths of wood to a

certain extent and significantly reduces the moisture absorption of wood. The difficulty of modification of the polymer monomer is how to accurately insert the monomer into the cell wall and improve the interface compatibility between the polymer and wood cell wall components. Therefore, in the modification process, some unsaturated monomers with functional groups are usually added to increase the interface bonding between organic monomers and wood. In order to solve the toxic and volatile problem of a traditional vinyl monomer, a water-soluble organic monomer can be used to replace the nontoxic, nonpungent odor, and nonvolatile monomer.

- (4) Modification of wood with urea-formaldehyde resin can improve its stability, durability, and strength. Phenolic resin modification can effectively improve the resistance of wood to white rot fungi and termites. The dimensional stability, water resistance, and swelling properties of melamine resin-modified wood are improved. The urea-formaldehyde modification solution is not easy to store and the modification of phenolic resin will produce free formaldehyde, which is harmful to the human body. Modification of formaldehyde resin has high production costs and this wood product is prone to cracking under dry and wet cycling conditions. It is the future development trend to optimize the process or material so as to control it simply and effectively.
- (5) Paraffin wax modification can significantly improve the dimensional stability, mechanical properties, and durability of wood. However, because paraffin wax is mainly used to modify wood by physical filling, there are common problems such as incomplete penetration and uneven penetration, which need to be improved by wood pretreatment. Second, the temperature of paraffin wax modification is not easy to control and the melting point of paraffin wax must be considered in conjunction with the high-temperature stability of the wood itself. The use of high melting point wax is a way to solve the shortcomings of wood's inability to withstand high temperatures.
- (6) The main applications of nanomaterials include three aspects: nanometal impregnated wood, nanocarrier, and nanocoating. The use of nanometal impregnated wood can give the wood more and more excellent properties. Nanocarriers can solve the problem that the activity of active substances is difficult to maintain for a long time, thereby improving the anticorrosion performance of the wood. Compared with traditional modification methods, nanochemical modification is more stable and durable, and more functional.

However, the potential risks to health and the environment require further research and assessment.

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