

Interfacial Compatibility of Wood Flour/Polypropylene Composites by Stress Relaxation Method

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ABSTRACT: To investigate the interfacial compatibility of wood flour/polypropylene (PP) composite, compressive stress relaxation curves at different temperatures (26, 40, and 60°C) were determined at various wood contents (0, 20, 30, 40, 50, 60, and 70%) without coupling agent and further determined at wood contents of 50 and 60% by using maleic anhydride grafted polypropylene (MAPP) and silane as coupling agents. The apparent activation energy (ΔE) was also calculated according to the Eyring's absolute rate reaction theory. The results showed that the stress relaxed faster at higher temperatures. ΔE of wood flour/polypropylene composites without coupling agent showed the highest value at wood content of 40%, suggesting that the best interfacial compatibility between wood and PP occurred at 40% wood content. Adding coupling agents increased ΔE obvi-

ously at higher wood contents (50 and 60%). With the increasing loading level of coupling agents, ΔE increased at first and then decreased gradually or finally reached a constant level depending on the type of coupling agent. The optimal loading of coupling agent corresponding to the highest ΔE value was related to wood content and the type of coupling agent. Within the experimental conditions used in this study, the optimal loading level for MAPP was 2% at both wood contents while for silane it was 1.5% and 2% at wood content of 50 and 60%, respectively. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 000: 000–000, 2012

Key words: wood flour/polypropylene composite; coupling agent; compressive stress relaxation; temperature; apparent activation energy

INTRODUCTION

Wood flour/polymer composites have been widely used in many fields such as construction, mobile industry, and packaging since they are cost-effective and exhibit good performance in many properties. Within all investigations on wood flour/polymer composites, interfacial compatibility is always the key problem that calls for solution.^{1–4} Many researchers have discussed the mechanisms of coupling agent modified wood flour/polymer composites by various methods such as environmental scanning electron microscope (ESEM),⁵ electron spectroscopy for chemical analysis (ESCA),¹ dynamic mechanical analysis (DMA),⁶ scanning electron microscope (SEM),⁷ differential scanning calorimetry (DSC),⁸ wide-angle X-ray diffraction (WAXD),⁹ and Fourier transform infrared spectroscopy (FTIR).¹⁰ It was found that coupling agent can form chemical bonds on the surface of wood and the interface between wood and polymer and it can well infiltrate the surface of wood, which

finally result in lower surface tension of wood material.^{1,2,5,11–14} It was also indicated that physical bonds and chemical bonds might both be formed between coupling agent, wood flour, and polymer in the composite.^{15–19} Recently, attempts were made to quantitatively evaluate the interfacial bonding between cellulosic material and polymer. For example, X-ray photoelectron spectroscopy (XPS) was used to investigate the reactivity of different coupling agents with surface hydroxyl groups on fibrous cellulose,²⁰ and many advanced imaging technologies such as full-field imaging techniques and X-ray computed tomography (CT) combined with numerical modeling tools were also used for better understanding of interaction between wood particles and polymer, as introduced in Muszynski.²¹ These methods usually depend on expensive equipments and the interpreting of the results tends to be complicated. Renner et al.²² applied micromechanical method to study the effect of particle characteristics on interfacial bonding of wood flour/PP composites and put forward a parameter B to represent the extent of reinforcement, which gave a simple way to describe the adhesion between the components in composite regardless of different mechanisms during the micromechanical deformation processes.

Stress relaxation approach is similar to the above mentioned micromechanical method theoretically,

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which will also not take into account the different mechanisms of the stress relaxation process but gives a parameter to describe the overall interfacial bonding state in the composite. Moreover, it is different from that micromechanical method because it can quantitatively evaluate the bonding in wood flour/polymer composites with different wood flour fractions. Stress relaxation refers to the fact that at a fixed strain, the stress resistance to the deformation decreases with increasing time.²³ When a material is elongated, compressed or undergoes other deformations within the elastic region, a stress would produce in the material to try to bring the material back to its original form. If the deformation is fixed, then the stress will decrease with the breakage of original bonding and the formation of new bonding. By using Eyring's absolute rate reaction theory,²⁴ the apparent activation energy of the reactions during this process can be evaluated to analyze the internal bonding quantitatively. Recently, Xie and Zhao²⁵ discussed the number of bridging and the type of cross linking reaction during axial tensile stress relaxation by measuring the intermittent stress relaxation. In recent years, the stress relaxation approach has been successfully applied to study the interaction between wood and chemicals used to modify wood. For example, Cao et al.²³ investigated the interaction between copper ethanolamine (Cu-EA) and hydroxyl groups of wood in Cu-EA treated wood by stress relaxation approach and found that the influence of Cu-EA treatment on stress relaxation of wood resulted from the combined effect of three mechanisms. Yu et al.²⁶ tested the tensile stress relaxation curves of wood during impregnation with different formulations of alkaline copper quat (ACQ) solutions and studied the interaction between ACQ constituents on wood. Yan et al.²⁷ calculated the apparent activation energy during the compressive stress relaxation of poplar samples pretreated with glycerin at temperatures ranging from 25 to 180°C and found that the apparent activation energies were 8.24 and 81.38 kJ/mol respectively for temperatures below and above 120°C, which corresponded to the hydrogen bonds formed between wood and glycerin molecules and the formation of new cross-linking with the degradation of hemicelluloses or lignin.

In our previous study, the tensile stress relaxation and compression stress relaxation of wood flour/PP composites were compared at room temperature and the results showed that the stress relaxation curves are similar from the two approaches.²⁸ Therefore, the compressive stress relaxation approach was explored to investigate the interaction between the elements in wood flour/PP composites and further the effect of wood content and the loading level of coupling agent content on it. Furthermore, the

results of stress relaxation of the composites at various temperatures can make reference for long-term performances of wood flour/polymer composites used at different conditions.

EXPERIMENTAL

Materials

The wood flour of poplar (*Populus tomentosa* Carr.) with a size passed 100 mesh sieve and polypropylene (PP) (K8303, Sinopec Chemical Products Sales Company, China) with a density of 0.9 g/cm³ were used as raw materials to prepare wood flour/polypropylene composite. The PP used is injection molding grade with a melt temperature around 165°C and a melt flow index (MFI) of 1.5–2.0 g/10 min at 230°C. The coupling agents used in this study included MAPP and silane [γ -methacryloxypropyltrimethoxysilane, CH₂=C(CH₃)COOCH₂CH₂CH₂Si(OCH₃)₃] with density of 1.04 g/cm³ which is designated as KH-570 by the Chinese Academy of Science. Polyfluotetraethylene (PTFE) membranes were used as demoulding materials to avoid sticking on boards during hot-pressing.

Wood flour/polypropylene composite manufacturing

Wood flour/polypropylene composite in this study was made by compression molding method after high speed blending to simplify manufacturing process and avoid the effect of manufacturing process on the properties. The size of the panels with was 270 mm × 270 mm × 3 mm and had a target density of 1.0 g/cm³. Wood flour/polypropylene composite without coupling agent were made at wood contents of 70, 60, 50, 40, 30, 20 and pure PP panel was also made as control samples. For wood contents of 50 and 60%, six different MAPP loading levels (0.5,1,1.5,2,4, and 8% of the total weight of wood flour and PP) and silane (0.5,1,1.5,2,3, and 5% of the total weight of wood flour and PP) were used.

Wood flour and PP are dried at 103 ± 2°C for 5 h to reach a moisture content lower than 5%. The wood flour, PP and coupling agents were weighed and then blended in a high speed blender (SHR-10, Beijing Zedao Machinery Ltd., China) at a rotation speed around 1000 r/min at about 20°C for 4 min to make the mixture more homogenous. The mixture was oven-dried and then taken out for hand matting. A hot press (SYSMEN-II, made by Chinese Academy of Forestry, China) was used to compress the mat at 180°C and 4 MPa for 6 min. After hot pressing, the formed mat was pressed at 4 MPa for another 6 min at room temperature in a cold press. The same method was used to prepare the PP control panels.

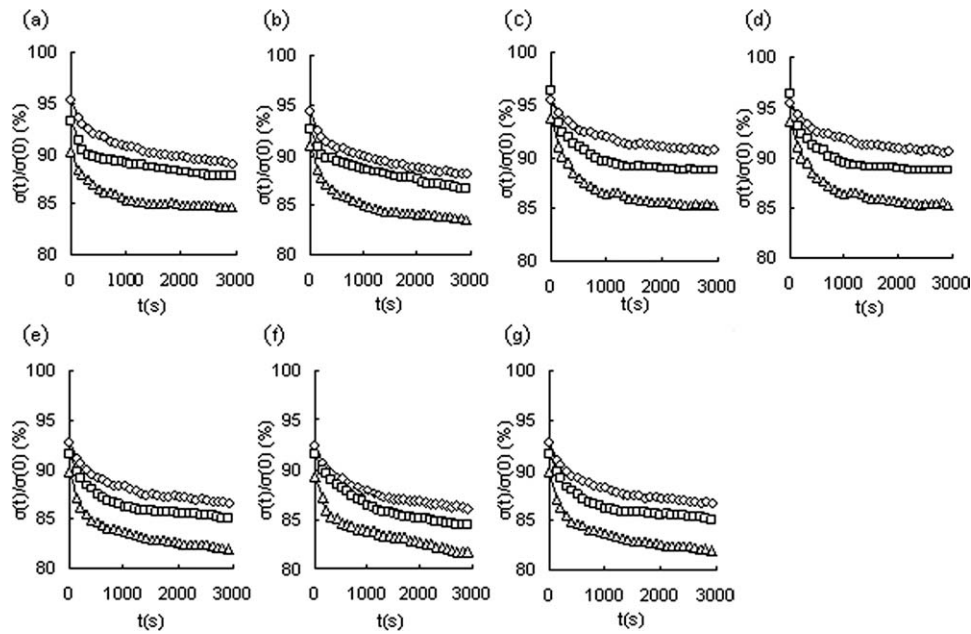


Figure 1 Compressive stress relaxation of wood flour/polypropylene composite without coupling agent at three temperatures of 26, 40, and 60°C Wood contents: (a) 0%; (b) 20%; (c) 30%; (d) 40%; (e) 50%; (f) 60%; and (g) 70%. Temperatures: \diamond , 26°C; \square , 40°C; \triangle , 60°C.

Determination of compressive stress relaxation curves

The compressive stress relaxation tests were performed on the self-assembled equipment at the temperature of 26 ± 1 , 40 ± 1 , and $60 \pm 1^\circ\text{C}$. The diagram of the equipment could reference to Cao et al.²³ The test for each sample lasted 3000 s. The size of samples is 10 mm \times 10 mm \times 3 mm (length \times width \times thickness). The compressive displacement is 20% of its original thickness. Three replicates were used for each condition.

Calculation of the apparent activation energy

Wood is a combination of polymers and has various relaxation mechanisms. The relationship between relaxation time and temperature conforms to Eyring’s absolute rate reaction theory,²⁴ as showed in eq. (1).

$$\tau_m = \tau_0 e^{\frac{\Delta E}{RT}} \tag{1}$$

where, τ_m is average relaxation time (s), τ_0 is a constant, R is gas constant, $R = 8.314 \text{ J}/(\text{mol}\cdot\text{K})$; T is absolute temperature (K) and ΔE is apparent activation energy (KJ/mol). Equation (1) can be logarithmically transformed to eq. (2).

$$\ln \tau_m = \ln \tau_0 + \frac{\Delta E}{RT} \tag{2}$$

where, τ_0 is a constant, so $\ln \tau_0 = 0$. Equation (2) can be differentially transformed to eq. (3).

$$d(\ln \tau_m) = \frac{\Delta E}{R} d\left(\frac{1}{T}\right) \tag{3}$$

Equation (3) can be rewritten as

$$\Delta E = R \frac{d(\ln \tau_m)}{d\left(\frac{1}{T}\right)} \tag{4}$$

Apparent activation energy can be calculated from the slope of the $\ln \tau_m - 1/T$ curve by plotting $1/T$ as abscissa axis and $1/T$ as ordinate axis according to eq. (4).

RESULTS AND DISCUSSION

Stress relaxation of wood flour/polypropylene composites at various temperatures

The stress relaxation curves of wood flour/PP composites without coupling agent for different wood contents at temperatures of 26, 40, and 60°C are showed in Figure 1. The stress relaxes faster at higher temperature, suggesting that not only pure PP but also wood flour/PP composites are sensitive to temperature. This is reasonable since wood is also composed by various polymers. With the increasing temperature, the molecules or chains of polymers obtain more energy and the “free volume” in the material became wider, which finally results in the breakage of bonds and accelerates the stress relaxation process.²³

To further analyze the stress relaxation curves of wood flour/PP composites, the rate of stress

TABLE I
Rates of Compressive Stress Relaxation of Wood Flour/PP Composites With or Without Coupling Agent

Type of coupling agent	Wood content (%)	Content of coupling agent (%)	Rate of stress relaxation ($\times 10^{-2}$)		
			26°C	40°C	60°C
No	0	–	2.37 (0.97 ^a)	2.61 (0.92)	2.85 (0.87)
	20	–	1.53 (0.91)	1.69 (0.99)	1.90 (0.98)
	30	–	1.50 (0.95)	1.65 (0.99)	1.85 (0.81)
	40	–	1.20 (0.92)	1.35 (0.95)	1.55 (0.97)
	50	–	1.60 (0.97)	1.73 (0.99)	1.91 (0.92)
	60	–	1.63 (0.91)	1.76 (0.98)	1.91 (0.90)
	70	–	1.93 (0.89)	1.95 (0.97)	2.01 (0.99)
MAPP	50	0.5	1.83 (0.96)	1.97 (0.93)	2.09 (0.87)
	50	1.0	1.49 (0.89)	1.64 (0.81)	1.88 (0.87)
	50	1.5	1.50 (0.92)	1.61 (0.87)	1.76 (0.93)
	50	2.0	1.43 (0.95)	1.51 (0.91)	1.60 (0.88)
	50	4.0	1.45 (0.93)	1.59 (0.87)	1.71 (0.84)
	50	8.0	1.49 (0.93)	1.67 (0.95)	1.77 (0.79)
	60	0.5	1.60 (0.98)	1.74 (0.99)	1.90 (0.80)
	60	1.0	1.49 (0.93)	1.73 (0.92)	1.83 (0.86)
	60	1.5	1.38 (0.98)	1.52 (0.81)	1.79 (0.84)
	60	2.0	1.35 (0.92)	1.41 (0.85)	1.60 (0.90)
	60	4.0	1.42 (0.97)	1.46 (0.99)	1.90 (0.96)
	60	8.0	1.47 (0.99)	1.65 (0.84)	1.79 (0.92)
Silane	50	0.5	1.53 (0.92)	1.65 (0.80)	1.89 (0.97)
	50	1.0	1.35 (0.99)	1.56 (0.83)	1.71 (0.99)
	50	1.5	1.31 (0.98)	1.47 (0.92)	1.60 (0.80)
	50	2.0	1.32 (0.95)	1.55 (0.96)	1.72 (0.80)
	50	3.0	1.57 (0.98)	1.61 (0.89)	1.85 (0.82)
	50	5.0	1.53 (0.99)	1.60 (0.89)	1.85 (0.80)
	60	0.5	1.66 (0.94)	1.78 (0.97)	1.84 (0.99)
	60	1.0	1.69 (0.93)	1.75 (0.80)	1.87 (0.84)
	60	1.5	1.41 (0.96)	1.48 (0.84)	1.69 (0.87)
	60	2.0	1.11 (0.98)	1.24 (0.85)	1.38 (0.96)
	60	3.0	1.14 (0.97)	1.37 (0.96)	1.56 (0.82)
	60	5.0	1.67 (0.98)	1.72 (0.94)	1.86 (0.91)

^a The values in parentheses represent the R^2 of the regression linear curves of double logarithm curve of $\sigma(t)/\sigma(0)$ versus t (0–1000 s).

relaxation was defined as the slope of double logarithm curve of stress relaxation curve. As showed in Figure 1, stress reduces with time increasing in a given period, which corresponds to a linear relationship while plotted in double logarithm form. The detailed procedure was described in Cao et al.²³ Before a relaxation time of 1000 s, the linear relationship is very clear and shows little sign of deviation. Therefore, the slope of the double logarithm curve of stress relaxation curve (t : 0–1000 s) is finally taken as the rate of stress relaxation. The rates of stress relaxation are listed in Table I, as well as the values of R^2 in Table I.

As showed in Figure 1 and Table I, the trend of stress relaxation of wood flour/PP composites without coupling agent at each temperature is consistent. Stress relaxes the fastest at wood content of 0% (pure PP) and the slowest at wood content of 40%. The rates of stress relaxation increases with temperature rising and the increment reduces with wood content increasing. The fact that PP show more significant change with increasing temperature than

wood flour leads to fairly larger displacement of molecular chains and finally faster stress relaxation. As a result, the difference of the stress relaxation rates between three temperatures narrows with the increasing wood content. For example, the difference between the rates of stress relaxation at 26 and 40°C for pure PP is 0.0024, but it is only 0.0002 at wood content of 70%.

After adding MAPP or silane coupling agent, stress relaxes slower, which can be clearly seen from the rates of stress relaxation as listed in Table I. At each temperature, the slowest rate of stress relaxation occurs at MAPP content of 2%. The trends of stress relaxation of wood flour/PP composites modified by silane is similar to those modified by MAPP except that the slowest rate of stress relaxation occurs at silane content of 1.5% for wood content of 50%. With temperature increasing, stress relaxes faster and the increment in stress relaxation rate of wood flour/PP composites modified by coupling agent is less than those without coupling agent. Coupling agent can react with wood flour molecule

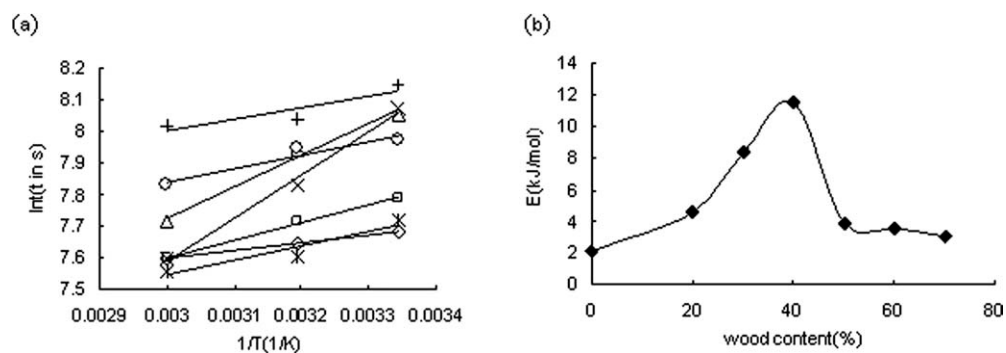


Figure 2 The apparent activation energy of wood flour/PP composites without coupling agent at various wood contents: \diamond : 0%, $y = 245.81x + 6.8604$, $R^2 = 0.99$; \square : 20%, $y = 558.2x + 5.9255$, $R^2 = 0.99$; \triangle : 30%, $y = 1008.2x + 4.6985$, $R^2 = 0.98$; \times : 40%, $y = 1382.9x + 3.4369$, $R^2 = 0.99$; $*$: 50%, $y = 464.98x + 6.1489$, $R^2 = 0.91$; \circ : 60%, $y = 429.43x + 6.5495$, $R^2 = 0.99$; $+$: 70%, $y = 391.05x + 6.8545$, $R^2 = 0.91$.

to form chemical bond and twist with PP,^{15,29} resulting in stronger bonding force and limitation of free movement of PP. Consequently, stress relaxation is slowed down.

Kinetics of stress relaxation of wood flour/polypropylene composite

The average relaxation time was taken as the point of the stress relaxation curve which starts contact with the tangent parallel to the abscissa axis in this study. Each temperature corresponds to a value of $\ln\tau_m$. The apparent activation energy (ΔE) can be calculated from the slope of the curves of $\ln\tau_m$ versus $1/T$ according to eq. (4).

In this study, the values of ΔE of wood flour/PP composites with all the conditions range from 2.04 to 12.94 kJ/mol, suggesting the occurrence of physical stress relaxation which is mainly caused by the sliding between wood molecules, the break-down of the intermolecular hydrogen bonds, the forming of new hydrogen bonds between wood molecules or molecules of wood and modifying agents.²⁷

The curves of $\ln\tau_m$ versus $1/T$ of wood flour/PP composites without coupling agent are given in Figure 2(a). It can be found that the values of R^2 range from 0.91 to 0.99, suggesting a good linearity

between $\ln\tau_m$ and $1/T$ at all wood contents used in this study. As showed in Figure 2(b), ΔE of wood flour/PP composites without coupling agent increases first and then decreases with wood content increasing and achieves the lowest at wood content of 40%, which is consistent with the result of stress relaxation. Higher apparent activation energy corresponds to the requirement of more energy to get the stress relaxed, which means a better internal bonding and compatibility between the molecules in the composite. Correspondingly, the bending modulus of rupture (MOR) and modulus of elasticity (MOE) at 40% wood content also reached their maximum according to our previous study.²⁸ Wood flour plays the role of a nucleating agent and provides sites for heterogeneous nucleation.^{4,9} At lower wood content, enough PP could encapsulate wood flour quite well. As a result, structure of wood flour/PP composites becomes more compact with wood content increasing. With further increases in the wood flour content, the nucleating effect of wood flour which agglomerates easily and the appearing voids cause poor interfacial adhesion and incompact structure between wood flour and PP.^{1,2,30}

ΔE of MAPP modified wood flour/PP composites at wood content of 50 and 60% are showed in Figure 3(a,b), respectively. ΔE increases first and

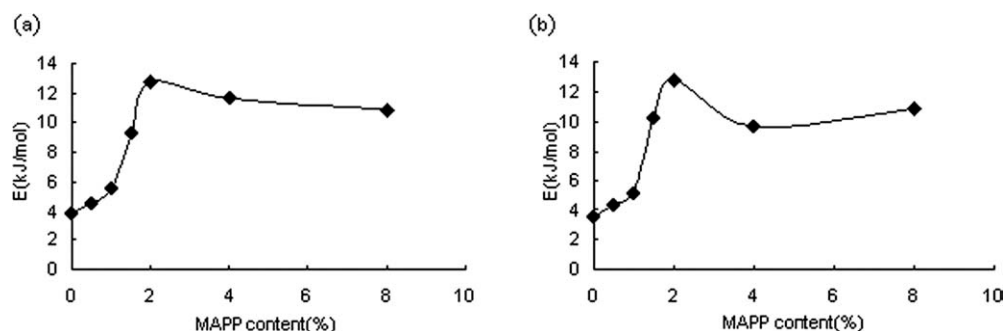


Figure 3 The apparent activation energy of wood flour/PP composites using different concentrations of MAPP as coupling agent at wood contents of 50% (a) and 60% (b).

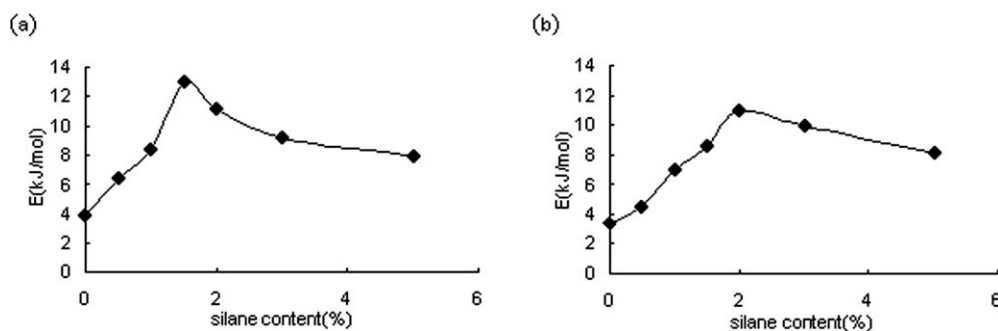


Figure 4 The apparent activation energy of wood flour/PP composites using different concentrations of silanes as coupling agent at wood contents of 50% (a) and 60% (b).

then decreases with MAPP content increasing and reaches the highest at MAPP content of 2%. After adding MAPP, MAPP may react with wood flour to form chemical bonding through esterification and twisted with PP resulting in stronger internal bonding and structure. During the stress relaxation process, the debonding of molecules between wood/MAPP/PP may take place with enough heating duration even under a low temperature below 100°C. The chemical debonding will require more energy than the physical stress relaxation processes. Therefore, the addition of suitable amount of MAPP increases the activation energy. However, if the amount of MAPP is higher than that required for reacting with wood molecules (2–4%), the excessive MAPP molecules will conglomerate together leading to weaker bonding between MAPP and PP, or MAPP molecules themselves, which finally causes faster stress relaxation. At high MAPP loading levels, excessive MAPP molecules play the role of a polymer and do not participate any further in the interaction between PP and wood flour when the wood flour content is kept constant.³¹ With further increase of MAPP loading ($\geq 4\%$), the fraction of chemical debonding between wood and MAPP molecules in all stress relaxation mechanisms becomes smaller and the physical debonding is predominant, resulting in gentle change of activation energy.

ΔE of silane modified wood flour/PP composites at wood content of 50 and 60% are showed in Figure 4(a,b), respectively. As similar to MAPP, ΔE reaches the highest at silane content of 2% except that at wood content of 50%, the highest apparent activation energy occurred at a silane loading of 1.5%. With silane content increasing, the interaction between wood flour and PP improves and the internal structure becomes more compact, but excessive silane molecules may also weaken the compatibility and bonding force between wood flour and PP, which is similar to MAPP. However, the trend of ΔE curve is different beyond the optimal loading while using different coupling agents (MAPP or

silane). As showed in Figure 3, ΔE of wood flour/PP composites decreases at first with 2–4% MAPP and then shows very small change at MAPP exceeding 4%. However, the ΔE of wood flour/PP composites using silane as coupling agent keeps decreasing beyond the optimal loading. This limited coupling effect of silane is considered to be related to the absence of covalent bonds between the functional groups of silane and the thermoplastic matrices. Besides, the residual silanol groups in the wood flour will further condense with each other and the covalent $-\text{Si}-\text{O}-\text{C}-$ bonds may not be stable.³² As a result, the negative effect of excessive silane molecules on the interfacial compatibility between wood flour and PP becomes obvious.

CONCLUSIONS

The result of this study showed that stress relaxation could be a good measure to study the interaction between the constituents in wood flour/polymer composites by evaluating the internal bonding quantitatively. The ΔE values of all wood flour/PP composites made in this study ranged from 2.04 to 12.94 kJ/mol, suggesting the predominance of physical stress relaxation and the occurrence of chemical debonding during stress relaxation of the composites with coupling agents. In addition, MAPP and silane both could effectively improve the interaction between wood flour and PP at higher wood contents, but excessive loading of coupling agent appeared negative effect on the properties. The wood flour/PP composites showed best properties at a MAPP loading of 2% or silane loadings of 1.5% and 2% at wood contents of 50% and 60%, respectively. This is corresponding to the lowest stress relaxation rate. The influence of the manufacturing process of wood flour/polymer composites would be considered in our further study.

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