

Study on the Effect of Organic Additives and Inorganic Fillers on Properties of Sodium Silicate Wood Adhesive Modified by Polyvinyl Alcohol

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To enhance the properties of sodium silicate wood adhesive modified by PVOH (polyvinyl alcohol), different organic additives (compound A) and inorganic fillers (compound B) were added into the system. Their effects on the thermal stability and molecular structure of the composite adhesive were investigated by scanning electron microscopy (SEM), spectroscopic analysis (FT-IR), differential scanning calorimetry (DSC), and thermogravimetric analysis (TGA). The results showed that when the compound A consisting of amino acid, TEOS (Tetraethyl orthosilicate), sodium dodecyl sulfate with the mass ratio of 5:3:3 and B consisting of 50% active magnesium oxide and 50% nano-silica were added into the cross-linked sodium silicate adhesive system with the ratio of 90:3, the bonding and water resistance properties were improved by 97.6% and 46.7%, or 13.7% and 36% relative to pure and cross-linked sodium silicate, respectively. The addition of organic additives and inorganic fillers had a good effect on both the bonding strength and thermal stability of sodium silicate wood adhesive modified by PVOH.

Keywords: Organic additives; Inorganic fillers; Bonding property; Water resistance; Thermal analysis

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INTRODUCTION

Developing environmentally friendly adhesives for the wood industry is a most critical issue that affects the trade between different countries and the public's acceptance of wood panel production. This problem has become more severe as traditional wood adhesives such as urea-formaldehyde resin, resorcinol-formaldehyde resin, and phenol formaldehyde resin have been reported in recent years to have significant adverse effects on the human body, especially on babies. Meanwhile, adhesives account for a substantial part of the cost of production during manufacturing wood products. With the background of intense competition in global trade, the development of low cost, multifunctional, and eco-friendly adhesives has become more urgent.

Inorganic adhesives have several benefits over traditional wood adhesive, including resistance to fire and other chemicals, non-toxic nature, low cost, and ease of manufacturing. Among all the existing inorganic adhesives, silicate adhesive, and especially the sodium silicate adhesive, is particularly prominent. Sodium silicate adhesive is non-toxic (Pizzi and Salvadó 2007) and has the least negative impact on the environment (Derelch 1958). Sodium silicate is abundantly available, has good adhesion performance and weather resistance, is difficult to burn, and is being easy to handle after being used (Schupp Jr. and Boller 1938; Weygandt 1938; Torkaman 2010; Zhang *et al.* 2012). For certain purposes, sodium silicate has been found to exhibit fairly high adhesion to many

substrates (metals, minerals, wood, and even plastic materials) (Kornev and Danilov 1996). As a wood adhesive, sodium silicate can solve the problems brought from the traditional wood adhesives in spite of its low bonding strength. As a kind of inorganic material, it is difficult to be bonded with wood whose surface is full of organic chemicals (Liu *et al.* 2012). The development of synthetic procedures for producing some polymers in the form of aqueous dispersions that manifest gluing properties can be considered to be a spectacular achievement of the mid-20th century (Voitovich 2008).

According to previous studies (Liu *et al.* 2012), polyvinyl alcohol (PVOH) can be cross-linked with sodium silicate, thus reducing the brittleness of the structure and improving its bonding strength. A lot of experiments have been conducted to explore the best cross-linked method and their proportions. Improvements in bonding strength have been found. However, the water resistance is another critical factor that needs to be considered in the case of wood adhesives. In this paper, the improvement of adhesive's properties by introducing some organic additives and inorganic fillers is studied.

EXPERIMENTAL

Materials

Sodium silicate was obtained from Shangyu Qiangsheng Chemical Co., Ltd. According to Liu *et al.* (2012), the best modulus of sodium silicate as wood adhesive is with a ratio of 2.8 of the inorganic compounds ($\text{Na}_2\text{O}\cdot 2.8\text{SiO}_2$) (Liu *et al.* 2012). PVOH of type 17-99 was obtained from Yunnan Yunwei Chemical Co.; this product can be dissolved in hot water with the temperature of 92 °C. Active magnesium oxide with particle size of less than 2 μm and nano-silica were supplied by Haicheng Meiling Chemical Co., Ltd. Amino acid, tetraethyl orthosilicate (TEOS), and sodium dodecyl sulfate were purchased from Guoyao Chemical Co., Ltd. The test materials was fast-growing poplar which was obtained from the Dongting Lake area in Hunan province of China.

Preparation of Sodium Silicate Adhesive

Cross-linked sodium silicate was modified by PVOH by using the method of Imam *et al.* (2001). PVOH was dissolved in cold water at a temperature of normal atmospheric temperature for 30 min and then in deionized water at a temperature of 90 to 95 °C for 2 h. Then the solution was cooled to be used.

According to the formula dosage design, the PVOH was dissolved as a 10% solution for addition to the four port flask. Then the sodium silicate and distilled water were added. All these were stirred uniformly in a water bath of 60 °C. After 10 min, emulsifier OP-10, citric acid, and phenol were added drop-wise in a water bath of 80 °C for 20 min. The sodium silicate and distilled water were added in drops again and stirred uniformly. The adhesive then could be used after aging for 24 h. When introducing the organic additives and inorganic fillers as designed, sodium silicate modified by PVOH was put in the water bath of 40 °C and a certain proportion of ammonium per sulfate was added as initiator for 30 min. When introducing the organic additives, the reaction system should be protected under inert gas such as nitrogen and maintain a constant reaction period with temperature of 60 °C. No matter what kinds of materials were added into the system, they were stirred until evenly dispersed.

Modified Sodium Silicate Adhesive with Different Methods

Cross-linked sodium silicate adhesive was modified using different methods. The samples modified by organics were treated with amino acid, TEOS, and sodium dodecyl sulfate addition to the cross-linked adhesive according to the designed proportion. Modification by inorganics indicated addition of magnesium oxide and nano-silica to the cross-linked adhesive according to the designed proportions. Modified by combined chemicals indicated that organic additives (compound A) and inorganic fillers (compound B) listed above were added with designed proportions.

Preparation of Wood Specimens

Wood specimens for mechanical testing obtained from 0.5 cm thick wood veneer (poplar, moisture content of 13%, tangential section) with dimensions of 300 mm × 300 mm × 10 mm were glued with adhesives under static pressures of 1.5 MPa at 30 °C for 24 h. The test specimens of plywood were prepared after storing them for 7 days at room temperature.

Physical and Mechanical Test of Wood Specimens

The bonding strength was determined according to GB/9846.8 using a MWD-50 microcomputer-controlled universal mechanical testing machine. The testing speed was 2 mm/min. All the tests were replicated 24 times, and results were presented as the averages.

Water Resistance Property

Water resistance property (evaluated with 24 h TS) was determined according to GB/T 17657-1999. The samples with dimensions of 50 mm × 50 mm × 3 mm (thickness) were completely immersed into distilled water at 20±2 °C, 24 h±15 min. After immersions for 6, 12, 18, and 24 h, the samples were taken out with removal of excess water on the surface and weighed. The water uptakes were calculated based on the weight percent gains at each stage. In order to avoid the contact between the samples and the bottom of the jar, a sieve was placed under them.

The accuracy of the measurement in case of thickness was 0.01 mm. A micrometer with an accuracy of 0.01 mm was used for the thickness measurement at point “O” (Coşoreanu and Lăzărescu 2013).

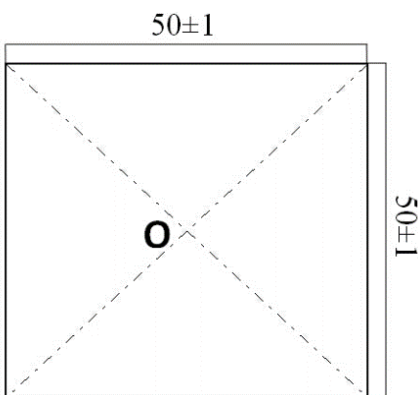


Fig. 1. Sample sizes and the point “O” of measuring the thickness swelling

All the tests were replicated 12 times, and results were presented as the averages.

Scanning Electron Microscope (SEM)

The modified adhesive was characterized using a QUANTA 450 SEM (FEI Inc., Netherlands). Each sample surface was sputter-coated with a layer of gold with the thickness of 20 nm. The maximum acceleration voltage was 30 kV, and the highest spatial resolution was 3 nm.

Fourier Transform Infrared Spectroscopy (FT-IR)

FT-IR spectra of modified adhesives were recorded on a K-Alpha XPS (monochromatic AlK α radiation, 1486.6eV, Thermo Fisher Scientific Co. Ltd, USA) at room temperature. The FT-IR samples were prepared by oven drying at 70 °C for two days after the curing of the adhesives and mixing the fine powder with KBr and compressing the mixture.

Differential Scanning Calorimetry (DSC)

The gelatinization properties of the samples were studied by using a differential scanning calorimetry (NETZSCH D204, Germany) as described by Sandhu and Singh (2007).

Thermogravimetric Analysis (TGA)

The thermal stability of the samples were studied with a thermogravimetric analysis system (TGA 209 F3, NETZSCH Co., Germany).

RESULTS AND DISCUSSION

Effect of Organic Additives on Properties of Modified Sodium Silicate Adhesive

The bonding strength of plywood bonded by sodium silicate adhesive modified by PVOH with different organic additives is shown in Fig 2. It is obvious that the bonding strength varied with different amounts of organic additives. It can be seen that the bonding strength of plywood increased when the additives amount increased from 0.6% to 1.2%, 0.2% to 0.8%, and 0.3% to 1.2%, respectively. And then it decreased when their amounts were further increased. For amino acid, when its adding amount accounted for 1% of the whole adhesive system, the bonding strength was the highest, and it was higher than the relative standard. TEOS and sodium dodecyl sulfate showed the same circumstances, and their best proportions were both 0.6%.

As shown in Fig. 2, compared to pure sodium silicate and sodium silicate modified by PVOH, the water resistance was also improved by the addition of organic additives. The 24 h TS results showed that the influences of amino acid, TEOS and sodium dodecyl sulfate on water resistance property were limited. Although on basis of the cross-linked modification of PVOH, the 24 TSs of amino acid, TEOS, and sodium dodecyl sulfate were 12%, 11.5%, and 11.5%, respectively. The improvements were just 4%, 8%, and 8%. However, when compared with the pure sodium silicate, they were decreased by 20%, 23%, and 23%. This indicates that these three kinds of organic additives that are always added into adhesives as water repellents have the potential to improve the PVOH modified sodium silicate adhesive's water resistance property. When these three organics were added into the adhesive system that was already modified by PVOH together with the best

formula, the improvement of mechanical and water resistance properties are listed in Table 1. Compared to that of sodium silicate modified with PVOH, the bonding strength increased by 2.7% and the 24 TSs decreased by 8%, respectively.

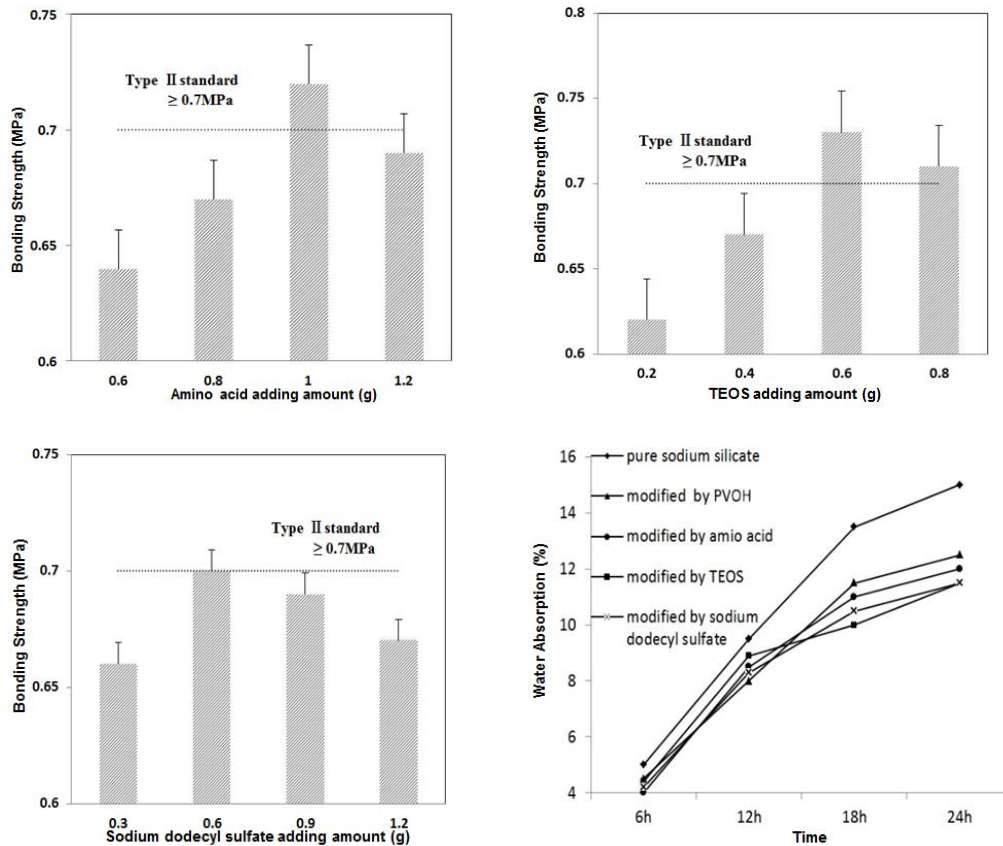


Fig. 2. Effect of organic additives on properties of modified sodium silicate adhesive

Table 1. Adhesive Performance Comparison Before and After Modification

Sodium silicate	Bonding strength (MPa)	24 TS (%)	Storage period (m)	Viscosity (mPa·s)
Pure	0.42	15	12	60
Modified by PVOH	0.73	12.5	6	86
Modified by organics	0.75	11	6	90

Effect of Inorganic Fillers on Properties of Modified Sodium Silicate Adhesive

Numerous preliminary experiments were conducted to look for the best inorganic fillers for sodium silicate adhesive. It was found that active magnesium oxide and nano-silica offered the greatest possibility. So their combinations with different proportions were applied as the filler in the modification system of cross-linked sodium silicate.

The bonding strength and water resistance properties of plywood bonded by sodium silicate adhesive modified by PVOH with different inorganic fillers are shown in Fig. 3. It is also obvious that the bonding strength and 24 TS varied with different proportions of

inorganic fillers. When the proportion of active magnesium oxide and nano-silica was 1:1, the bonding strength was 0.73 MPa, which is the same with that of the adhesive after being modified by PVOH and the 24 TS is 10.5% which is decreased by 16%.

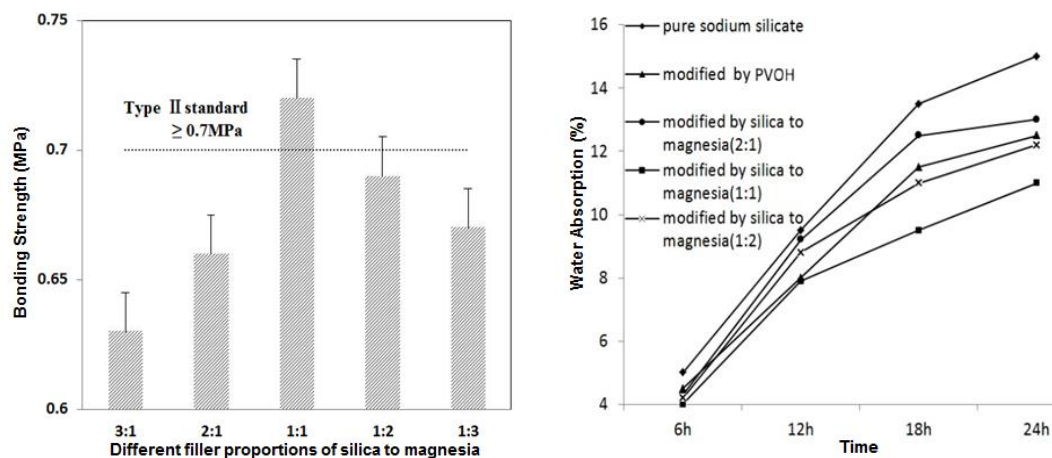


Fig. 3. Effect of inorganic fillers on properties of modified sodium silicate adhesive

Table 2. Adhesive Performance Comparison Before and After Modification

Sodium silicate	Bonding strength (MPa)	24 TS (%)	Storage period (m)	Viscosity (mPa•s)
Pure	0.42	15	12	60
Modified by PVOH	0.73	12.5	6	86
Modified by organics	0.75	11	6	90
Modified by inorganics	0.73	10.5	6	88

Combined Effects of Organic Additives and Inorganic Fillers on Properties of Modified Sodium Silicate Adhesive

To further explore the combined effects of organic additives and inorganic fillers on properties of modified sodium silicate adhesive, with nano-silica and magnesium oxide activity ratio of 1: 1 ratio as a compound A, amino acid, TEOS and sodium dodecyl sulfate 1: 0.6: 0.6 ratio as the compound B, the influence of different proportions of A and B (combined chemicals) on mechanical and water resistance properties of modified sodium silicate was studied. According to the results of the pre-experimental data, the proportions of 70: 3, 80: 3, 90: 3, 100: 3, 110: 3, 120: 3 of compound A and B were selected. The results are shown in Fig. 4.

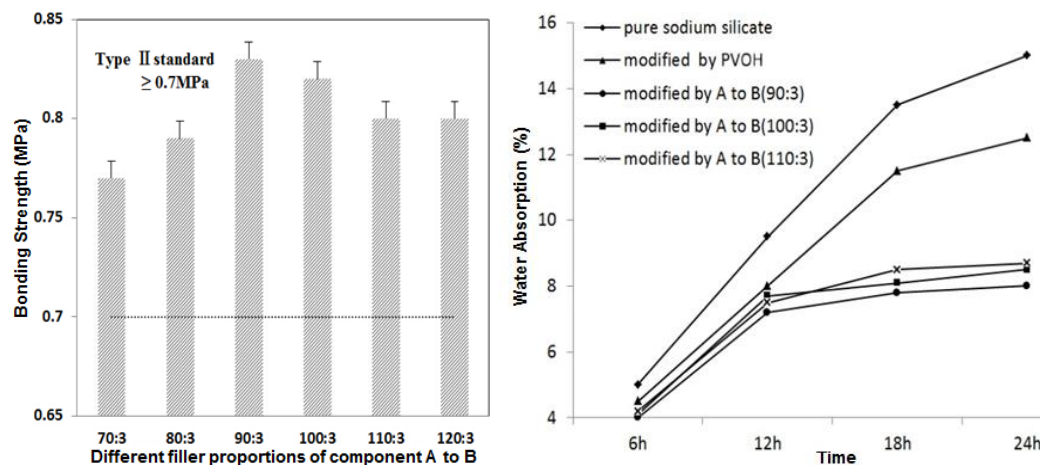


Fig. 4. Combined effects of organic additives and inorganic fillers on properties of modified sodium silicate adhesive

It could be concluded from these figures that the combination of organic and inorganic chemicals (compound modification) had a great influence on both the mechanical and water resistance of modified sodium silicate. All the bonding strengths were above the Type II standard and ranged from 0.77 MPa to 0.85 MPa. The average bonding strength under the best proportion of 90:3 was 0.83 MPa, which represented an improvement of 13.7% compared with that of modification sodium silicate by PVOH. As for the water resistance property, the 24 TS was 8% and decreased by 36% relative to that of modification sodium silicate by PVOH.

It is assumed that when inorganic fillers are added into the system they cannot be well integrated. Organic additives that contain hydrophilic groups and hydrophobic groups can help to protect the cross-linked sodium silicate so that the inorganic fillers and silicate form masked monomers, *i.e.* hydroxyl groups between them are isolated and the adhesive system can be more uniform. So the combination of organic and inorganic chemicals improved the properties modified sodium silicate. Table 3 shows the different properties of sodium silicate adhesive after different kinds of modification.

Table 3. Adhesive Performance Comparison Before and After Modification

Sodium silicate	Bonding strength (MPa)	24 TS (%)	Storage period (m)	Viscosity (mPa·s)
Pure	0.42	15	12	60
Modified by PVOH	0.73	12.5	6	86
Modified by organics	0.75	11.0	6	90
Modified by inorganics	0.73	10.5	6	88
Modified by combined chemicals	0.83	8.0	6	90

SEM Analysis

Figures 5 and 6 show SEM images of pure sodium silicate, cross-linked sodium silicate by PVOH, and modified cross-linked sodium silicate by organic additives and inorganic fillers (combined chemicals). It can be seen that the surface of pure sodium silicate was full of cracks due to its natural brittleness, and this led to a relatively low bonding strength.

When it was cross-linked by PVOH, the surface showed an obvious network structure and became more uniform. No cracks can be seen on its surface. And the space between the cross-linking structures had similar size, which can provide a relatively stable performance when it is used as wood adhesive. When combined organic additives and inorganic fillers were added, the shape and size of the particles were relatively uniform and the space inside the cross-linked network was filled, which can supply the adhesive a strong backbone. That means organic additives can help to isolate hydroxyl groups and inorganic fillers holds the whole system, so their combination can improve the bonding strength and water resistance properties significantly.

FTIR Analysis

FTIR test is a very useful method to analyze strand conformation of an adhesive, and it provides a sensitive indication of changes in molecular structures (Madejová 2003). Figure 7 shows the IR spectra of sodium silicate after different modification methods, and Table 4 shows different infrared absorption spectrums of these materials.

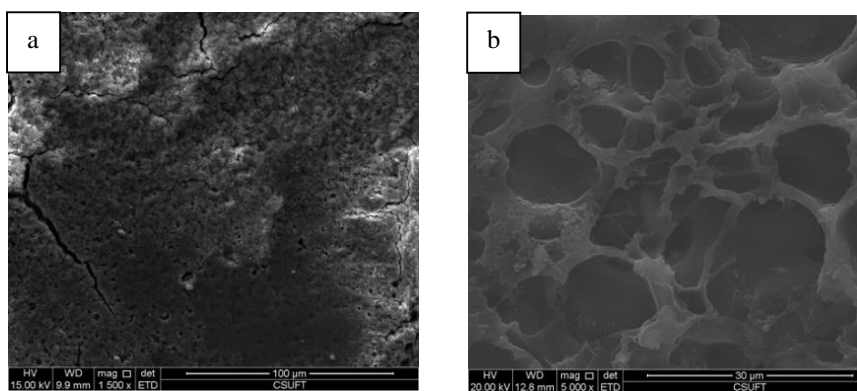


Fig. 5. SEM images of pure sodium silicate before and after modified by PVOH (a 1500x,b 5000x)

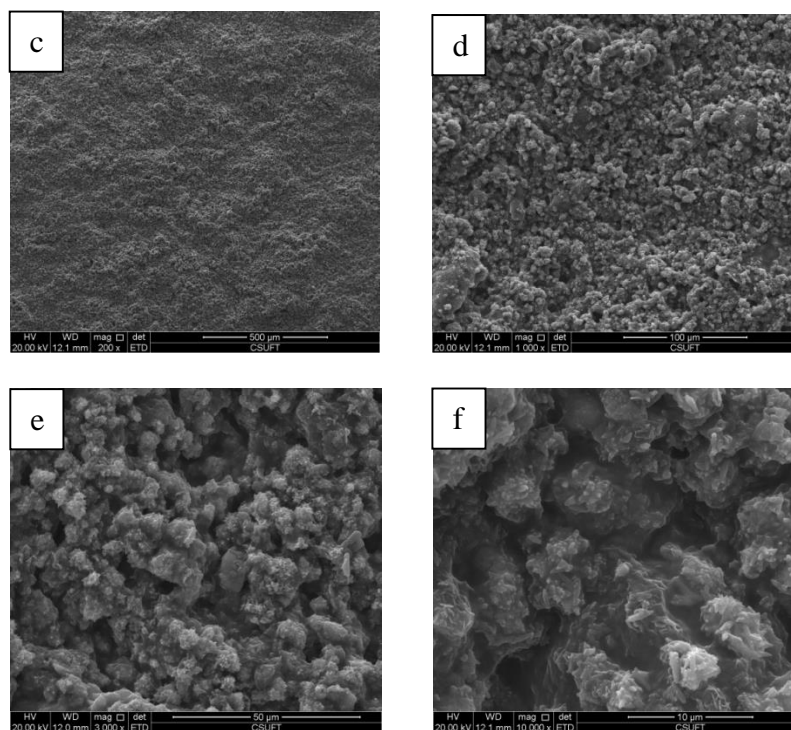


Fig. 6. SEM images of cross-linked sodium silicate modified by combined chemicals (c 200x,d 1000x,e 3000x,f 10000x)

Table 4. Infrared Absorption Spectral Data

No.	Wavenumber/cm ⁻¹	Designation
1	3440	Si-OH stretching vibration band
2	2940	-CH ₂ - stretching vibration band
3	2360	CO ₂ absorption peak
4	1590	Si-O-Si stretching vibration band
5	1426	CO ₃ ²⁻ characteristic peak
6	1080	C-O-O stretching vibration band
7	874	Si-OH bending vibration
8	797	Si-O-Si bending vibration

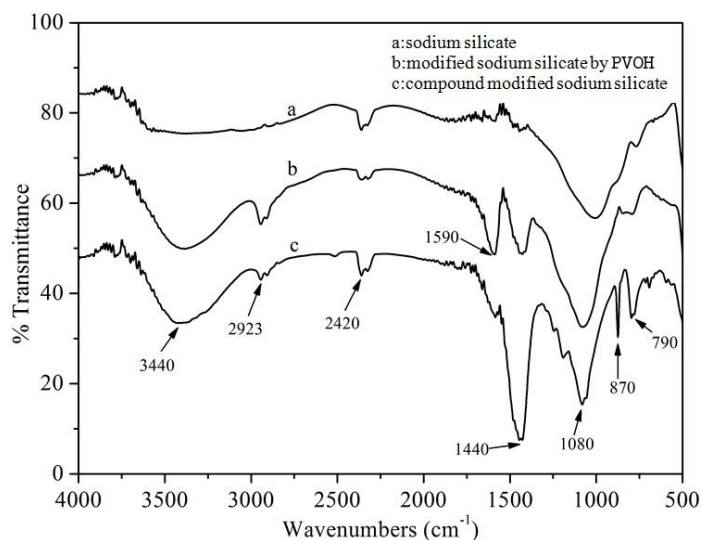


Fig. 7. IR spectra of sodium silicate after different modification methods

Notably, after modification by PVOH, the IR spectra of sodium silicate exhibited more absorption peaks than before. The new peaks corresponded to the stretching vibration band of Si-OH, the stretching vibration band of $-\text{CH}_2-$, and the stretching vibration band of Si-O-Si at 3440 cm^{-1} , 2940 cm^{-1} , and 1590 cm^{-1} , respectively. However, after adding organic additives and inorganic fillers, the CO_3^{2-} characteristic peak was much stronger than that of modification by PVOH, and this led to a lower stretching vibration band of Si-O-Si. Meanwhile, the bending vibration of Si-OH at 874 cm^{-1} was also noticeably stronger. These findings indicate that, compared to modification by PVOH, combined chemicals can further promote the curing of adhesive to improve its bonding strength.

Curing Properties by DSC Analysis

The curing behaviors of sodium silicate before and after different modification were tested by differential scanning calorimetry (DSC), and the results are presented in Figs. 8 and 9. The sample pans were heated from 30 to $1000\text{ }^\circ\text{C}$ at a rate of $15\text{ }^\circ\text{C}/\text{min}$. The thermal transitions of the samples are defined as T_o (onset), T_p (peak of curing), and T_c (conclusion), and the enthalpy of curing is referred to as ΔH_{gel} . The enthalpies were calculated on a dry sodium silicate basis. The curing temperature range (R) was calculated as $2(T_p - T_o)$, as described by Krueger *et al.* (1987).

It is apparent that the curing temperature of modified sodium silicate by compound was higher than that of pure sodium silicate and sodium silicate modified by PVOH. Compound modified sodium silicate showed T_o and T_p between 180.67 and $254\text{ }^\circ\text{C}$, whereas pure sodium silicate and the same after PVOH modification just showed values between 114.06 and $187.05\text{ }^\circ\text{C}$ and between 193.7 and $247.2\text{ }^\circ\text{C}$, respectively. After compound modification, the necessary enthalpy for the adhesive curing (DSC exothermic peak area) was 48.45 J/g and 8.74 J/g lower than for pure sodium silicate and modified sodium silicate by PVOH. This result indicates that lower enthalpy is needed for silicate adhesive system when organic additives and inorganic fillers are added, which means that it is easier to be cured. Meanwhile, the initial curing temperature of compound modified sodium silicate is $13.03\text{ }^\circ\text{C}$ lower than that of the one after PVOH modification. This means that the compound modified adhesive began to cure at a lower temperature.

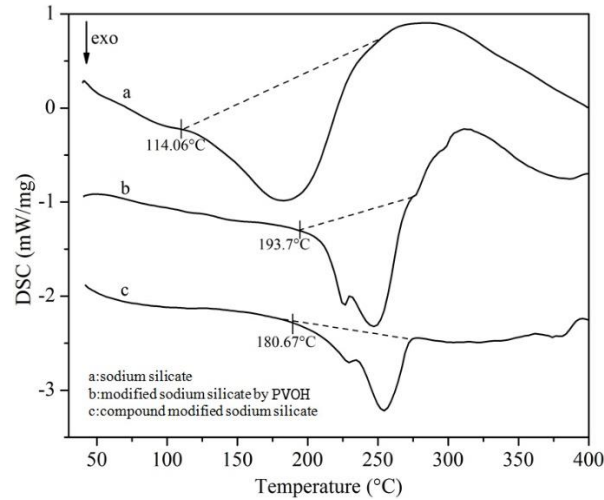


Fig. 8. DSC curves of sodium silicate after different modification methods

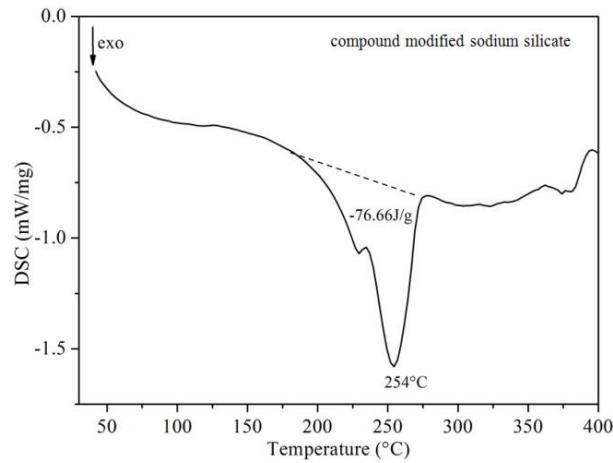


Fig. 9. DSC curves of sodium silicate after compound modification method

Thermal Stability

The thermal stability of the pure sodium silicate and modified silicate with different methods are shown in Figs.10 to 13.

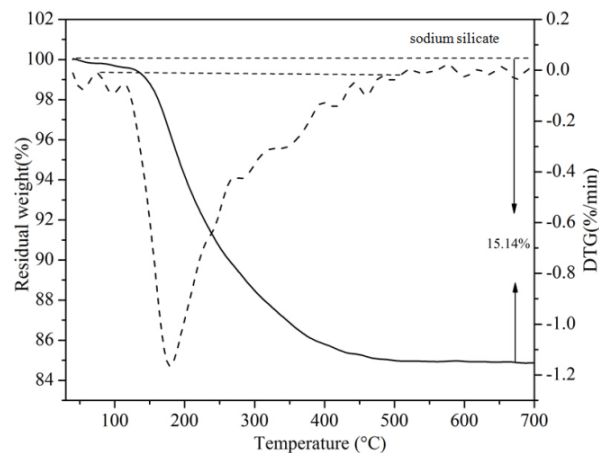


Fig. 10. TGA curves of pure sodium silicate

The samples (5 mg, dry weight) were loaded into an aluminum oxide pan. Experiments were tested from 30 °C to 1000 °C at a heating rate of 15°C/min and with a 30 mL/min argon gas flow. The thermo-gravimetric parameter of the native sodium silicate samples were defined as T_o (onset), T_i (inflection), T_c (conclusion), and residual mass.

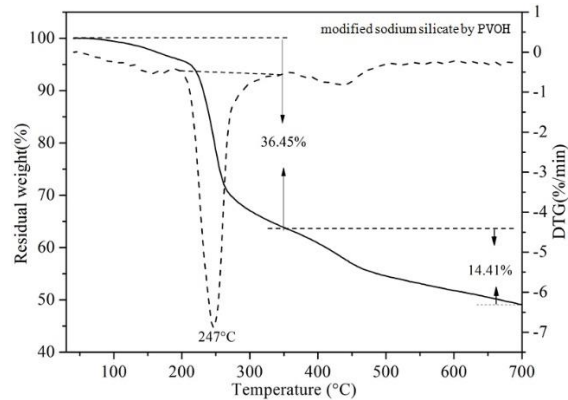


Fig. 11. TGA curves of sodium silicate modified by PVOH

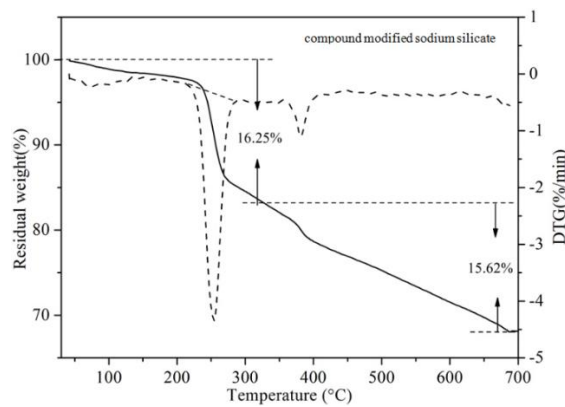


Fig. 12. TGA curves of compound modified sodium silicate

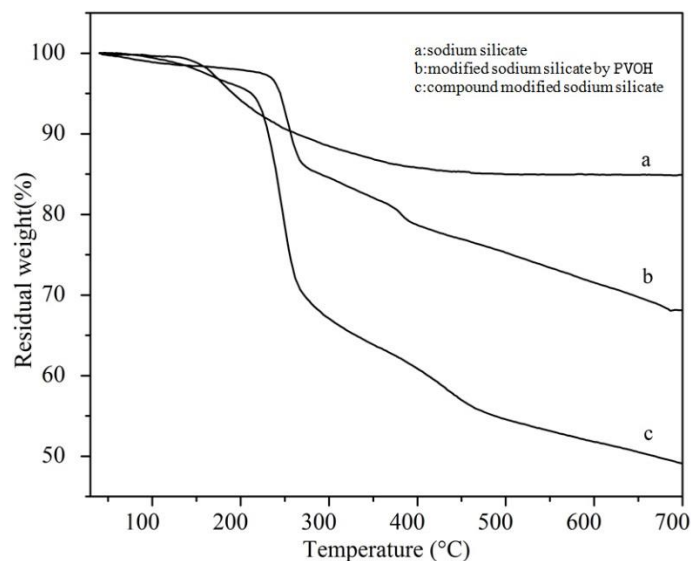


Fig. 13. Comparison of TGA curves of sodium silicate before and after modification

As shown in the above figures, the TGA curves of sodium silicate before and after modification were remarkably different. For pure sodium silicate, there was only one stage of decomposition and the total weight loss rate was just 15.14%. However, after being modified by PVOH, there were two stages of decomposition. Obviously, after introducing organic material (PVOH) into the adhesive system, although the bonding property was improved, its total weight loss rate increased sharply to 50.86%. That means that the thermal stability decreased in an overall sense. On the other hand, for compound modified sodium silicate, there were also two stages of decomposition with the total weight loss rate of 31.87%. So the thermal stability is improved.

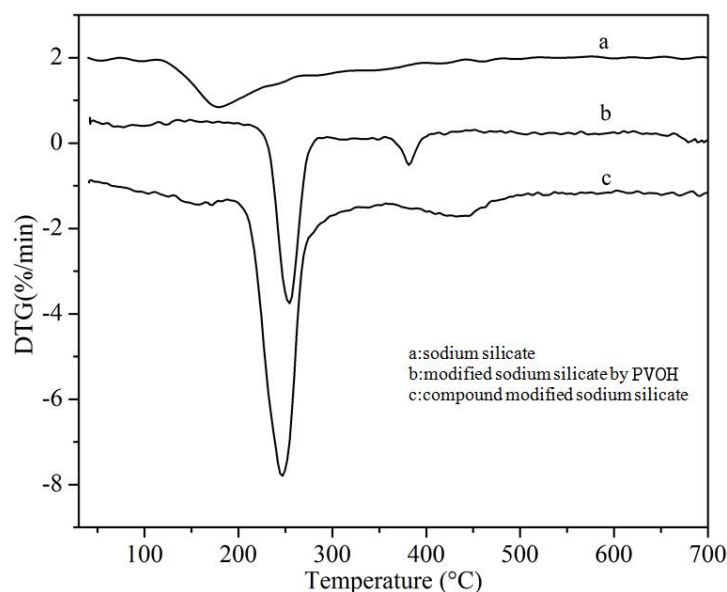


Fig. 14. Comparison of DTG curves of sodium silicate before and after modification

From the data presented above, it can be easily seen that sodium silicate modified by PVOH and compound reached the maximum rates of thermal decomposition at almost the same temperature. However, the peak height and peak areas varied remarkably. The DTG peak height and areas of compound modified adhesive were both smaller than that of PVOH-modified sodium silicate. This indicates that the adhesive system after compound modification had a lower thermal decomposition rate and it had a higher level of thermal stability.

CONCLUSIONS

1. For organic additives, the best ratio of amino acid, TEOS, and sodium dodecyl sulfate to modify sodium silicate adhesive cross-linked by PVOH is 5:3:3.
2. For inorganic fillers, the best ratio of active magnesium oxide and nano-silica is 1:1.
3. Compound A consists of 5:3:3 of amino acid, TEOS, and sodium dodecyl sulfate. Compound B consists of 1:1 of active magnesium oxide and nano-silica. When the ratio

of A and B is 90:3, the bonding strength and water resistance properties of compound modified sodium silicate adhesive are the best.

4. Compared to pure sodium silicate, the bonding strength and water resistance properties of compound modified sodium silicate improved by 97.6% and 46.7%, respectively. Compared to cross-linked sodium silicate by PVOH, the improvements are 13.7% and 36.0%, respectively.
5. The synergistic effect of the compound also improves the thermal stability of sodium silicate adhesive, which decreases sharply after being modified by PVOH.
6. Based on the cross-linked structure of sodium silicate and PVOH, the adhesive after compound modification shows the potential to be an environmental friendly wood adhesive to replace traditional adhesives in particular circumstances.

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