1	Reinforcement of continuous fibers for extruded wood-flour/HDPE
2	composites: effects of fiber type and amount
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10	Abstract
11	Continuous-fiber reinforced WPCs were prepared using an extruder with a special
12	die. The effects of the amount (1 to 7 bundles) and type of fiber (aramid rovings and
13	yarns and carbon and glass yarns) on the mechanical properties of WPCs were studied.
14	The adding of continuous fibers led the tensile, flexural and impact strength of the
15	composites to increase by up to 47.3%, 83.1% and 713.4%, respectively. The damping
16	ratio analysis revealed that the interfacial bonding of glass-yarn reinforced WPCs was
17	the best among the tested samples. Adding continuous fibers to WPCs at a low volume
18	fraction can promote their use as load-bearing engineered materials.

19 **Graphical abstract:** 



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# 21 Highlights:

- 22 23
- Continuous-fiber reinforced WPCs were prepared in plate profiles using an extruder with a special die.
- The continuous fibers significantly improved the mechanical strength and 25 toughness of wood-flour/HDPE composites.

• The damping ratio of the composites is used to analyze their interfacial bonding.

27 Keywords:

Wood-polymer composites; reinforcement; continuous fiber; fiber type; interface;
 mechanical properties

## 30 1. Introduction

31 Wood-polymer composites (WPCs) have attained commercial success due to their advantages such as a relative low water absorption, recyclability, high yield efficiency, 32 corrosion prevention, low cost, etc. [1, 2]. They have been widely used in the 33 34 construction, decorative, automotive and transportation fields [3]. However, in terms of 35 mechanical strength, WPCs are not comparable to wood for building structure or steel due to the incompatibility between the polar wood-flour and the hydrophobic polymers. 36 Inadequate strength, impact strength in particular, restricts the utilization of WPCs in 37 load bearing materials [4]. It is therefore very important to improve the strength of 38 39 WPCs to expand their application.

Numerous papers devoted to the improvement of WPCs mechanical properties 40 have been published [4-7]. The main methods used include adding capitalizer [8, 9], 41 42 treating wood flour [10, 11] and adding reinforcement [12, 13]. Coupling agents have become an indispensable component for commercial WPCs. Meanwhile, adding 43 44 reinforcing fibers can further increase the strength of WPCs. Carbon, Kevlar and glass fibers were able to improve the strength and toughness of WPCs, especially long fibers 45 [14]. Short carbon, Kevlar and glass fibers have been widely used to reinforce WPCs 46 in previous studies [4, 15-17]. All these fibers increased the mechanical strength of 47 WPCs. Furthermore, the mechanical strength of fiber-reinforced WPCs varies with 48 49 fiber type and content. Compared to glass and basalt fibers, carbon fibers showed the best enhancement under the same conditions [18]. However, fiber agglomeration is a 50 major challenge to getting good enhancement in short-fiber reinforced WPCs. 51

52 The mechanical properties of long-/continuous-fiber reinforced WPCs are higher 53 than that of short-fiber reinforced WPCs [19]. However, previous studies on long-54 /continuous-fiber reinforced WPCs are inadequate due to processing technology 55 limitations. The long-fiber reinforced WPCs are generally prepared using hot-press 56 processing. Their structure usually resembles a sandwich. Indeed, it is difficult to 57 manufacture continuous-fiber reinforced WPCs directly. In another word, it is a 58 current challenge that needs to be tackle.

In Dura's study [20], a unidirectional laminate of glass fibers was glued onto both surfaces of WPCs at a 2.7% volume fraction. The results indicated that the tensile strength of the resulting composites increased by up to 103%. Carbon-fiber cloth was glued to the surface of wood-flour/HDPE composites to improve the mechanical 63 properties of the composites [21]. This resulted in an interesting phenomenon where the location of long carbon fibers in the WPCs significantly affected their mechanical 64 properties. When fibers were attached to the surface of the WPCs, the increase in 65 flexural strength was greater than the increase in tensile and impact strength. Whereas, 66 the increase in tensile strength was greater than the increase in flexural strength when 67 the fibers were embedded in the WPC [22]. In addition, some attempts to significantly 68 enhance the mechanical strength of WPCs with a similar sandwich structure were 69 presented. A high carbon-steel flat bar was glued onto the surfaces of commercial WPCs 70 and produced an 82% increase in flexural strength [23]. The volume fraction of the steel 71 strips was equivalent to 2.3%. However, the specific gravity of the resulting composites 72 increased. Although these methods can improve the mechanical strength of WPCs, the 73 resulting composites were semi-continuous with masked surface textures and the 74 75 method was not cost-effective.

A few studies have explored the use of continuous fibers in composite materials. Reinforced continuous-glass fiber/polypropylene panels were attached to the surface of WPCs via a double belt pressing program [24]. Glass-fiber rovings were embedded into WPCs using an extruder with a die designed by the authors. The result showed that the mechanical strength of the WPCs increased after adding continuous-glass rovings [14]. It would be very interesting to study the effect of continuous fibers on the mechanical properties of WPCs.

83 The performance of WPCs reinforced by different kinds of continuous fibers was studied with the goal of promoting the use of WPCs in engineering applications. In the 84 current study, aramid rovings and aramid, carbon and glass yarns were used as 85 reinforcements. The effects of continuous-fiber type, amount and structure on the 86 mechanical properties of wood-flour/HDPE composites were studied. Flexural, tensile 87 and impact tests of the reinforced WPCs were undertaken. A statistical analysis of the 88 89 mechanical test results was performed using Statistical Product and Service Solutions (SPSS) software. Meanwhile, their failure modes of the resulting composites in tensile, 90 flexural and impact tests, were analyzed respectively. 91

- 92 2. Materials and methods
- 93 2.1 Materials

Poplar-wood veneer (Harbin, China) was ground into wood flour (40~80 mesh) using a hammer mill. High density polyethylene (HDPE) pellets (5000S) with a melt flow index of 0.7 g $\cdot$ 10 min<sup>-1</sup> (according to ASTM D1238) were provided by Daqing Petrochemical Co., China. Its density is 0.954 g $\cdot$ cm<sup>-3</sup>. Maleic anhydride grafted polyethylene (MAPE) with a melt flow index of 4.85 g $\cdot$ 10 min<sup>-1</sup> and a graft ratio of 0.88 wt% was purchased from Nanjing Juxing Polymer Materials Co., Ltd. The

100 continuous fibers (carbon, glass and aramid yarns and aramid rovings) were purchased from YuShun Textile Co., Ltd., Dongguan, China. The carbon yarns were prepared 101 using the carbon fiber of T300-3000-50A (TORAY, Japan). Two carbon fiber rovings 102 are wound into a twisted fiber bundle. The glass fiber (YS0886), having an average 103 linear density of 2000 tex. The average linear density of the aramid yarn and roving are 104  $1000D \times 3$ . Denier (D), the unit of linear density, refers to the mass (g) of a 9000 m 105 long fiber bundle. Generally, a fiber bundle of 1000D includes 666 single fibers. All of 106 107 them have an equivalent diameter of 0.6 mm per bundle.

### 108 2.2 Preparation of wood-flour/HDPE composites

Wood flour was dried in an oven before blending for 12 hours and its moisture 109 content was about 2%. The wood flour, HDPE and additives (MAPE and polyethylene 110 wax) were compounded in a high-speed mixer (SHR-10A, Zhangjiagang Tongsha 111 112 Plastic Machinery Company, China) for 5 minutes at ambient temperature. The mixture was then extruded using a co-rotating twin screw extruder (Nanjing Rubber and Plastics 113 Machinery Co., Ltd., Nanjing, China) with a screw blade measuring 30 mm in diameter 114 and L/D = 36. The processing temperature ranged from 155 to 175 °C. The wood flour 115 content was 55 wt% while the weight proportion of HDPE was 40%. The addition of 116 MAPE was 4 wt% while polyethylene wax of 1 wt% was added into composites as a 117 lubricant. The mixture was then pelletized into lengths of less than 2 mm. The 118 continuous-fiber reinforced WPC panels were prepared by an SJ-45 single screw 119 extruder (Nanjing Rubber and Plastics Machinery Co., Ltd., Nanjing, China). A special 120 die, being allowed fibers and WPC fluid to pass through, was designed and used, and 121 the continuous fibers were coated by the WPC matrix inside the die (Fig. 1). The die 122 temperature was set as 170 °C. The resulting composites were shaped into a continuous 123  $4 \times 50$  mm plate (thickness  $\times$  width). The amount of fibers in the resulting composites 124 ranged from 1 to 7 bundles. Converting into volume fraction, it varies from 0.14% of 125 126 one bundle to 0.98% of 7 bundles. The number of fibers decreases from the sides to the middle, which means that the fiber bundle is located in the middle of the composites 127 when the amount is one (Fig. S1). 128



#### 130 Fig. 1. Illustration of processing of the continuous fiber reinforced WPCs and their shape.

## 131 *2.3 Dynamic mechanical analysis (DMA)*

132 The DMA test was conducted using a dynamic mechanical analyzer (DMA Q800, 133 TA Instruments, New Castle, USA). A single cantilever strain-controlled mode with an 134 oscillating amplitude of 50 $\mu$ m was used. The frequency was 1 Hz. DMA tests were 135 performed using a temperature range of -20°C to 130°C with a rate change of 3 °C min<sup>-</sup> 136 <sup>1</sup>. Three 35 × 12 × 4 mm test specimens were analyzed. The composites contained three 137 bundles of fibers.

#### 138 2.4 Microscopic characterization

The fracture surface of the resulting composites was obtained using a scanning electron microscope (SEM) (Hitachi S-3500N, Japan). Before testing, the samples were frozen in liquid nitrogen for approximately 20 minutes and then broken using vises. However, the aramid yarns and rovings did not break, the WPC matrix broke instead. They were therefore cut off using scissors after the fracture of the WPC matrix. The fracture-surface was then sputter coated with a layer of gold. The test was carried out with an accelerating voltage of 20 kV.

146 2.5 Mechanical Tests

147 The tensile test was carried out following the ASTM D638-2004 standard at room 148 temperature. The sample dimension was 200 mm in length, 32 mm in narrow width and 149 4 mm in thickness. The measuring length was 50 mm. Samples were prepared using a 150 saw. The test speed rate was 5.0 mm $\cdot$ min<sup>-1</sup>. Six samples were tested for each type of 151 composite. All mechanical tests were carried out at room temperature.

152 Three-point flexure tests were conducted on  $100 \times 30 \times 4$  mm samples. A crosshead 153 speed of 2.0 mm·min<sup>-1</sup> was set for the test. Six samples were used for the flexural test. These samples were prepared using a small table saw. A universal mechanical testing
machine (CMT5504, MTS Systems Co., Ltd., USA) was used for flexural and tensile
tests.

157 The unnotched Izod impact strength based on ISO179-2000 was determined using 158 an impact instrument (CJ5 Chengde Testing Machine Co., Chengde, China). An impact 159 velocity of  $3.8 \text{ m} \cdot \text{s}^{-1}$  was used. The sample dimension was  $80 \times 30 \times 4$  mm. Ten 160 replicates for each composition were tested.

- 161 **3. Results and discussions**
- 162 *3.1the surface chemical and morphology of fibers*

The utilized fibers were analyzed trough FTIR and SEM tests. The FTIR spectra 163 showed that the chemical composition of carbon, glass and aramid fibers greatly varied 164 with their types (Fig. S2). No functional group was observed for carbon fiber while 165 only a peak at 900 cm<sup>-1</sup>, being attributed to Si-O bond, appeared on the glass fiber 166 spectrum. However, there were a lot of absorb peaks on the FTIR spectrum of aramid 167 fibers. The result showed that some polar functional groups, such as amino and carbonyl, 168 are present in aramid fiber. Thereby, the compatibility of polar aramid and non-polar 169 170 HDPE would be worse comparing with carbon fiber and glass fiber.

The surface morphology of fibers were shown in Fig. 2. There were many grooves on the surface of carbon fiber whereas the surface of aramid fiber was smooth. Some adherents were observed on the glass fiber surface. In summary, the roughness of carbon fiber surface was bigger than that of glass and aramid fibers, which may lead to a better adhesion for HDPE on carbon fibers surface.



- 177 Fig. 2. The micromorphology of fibers. a) glass fibers, b) carbon fibers, and c) aramid fibers.
- 178 *3.1 Dynamic mechanical analysis (DMA)*



Fig. 3. Storage modulus of the wood-flour/HDPE composites reinforced with different continuous fibers.



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Storage modulus (E') is the contribution of the elastic component of composites 181 [25]. The variation of storage modulus as a function of temperature for the different 182 fiber-reinforced WPCs was shown in Fig. 3. As temperature increased, the E' of the 183 resulting composites decreased due to the relaxation process of the polymer matrix, 184 which is attributable to the presence of polyethylene in the matrix. In comparison with 185 the original WPCs, the storage modulus of the fiber reinforced WPCs was higher over 186 the entire temperature range. The storage modulus of the fiber reinforced WPCs is 187 188 primarily determined by the nature of both the WPCs and the reinforcing fiber and also by the interfacial bonding. When continuous fibers are incorporated into WPCs, the 189 increase in the storage modulus is attributed to the stiffness of the reinforcing fibers. 190 The difference in the storage modulus of WPCs reinforced with different fiber types is 191 controlled by the interfacial strength of the composites and the fibers' nature. Glass-192 roving reinforced WPCs had the biggest E' due to their good interfacial adhesion and 193 stiffness. 194



Fig. 4 Damping ratio analysis of the resulting composites. a) the resulting composites, b) the system
(pure wood-flour/HDPE) and c) the interface between continuous fibers and wood-flour/HDPE.

Tan $\delta$  is defined as the ratio of the loss modulus to the storage modulus and 199 represents the ability of materials to lose energy during deformation. The damping 200 effect of composites is attributable to the nature of the matrix, the stiffness of the fibers, 201 the interfacial friction and energy dissipation at cracked and delaminated sites [26]. Fig. 202 4a showed the variation of tand with temperature for the continuous-fiber reinforced 203 WPCs in this study. Tand improved with increasing temperature and it barely varied 204 with respect to fiber type. This can be explained by the fact that the interfacial friction 205 between WPC matrixes and fibers increased the damping, although the stiffness of the 206 fibers can cause the storage modulus to increase. 207

Tanδ can be used to characterize a material's viscoelasticity and the interfacial bonding of composites [27]. The viscoelastic behavior of the continuous-fiber reinforced WPCs is more complicated than that of pure polymers and the original WPCs. The energy dissipation of the resulting composites depends on the nature of the matrix resin (HDPE), wood-flour characteristics, the reinforcing fiber, composite structure, the interfacial bonding of the wood-flour/HDPE composites and the interfacial friction between the continuous fibers and the matrix. To study the effect of continuous fibers,
WPCs were considered as simple individuals. To further simplify the study of their
viscoelastic behavior, the complex modulus and damping ratio of two factors are
considered. The damping ratio can be stated as: [28]

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$$\tan \delta_c = \tan \delta_s + \tan \delta_{in} \tag{1}$$

where  $tan\delta_c$ ,  $tan\delta_s$ , and  $Tan\delta_{in}$  are damping ratios of the resulting composite, the system/materials (WPCs and fibers) and the interface, respectively. The damping ratio of the system can be obtained from the complex modulus of the fiber and matrix according to the following formula:

$$\tan \delta_s = \frac{\tan \delta_f E'_f V_f}{E'_c} + \frac{\tan \delta_m E'_m V_m}{E'_c}$$
(2)

Where the  $tan\delta_m$ , E'<sub>m</sub> and V<sub>m</sub> are the damping ratio, storage modulus and volume 224 225 fraction of WPC matrix respectively. Therefore, system damping ratio  $(tan \delta_s)$  can be estimated based on the storage modulus (E'<sub>f</sub>), damping ratio (tan $\delta_f$ ) and volume fraction 226 (V<sub>f</sub>) of the fiber and WPCs and on the storage modulus (E'<sub>c</sub>) of the composites [28]. 227 Hence, the interfacial damping  $(\tan \delta_{in})$  can be calculated by subtracting the system 228 229 damping ratio from the composite damping ratio (tan\deltac). Compared to WPCs, the 230 stiffness of reinforcing fibers is much higher. Therefore, the damping ratio of reinforcing fibers with a high stiffness would be almost zero. Therefore,  $tan\delta_s$  is only 231 determined by the damping ratio and storage modulus of the WPCs and the storage 232 modulus of the resulting composites. The system damping ratio as a function of 233 234 temperature was shown in Fig. 4b. The addition of continuous fibers obviously reduced the system damping ratio. The  $tan\delta_s$  of glass-yarn reinforced WPCs was the smallest 235 among all fiber reinforced composites, especially at high temperatures, because of the 236 effective stress transfer at the interface. Fig. 4c showed the interfacial damping ratio of 237 238 the resulting composites as a function of temperature. For an ideal interface, there is no 239 relative movement between wood-flour/HDPE and fibers. Thereby, in this case, the interfacial friction damping would be zero. Without bonding between continuous fibers 240 241 and WPCs, the interfacial damping would also be zero. However, most of the interface would dissipate energy and contribute to the damping. The interfacial bonding between 242 the test fibers and wood-flour/HDPE composites was poor. Fig. 4 showed that the 243 244 interfacial damping of the composites varied with fiber type. There is no doubt that the better the interfacial adhesion, the higher interfacial friction and the energy dissipation. 245 Thereby, it can be seen that the interfacial adhesion between glass yarns and WPCs was 246 the best among all the resulting WPCs. According to their interfacial adhesion strength, 247 the resulting reinforced composites could be ranked from highest to lowest as follows: 248 glass yarns, carbon yarns, aramid rovings and aramid yarns. Differences in the 249 interfacial adhesion strength of the resulting composites would lead to a change in their 250 properties and failure mechanisms. 251

#### 252 *3.2 Fracture surface morphological analysis*

The morphological characteristics of the continous-fiber reinforced wood-253 flour/HDPE composites on the fracture surface (vertical and parallel to the fiber) was 254 255 observed using a SEM (Fig. 5 and S3). The interfacial compatibility of the continuousfiber reinforced composites varied depending on fiber type. The glass varn bundles 256 were pulled off and the glass fibers were covered by matrix resin (Fig. 5a and S3a), 257 258 which indicated that there was a good interfacial adhesion between the WPCs and glass 259 varns. This showed a good correlation with the DMA results. Meanwhile, a small 260 amount matrix resin infiltrated into the glass yarn bundles as show in Fig. 5a, increasing the contact area. However, wood flour was not found among the glass fibers. This can 261 be explained by the fact that wood flour is difficult to flow into the fiber bundles due to 262 its big size. Although there was also a good compatibility between the carbon fibers and 263 the WPC matrix, the HDPE resin could not iniltrate the carbon-yarn bundles because 264 of the close arrangement of the carbon fibers. However, there were many branched 265 fibers on the carbon-yarn bundles (Fig. 5b). This can increase the contact area between 266 the fibera and the WPC matrix, leading to the improvement of the interfacical strength. 267 The resin covered on the surface of fibers for both carbon yarns and glass yarns 268 269 reinforced composites. However, there were obvious cracks between the aramid fiber bundles and the WPC matrixes (Fig. 5c and d). The matrix resin of WPCs did not 270 impregnate into the aramid rovings and yarns yet. The WPC matrixes have more 271 difficulty infiltrating aramid yarns than aramid rovings due to the twisted structure of 272 273 the yarns. It was worth noting that aramide fiers were closed to wood flour instead of HDPE resin (Fig. S3c and f). Generally, reinforced fiebrs exhibit different interfacial 274 interactions to components of the composites and selectively locate in the component 275 which exhibits relatively high interfacial affinity to them [29]. In summary, the 276 interfacial compatibility of glass-yarn reinforced wood-flour/HDPE composites is the 277 best among the studied composites. This result is in great agreement with the results 278 from DMA test. 279



Fig. 5 Fracture surface (Vertical to fiber) morphological of wood-flour/HDPE composites reinforced by
 glass yarns (a), carbon yarns (b), aramid rovings (c) and aramid yarns (d).

283 *3.3 The flexural properties* 

284 The addition of continuous fibers significantly improved WPC flexural strength (Fig. 6). The largest increase was 47.3% for WPCs reinforced by seven glass yarns. 285 This can be explained by the fact that the load is transmitted to the reinforcing fibers 286 through the interfacial shear force [30]. The fiber bundles withstood part of the load, 287 indicating that the flexural strength of continuous-fiber reinforced composites increased. 288 With the load increasing, the interfacial shear force exceeded the interfacial bonding 289 strength, resulting in breakage of the interfacial bonding, consequently, in the rupture 290 of the composites. With increasing amounts of fiber, the composites' flexural strength 291 tended to increase. This can be explained by the fact that more load is transferred to the 292 293 fibers due to the increase in the interfacial area between the fibers and the WPC matrixes. The results also showed that the flexural strength of the composites varied 294 depending on the type of fibers. Also, glass yarns have the greatest effect on flexural 295 strength among the studied fibers due to their best interfacial bonding (Fig. 4 and 5). 296 297 The flexural strength of fiber reinforced WPCs is mainly related to their interfacial 298 strength.



Fig. 6. Effect of fiber amount and type on the flexural strength of continuous-fiber reinforced wood-flour/HDPE composites.

Fig. 7 showed the load-displacement curves obtained from the flexural tests. In 302 contrast to the original WPCs that exhibited sudden breakage, the continuous-fiber 303 reinforced WPCs were still able to withstand a certain load even after damage to their 304 matrix. This can be attributed to the fact that the continuous fibers were encapsulated 305 in the WPC matrix in a longitudinal direction. Therefore, the continuous-fiber bundles 306 were not pulled off when the composite was damaged since the fiber bundles were still 307 embedded in the composite and kept the broken parts of the WPC matrix together (Fig. 308 8). There was still friction between the fiber and the substrate when an external load 309 310 was applied and this friction force can still offset some of the load. Hence, those curves 311 showed a gradual phase after the decline. With increasing amounts of fibers, the friction force between the fiber bundles and WPCs increased, resulting in an improvement in 312 the remaining load (Fig. 7). The load-displacement curves of glass-yarn reinforced 313 WPCs were different from that of the other composites, given that their load exhibited 314 315 a step-wise decrease after the failure point. Based on the results of DMA tests, the best interfacial bonding occurred between glass yarns and WPC matrixes, which is good for 316 flexural strength. However, the toughness of glass fibers is poor and they gradually 317 break as the bending deflection increases. The carbon-yarn reinforced WPCs exhibited 318 the highest load values after failure as a result of good interfacial bonding and great 319 320 fiber strength. The aramid-yarn reinforced WPCs withstood a greater load after the failure point compared to aramid-roving reinforced WPCs (Fig. 7e). There was no 321 bonding between fibers in the center of fiber bundles for aramid rovings and, due to 322 poor fluidity, the WPCs' matrix could not enter their center and act as binder. Hence, 323 324 the aramid fibers which are in the center of the fiber bundle were easily pulled out, negating their potential enhancement effect. These results indicate that the flexural strength of the resulting composites is affected by both interfacial adhesion strength and the nature of the reinforcing fibers.

The failure mechanism of the continuous-fiber reinforced WPCs in flexural tests 328 was illustrated in Fig. 8. First, the continuous-fiber reinforced WPCs deform after being 329 subjected to an external force. Most of the load is transmitted to the fibers at the same 330 time. With the increasing load, cracks initiate in the interface area and in the matrix, 331 332 thereby absorbing a lot of energy. The cracks then propagate and those in the interfacial 333 area expand along the poor interface. Cracks that encounter good interfacial bonding will change direction, extending up around the fiber. Fibers crossing the cracks will 334 delay crack expansion. Cracks continue to expand as the load increases, resulting in 335 composite failure. However, the fibers are not completely pulled out when the WPC 336 337 matrix fails, the friction between the fibers and the matrix is still able to resist certain levels of applied load. In contrast to this, the pure WPCs break to two parts directly 338 after damage (Fig. S4). This phenomenon indicates that the safety level of WPCs may 339 be increased by continuous fiber reinforcing when they are used in construction 340 341 engineering applications.





343 Fig. 7 Load-displacement curves in flexural tests for aramid-roving reinforced WPCs wood-

- 344 flour/HDPE (a), aramid-yarn reinforced wood-flour/HDPE (b), glass-yarn reinforced WPCs (c),
- 345 carbon-yarn reinforced wood-flour/HDPE (d), and wood-flour/HDPE reinforced with different fiber
- 346 bundles (6 bundles) (e).



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Fig. 8 Illustration of the damage mechanism for continuous-fiber reinforced wood-flour/HDPEcomposites in flexural tests.

### 350 3.4 Tensile tests

A large impressive increase in tensile strength was achieved by adding the 351 352 continuous fibers to the WPCs (Fig. 9). The tensile strength of WPCs reinforced with seven bundles of carbon yarns increased by 83.1 %. This is quite stunning when 353 compared to enhancements reported in previous studies [6, 31-33]. In tensile tests, 354 stress is transferred from the matrix to the fibers through interfacial shear, resulting in 355 an increase in tensile strength. The amount of stress is determined by fiber-matrix 356 357 interfacial adhesion strength, fiber orientation, fiber length and fiber volume content. The stress increases with increasing fiber length, until the fiber length reaches a critical 358 length [34]. Contrary to short-fiber reinforced WPCs, all the fibers in the researched 359 composites are longer than the critical length. Therefore, the tensile strength of the 360 resulting composites was affected by both interfacial bonding and the nature of fibers 361 with a certain level of fiber volume content. Furthermore, The tensile strength of the 362 resulting composites increased with an increase in fiber amount. This phenomenon 363 applies to all fiber reinforced WPCs in the current study. The tensile strength of the 364 resulting composites varied with the type of continuous fibers. The impact of carbon 365 fibers on the enhancement of tensile strength was the greatest among all studied fibers. 366 367 Although the interfacial adhesion strength of glass-yarn reinforced WPCs was the best (Fig. 4 and 5), the strength of glass fibers was the lowest. This leaded to that the tensile 368 strength of glass-yarn reinforced WPCs was the lowest among all fiber reinforced 369 composites. The difference in tensile strength between aramid-roving and aramid-yarn 370 371 reinforcement was very small. Moreover, it was observed that, as the fiber amount increased, the tensile strength of the composites increased. This observation illustrates 372 that the effect of fiber amount on composite tensile strength is larger than the effect of 373 fiber type. 374

The effect of fiber type on WPC enhancement is different for flexural tests compared to tensile tests due to differences in stress patterns. The direction of the tensile stress is the same as fiber direction in tensile tests (Fig. 10). The fiber length is longer 378 than the critical length, so the load is transmitted to the fibers by the interfacial shear force when the composites are subjected to a tensile stress. Cracks appear in the matrix 379 and in the interface between the matrix and the fiber bundles with an increase in the 380 load. Fiber orientation is perpendicular to the main direction of the cracks, stopping the 381 cracks from propagating. However, as the load continues to increase, the cracks expand 382 but the fibers can still withstand most of the load until the composite is damaged. The 383 failure mode of continuous-fiber reinforced WPCs is attributed to the failure of 384 interfacial bonds or the breakage of the fiber. In this case, the interfacial strength of 385 glass-yarn reinforced WPCs was enough, whereas the strength of glass fibers was 386 insufficient. The failure of glass-fiber reinforced composites occurs because of fiber 387 breakage instead of fibers debonding (Fig. S5a). This means that the tensile strength of 388 glass fiber is not enough for fiber reinforced WPCs. On the other hand, when WPCs are 389 reinforced by aramid rovings, aramid yarns or carbon yarns, which have great strength 390 and modulus of elasticity, interfacial bonding failure leads to damage in the resulting 391 composites (Fig. S5b). The debonding of the fibers from the substrate stops the stress 392 from being transmitted to the fibers. The stress withstood by fibers is smaller than the 393 tensile strength of the fibers when the composite fails. 394



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- 396 Fig. 9 Variation in tensile strength for continuous-fiber reinforced wood-flour/HDPE with respect to fiber
- 397 type and amount.



400 Fig. 10. Illustration of the damage mode of continuous-fiber reinforced wood-flour/HDPE in tensile tests.

## 401 *3.5 Impact tests*

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402 Adding continuous fibers led to an important improvement in the impact strength of WPCs (Fig. 11). The result of the independent sample T-test shows that the 403 probability of the fiber having no effect on impact strength is 0.007 (Table S1). In other 404 words, the addition of continuous fibers significantly increased the impact strength of 405 the wood-flour/HDPE composites. Adding seven carbon fibers increased the impact 406 strength of WPCs by 713.4%. The impact strengths of fiber reinforced WPCs 407 significantly increased with the increasing of fiber amount. This rule applies to all types 408 of fibers tested. Impact strength is more sensitive to fiber quantity in continuous-fiber 409 reinforced wood-flour/HDPE composites, compared to flexural and tensile strength. 410 The variation in impact strength with fiber type was also obvious. The impact strength 411 of the carbon-yarn reinforced WPCs was the greatest among the tested composites, and 412 similar to the results of the tensile test. The improvement in the impact strength of glass-413 varn reinforced WPCs was the smallest due to its poor shear strength. The effect of 414 carbon yarns on impact strength was significantly higher than that of glass fibers. 415 Comparing of Fig. 6, 9 and 11, the effect of fiber type on impact strength is more 416 obvious than that for tensile and flexural strength. 417

The impact strength of short-fiber reinforced composites is affected by fiber length, 418 fiber nature, dispersion, orientation, interfacial bonding and flaws formed at the end of 419 the fibers [30]. Compared with short-fiber reinforced composites, there are fewer 420 factors that influences the impact strength of continuous-fiber reinforced WPCs. Fiber 421 422 nature and interfacial bonding are the major factors controlling their impact strength. Generally, the stress dissipation mechanisms are confined due to the fibers limiting the 423 movement of the plastic molecular chain. However, when the composites were 424 subjected to an external impact load, the fibers were subjected to a tensile stress and the 425

load was transmitted to the continuous fibers by the interfacial shear. Meanwhile, a lot 426 of cracks quickly generated and propagated in the interface and the matrix. According 427 to the crack growth mechanism, this process will absorb lots of strength [35]. Also, the 428 continuous fibers are subjected to a shearing stress in impact tests. Two modes of 429 fracture, fiber fracture and pulling out, appeared for the different continuous-fiber 430 reinforced WPCs in this study (Fig. 12). The toughness and strength of glass yarns were 431 smaller than that of the other three kinds of fibers. It is well known that glass fibers 432 break easily when subjected to shear stress. So, the glass-yarn bundles break along with 433 the composite fractures. Unlike glass yarns, aramid yarns, aramid rovings and carbon 434 yarns are pulled during composite ruptures. These fibers have sufficient toughness to 435 withstand the shear stress. Damage to these composites results from interfacial bonding 436 failure rather than from fiber breakage. The impact strength of carbon-yarn reinforced 437 WPCs was the highest among the studied fiber reinforced WPCs. This is mainly due to 438 the fact that the interfacial bonding of carbon-yarn reinforced WPCs was better than 439 that in aramid-yarn and aramid-roving reinforced WPCs. The impact test failure mode 440 of continuous-fiber reinforced WPCs is like that of tensile tests. However, the effect of 441 fiber nature on the impact strength is more pronounced compared to tensile strength, 442 due to the presence of shear stress. 443



Fig. 11. Variation, with respect to fiber type and amount, of Izod impact strength of continuous-fiberreinforced wood-flour/HDPE composites.



447

448 Fig. 12. Illustration of the damage mode of continuous-fiber reinforced wood-flour/HDPE composites in
449 Izod impact tests: pulling out (a) and breaking (b).

## 450 **4. Conclusion**

This study examined the effects of fiber content and type on the mechanical 451 properties of WPCs reinforced by continuous fibers and analyzes their failure 452 mechanism. The results show obvious improvement in all composites when continuous-453 fiber reinforcement is used. The flexural, tensile and impact strength of the composites 454 are improved by as much as 47.3 %, 83.1% and 713.4%, respectively. The interfacial 455 compatibility of glass-yarn reinforced WPCs is the best among the studied samples. 456 However, carbon yarns had the largest contribution to the increase in tensile and impact 457 strengths. The bonding strength between carbon fibers and WPCs needs to be improved 458 to further enhance the properties of the resulting composites as well as aramid fibers. 459 Using continuous reinforcing fibers was incomparably much more effective than other 460 461 treatments.

# 462 Acknowledgements

This work was supported by a grant from the Fundamental Research Funds for the Central Universities (No. 2572017AB09), the National Key R&D Program of China (No. 2017YFD0600802) and the author Jingfa Zhang (201706600026) is supported by the China Scholarship Council.

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