Organosilane-modified Wood Materials: A Review of Research and Applications

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This review article discusses the use of organosilane (OS) compounds and their derivatives to modify wood materials. The emphasis in this work is on trialkoxysilanes and effects of their reactions with hydroxyl groups at wood surfaces. The versatile properties of OS make them ideal for improving the water resistance, weather resistance, antibacterial properties, and dimensional stability of wood. This article provides an overview of recent techniques and methods used to modify wood substrates with OS, highlighting the main findings and advancements in the field. Additionally, the article suggests future research directions, including innovative modification mechanisms, strategies to control the pollution caused by biomaterial-based silanes, and the investigation of the influence of wood permeability on silane modification.

DOI: 10.15376/biores.18.3.Liu

Keywords: Organosilane; Trialkoxysilanes; Modification; Wood; Dimensional stability

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INTRODUCTION

As a natural, green, and sustainable material, wood is widely used in indoor and outdoor conditions. However, wood has some disadvantages, such as color change, deformation, decay, and insect attacks, which can be made worse by changes of moisture in wood, as well as changes in temperature (Liu *et al*. 2019). Physical and chemical modifications have been developed to improve properties and functions of wood (Liu *et al*. 2021; Yang and Jin 2021; Yang *et al*. 2021; Tu *et al*. 2021). Among them, wood treated with organosilane (OS) is a relatively new wood modification method to improve the physical and chemical properties of the wood. Within this article, the term OS almost always will be referring to a specific class of compound, the trialkoxysilane family of reagents. Research on this technology started in the 1970s, and it has been gradually widely considered and applied recently. The organosilane-modified wood materials have advantages in their resistance to water absorption, fire, some microorganisms, and weather (Zucchi *et al.* 2006; Huang *et al.* 2017; Aziz *et al.* 2021), and they are commonly used in construction, decoration, furniture, electronics, and aviation. There are many challenges in the research of organic silane-modified wood materials, including improving the modification efficiency, studying the environmental impact of the modification, and prolonging its service life. Therefore, much research must be done to improve the performance and sustainability of organosilane-modified wood materials.

Silane coupling agents are a commonly used class of OS modifier. One end of the molecule reacts with inorganic particles or wood after hydrolysis, and in some cases the other end reacts with polymers. This class of amphiphilic (*i.e.* surface-active) molecules can significantly improve the interfacial performance between nanoparticles and polymers (Jamil *et al.* 2018; Aziz *et al.* 2020). In addition, using this type of modifier in natural fiber composites can improve fiber adhesion (Zheng *et al.* 2021); thus researchers have been focusing on cellulose composite preparation (Sabarinathan *et al.* 2020).

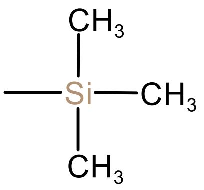
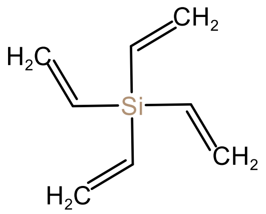
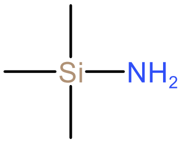
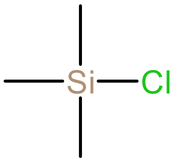
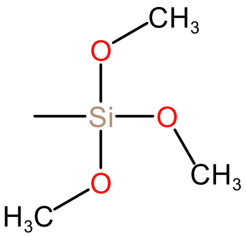
In the modification of logs, silane derivatives (especially alkoxysilanes) act as efficient and straightforward additives on the surface of or inside the wood (Wang *et al.* 2020a; Zhou and Liu 2022). Through hydrolysis and condensation reactions, alkoxy-silane reacts with the two hydroxyl groups on cellulose simultaneously to form stable silico-oxygen bonds and establish Si-O-Si bonds with other silane molecules (Donath *et al.* 2004; Panov and Terziev 2009; Xie *et al.* 2011; Broda *et al.* 2019b). Also, certain silane derivatives, such as alkyl trialkoxy silanes, are ideal wood waterproofing agents due to their hydrophobic properties and ability to stabilize the wood dimensions through cell-wall expansion (Hill 2007; De Vetter *et al.* 2010; Xie *et al.* 2011; Canosa *et al.* 2018).

Because of its unique chemical properties, silane can replace one end of its molecule with other organic or inorganic groups, which determines whether it can modify wood through different reactions. However, the most common goal of OS modification of wood is to try to change the relationship between the wood components and moisture to limit the dimensional changes in wood or changes in other properties of the material (Donath *et al.* 2006).

ORGANOSILANE CHARACTERISTICS

OSs are compounds in which one of the hydrogen atoms in the simplest silane (SiH_4) molecule is replaced by another atom or group of atoms. Their general formula is R_nSiH_{4-n}. Here R is an organic functional group, often including alkyl, amino, sulfhydryl, glycidyl oxyl, methoxy, or vinyl functional groups, which can provide organic compatibility (Table 1). Silanes can have organic and inorganic groups and organic and inorganic commonalities. They can build links between two substances with different properties, reducing interfacial defects and tensions in composites (Xu and Zhou 2023). There can be a very wide range for the fourth substituent on the silicon atom, it can be methyl or long-change alkyl, and obtain different effects, or different groups to produce the same effect. After the substitution of hydrogen atoms in the Si-H bonds with alkoxy groups in silanes, the resulting compounds can be classified as monoalkoxysilanes, dialkoxysilanes, trialkoxysilanes, or tetraalkoxysilanes based on the degree of substitution. Similarly, H can be substituted by other elements and groups, such as chlorine, which gives the chlorosilanes. Alkoxysilanes typically require some water to complete the bonding with the substrate, while reactions with chlorosilanes must be carried out in the complete absence of water (Szkek *et al.* 2022). Since the focus of this article involves plant-based materials, which typically contain some water at intermediate values of relative humidity, emphasis will be placed on trialkoxysilane treatments.

Table 1. Some Examples of Silanes with Simple Structures

Substituent	Example	Molecular Formula	Structural Formula
Alkyl	Trimethylsilane	$C_3H_{10}Si$	
Alkenyl	Tetravinylsilane	$C_8H_{12}Si$	
Amino	Trimethylsilylamine	$C_3H_{11}NSi$	
Halogen	Chlorosilane	ClH_3Si	
Alkoxy	Trimethoxysilane	$C_3H_{10}O_3Si$	

Modification Mechanism

OS and its derivatives can be used in a variety of conditions. The temperature, pressure, and pH of the conditions all affect the reaction of silane. The primary reaction of silane with wood includes two aspects (Fig. 1). The first is the hydrolysis of silane derivatives by reaction with a small amount of water, after which the hydrogen bond of silanol is more electrophilic and reactive. At this stage, the reaction tendency can be controlled by the solvent, alcohol, water, and pH.

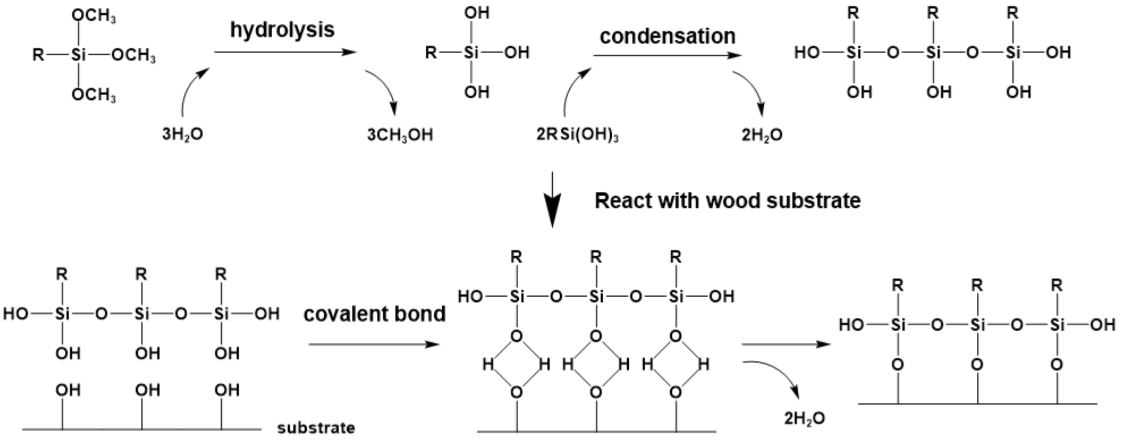


Fig. 1. Mechanism of wood modification by silane (re-used with permission from Taylor & Francis, Runumi and Ajay 2021)

The second is that in which the hydrolyzed product combines with the hydroxyl group on the substrate. The reaction initially involves hydrogen bonds, which become transformed to covalent bonds when the material is dried or thermally cured, and water is released (Parr and Finzel 1979). In most cases, silane binds to the hydroxyl group on cellulose or other wood polymers.

Several potential reactive groups in wood-based materials and OS can form covalent or hydrogen bonds between their molecules. The main constituents of wood-based materials (cellulose, lignin, and hemicellulose) all contain "potential reactive moieties". For example, cellulose contains hydroxyl groups, hemicellulose contains hydroxyl groups along with some carboxyl groups, and lignin contains some ester and phenolic hydroxyl groups. In OS, highly reactive alkoxy groups can polymerize by reacting with other silane molecules or different chemicals (Donath *et al.* 2004). In addition, OS can contain several different functional groups that facilitate interactions with wood polymers (Tingaut *et al.* 2006). Various silanes have been shown to react with cellulose, hemicellulose, and lignin (Robles *et al.* 2018; Siuda *et al.* 2019; Li *et al.* 2020; Neves *et al.* 2020; Popescu and Broda 2021). The reaction between the OSs and the constituents of wood, including lignin, cellulose, and hemicellulose, is easily initiated due to the presence of numerous electrophilic and nucleophilic functional groups within these wood components, which are also present in OS molecules. Additionally, the complex structure of wood generates a variety of intricate interactions and competitive reactions, leading to a more complex and harder-to-control reaction process.

The interaction mechanism of OS involved in wood materials mainly includes four types: 1) Binding: OS can be combined on the surface of wood to form a protective film and improve the water resistance and thermal stability of wood (Yeo *et al.* 2015). 2) Filling: OS can fill the voids of wood CW (cell wall), improve the density of wood, and thus reduce the water absorption of wood (Chen *et al.* 2022). 3) Biodegradative resistance: OS has specific antibacterial and mildew resistance, which can reduce the attraction of wood to microorganisms and prevent wood from being corroded (Odalanowska *et al.* 2022; Shilova *et al.* 2022). 4) Change the structure of wood: OS can change the structure of wood and thus improve its mechanical properties (Chang *et al.* 2022).

Factors in the Modification Process

Effect of kinds of silanes

The silane type has the most prominent influence on the whole modification because there are many types of silanes, and the difference in side chains can significantly affect the entire modification reaction mechanism. Generally speaking, the molecular structure of silane has a significant influence on its modification. For example, linear silane molecules typically penetrate the structure of wood materials more efficiently than branched silane molecules and thus have a better modification effect.

Wang *et al.* (2019) investigated the effects of different silane coupling agents (KH550: 3-aminopropyltriethoxysilane, KH560: 3-methacryloxypropyltrimethoxysilane, and KH570: (vinyltris(methylethylketoximino)) silane) on the modification of cellulose models with nano-SiO_2 under the same conditions (Fig. 2). The results indicated that KH550 exhibits the best modification effect on the nano-SiO_2-cellulose system. They speculated that the reason for this effect is that KH550 can form more hydrogen bonds in the nano-SiO_2-cellulose system. This modification also led to improvements in the mechanical properties, free volume, interaction energy, and thermodynamic properties of hydrogen bonds. Some of these silane coupling agents possess certain substituents that confer specific wettability effects. Consequently, the amine group would become more hydrophilic at a lower pH, where it acquires a positive charge. Zhang *et al.* (2022a,b) investigated the effects of different silane coupling agents (KH550, KH560, KH570, and KH580: trimethoxysilylpropyl-ureidoethoxyethylimidazole) on the wettability, surface free energy, and film adhesion of silicate-modified poplar wood. The silanol hydrolyzed by the silane coupling agent and the silanol on the surface of the substrate undergo a dehydration condensation reaction to form a Si-O-Si bond. The silane coupling agent binds to the surface of silicate-modified poplar wood, among which KH560 has the best modification and adhesion to the wood surface.

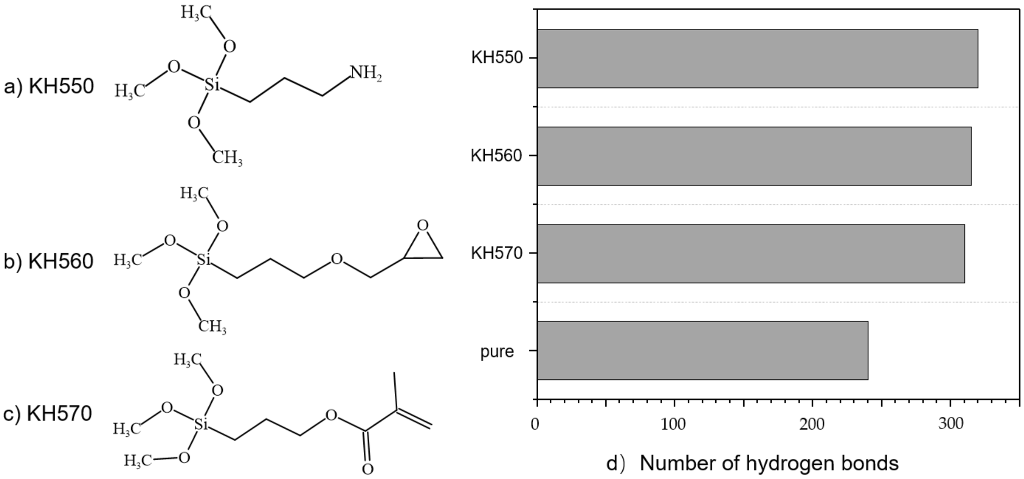


Fig. 2. a) Chemical structure of KH550; b) KH560; c) KH570; and d) the number of hydrogen bonds under different conditions ("pure" stands for unprocessed), Wang *et al.* 2019, CC BY 4.0

Effect of alkaline condition

Khamtree *et al.* (2020) treated rubber wood powder with alkaline silane at a concentration of 5% silane to obtain recycled polypropylene composite materials. The effects of silane treatment with different concentrations and treatment times on water absorption, mechanical properties, and thermal properties of WPCs (wood plastic composites) were studied. The results showed that the properties (as mentioned above) modified by alkaline silane were higher than those of non-alkaline silane and pure alkaline silane. In addition, the WPCs treated with 5% silane concentration for 2 h had the best performance. Alkali treatment can effectively remove surface impurities, destroy hydrogen bonds in wood, and increase surface roughness. There are also examples of improving the tensile strength of composite materials made by alkaline treatment of wood fiber (Gwon *et al.* 2010). In addition to incorporating alkaline substances into silane modification agents, alkaline pretreatment of the fibers can also be carried out. Rajini *et al.* (2012) studied the effect of alkaline treatment on silane composites. They showed that alkali treatment of fibers improves the mechanical properties of the composites. Liu *et al.* (2022) mixed modified wood chips with NaOH (mass fraction of 2 to 8%) and silane coupling agent (mass fraction of 1 to 5%) to enhance the surface roughness of the wood chips, so that they can better combine with the cement matrix to form cement particle board. The alkali coupling agent used removed most amorphous components such as those of hemicellulose and cellulose in wood chips. Such removal resulted in higher crystallinity, higher relative cellulose content, and higher surface roughness of the wood surfaces. The modification results show that this change can make the interface between the wood chips and cement matrix have better performance, and the flexural strength of the treated cement particle board is 27.1% higher than that of the untreated composite. The reaction between OS and wood substrate can be well promoted by hydroxide ions under alkaline conditions. The interfacial properties of the material substrate can also be increased.

Effect of efficiency of reaction

Fathi *et al.* (2017) conducted a more complex and effective reaction design, using a silane-peroxide cross-linking system to toughen WPCs. After the reaction of cellulose with silane, it continues to react with peroxides, and the peroxides in different branch chains undergo a condensation reaction, which effectively enhances the structure of cellulose and improves the impact performance and toughness of WPCs. Starting from the types of silane, reaction conditions, and other more complex reaction systems, scholars have carried out a large number of studies on the modification mechanism of silane, the main purpose of which is to enhance the reaction efficiency of silane, generate more chemical bonds, and better enhance the substrate performance (Ouyang *et al.* 2014; Zhang *et al.* 2022).

Rao *et al.* (2018) studied the interfacial structure and bonding mechanism of a silane coupling agent when it modified wood flour and polyethylene composites. They studied the effect of the addition of the coupling agent on the chemical structure of the composite materials through attenuated total reflectance Fourier-transform infrared spectroscopy and nuclear magnetic resonance spectroscopy analysis. The results showed that there is a transition from crystalline cellulose to a non-crystalline state during the treatment of the coupling agent, and the optimal interface of the composite is obtained through mutual diffusion, electrostatic adhesion, chemical reaction, and mechanical interlocking binding mechanism. Fathi *et al.* (2017) conducted a study where they employed 2,2,6,6-tetramethylpiperidine-1-oxyl radical to oxidize flax fibers, thereby converting the primary OH groups on the fiber surface to carboxyl groups. This modification enabled subsequent silane treatment processes by transforming the groups that were initially unsuitable for silane reaction into suitable ones, which significantly enhanced the silane modification efficiency (Ouyang *et al.* 2014). Meekum and Khongrit (2018) studied the effect of different chain lengths of alkoxysilanes on wood fiber modification. First of all, the chain length of the compound will affect its distribution in the wood fiber, thus affecting the properties of the wood fiber. Silanes with shorter molecular chains are more likely to penetrate the CW, react with the CW components, and perform better in dimensional stability and hydrophobicity. Silanes with longer molecular chains cannot enter wood fiber CWs, which limits their ability to react with hydroxyl groups for better performance. Ryu and Kim (2022) studied the hydrolysis and condensation reaction mechanism of alkoxysilane and used zinc ions to accelerate the reaction of OS, which significantly improved the reaction efficiency.

In order to reduce the modification time and cost, it is also very important to study the modification efficiency of silane, whether it is to improve the reaction conditions, pretreatment, or to control the permeability of silane by different chain lengths. Although there are different applications in different fields, the silane reaction mechanism is almost the same, which seems to prove the strong versatility of silane.

WOOD MODIFICATION BY OS

OS compounds are used as a surface modifiers, consolidation agents, and accelerators in construction, glass, plastic, papermaking, coating, medical treatment, and food production (Moradllo *et al.* 2016).

Although applied in different fields, silane has a variety of functional capabilities, such as anti-corrosion, water resistance, fire resistance. For example, Ratajczak *et al.* (2020) grafted [3-(2-aminoethylamino)propyl]-trimethoxy-silane to wood and studied some conditions for effective binding of silane to wood during the grafting process. In their studies, features in the infrared spectrum of treated wood demonstrated the reactivity of silane with the wood matrix.

In the subsequent leaching experiment, the stability of the binding of organic silane bonds with wood was confirmed through the detection of the chemical element content of wood before and after leaching (Ratajczak *et al.* 2020). This confirmation demonstrates the ability of silane to combine with wood, and has been shown in some other studies to improve the dimensional stability of wood (Broda and Mazela 2017). However, there are few studies directly related to wood (modifying wood itself through silane directly), which should be a more worthwhile area of research. Some typical silane applications are described in Table 2.

Table 2. Application and Treatment Result of Silane

Silane	Method	Result	References
MTMOS	Oscillated vacuum–pressure method	Anti-shrink efficiency increased by 25.1%	(Broda and Mazela 2017)
PEG-silane	Immersed for 12 h	Anti-shrink efficiency increased to 80%	(Meints *et al.* 2018b)
MPTES, DEAPTMDS	Oscillated vacuum–pressure method	Equilibrium moisture content reduce and well combine	(Popescu and Broda 2021)
AEAPTMOS, MTMOS	Immersion or brushing	Enhanced resistance to the weathering process and color stability	(Hochmańska *et al.* 2014)
VTMOS	Immersed at vacuum for 15 min and 0.8 kPa for 2h	The contact angle increased to a range of 65.20%-73.07%	(Woźniak *et al.* 2018)
HDTMSM, TMS	Immersed for 6 h	The contact angle increased to 150%	(Ou *et al.* 2021)

OFTES	Sol–gel method	The contact angle increased to 131%	(Szubert *et al.* 2019)
D4H	Immersed for 5 min	The contact angle increased to 140.1%	(Tang *et al.* 2022)
PMHS	Immersed for less than 1 min	The contact angle increased to 139% and the water absorption decreased to 30.72%.	(Lin *et al.* 2018)
MTMOS: Methyltrimethoxysilane; **PEG**: Polyethylene glycol; **MPTES**: 3-Mercaptopropyltriethoxysilane; **DEAPTMDS**: 1,3-bis(diethylamino)-3-propoxypropanol-1,1,3,3-tetramethyldi-siloxane; **AEAPTMOS**: Aminoethylaminopropyltrimethoxysilane; **VTMOS**: Vinyltrimethoxysilane; **HDTMS**: Hexadecyltrimethoxysilane; **MTMS**: Methyltrimethoxysilane; **OFTES**: 3-(2,2,3,3,4,4,5,5-octafluoropentyloxy)propyltriethoxysilane; **D4H**: Tetramethylcyclotetrasiloxane; **PMHS**: Poly (methylhydrogen) siloxane.			

Effects of Wood Modification

Dimensional stability

Organosilane-modified wood usually brings silane and its reaction products into the cell wall (CW), so that OS reacts in the CW and is mechanically fixed, to achieve enhanced dimensional stability of wood. To achieve this, there are two strategies: One is to impregnate the wood in an ethanol solution (the water required for silane hydrolysis is only present in the CW). The other is that silane is hydrolyzed before impregnation (Donath *et al.* 2004), which is the determining factor in silane stabilizing the wood (Schneider and Brebner 1985).

Broda and Mazela (2017) used a soaking method and oscillating pressure method to study the stability of silane modification to waterlogged oak. They found there was no significant difference, but the silane (methyl dimethoxy-silane) effectively penetrated the wood CW and caused its expansion. Thus, the dimensional stability of wood is improved (Broda and Mazela 2017; Broda *et al.* 2019c). In addition to dipping in solution, there are cases of using resins to bind wood to silanes. Ratajczak *et al.* (2020) studied the effect of [3-(2-aminoethylamino)propyl]trimethoxysilane on the wood substrate during grafting. To solve the phenomenon of silane precipitation from wood after impregnation, the researcher determined the stabilizing effect of alkyd resin on silane by analyzing the contents of silicon and nitrogen in wood before and after leaching. The degree of silicon and nitrogen leaching from wood using alkyd resin was lower, and the results showed that alkyd resin improved the binding of silane to the wood matrix.

In some instances, silane has been used as an additional additive in PEG-impregnated wood. Meints *et al.* (2018a) used a PEG with methoxy and trimethoxy silane functional groups during the vacuum-impregnation process of wood, and their experimental results demonstrated that the use of PEG-silane resulted in greater toughness than ordinary PEG-modified wood. The purpose of the modification changes is to make silane better enter the wood CW or better react with the wood CW. Silane participates in the chemical reaction as a modifier or additive to enhance the wood properties. The permeability of wood CWs is very important for wood modification. For example, the pretreatment of wood using deep eutectic solvents can increase the permeability of wood while preserving the structure of wood (Li *et al.* 2022).

Popescu and Broda (2021) used two alkoxysilanes, methyltrimethoxysilane and (3-mercaptopropyl) trimethoxysilane and 1,3-bis-(diethylamino)-3-propoxypropanol)-1,1,3,3-tetramethyldisiloxane to modify the surface of the dried waterlogged wood and found they can effectively stabilize wood. They also found that the mechanism of stabilizing wood using alkoxysilane is based on the expansion of CWs by silane molecules and wood chemical modification, while polysiloxane fills the CWs. This difference may be due to additional reactions between different groups and wood hydroxyl groups, which enhance the silane-wood interaction and assist in wood dimensional stabilization.

Broda *et al.* (2020) investigated the use of OSs as wood consolidation agents with different molecular sizes and chemical structures. Their research showed that low molecular weight organosilicon is a better wood stabilizer. It was able to penetrate CWs while having functional groups interacting with wood polymers. In their study, (3-mercaptopropyl) trimethoxysilane provided the best shrinkage resistance by penetrating CWs to strengthen the wood structure. In more in-depth studies, demethoxylation of lignin was observed in the case of alkoxysilane treatment, whereas (3-mercaptopropyl) trimethoxysilane treatment was most effective in stabilizing the wood, and severe changes in wood composition, such as decomposition of lignin, could be observed (Broda and Yelle 2022).

Hydrophobic property

The hydrophobic property of wood is closely related to its dimensional stability (Thybring *et al*. 2022). Generally, the stronger the hydrophobic property is, the better the dimensional stability of the wood. Similarly, the susceptibility of wood to microbes comes from the water in the wood. When moisture approaches the fiber saturation point, the wood becomes suitable for microbial growth, which reduces the durability of the wood, thus the hydrophobic properties are particularly important for the application such as self-cleaning and anti-corrosion (Sun *et al.* 2021).

Hochmańska *et al.* (2014) investigated the properties of methyltrimethoxysilane, AEAPTMOS and glycidoxypropyltrimethoxy-silane for wood impregnation. Among them, the wood modified with 5% concentration of MTMOS exhibited the highest hydrophobic and aging resistance, while the wood modified with AEAPTMOS had enhanced color stability. Woźniak *et al.* (2018) obtained very effective hydrophobic properties of Scots pine by impregnating it with a formulation solvent of ethanolic extracts of propolis, VTMOS, and tetraethyl orthosilicate. The addition of silane may only make up for the lack of groups in the ethanol extract, which cannot fully react with the hydroxyl group in wood, leading to the improvement of hydrophobic properties. Ou *et al.* (2021) prepared a single-use superhydrophobic composite solution for wood modification. Modified wood can remain hydrophobic after long-time subjection to mechanical damage, chemical damage, and environmental exposure. This solution is made from a mixture of HDTMS and MTMS. The deposition and solidification of the compound silane on the surface and inside of the wood give the wood superhydrophobicity. Due to the infiltration of silane, new silane deposits will appear inside the wood when the surface is worn down and these maintain the superhydrophobicity of wood. Szubert *et al.* (2019) studied a fluorosilane-based wood protectant(paint), using 3-(2,2,3,3,4,4,5,5-octafluoropentyloxy)propyltriethoxysilane and other silane additives to prepare a hydrophobic protective layer on wood surfaces. This paint forms a durable protective coating that permanently increases the hydrophobicity of the wood. Fluorocarbon silane is hydrogen-bonded with wood to form a stable structure. The fluorocarbon chain attached to the silicon atom is an effective barrier to prevent water from entering and contacting the wood surface. An increase in hydrophobicity is usually accompanied by an increase in dimensional stability. The hydrophobic properties and dimensional stability properties work in a similar way, which increases dimensional stability when water is isolated.

Tang *et al.* (2022) developed a simple method to modify wood by soaking wood in a solution of D4H for 5 min. The hydrophilic groups on the surface of wood and the S-H bond on D4H undergo a dehydrogenation reaction under the catalyst; thus the D4H with a hydrophobic methyl group is grafted to the surface of the wood, and the wood obtains good hydrophobicity due to the reduced the number of hydroxyl groups. Similarly, polysiloxane can also be used as a flame retardant or protective agent to modify the wood surface, which is almost similar to most silane applications (Wang *et al.* 2020b; Papachristou *et al.* 2022). Lin *et al.* (2018) prepared a highly hydrophobic wood based on poly(methylhydrogen) siloxane, in which the hydrophobic group ($-CH_3$) on the poly(methylhydrogen) siloxane chain was covalently grafted onto the surface of the wood, leading to the wood having good hydrophobic and dimensional stability. At the same time, the mechanical property, thermal stability (slow decomposition at high temperatures), and ultraviolet resistance of wood increased after treatment by this method. Grafting polysiloxane onto wood surfaces can improve its applicability to wood materials by providing a protective and functional coating that enhances the wood's durability and resistance to environmental factors such as moisture, fungi, and ultraviolet light.

Biological resistance

As a biological material, wood is prone to decay and degradation by fungi and microorganisms, which is unfavorable in wood applications (Goodell *et al.* 2020). To study the effect of OS on wood biological resistance, a large number of scholars have made efforts in this regard.

Alfieri *et al.* (2021) used nanoparticles in conjunction with an OS to impregnate and modify wood. The hydrophobic properties of wood modified by this system were improved, even better than those modified by some short-chain silanes. At the same time, this system showed excellent protection against both biological and abiotic factors. Kamperidou (2019) treated the surface of thermally modified wood and found that OS provided effective biological resistance to wood. After thermal modification, wood moisture is lost and biological resistance is improved. However, such improvement is not permanent. After a period of exposure to the biological environment, wood will still be corroded or damaged. The thermal modification combined with a silane treatment further enhances the biological resistance of wood.

Broda and Plaza (2023) studied the biological resistance of the Scots pine (*Pinus sylvestris* L.) after impregnation with three different OSs. The wood modified by the oscillating impregnation method obtained a certain degree of enhanced water resistance and durability and biological resistance. Fungal growth is inhibited due to bis(diethyl-amino)-3-propoxypropanol-1,1,3,3-tetramethyldisiloxane having complete filamentous fungal resistance. It is inferred that the amino groups carried on the OS have these antifungal functions (Reinprecht and Grznárik 2015). Woźniak *et al.* (2020) impregnated *Pinus sylpholides* with a mixed modifier prepared with propolis extract and silane, and found that the modified wood also obtained resistance to biodegradation and enhanced mechanical strength and hydrophobicity. This biological resistance is mainly derived from the inhibition of propolis extract on microorganisms.

The improvement of the biological resistance of wood can be attributed to the loss of wood moisture and nutrient sources. In addition, the toxicity of certain chemicals to biological systems can inhibit the growth of microorganisms. Wood modification involving organic silane can be well combined with the change of silane side-chain groups. Under the combined action of them, wood can obtain good biological resistance. However, people are more inclined to use non-toxic and pollution-free materials at present, and as such, attention should be paid to the selection of organic silane to make it more environmentally friendly.

Effects of Fiber Modification

Studies have been reported on wood cellulose modified by silane and composite material synthesis. In these cases, silane can improve the surface properties of fibers, make them better bonded, improve the interfacial strength between the fibers and matrix, and increase the hydrophobicity, tensile strength, and shear strength of composites to some extent (Ying *et al.* 2017; Qiu *et al.* 2022) (see Table 3).

Gao *et al.* (2017) used high temperature and high-pressure steam and a silane coupling agent to chemically modify poplar fiber. The results showed that the mechanical properties of the composite were the best when the content of silane was 2%, the temperature was 170 ℃, and the pressure was 0.8 MPa. Using infrared spectroscopy analysis, it was found that under high temperature and pressure, the free hydroxyl group on the cellulose formed a hydrogen bond with the hydrolyzed silane, reducing the number of hydroxyl groups. At the same time, during the heat treatment process, the silanol group generated by the hydrolysis of silane forms a covalent Si-O-C bond with the wood fiber, which can indicate that the coupling agent is grafted to the wood fiber through the copolymerization reaction. The results showed that the addition of coupling agents further improved the storage modulus, loss modulus, and mechanical properties of wood fiber composites.

Ismail *et al.* (2012) studied the changes of mechanical properties of straw fiber and polymer composites modified by different silane coupling agents and irradiated by a constant electron beam. Highly polar cellulose fibers are inherently incompatible with hydrophobic polymers; thus, silane coupling agents are added to the composite to achieve the compatibility. At the same zero electron beam irradiation, the properties of the composites containing N-(2-aminoethyl)-3-aminopropyltrimethoxysilane is higher. In addition, electron beam irradiation increases the active component (more free radicals) of the composite material, which greatly increases the probability of chemical bond formation and the strength of the composite material. Koohestani *et al.* (2017) investigated the effects of silane on the mechanical, thermal, and rheological properties of maple polyethylene composites. The vinyltrimethoxy and aminoethyl aminopropyl trimethoxy silanes have different effects on the composites. The tensile strength and flexural strength modified by vinyl silane are better, and the stiffness modified by aminosilane is lower, but both of them improve the thermal stability of WPCs. For natural fibers, their hydrophilicity is the most important factor affecting the technical performance of green composite materials (Anbupalani *et al.* 2020). Organic compounds such as silane can greatly reduce the hydrophilicity of natural fibers and improve more properties of green composite materials on a macro scale.

Using silane modified wood fiber as a filler, Chen *et al.* (2022) prepared an ethylene propylene diene monomer (EPDM) bio-composite material by melt mixing and compression molding. This silane-modified wood fiber has low water absorption and significantly enhanced thermal stability. Compared to pure EPDM, the tensile strength and modulus of the composite were increased by 115% and 259% respectively.

Sun *et al.* (2019) studied preparing bio-composite materials from rice husk and the polymer polyethylene. To improve the interface bonding performance of composite materials, the effects of different modifiers on the modification of composite materials were investigated. In this paper, KH550 silane coupling agent and compatibilizer maleic anhydride were used for complementary modification to improve the interfacial properties of rice husk fiber and other composites. The silane coupling agent crosslinked with the hydroxyl group on the surface of rice husk fiber to generate a nitrogen hydrogen bond and carbon oxygen double bond. Similarly, through the chemical connection between the modifier and rice husk, the amount of hydroxyl groups and surface energy on the fiber surface were reduced, which promoted the dispersion and compatibility of the fiber in the plastic matrix.

Khan *et al.* (2021) used a chemical pretreatment to improve the mechanical properties, thermal stability, and water resistance of wood pellet composites. Solid wood particles were pretreated with different concentrations of silane and NaOH, which improved the interface bonding between wood particles and epoxy resin, and thus improved the properties of wood composites. Silane under alkaline conditions can decrease the number of hydroxyl group of wood particles and increase the surface roughness of wood particles and improve the binding ability of wood particles with resins.

Table 3. Silane Modified Wood Fiber Composites

Silane	Method	Result	Reference
HDPE	Exposed to steam at 170 °C and 0.8 MPa	Thermal stability, storage modulus and mechanical properties improved	(Gao *et al.* 2017)
A700	130 °C and electron beam irradiation	Flexural strength, modulus of elasticity and impact strength increased	(Ismail *et al.* 2012)
VTMS	125 °C	Thermal stability, modulus of elasticity, tensile and flexural strength increased	(Koohestani *et al.* 2017)
VTES	70 °C for 6h	Thermal stability, tensile strength and modulus increased	(Chen *et al.* 2022)
KH550	Hot-press for 9 min, cold press for 5 min	Tensile strength, water-resistance, elastic modulus and toughness increased	(Sun *et al.* 2019)
APTES	Pretreatment	The mechanical properties, thermal stability, and water resistance increased	(Khan *et al.* 2021)

HDPE: High-density polyethylene;
A700: N-(2aminoethyl)-3-amino propyltrimethoxy silane;
VTMS: Vinyl-trimethoxy silane;
VTES: Vinyltriethoxysilane;
APTES: Aminopropyltriethoxysilane.

In the modification process of cellulose (the raw material of composite materials), first, the high temperature and high-pressure steam treatment can reduce the number of fiber surface hydroxyl groups and the polarity of the fiber surface and improve the thermal stability and crystallinity of composite materials. When silane is added, the storage modulus, loss modulus, and mechanical properties of the composite are further enhanced (Gao *et al.* 2017). Natural fibers can enhance the properties of fiber composites from many aspects, such as natural fiber properties, fiber size and direction, fiber stacking sequence, interface bonding degree, additives, and coupling agents (Khan *et al.* 2018). The direct application of silane on wood also occurred, such as some studies using silane impregnation to reduce the hydrophilic properties of waterlogged wood (Broda *et al.* 2019a).

Effects of Coatings Modification

Kumar *et al.* (2018) used a self-assembled monomolecular membrane based on octadecyl trichlorosilane to perform nanocoatings on the surface of alkaline resistant glass fibers. This system successfully formed superhydrophobic nanocoatings on the surface of alkali resistant glass while increasing its tensile strength. Based on the lamination and film-forming characteristics of OTS, functional improvement was made for glass fibers. A similar coating technique is the waterproofing coating based on hexamethyldisilazane studied by Subramanian *et al.* (2020). Silica has a certain degree of similarity with wood (both have hydroxyl groups) and strong hydrophilicity due to its rich hydroxyl group. They first modified the silica surface with hexamethyldisilazane, replacing two hydroxyl groups, and then further reacted with other silane coupling agents and resins. The water contact angle of the product was greater than 150° and the materials also exhibited better mechanical stability, durability, and adhesion. Complex chemical reactions occur between the silane coupling agent and modified silica and resin, forming a strong covalent structure, and finally forming multifunctional superhydrophobic nanocomposites. Reverdy *et al.* (2018) reported a one-step formulation for superhydrophobic coatings. The biologically based cellulose fibers were treated with alkyl ketene dimer or amino propyl trimethoxy silane as a binder. The water contact angle also reached 150°. In terms of surface modification of transparent wood (wood-based material impregnated with resin after lignin has been removed), Li *et al.* (2022) successfully prepared transparent wood, coated the wood surface with fluorinated alkyl silane, resulting in high hydrophobic properties of the wood. According to Fourier transform infrared spectroscopy and X-ray photoelectron spectroscopy test results, FAS forms Si-O-C bond combined with transparent wood, which also polymerizes to form Si-O-Si bonds (Ding *et al.* 2022).

In addition to adding hydrophobic properties, silyl coatings can also provide other functions. Zhou *et al.* (2017) reported the stable dispersion of hydrophobic nanoparticles, fluorinated alkyl silane, and fluorocarbon surfactants in water, and prepared water-based superhydrophobic surface coatings suitable for various solid substrates. Zheng *et al.* (2020) also modified the water-based acrylic resin with KH550, effectively improving the crosslinking density of the resin, and forming a silane molecular layer with the substrate. The modified coating has better mechanical and thermal properties, as well as improved wear resistance, water resistance, impact strength, and hardness.

Almost all these coating technologies can be applied to wood. Gao *et al.* (2015) prepared titanium dioxide thin films modified by (sevofluoro-1,1,2,2-tetradecyl)-trimethoxysilane on the wood surface by a hydrothermal method, which obtained a very strong superhydrophobic surface that was resistant to acidic solutions. Szubert (2018) synthesized a rapeseed oil-based silane from rapeseed oil and used it as a wood surface coating. By soaking the silane solution, a thin film with an O-Si bond is deposited on the surface of the wood, which provides a relatively durable modification for wood and has a function of hydrophobicity. Silane coatings are also commonly used for wood, but unlike earlier wood coatings, silane coatings are more environmentally friendly and non-toxic (Li *et al.* 2019).

Effects of Adhesive Modification

Similar to coating modification, silane is often used to modify adhesives. Adhesives commonly used in wood products are often added with additives to improve their flame-retardant, water-resistant, and wear-resistant properties (Su *et al.* 1998; Li *et al.* 2018). However, silane can meet these requirements. Silane is a commonly used crosslinking agent in adhesives, which can be used to modify and improve the properties between different materials. It is typically incorporated into adhesives and used as a bonding agent after quick mixing. This enhances the strength and adhesion properties of the materials.

Chang *et al.* (2022) modified sodium lignosulfonate by adding two different silanes to improve the surface bonding strength of polypropylene decorative film. The results showed that both silanes can be grafted onto hydrolyzed Si-OH and -OH groups. The long chains of silanes can be physically twisted, contributing to the formation of a network structure and enhancing the bonding strength on the surface of plywood (Onat and Özan 2017). Wang *et al.* (2018) successfully improved the fire resistance of plywood by using silicon-based (vinyl silane additive) in adhesive bonding. The role of silane in this process is to oxidize and form a protective layer to insulate the plywood from oxygen. Unlike traditional plywood adhesives, Chen *et al.* (2017) studied the effect of silane coupling agents on the properties of starch-based wood adhesives. They added a certain amount of KH570 into the adhesive sample, and the results showed the silane coupling agent increased the covalent bond of the adhesive, enhanced thermal stability, and increased bonding strength. The silane coupling agent acts as an amphiphilic surfactant (Zheng *et al.* 2019). The organic end reacts with starch, and the other end grafts wood to form a chemical bond, which increases the bonding strength and improves the interface property between the wood and starch adhesive. Zhang *et al.* (2015) also prepared a starch-based silane adhesive for wood, focusing on improving the hydrophilicity of starch. Starch-based adhesives reduce the number of exposed hydroxyl groups and enhance the molecular structure through a series of silane and olefin substances reactions, significantly enhancing the thermal stability, bonding strength, and water resistance. Liu *et al.* (2019) studied two silane coupling agents (KH550 and A171) that improved the bonding between WPCs and wood veneer. The silane coupling agents were grafted onto the wood surface, and the water contact angle increased to 120° after treatment, while the bonding strength was also significantly improved.

CONCLUDING REMARKS

This paper has introduced some applications of organosilane (OS) related to wood and the effects of OS on various materials. Many properties of modified materials are improved by silane. When OS is used as a coupling agent in coating, adhesive and other fields, it can effectively improve the interface properties of materials. When OS is used for wood modification, the hydrolyzed products combine with the groups on the cell wall (CW) to improve wood properties. The following points follow, in general, from the work cited in this article:

1. When OS is used as a wood modifier, usually in the form of a trialkoxysilane, its initial modification mechanism is to combine with the hydroxyl groups on the wood matrix to form hydrogen bonds. Thus, the type of OS and the ability to produce hydrogen bonds are important factors affecting its modified wood. In addition, the higher the degree of reaction between silane and substrate, the more chemical bonds are produced during the curing phase, and the better the modification. Therefore, the selection of OS and the control of reaction conditions are essential for wood modification. Meanwhile, the modification mechanisms should be deeply investigated in a future study.
2. In terms of pollution control, especially when OS is used as a coating on the wood surface, its contact with the human body will be more frequent. This requires silane to have high non-toxicity. There have been cases of the synthesis of non-toxic silanes from rapeseed oil, which also inspires the synthesis and improvement of OS from existing biomass materials. Thus, the research on silane modification must stay within the research on the new non-toxic and pollution-free silane. Study on the biological OS can improve most of the pollution problems caused by OS.
3. A better modification requires silane to penetrate deeply into the wood CW and complete bonding and curing inside the CW. Research on improving wood CW permeability may bring new ideas to the application of silane.

ACKNOWLEDGMENTS

The authors are grateful for the support of the National Natural Science Foundation of China (Grant Nos. 31870545 and 31570558) and Nanjing Forestry University Undergraduate Innovation Training Project (2022NFUSPITP0330).

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Article submitted: March 22, 2023; Peer review completed: May 6, 2023; Revised version received: May 15, 2023; Accepted: May 21, 2023; Published: June 1, 2023.
DOI: 10.15376/biores.18.3.Liu