

# Characterization of Mechanical Properties of Jute/PLA Composites Containing Nano SiO<sub>2</sub> Modified By Coupling Agents

**Xueyang Song**

Soochow University

**Cuicui Fang**

Soochow University

**Yuanyuan Li**

Soochow University

**Ping Wang** (✉ [pingwang@suda.edu.cn](mailto:pingwang@suda.edu.cn))

Soochow University <https://orcid.org/0000-0001-6692-3850>

**Yan Zhang**

Soochow University

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## Research Article

**Keywords:** laminating composite materials, Jute non-woven, PLA, biodegradable, nano-SiO<sub>2</sub>

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1           Characterization of mechanical properties of  
2           jute/PLA composites containing nano SiO<sub>2</sub>  
3           modified by coupling agents

4           Xueyang Song, Cuicui Fang, Yuanyuan Li, Ping Wang, Yan Zhang\*

5           National Engineering Laboratory for Modern Silk, College of Textile and Clothing  
6           Engineering, Soochow University, Suzhou, China

7           \* Corresponding author: Yan Zhang (yanzhng86@suda.edu.cn)

8  
9           **Abstract:** Although jute fibers-reinforced PLA composites shows strong application  
10          prospects, their low mechanical properties limit their applications to some extent. In  
11          this paper, nano-SiO<sub>2</sub> particles as well as modified nano SiO<sub>2</sub> by coupling agents  
12          which can efficiently improve the strength and toughness of composite materials are  
13          introduced into the PLA matrix. The bending, stretching and thermal properties of  
14          designed jute/PLA nonwoven composites were studied. The study shows that the  
15          nano-SiO<sub>2</sub> particles are beneficial to the interface performance between the PLA  
16          matrix and jute leading to improvement in the mechanical property and thermal  
17          stability. Moreover, thermomechanical properties indicate that the addition of SiO<sub>2</sub>  
18          can improve the jute/PLA interfacial adhesion and increase the glass transition  
19          temperature of the material. Finally, toughening mechanism of nano-SiO<sub>2</sub> particles in  
20          the jute/PLA composite was analyzed.

21          **Keywords:** laminating composite materials; Jute non-woven; PLA; biodegradable;  
22          nano-SiO<sub>2</sub>

23

## 24 **Introduction**

25 In recent years, thermoplastic composites are increasingly applied in the  
26 high-performance engineering field due to their low density, good recyclability, and  
27 high production efficiency (Faruk et al. 2012). However, environmental compatibility  
28 of these materials is still a huge challenge. In this case, fully biodegradable green  
29 composite materials provide a feasible solution. To prepare a completely  
30 biodegradable composite material, natural fiber is a desirable reinforcing material and  
31 natural polymer can be selected as matrix material, such as jute/polylactic acid (PLA)  
32 composite materials. PLA, a polymer material, is produced from renewable  
33 agricultural raw materials. It has tremendous advantages such as high strength, high  
34 hardness, transparency and industrial production, and is one of the most promising  
35 biodegradable polymers (Qu et al. 2015). Among all biodegradable reinforced fibers,  
36 jute fiber has low cost, biodegradability and excellent mechanical property, which has  
37 made it as one of the most widely used natural plant fibers in the research of green  
38 bio-based composites (Dong et al. 2014). There are wide possible applications for  
39 jute/PLA composite, and a lot of research have been proposed to investigate its  
40 mechanical properties (Bax and Mussig 2008; Mohammed et al. 2019; Nishino et al.  
41 2003; Pan et al. 2007). However, PLA resin matrix lacks chemical bonds and  
42 functional groups that react with jute. Therefore, the interface compatibility between  
43 jute fiber and PLA is poor, leading to a poor mechanical property of final composite.  
44 (Balla et al. 2019; Costa et al. 2012; He et al. 2019; Hossen et al. 2020; Pereira et al.  
45 2019). At present, some physical and chemical methods have been proposed,  
46 including fiber treatment technologies such as acylation, alkali treatment,  
47 etherification and isocyanate treatment, as well as the use of coupling agents to  
48 improve the interfacial bonding properties of jute/PLA composites (Dong et al. 2020;  
49 Fang et al. 2020; Huda et al. 2007; Kumar et al. 2010; Kakroodi et al. 2012). Nano  
50 particle is also a feasible option to toughen the polymer matrix and improve the  
51 mechanical properties of fiber-reinforced composites.

52 In recent years, nano particles have been extensively applied to prepared  
53 composites, which has significantly improved the mechanical properties of

54 composites materials (Gauvin et al. 2015; Jumahat et al. 2012; Jumahat et al. 2010).  
55 Among the nanoparticles used as reinforcing materials, many researchers chose  
56 doping nano-silica (nano-SiO<sub>2</sub>) to improve the mechanical properties of  
57 fiber-reinforced composite materials (Jumahat et al. 2015; Jumahat et al. 2012;  
58 Jumahat et al. 2010). Luo et al. (2018) used technology-resin film infusion (RFI)  
59 technology to prepare nano-SiO<sub>2</sub> modified carbon fiber reinforced composite  
60 materials. When the content of nano-SiO<sub>2</sub> is 4 wt%, the toughening and strengthening  
61 effect is the best. The tensile strength and flexural strength increased to 595.69 and  
62 703.76 MPa, respectively, which were 86.30% and 126.98% higher than the tensile  
63 strength and flexural strength of intact sample without SiO<sub>2</sub>. Hashim et al. (2019)  
64 studied the effect of SiO<sub>2</sub> nanoparticles as a reinforcing filler on the tensile response  
65 of basalt fiber reinforced polymer (BFRP) composites. It was found that the addition  
66 of SiO<sub>2</sub> nanoparticles showed significant improvement in tensile modulus with 6%,  
67 14% and 19% for 5 wt%, 15 wt% and 25 wt% nano-SiO<sub>2</sub> content, respectively. Reddy  
68 et al. (2010) used traditional melt blending technology to prepare low nanoparticle  
69 loaded polymer composites with improved mechanical properties. The results of  
70 tensile tests indicate that the addition of SiO<sub>2</sub>-g-DGEBA particles to the polymer  
71 matrix led to an increase of both elastic modulus and toughness (from 0.36 to 0.54  
72 GPa, and 19.06 to 21.05 MJ/m<sup>3</sup>, respectively). Filling nano-SiO<sub>2</sub> into thermoplastic  
73 matrix can combine the rigidity of nano-SiO<sub>2</sub> with the toughness of thermoplastic  
74 matrix, which makes ultimate composite have comprehensive mechanical  
75 performance (Hashim et al. 2019; Luo et al. 2018; Park et al. 2020).

76 In this study, nano-SiO<sub>2</sub> particles were adopted and dispersed into the PLA  
77 matrix aiming to improve the mechanical property of jute/PLA composite. Moreover,  
78 considering the dispersibility, the nano-SiO<sub>2</sub> was first modified with different silane  
79 coupling agents, then combined into PLA-based composite films and finally  
80 incorporated in final laminated composites. This use of sandwich-like lamination  
81 structure layer can mix PLA and jute as evenly as possible. Therefore, the effect of  
82 silane coupling agents on the mechanical property of as-prepared composites via  
83 hot-pressing technology was comprehensively analyzed.

84

## 85 **Materials and methods**

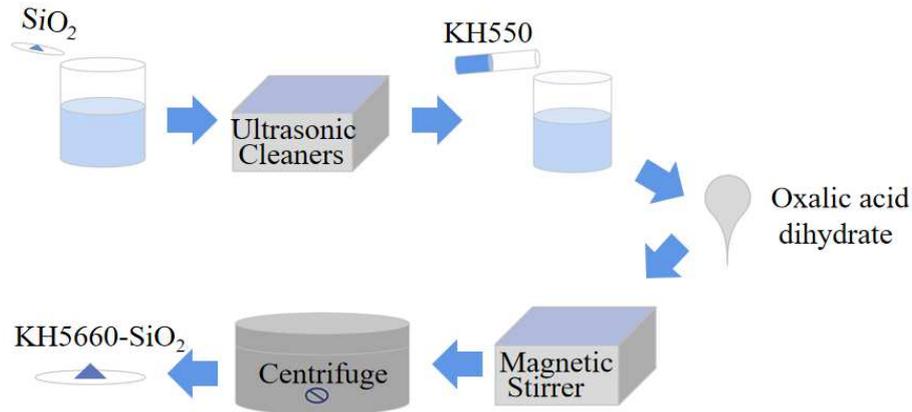
### 86 *Materials*

87 PLA (4032D; 1.24 g/cm<sup>3</sup>) was supplied by Nature Works, USA; Jute fibers (1.45  
88 g/cm<sup>3</sup>) was purchased from Shandong Jiangke Linyi Co., Ltd; Nano silica (SiO<sub>2</sub>,  
89 99.5%, 50±5 nm) was provided by Suzhou Great Medical Technology Co., Ltd;  
90 Silane coupling agent (KH550, KH560, KH570) chemicals were provided by Jiangsu  
91 Argon Krypton Xenon Material Technology Co., Ltd. KH550  
92 (NH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>CH<sub>2</sub>Si(OC<sub>2</sub>H<sub>5</sub>)<sub>3</sub>) belongs to aminosilane, KH560  
93 (CH<sub>2</sub>OCHCH<sub>2</sub>O(CH<sub>2</sub>)<sub>3</sub>Si(OCH<sub>3</sub>)<sub>3</sub>) belongs to epoxy silane, KH570  
94 (CH<sub>3</sub>CCH<sub>2</sub>COO(CH<sub>2</sub>)<sub>3</sub>Si(OCH<sub>3</sub>)<sub>3</sub>) belongs to formaldehyde acryloyloxy functional  
95 group silane.

96

### 97 *Preparation of modified SiO<sub>2</sub>*

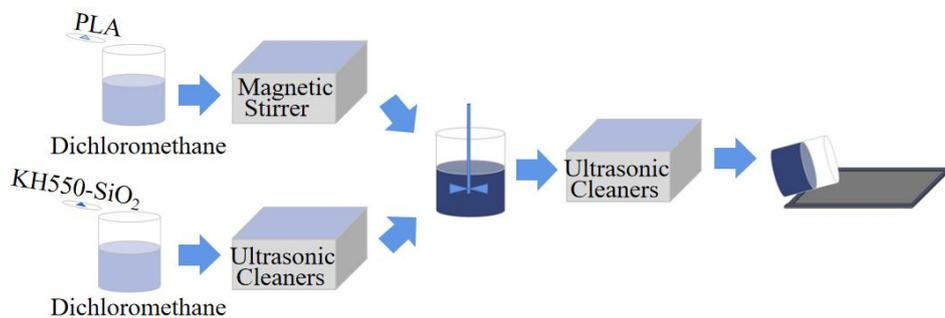
98 The absolute ethyl alcohol and deionized water were first mixed with a mass  
99 ratio of 9:1, and 5 g of SiO<sub>2</sub> was added into 100 mL of mixture. The SiO<sub>2</sub> solution was  
100 then sonicated in an ultrasonic cleaner for 1 h. The coupling agent, e.g. KH550, with a  
101 volume of 5% of the mixture was prepared and adjusted to the pH of 4 with oxalic  
102 acid. The above mixed solutions were magnetically stirred for 3 h at 60 °C for  
103 reaction. Finally, it was centrifuged (model: TG16G) and then washed with absolute  
104 ethanol solution for 3 times. The obtained precipitate was dried in an oven at 60 °C  
105 for 4 h to obtain modified SiO<sub>2</sub> powders, named as KH550-SiO<sub>2</sub>, KH560-SiO<sub>2</sub> and  
106 KH570-SiO<sub>2</sub> respectively. The scheme of preparation process of modified SiO<sub>2</sub> is  
107 illustrated in Figure 1.



**Fig. 1** The scheme of preparation process of modified SiO<sub>2</sub>

*Preparation of composite SiO<sub>2</sub>-PLA films*

3 g of PLA raw material was dissolved in dichloromethane, stirred magnetically (model: DF-101S) for 1.5 h at room temperature; different mass fractions of modified SiO<sub>2</sub> (0.5 wt%, 1 wt%, 2 wt%, 4 wt% and 8 wt%) were mixed into dichloromethane and ultrasonicated (model: VGT-1730QT) for 40 min. These two solutions were mixed and magnetically stirred for 2 h, followed by ultrasonication for 1 h at 35 °C. Then, the solution was poured into a glass plate and placed in a fume hood at room temperature to make the methylene chloride solvent completely volatilize. Finally, the composite SiO<sub>2</sub>-PLA film was peeled from the mold and placed in a vacuum oven at a temperature of 45 °C for 3 h, as shown in Figure 2.

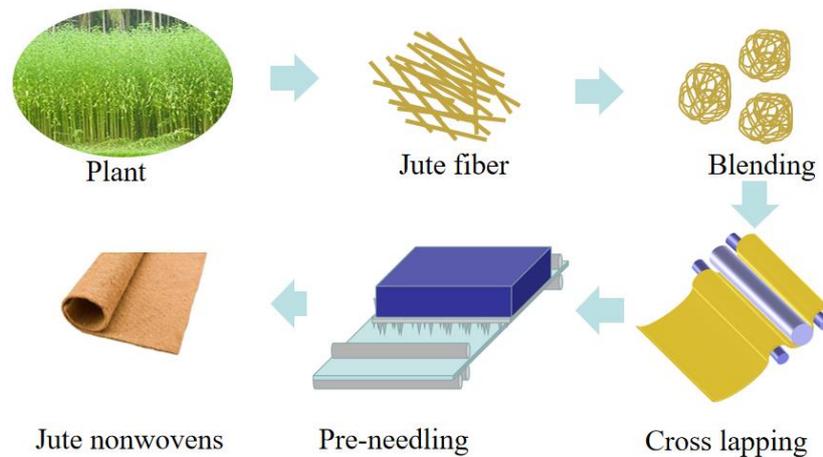


**Fig. 2** The scheme of preparation process of composite SiO<sub>2</sub>-PLA film

*Preparation of jute non-woven*

The preparation process of jute non-woven is shown in Figure 3. The aspect ratio

127 of jute fiber is about 314:1. First, the fluffy jute fibers was combed, opened and mixed  
128 through a coarse-garge carding machine (model: SSL-II) to make the fibers looser and  
129 more uniform. Then the loose jute fibers were sent to the blender for further fine  
130 opening. The fine opening and carding process make the jute fibers finally become a  
131 fluffy and uniform fiber web. The fiber web obtained by cross-netting has higher  
132 quality. In order to obtain a certain strength of non-woven fabrics, the four-layer jute  
133 net were finally reinforced with a needle punch (model: CSZC). The final thickness of  
134 the jute nonwovens is  $0.51\pm 0.04$  mm, and the surface density is  $200\pm 2$  g/m<sup>2</sup>.



135

136

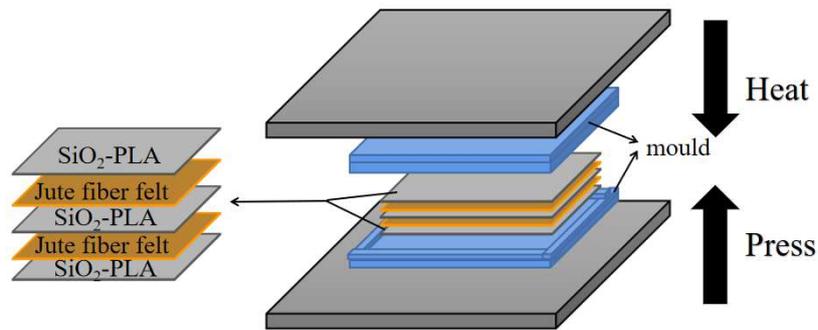
**Fig. 3** The preparation process of jute non-woven fabric

137

### 138 *Preparation of composite material*

139 To remove all moisture, SiO<sub>2</sub>-PLA films and jute nonwoven were first dried in a  
140 vacuum oven at 80 °C for 6 h before hot pressing. The laminated composite material  
141 was processed through a compression mold using a film stacking method, in which  
142 two jute pads were arranged between three PLA films in parallel, as demonstrated in  
143 Figure 4. The entire assembly was then placed in a steel mold controlled under  
144 different time and temperature. The laminated material is compacted for 0.5 h under  
145 the conditions of a temperature of 180 °C and a pressure of 1 MPa, and then is  
146 naturally cooled in the vulcanization machine(model: HG-3621) for 8 h. After 8 h, the  
147 sample was taken out of the laminating mold, and the width and length were 150 mm  
148 × 150 mm and the thickness was 2 mm, and the mass ratio of PLA to jute was 10:3.

149 The samples are numbered according to the type of coupling agent and concentration.  
150 For example, 550-0.5, in which 550 represents the silane coupling agent of KH550,  
151 the sample untreated with the coupling agent is directly represented by SiO<sub>2</sub>, and 0.5  
152 represents 0.5 wt% of the modified SiO<sub>2</sub> mass fraction in SiO<sub>2</sub>-PLA film.



153

154

**Fig. 1** The preparation process of composite material

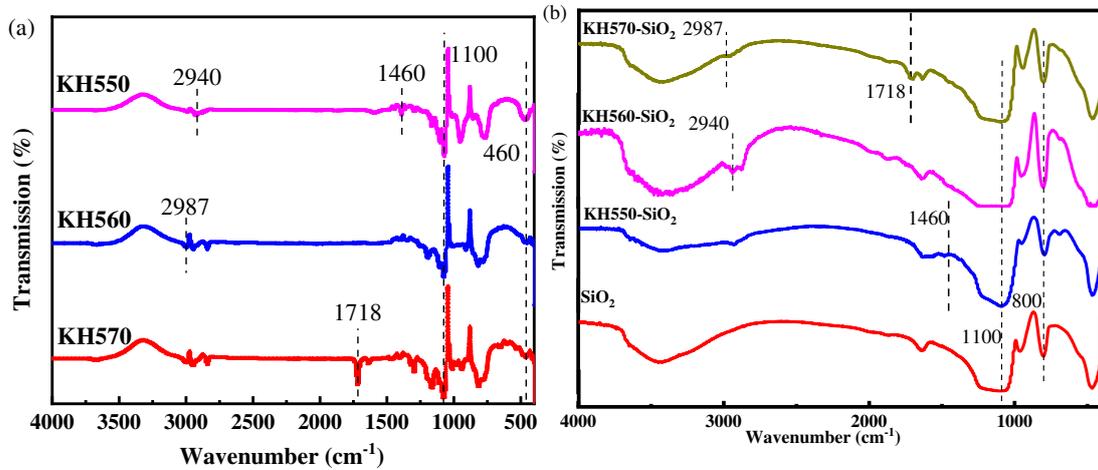
155

#### 156 *Characterization of jute / PLA composites*

157 In order to study that effect of different silicon dioxide on the mechanical  
158 properties of jute/PLA composite, the SiO<sub>2</sub> modified by silane coupling agent was  
159 characterized by infrared transmission spectroscopy (5700, Nicli Instruments, USA)  
160 and particle size analyzer (LS-909). At the same time, the mechanics of composite  
161 materials were characterized by scanning electron microscope (TM3030, Hitachi,  
162 Japan), dynamic mechanical thermal analysis (SDTQ800, V21.3, Build 96, USA) and  
163 related mechanical testing methods (Instron 336, USA).

164

#### 165 **Results and discussions**



**Fig. 5** (a) FTIR spectra of KH550, KH560 and KH570; (b) FTIR spectra of SiO<sub>2</sub>, KH550-SiO<sub>2</sub>, KH560-SiO<sub>2</sub> and KH570-SiO<sub>2</sub> nanoparticle

Fourier-transform infrared spectroscopy (FTIR) can be used to verify the graft polymerization of silane coupling agents on SBR surface. Figure 5 shows the infrared spectra of KH550, KH560 and KH570 as well as corresponding silane coupling agent grafted SiO<sub>2</sub>. It can be seen from Figure 5 (b) that the infrared spectrum of the SiO<sub>2</sub> before and after modification has a large absorption peak near 1100 cm<sup>-1</sup>, which is the antisymmetric absorption peak of Si-O-Si bond, and near 800 cm<sup>-1</sup> is the symmetrical contraction vibration of the Si-O-Si bond. The infrared spectrum of KH550-SiO<sub>2</sub> is relatively flat compared to others in the range of 3500-3100 cm<sup>-1</sup>, indicating that -NH<sub>2</sub> in KH550 has been successfully grafted and modified on the surface of nano-SiO<sub>2</sub>. In Figure 5 (a), KH550 shows characteristic peaks of -CH<sub>2</sub>- at 2940 cm<sup>-1</sup> and 1640 cm<sup>-1</sup>, while KH550-SiO<sub>2</sub> also has a shoulder peak at 2940 cm<sup>-1</sup> and 1460 cm<sup>-1</sup>, which further indicates that KH550 has successfully modified nano-SiO<sub>2</sub>. Compared with FTIR spectrum of SiO<sub>2</sub>, in the spectrum modified by KH560-SiO<sub>2</sub>, the stretching vibration absorption peak of methyl appears near 2987 cm<sup>-1</sup> which is related to characteristic methyl peak of KH560, indicating that KH560 silane coupling agent has been successfully grafted to the second Si-O surface (He et al. 2019). In the infrared spectrum of KH570 and KH570-SiO<sub>2</sub>, there is a C=O stretching vibration absorption

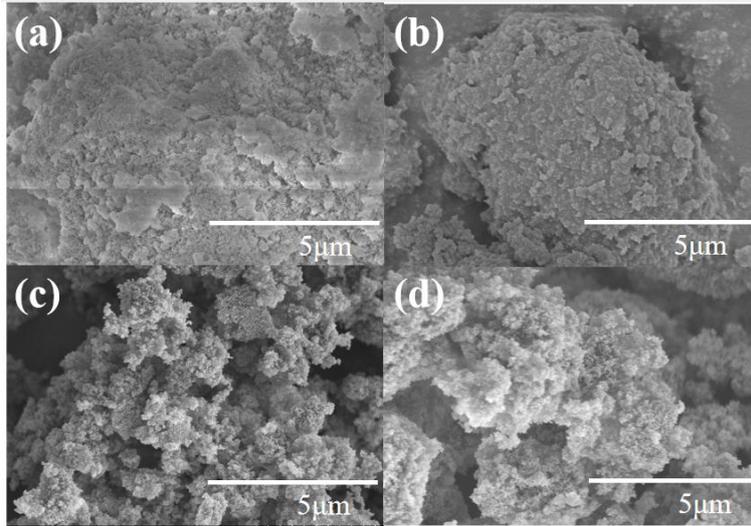
188 peak on the carbonyl group at  $1718\text{ cm}^{-1}$ , which indicates that KH570 has been  
189 successfully grafted onto nano-SiO<sub>2</sub>.

190 The particle size of original and modified SiO<sub>2</sub> is shown in Table 1. The surface  
191 morphology of original and modified SiO<sub>2</sub> is shown in Figure 6. Nano-SiO<sub>2</sub> has a  
192 small particle size, a large specific surface area. Due to the large specific surface area,  
193 the powder is easy to agglomerate together, so that the total surface area is reduced,  
194 and the total energy is reduced. It can be clearly seen that the unmodified particles are  
195 larger than modified particles, and KH560 has the best modification effect on SiO<sub>2</sub>  
196 particles. Unmodified nano-SiO<sub>2</sub> has a relatively high polydispersity index (PDI) and  
197 the particle size, particle size, and unevenness appeared and severe aggregation. The  
198 particle size and PDI of the modified nano-SiO<sub>2</sub> powder are reduced, indicating that  
199 the nano-SiO<sub>2</sub> modified by the silane coupling agent can effectively prevent its  
200 agglomeration and achieve the modification purpose. On the one hand, the hydroxyl  
201 groups on the particle surfaces are replaced by organic functional groups, reducing the  
202 number of active silanol groups, thereby reducing the tendency of nanoparticles to  
203 agglomerate. On the other hand, the grafted long carbon chain increases the distance  
204 between particles and the steric hindrance of the hydroxysilyl polycondensation  
205 reaction on the particle surface, which increases the difficulty of effective collisions  
206 and more conducive to the dispersion of nanoparticles.

207

208 **Table 1** The specification of prepared nano SiO<sub>2</sub>

Sample	Z-Average (nm)	Peak Size (nm)	Peak Intensity (%)	PDI
SiO <sub>2</sub>	300	220.8	65.2	0.343
KH550-SiO <sub>2</sub>	107.7	143.3	69.3	0.287
KH560-SiO <sub>2</sub>	67.6	92.7	100	0.210
KH570-SiO <sub>2</sub>	91	136.6	100	0.229

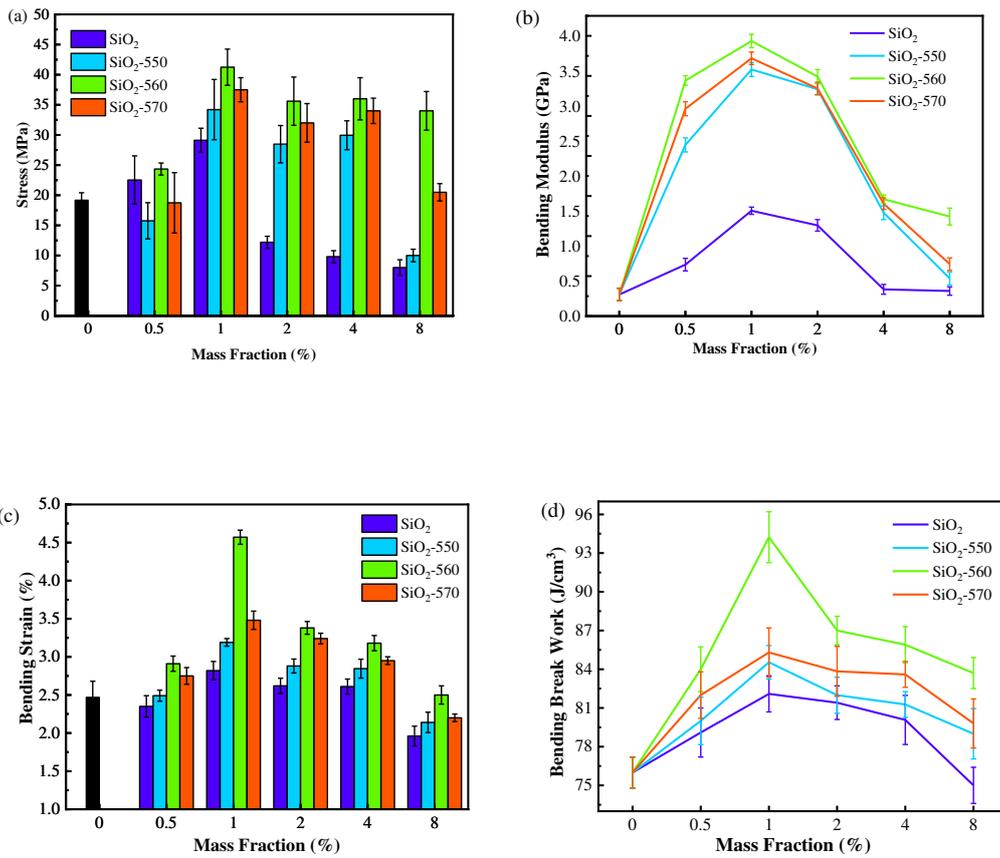


209

210 **Fig. 6** The surface morphology of SiO<sub>2</sub>. (a) SiO<sub>2</sub>; (b) SiO<sub>2</sub>-550; (c) SiO<sub>2</sub>-560; (d) SiO<sub>2</sub>-570

211

212 *Bending property*



213

214

215 **Fig. 7** The effect of nano-SiO<sub>2</sub> on tensile properties of jute/PLA composite bending

216 performance. (a) Bending strength; (b) Modulus; (c) Bending strain; (d) Bending break work

217

218 The experiment was repeated at least 5 times for each sample. Figure 7 (a) and (b)  
219 shows the bending strength and modulus of pure jute/PLA fibers composites involving  
220 SiO<sub>2</sub> and modified SiO<sub>2</sub> nanoparticles with different mass fractions. It can be seen that  
221 the bending modulus of jute/PLA fibers composites is relatively low. With  
222 incorporation of SiO<sub>2</sub>, the bending strength and modulus of the composites is more or  
223 less improved. Compared with pure jute/PLA composite material, there is no  
224 significant improvement in the bending performance of the sample SiO<sub>2</sub>-4. However,  
225 after adding the nano-SiO<sub>2</sub> particles modified by the coupling agents, the bending  
226 strength and modulus of 550-4 increased to 29.95 MPa and 1.25 GPa respectively,  
227 which were 149.5% and 76.8% higher than those of the jute/PLA composite, and  
228 increased by 137.4% and 72.0% compared with the unmodified SiO<sub>2</sub> jute/PLA  
229 composite. The bending strength and modulus of 560-4 were increased to 36 MPa and  
230 2.17 GPa respectively, and the bending strength and modulus of 570-4 were 34 MPa  
231 and 2.01 GPa. Compared with the jute/PLA fibers composites, the bending strength of  
232 560-0.5, 560-1, 560-2, 560-4 and 560-8 composites increased by 2.8%, 96.6%, 74.7%,  
233 74.2% and 68.3% respectively. It is also found from Figure 7 (a) and (b) that as the  
234 mass fraction of different types of added SiO<sub>2</sub> increases, the change trend of material  
235 bending strength and modulus is basically same.

236 The similar phenomena can be also observed in the bending strain and bending  
237 break work of jute/PLA composites. The bending break work was calculated by  
238 integrating the tensile stress–strain curves. As shown in Figure 7 (b), the increase of  
239 SiO<sub>2</sub> improves the bending strain and bending break work of jute/PLA composites.

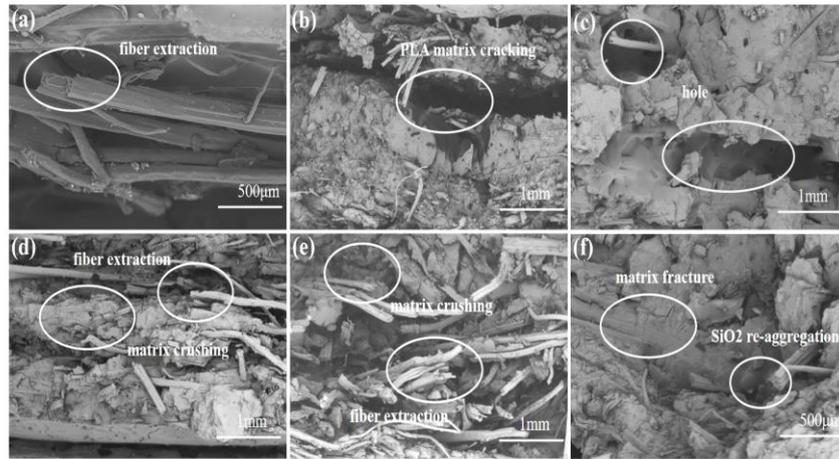
240 In short, the bending performance of the sample increases with the increase of  
241 the SiO<sub>2</sub> mass fraction, peaks at about 1wt%, and then decreases. That is, under the  
242 same mass fraction, SiO<sub>2</sub> treated with different silane coupling agents has the same  
243 effect on the jute/PLA composite. The SiO<sub>2</sub> sample modified with KH560 has the best  
244 bending strength and modulus. It is mainly due to the fact that realize the interface  
245 combination through the coupling agent, forming a better entirety, and then  
246 strengthening the effect of stress transmission(Yang et al. 2009; Sanivada et al. 2020;  
247 Fang et al. 2020; Li et al. 2013). Sample 560-1 has best bending performance

248 resulting from the excellent dispersibility and uniform particle size as illustrated in  
249 Table 1.

250

### 251 *The fracture morphology*

252 In order to illustrate the toughening mechanism, the fracture morphology of the  
253 reinforced PLA-based composite after the tensile test is shown in Figure 8. The tensile  
254 fracture surface of the jute/PLA composite, illustrated in Figure 8 (a), shows large  
255 amounts of fiber extraction leading to a lower mechanical resistance to some extent.  
256 This is consistent with comparatively large elongation ratio of the composite material  
257 which also indicates a poor interfacial cohesiveness. Figure 8 (b) shows the SiO<sub>2</sub>  
258 particles modified by KH560 are evenly distributed in PLA matrix. At this time,  
259 delamination occurs in the tensile section, and the bending strength is reduced. With  
260 the increase of SiO<sub>2</sub>, there are more holes appeared which are prone to cause fracture.  
261 Nano-SiO<sub>2</sub> particles induce local plastic deformation of the PLA matrix, showing a  
262 rougher fracture surface. And as the stress concentration point, SiO<sub>2</sub> particles produce  
263 a large number of small cracks, and has a small amount of fibers extraction, as shown  
264 in the Figure 8 (d). There are a large number of cracks and fiber extraction at the  
265 break of sample 560-4. Therefore, sample 560-4 consumes a lot of energy when it  
266 breaks. Due to the poor dispersibility of SiO<sub>2</sub>, the modified SiO<sub>2</sub> re-aggregated at 8%  
267 by mass fraction. As the mass fraction of SiO<sub>2</sub> increases, there are more contact points  
268 between SiO<sub>2</sub> and jute, which reduces the adhesion between jute and PLA, and the  
269 mechanical properties dropped rapidly, as shown in Figure 8 (f).



270

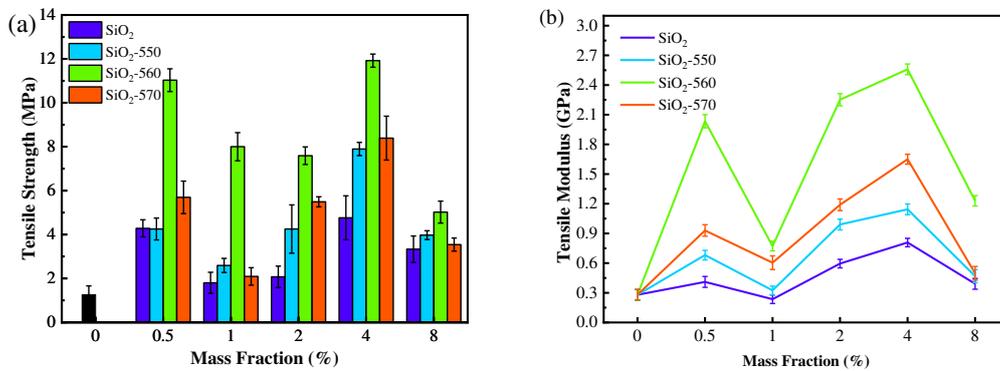
271 **Fig. 8** The fracture surface morphologies of jute/PLA composite ( $\times 200$ ). (a) jute/PLA

272 composite. (b) Sample 560-0.5. (c) Sample 560-1. (d) Sample 560-2. (e) Sample 560-4. (f)

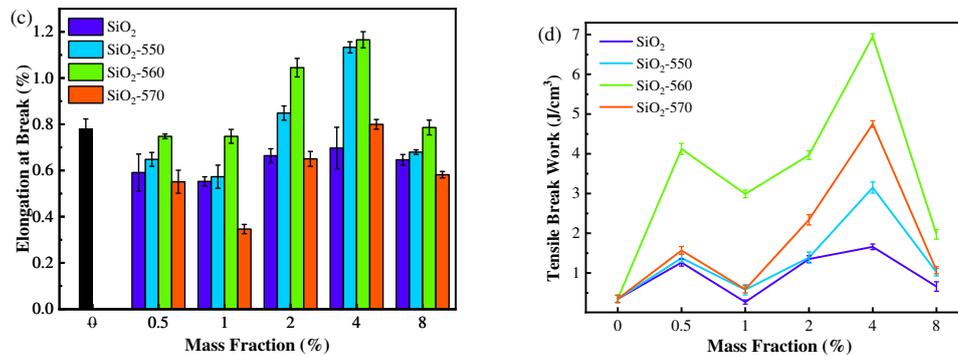
273 Sample 560-8

274

275 *Stretching property*



276



277

278 **Fig. 9** The effect of nano-SiO<sub>2</sub> on tensile properties of jute/PLA fibers composites. (a) Tensile  
279 strength; (b) modulus; (c) Elongation at break; (d) Tensile break work

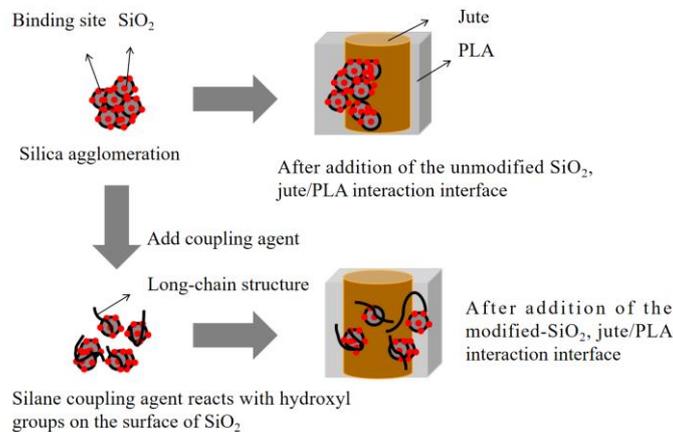
280

281 The tensile property of the jute/PLA composite and the nano-SiO<sub>2</sub> jute/PLA  
282 composites is shown in Figure 9. The tensile strength of 560-0.5 increased from 1.47  
283 MPa in the jute/PLA composites to 11.03 MPa. The elastic modulus of 560-0.5 is as  
284 high as 2.01 GPa, which is 614.2% higher than the 0.28 GPa of the jute/PLA  
285 composites. The tensile strength of 560-1 is 8 MPa and the elastic modulus of 560-1 is  
286 0.81 GPa. The tensile strength of 560-2 is 7.59 MPa and the elastic modulus is 2.32  
287 GPa. The tensile strength of 560-4 is 11.92 MPa and the elastic modulus is 2.57 GPa.  
288 The tensile strength of 560-8 is 5.02 MPa and the elastic modulus is 1.20 GPa. The  
289 tensile break work of SiO<sub>2</sub>-4, 550-4, 560-4, and 570-4 is increased by 138.0%,  
290 389.7%, 609.5%, 417.0% compared with the jute/PLA fibers composites.

291 It is concluded from Figure 9 that the cooperation of nano-SiO<sub>2</sub> can effectively  
292 improve the tensile tolerance of composite materials. With the increase of nano-SiO<sub>2</sub>  
293 content, the change trend of the tensile properties of the composite material before  
294 and after modification is basically the same. And when the SiO<sub>2</sub> mass fraction is same,  
295 the sample modified by KH560 has the best tensile strength. This can be attributed to  
296 the small particle size and uniform shape of SiO<sub>2</sub> modified by KH560, which is  
297 consistent with the DLS data.

298 When preparing jute/PLA laminates, SiO<sub>2</sub> can be more evenly dispersed in the  
299 matrix, thereby enhancing the effective load transfer between jute and PLA,  
300 improving the tensile properties of the composite material. The silane coupling agent  
301 can serve as a bridge to improve the compatibility of the interface between SiO<sub>2</sub> and  
302 jute/PLA, forming a better whole, and then strengthening the stress transmission.  
303 Figure 10 shows the interaction between the silica particles and the interface of the  
304 jute/PLA composite before and after the modification. The silane coupling agent can  
305 react with the silicon hydroxyl group on the surface of SiO<sub>2</sub> after being hydrolyzed.  
306 The degree of aggregation of modified-SiO<sub>2</sub> is reduced, exposing more binding sites  
307 that can interact with PLA. At the same time, the long-chain structure of the coupling

308 agent will generate entanglement on the interface between jute fiber and PLA, which  
 309 efficiently enhances the interface bonding performance, expedites interfacial stress  
 310 transfer, and ultimately improves the mechanical property of the composite material.



311

312 **Fig. 10** The interaction between the silica particles and the interface of the jute/PLA

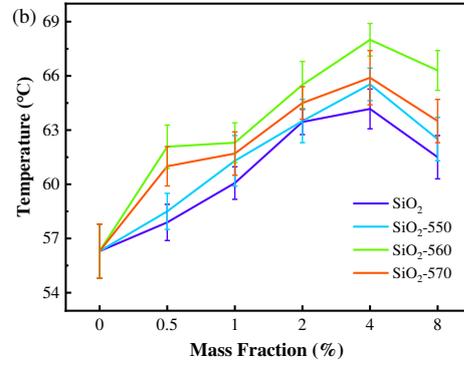
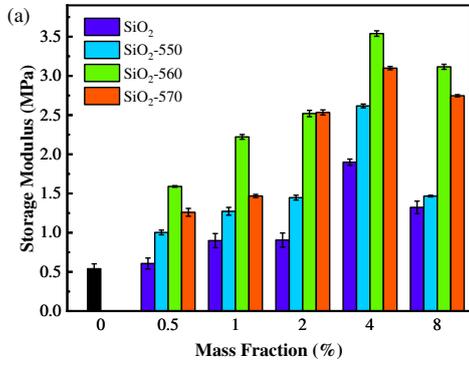
313 composite before and after the modification of the silane coupling agent.

314

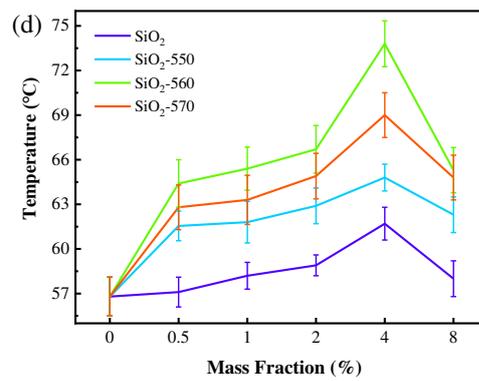
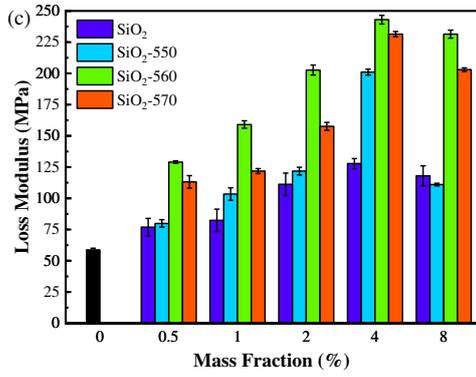
315 *Dynamic mechanical thermal analysis (DMTA)*

316 Figure 11 respectively show the dynamic storage modulus ( $E'$ ), loss modulus ( $E''$ )  
 317 and loss coefficient  $\tan\delta$  ( $E''/E'$ ) of jute/PLA composites with different contents of  
 318 SiO<sub>2</sub> modified from room temperature to 180 °C. As shown in Figure 11 (a) and (b),  
 319 the stored modulus values and corresponding temperatures of different samples at the  
 320 beginning of chain segment movement. In the temperature range studied, the storage  
 321 modulus of the jute/PLA composite modified by SiO<sub>2</sub> is higher than that of the  
 322 untreated jute/PLA composite. In addition, different variables showed the same  
 323 change pattern. With the increase of SiO<sub>2</sub> content, the storage modulus of the  
 324 modified jute/PLA composite material increases first and then decreases. The increase  
 325 in storage modulus and corresponding temperature of chain segment starting to move  
 326 indicates that the interface adhesion between jute and PLA has been improved,  
 327 resulting in greater stress transfer between them (Porrás and Marañón, 2012). That is,  
 328 the increase of the SiO<sub>2</sub> content can improve the interfacial adhesion, but because of  
 329 the poor dispersibility of SiO<sub>2</sub>, when the content is greater than 4%, the effect of the

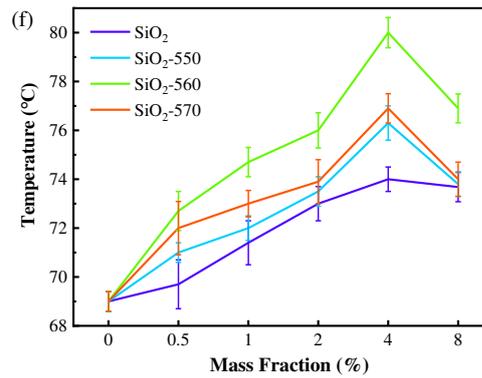
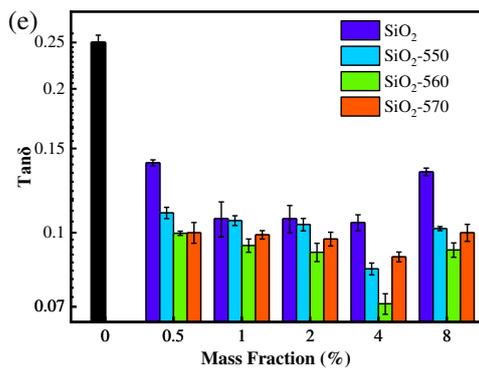
330 coupling agent on SiO<sub>2</sub> modification is not obvious, and the jute/PLA interface  
 331 adhesion begins to decrease.



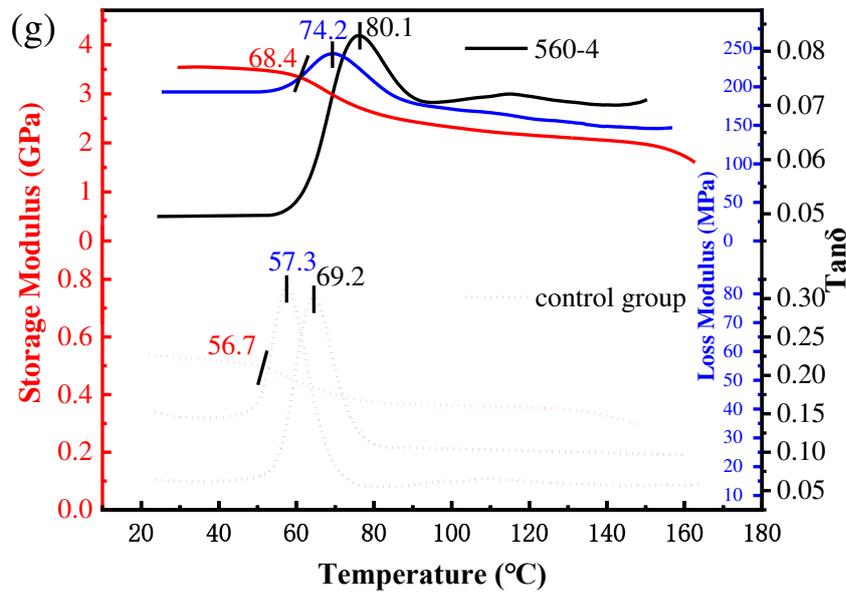
332



333



334



335

336 **Fig. 11** The DMTA of jute/PLA composite. (a) Storage modulus; (b) The temperature  
 337 corresponding to storage modulus; (c) The maximum loss modulus; (d) The  
 338 temperature corresponding to the maximum loss modulus; (e) The maximum  $\tan\delta$ ; (f)  
 339 The temperature corresponding to the maximum  $\tan\delta$ ; (g) The comparison of DMTA  
 340 between jute/PLA composite and sample 560-4

341

342 The next study parameter is the loss modulus, which represents the energy  
 343 dissipated by the jute/PLA composite under stress (Laly et al. 2003; Doan et al. 2007).  
 344 It is observed in Figure 11 (c) and (d) that the loss modulus value of the  
 345  $\text{SiO}_2$ -modified jute/PLA composite is much higher than that of the untreated jute/PLA  
 346 composite. It is known that the maximum value of loss modulus corresponds to the  
 347 glass transition temperature ( $T_g$ ) of the composite material. The  $T_g$  of the untreated  
 348 jute/PLA composite is around 57.3 °C. In contrast, the  $T_g$  of the  $\text{SiO}_2$ -modified  
 349 jute/PLA composite material moved to the high temperature zone, reaching a  
 350 maximum of about 74.2 °C.

351

352 Finally, loss factor,  $\tan\delta$ , which refers to the ratio of the loss modulus of the  
 composite material to the storage modulus of the composite material, and represents

353 the damping energy of the material. The decrease of the  $\tan\delta$  value indicates  $\text{SiO}_2$   
354 improves the bonding strength between PLA and jute, and reduces the fluidity of PLA  
355 macromolecules in the composite material. Due to the improvement of the  
356 hydrophobicity of the  $\text{SiO}_2$ -modified Jute/PLA, the interfacial adhesion is enhanced,  
357 and the fluidity of the polymer chains at the jute/PLA composite interface is reduced.  
358 Among the unmodified  $\text{SiO}_2$  and  $\text{SiO}_2$  jute/PLA composites modified by KH550,  
359 KH560 and KH570, the unmodified jute/PLA composites have the highest  $\tan\delta$  value,  
360 and the KH560 modified  $\text{SiO}_2$  jute/PLA composites has the highest  $\tan\delta$  value,  
361 indicating the dispersibility of  $\text{SiO}_2$  modified by KH560 is best. Among all  $\text{SiO}_2$   
362 composite material modified by KH560, jute/PLA with a  $\text{SiO}_2$  content of 4% has the  
363 best interface bonding. As shown in Figure 11 (e) and (f).

364 Comprehensive analysis of sample 560-4 has the best thermomechanical  
365 properties. Compared with the jute/PLA composite, the storage modulus, glass  
366 transition temperature and loss modulus have increased;  $\tan\delta$  has decreased. As shown  
367 in Figure 11 (g), the comparison of DMTA between jute/PLA composite and sample  
368 560-4. The glass transition temperature of sample 560-4 is 29.5% higher than that of  
369 the jute/PLA composite, indicating that the experimental optimization has improved  
370 the bonding strength of the interface between PLA and jute. The storage modulus  
371 values of different samples decrease with increasing temperature. The  $\tan\delta$  value of  
372 the jute/PLA composite material increased with the increase of temperature when the  
373 mass fraction of  $\text{SiO}_2$  treated by KH560 was 4%, until it reached the maximum at  
374  $69.2^\circ\text{C}$ , and then the opposite trend was observed in the rubber area.

375

## 376 **Conclusions**

377 In order to enhance the mechanical properties of the jute/PLA composite,  
378 nano- $\text{SiO}_2$  particles modified by different coupling agents were introduced into the  
379 PLA matrix. The effect of the type of coupling agent and the mass fraction of  $\text{SiO}_2$  on  
380 the bending, stretching and thermomechanical properties of the composite was studied.  
381 The study shows that the addition of  $\text{SiO}_2$  improves the interface performance  
382 between PLA and jute and improves the mechanical properties of the composite

383 material. Meanwhile, the smaller the particle size of SiO<sub>2</sub> and the more uniform the  
384 particles, the better the mechanical properties of the composite material. The analysis  
385 of thermomechanical properties shows that the addition of SiO<sub>2</sub> can improve the  
386 thermomechanical properties and glass transition temperature of composite materials.  
387 In addition, as shown in the tensile and bending tests, the strength, stiffness and  
388 toughness of SiO<sub>2</sub> composite material with mass fraction of 4% modified by the  
389 KH560 coupling agent are significantly improved, and its elongation at break is  
390 slightly affected. Compared with intact jute/PLA composite, the flexural strength and  
391 flexural modulus of sample 560-4 increase by 150.3% and 77.4%, respectively, while  
392 the tensile strength and tensile modulus increase by 816.9% and 700.1%, respectively.  
393 Its excellent mechanical properties are the result of the better interface compatibility  
394 of jute/PLA caused by the SiO<sub>2</sub>-CN bond. In addition, the fracture morphology of the  
395 composite material shows that the plastic deformation of the matrix caused by SiO<sub>2</sub>  
396 are the main toughening mechanisms.

397 In summary, adding SiO<sub>2</sub> modified by coupling agent to jute/PLA composites  
398 can improve the mechanical properties of composites, which can be used in packaging,  
399 automotive and other industries. Make products more green and environmentally  
400 friendly.

401

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406

#### 407 **Declaration of competing interest**

408 The authors declare that they have no known competing financial interests or personal  
409 relationships that could have appeared to influence the work reported in this paper.

410

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## Figures

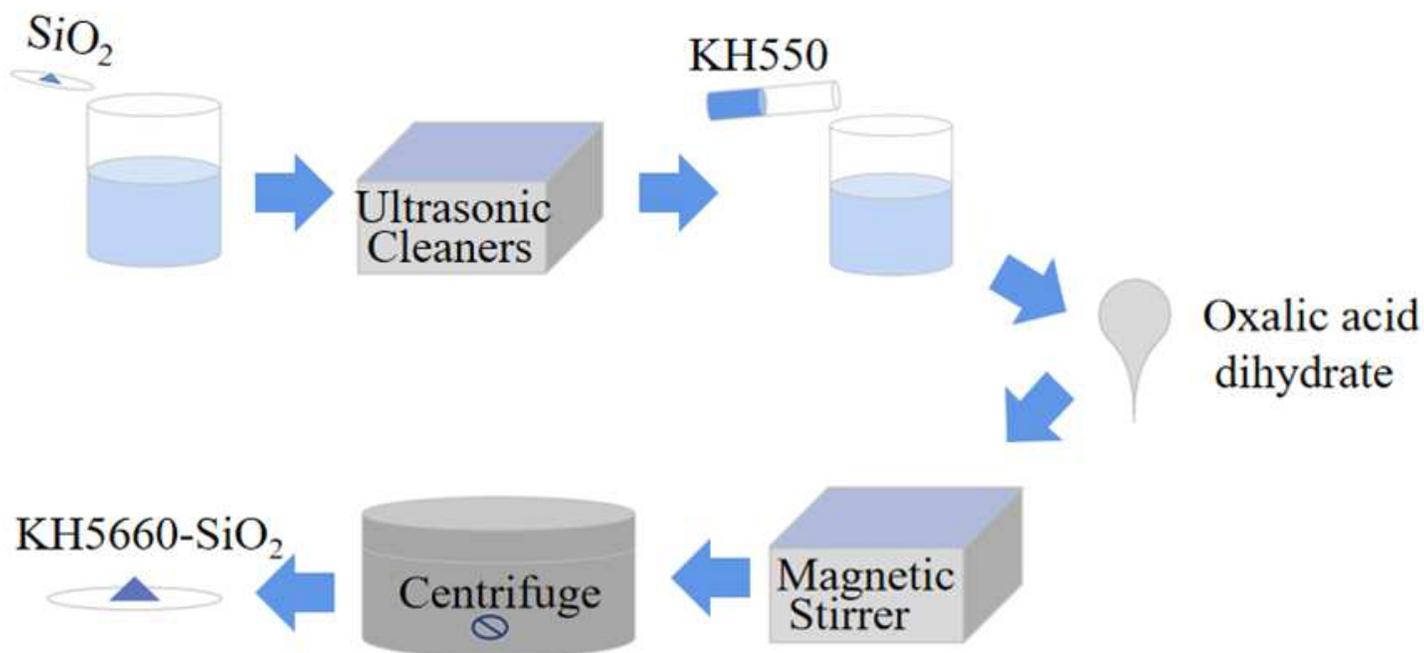


Figure 1

The scheme of preparation process of modified SiO<sub>2</sub>

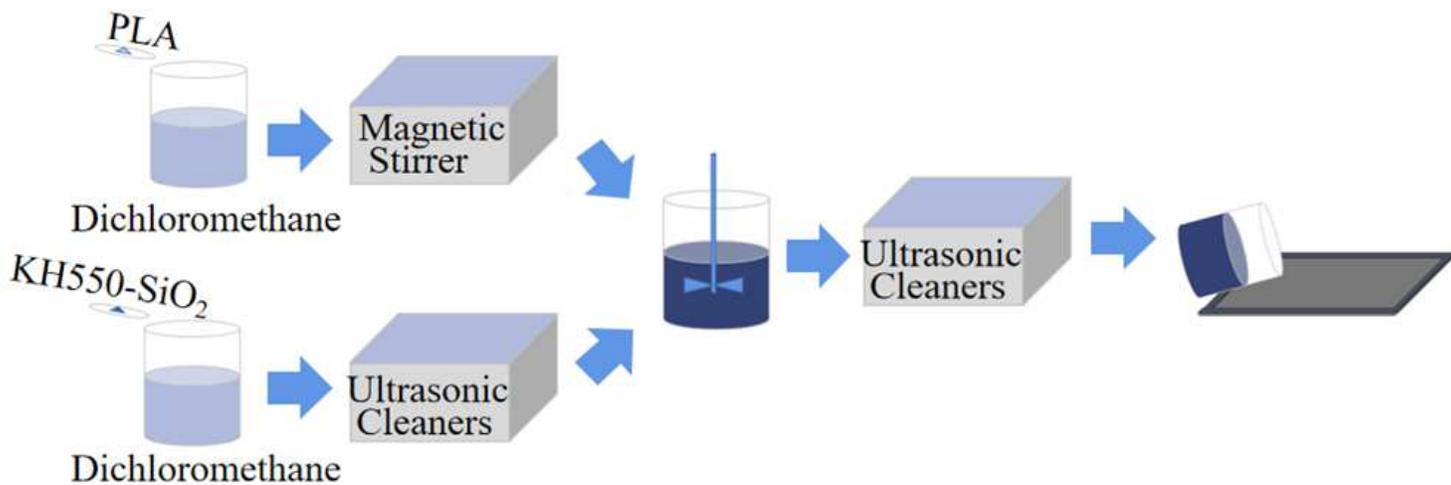


Figure 2

The scheme of preparation process of composite SiO<sub>2</sub>-PLA film

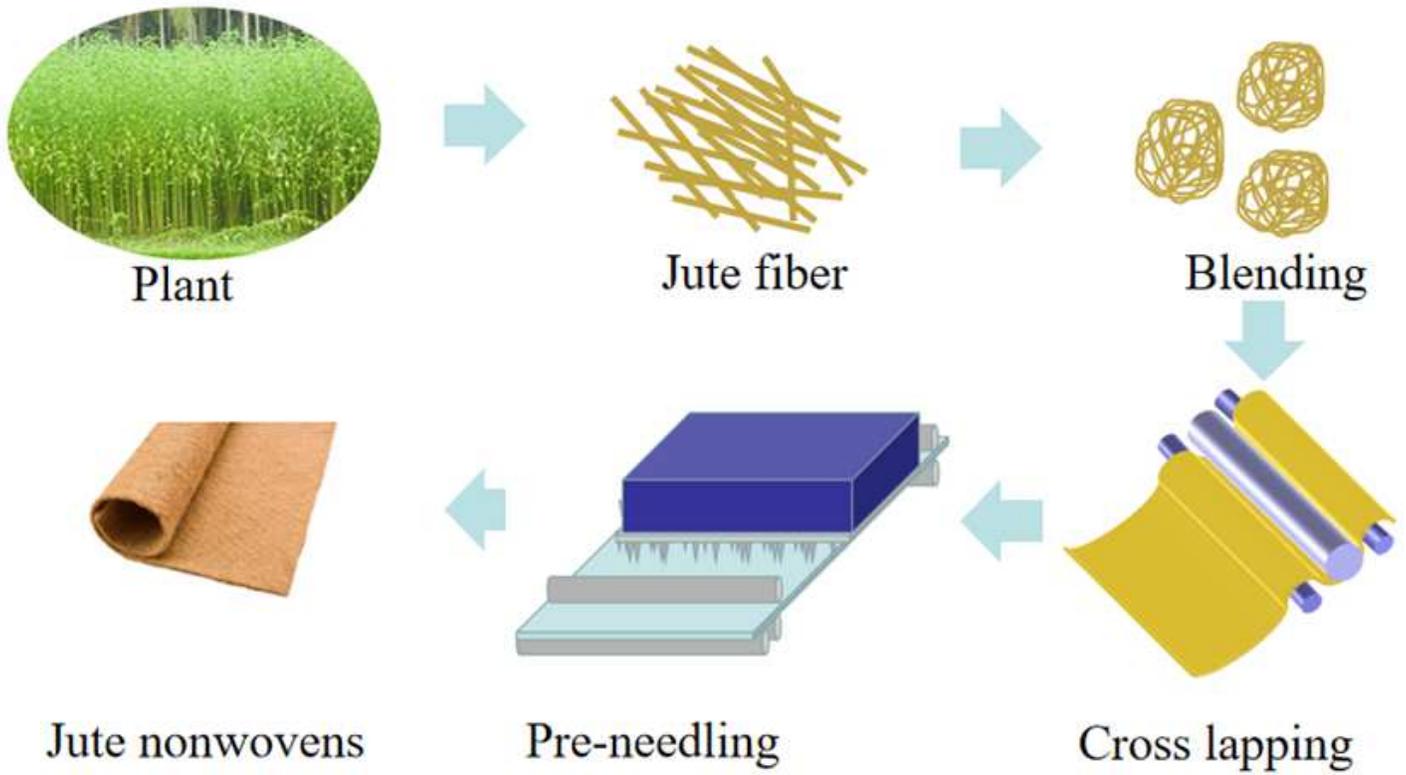


Figure 3

The preparation process of jute non-woven fabric

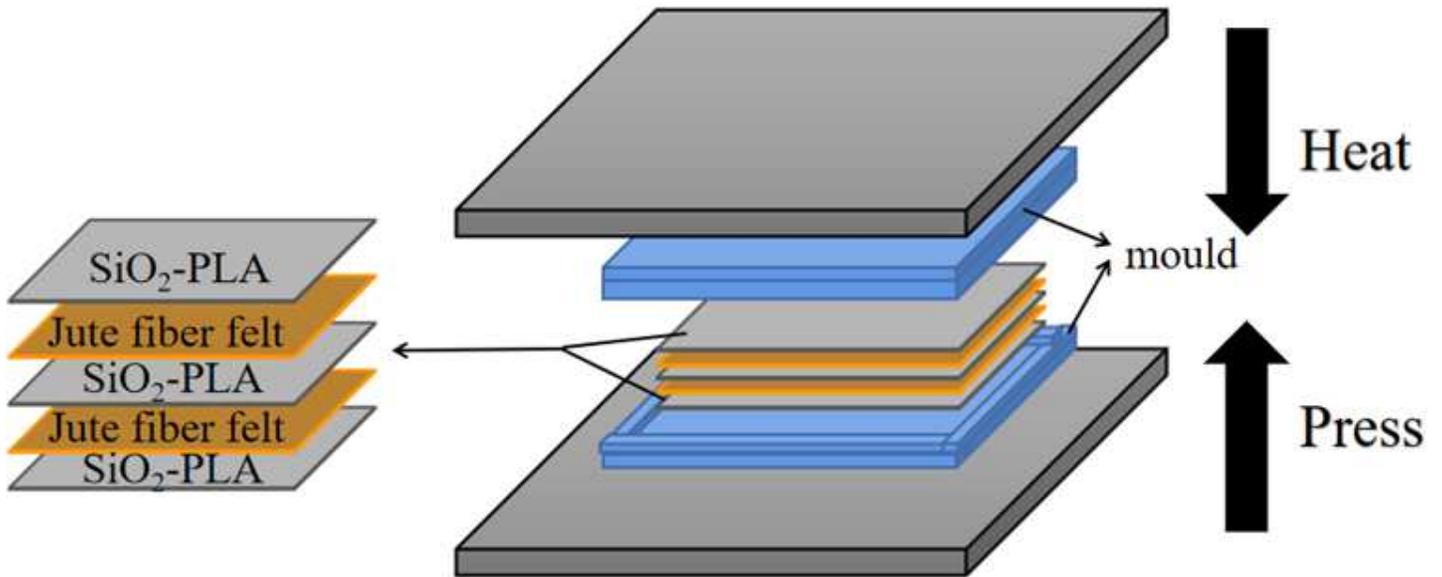


Figure 4

The preparation process of composite material

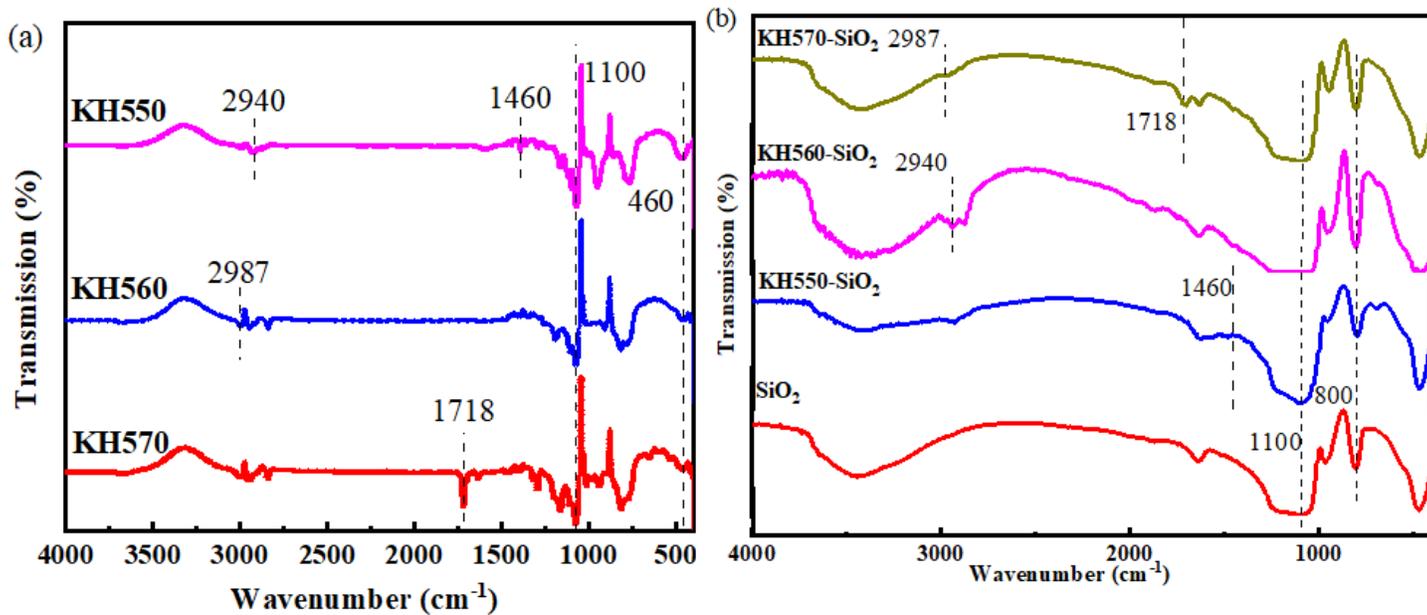
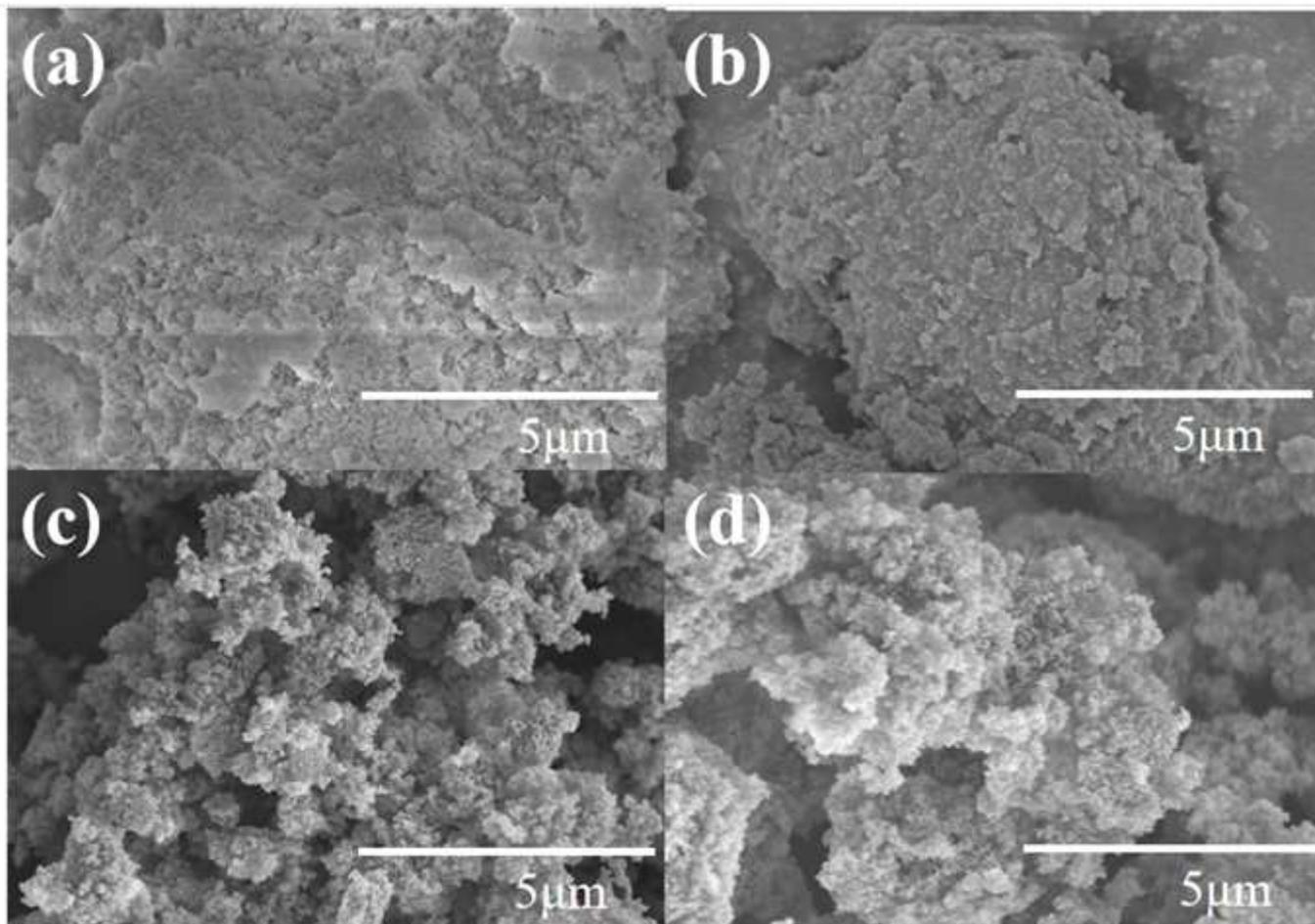


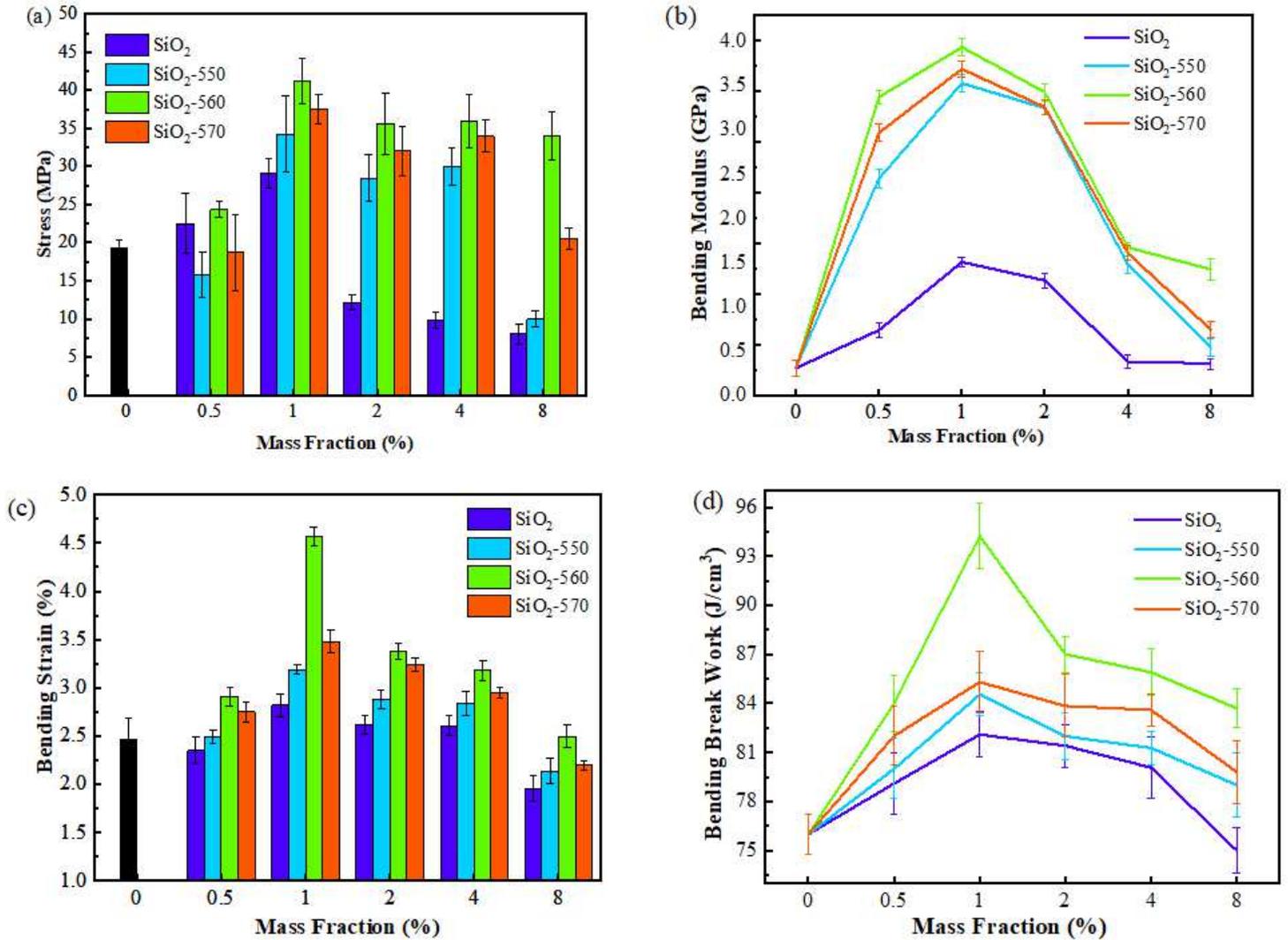
Figure 5

(a) FTIR spectra of KH550, KH560 and KH570; (b) FTIR spectra of SiO<sub>2</sub>, KH550-SiO<sub>2</sub>, KH560-SiO<sub>2</sub> and KH570-SiO<sub>2</sub> nanoparticle



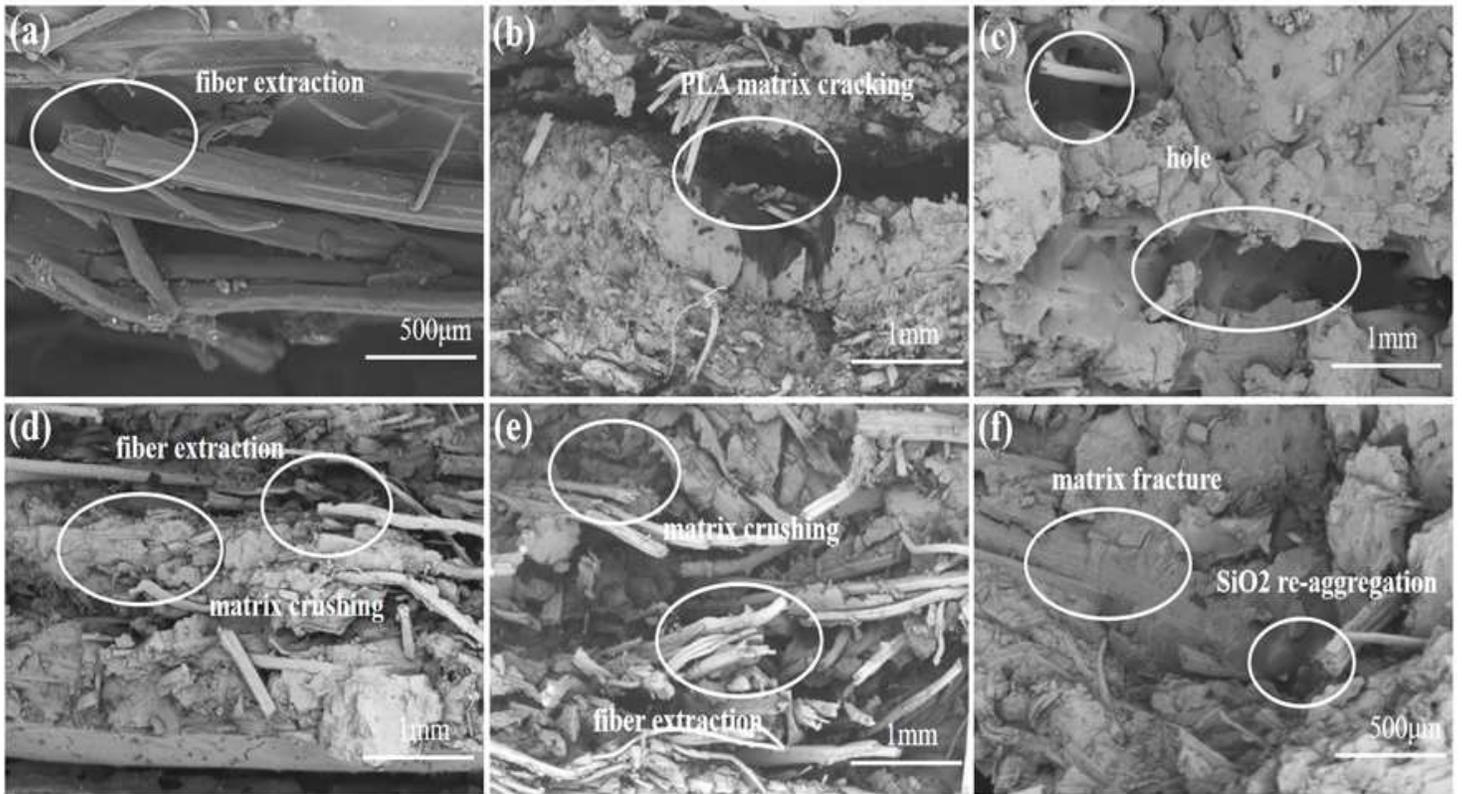
**Figure 6**

The surface morphology of SiO<sub>2</sub>. (a) SiO<sub>2</sub>; (b) SiO<sub>2</sub>-550; (c) SiO<sub>2</sub>-560; (d) SiO<sub>2</sub>-570



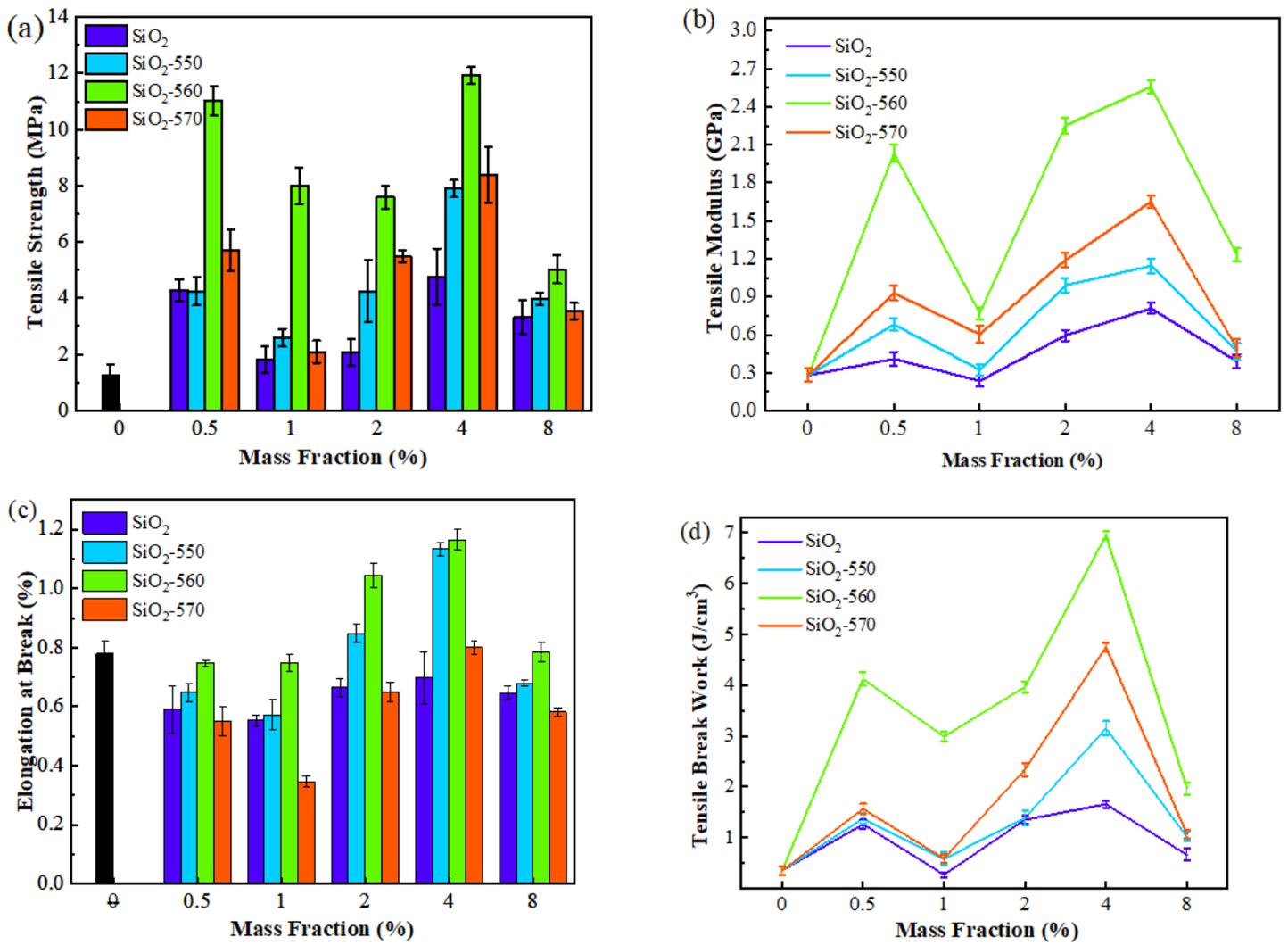
**Figure 7**

The effect of nano-SiO<sub>2</sub> on tensile properties of jute/PLA composite bending performance. (a) Bending strength; (b) Modulus; (c) Bending strain; (d) Bending break work



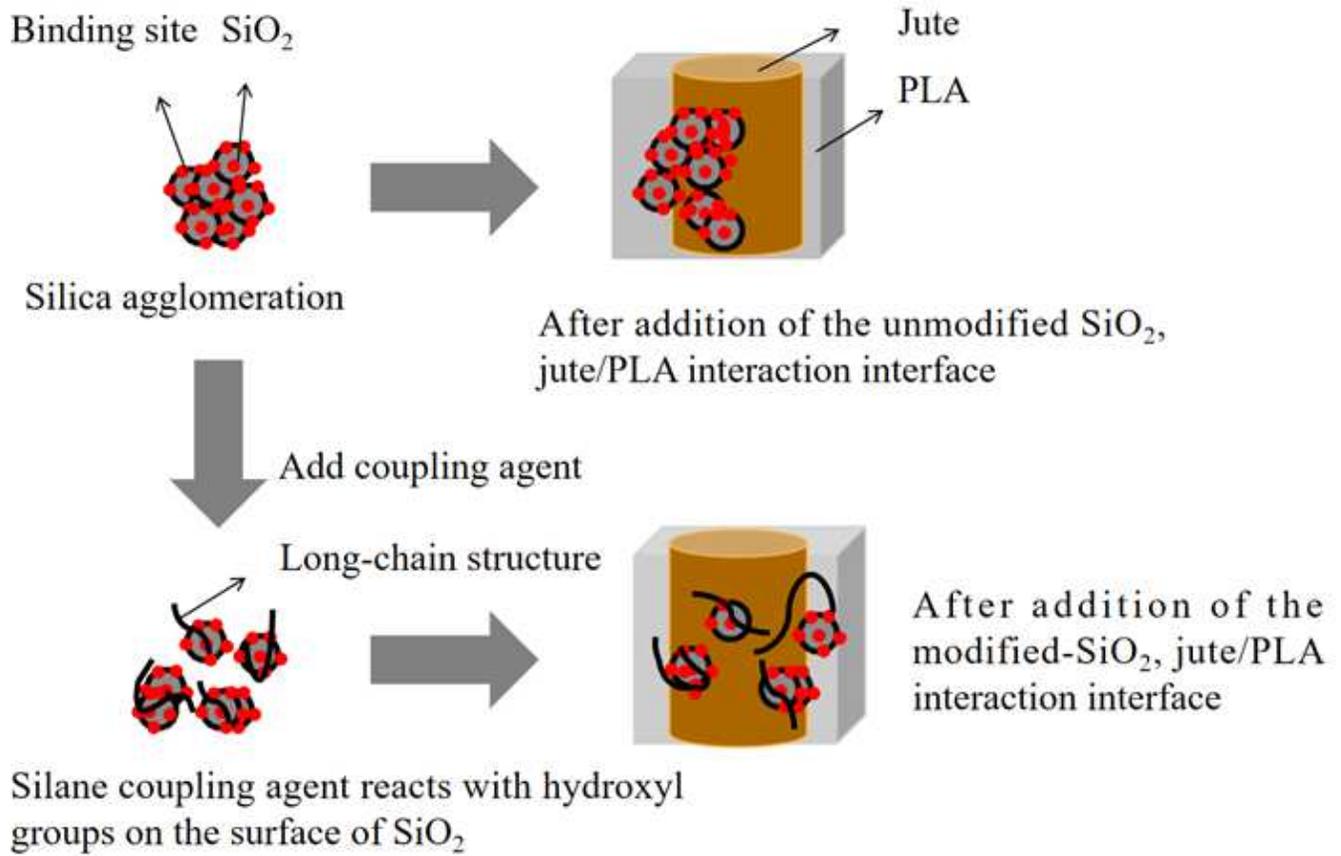
**Figure 8**

The fracture surface morphologies of jute/PLA composite (×200). (a) jute/PLA composite. (b) Sample 560-0.5. (c) Sample 560-1. (d) Sample 560-2. (e) Sample 560-4. (f) Sample 560-8



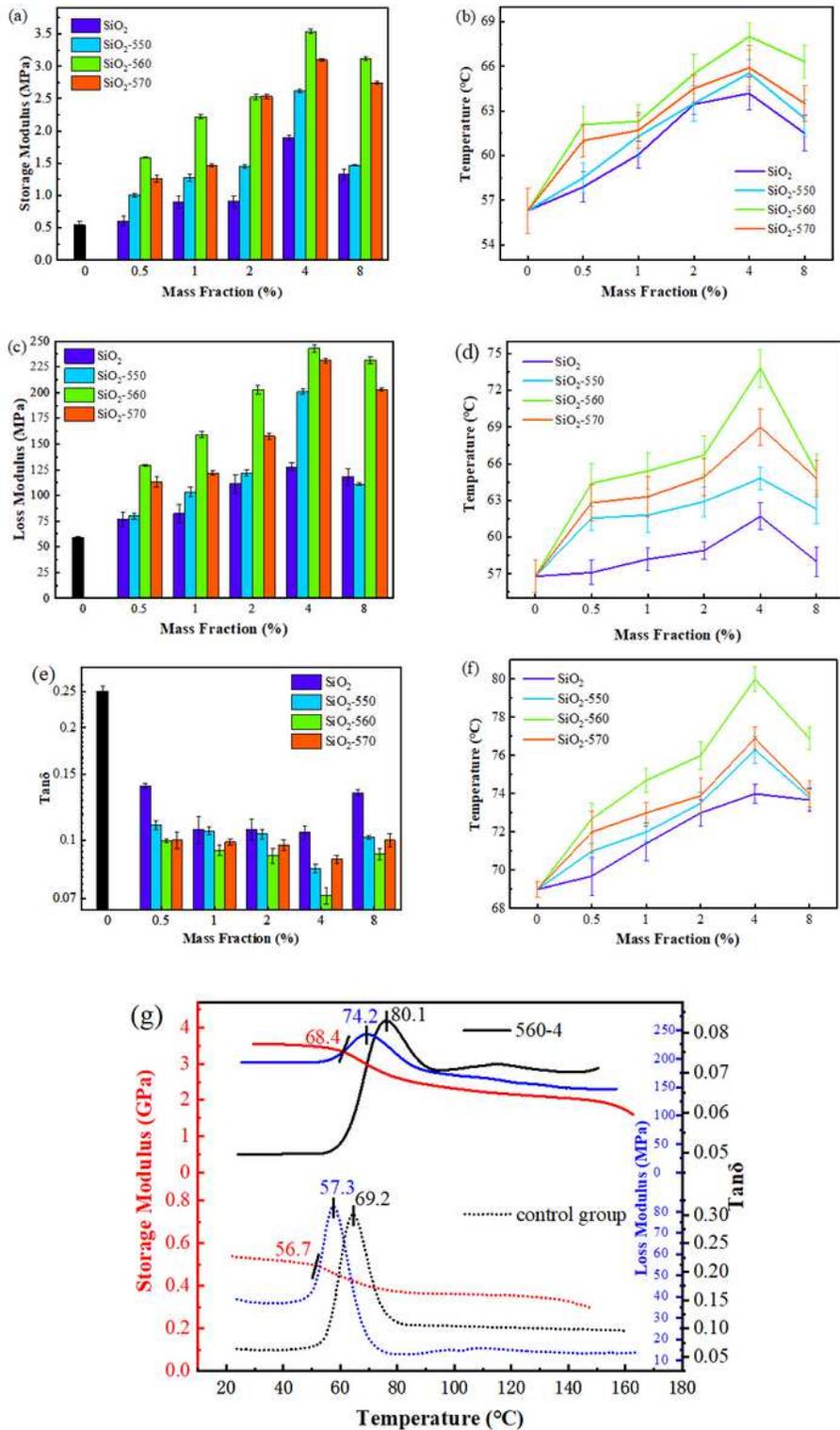
**Figure 9**

The effect of nano-SiO<sub>2</sub> on tensile properties of jute/PLA fibers composites. (a) Tensile strength; (b) modulus; (c) Elongation at break; (d) Tensile break work



**Figure 10**

The interaction between the silica particles and the interface of the jute/PLA composite before and after the modification of the silane coupling agent.



**Figure 11**

The DMTA of jute/PLA composite. (a) Storage modulus; (b) The temperature corresponding to storage modulus; (c) The maximum loss modulus; (d) The temperature corresponding to the maximum loss modulus; (e) The maximum  $\tan\delta$ ; (f) The temperature corresponding to the maximum  $\tan\delta$ ; (g) The comparison of DMTA between jute/PLA composite and sample 560-4

## Supplementary Files

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