REVIEW PAPER

PLA composites reinforced with rice residues or glass fiber—a review of mechanical properties, thermal properties, and biodegradation properties

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Abstract

Fiber reinforced polylactic acid (PLA) composites are one of the current research hotspots. Rice residues (RR) and glass fiber (GF) are commonly used as raw materials for strengthening PLA composites. In this paper, the mechanical properties, thermal properties and degradation properties of RR/PLA and GF/PLA composites were reviewed. The results showed that the defect in fiber (RR, GF) reinforced PLA is significantly improved by modification and the mechanical properties are improved by about 40%. The main reason is the unification of the surface polarity of fibers and PLA, as well as the role of connection established by the functional groups. Meanwhile, the surface modification of RR/PLA and GF/PLA composites can also improve their thermal and degradation properties. RR/PLA composites has worse heat resistance than pure PLA, whereas GF/PLA composites has better thermal stability than pure PLA. The crystallinity of RR/PLA composites was generally higher than that of pure PLA, indicating that natural fibers played the role of nucleating agent and promoted the nucleation of PLA. The pyrolysis loss of natural fiber reinforced PLA composites begins at 100 °C and can be divided into three stages: evaporation of water within the fiber, degradation of cellulose and hemicellulose, and degradation of non-fiber components. At about 300 °C, the degradation rate is the highest, and the weight loss ratio is above 90% at the end of the pyrolysis process. In terms of degradation performance, RR/PLA composites has excellent biodegradability, and its biodegradability increases with the increasement of RR content. Degradation of GF/PLA composites is only degradation of PLA, so the decline of mechanical properties of GF/PLA composites is weakened. It can be predicted that with the advent of the era of carbon neutrality, the green degradable PLA composite technology will be developed deeply and quickly.

Keywords PLA composites · Rice residues · Glass fiber · Surface treatment · Mechanical properties · Thermal properties · Degradation properties

Introduction

As the climate change in global generates a major problem to human society, more countries have elevated "carbon neutrality" as a national strategy and proposed a vision for a carbon-free future [1], which restricts the use of natural resources such as oil and coal [2]. It has been understood that the petroleum-based plastics are widely used in various industries due to their high quality and low price [3].

With the development of society, the usage in plastics was grown quickly and caused a serious environmental pollution [4–6]. Significant attention has been focused on the application of transportation, aviation, and construction of the biodegradable composite [7, 8]. Polylactic acid (PLA), which is the most commonly used biodegradable thermoplastic aliphatic polyester, produces from glucose derived from renewable corn, sugar cane and sugar beets [9]. It has the characteristics of recyclability and composability widely used for 3D printing applications primarily, and it is one of the most hopefully replaced petroleum-based polymers [10–14]. The tensile strength and Young's modulus of PLA are equivalent to polypropylene (PP), polyethylene (PE) and polystyrene (PS), but low strength, poor heat resistance and high-cost limits its application [9, 15]. Many methods are developed to improve the performance

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of PLA, which has a research hotspot utilized fiber reinforcement [16–20]. PLA is reinforced by two types of fiber that is natural fiber and artificial fiber [21–23]. Natural fibers are more environmentally friendly, while artificial fibers have stronger mechanical and thermal performance [24, 25].

Natural fibers have the characteristics of natural growth, light weight and recyclability that can reduce the health problems caused to operators such as skin irritation, etc. [26–32]. As a kind of natural fiber, rice residue (RR), including rice straw (RS) and rice husk (RH), a remaining biomass resources after rice harvest, has the advantage of abundant content, cheap cost, and completely degradable [33, 34]. The incineration of RR usually exhausts a large amount of greenhouse gas, which will have a great negative impact for the environment [35]. According to research, the RS and RH cellulose content keeps 41–57% and 35–45%, respectively, suggests that they have the function in enhancing PLA [36–38]. Comprehensive literature shows that the tensile strength of RR/PLA composites is mostly around 50 MPa and the bending strength of RR/PLA composites is mostly around 80 MPa [39–41]. The performance can meet the less demanding decorative parts such as the interior decorative parts of automobiles or airplanes [22, 42, 43]. For secondary load-bearing parts and load-bearing parts that require higher mechanical properties, they are usually reinforced by artificial fibers [44, 45]. Carbon fiber (CF) reinforced composites are the mainstream of research and development in high-performance composites, and it have been widely applied in the primary and secondary bearing structures of aircraft [46, 47]. However, fiber reinforced composites have been used a few in load-bearing parts and secondary load-bearing parts of automobiles due to the limitations of cost and performance. Glass fiber (GF), which often manufactured high-performance composites, is cheaper than carbon fiber and has the characteristics that natural fiber does not possess (high temperature resistance, corrosion resistance, strong mechanical properties, etc.) [48–50]. The tensile strength and bending strength of GF/PLA composites can reach 120 MPa and 170 MPa, respectively. The value greatly exceeded the properties of modified polypropylene widely used in automobiles and met the performance requirements in some main and secondary load-bearing components in transportation and aviation [51, 52]. In conclusion, the industrial application of RR/PLA and GF/PLA composites has great potential and it is very necessary to summarize the factors that affect the mechanical properties to obtain better properties [53, 54]. In addition, the fiber composites need heat resistance adequately, as it would inevitably contact with the environment in application. The degradation of fiber composite is also considered after the service cycle [55, 56]. It is beneficial to summarize the thermal properties and biodegradation properties of fiber composites [57, 58].

In the past period of time, fiber reinforced PLA composites have undergone significant development, and have many applications in some fields. These applications are attributed to the improved performance of composites. In this study, a literature review is done with the intent of reviewing the performance of RR/PLA and GF/PLA composites in term of mechanical properties, thermal properties and degradation properties. The purpose of this study is to explore the RR/PLA and GF/PLA composites technology, and it is hoped to be helpful with the future research.

Mechanical properties of RR/PLA composites

Pretreatment

The components of RR are mainly cellulose and hemicellulose, so it has a strong hydrophilic prosperity due to a large number of hydroxyl groups within the surface [53, 59]. PLA molecule has hydrophobic property, which resulted in inferior interface compatibility [59, 60]. In the study of Wu et al. [61], the tensile strength of RH/PLA composite was gradually decreased with an increase of the RH content. The fiber weight content increased by 40%, the tensile strength of the RH/PLA composites was reduced by 57.6%. Yussuf et al. [62] suggested that PLA composites filled by RH had a negative impact. Compared with pure PLA, the flexural strength of RH/PLA composites was decreased by 55.6%. As shown in Fig. 1, the interface strength of RR/PLA composite was very weak with many voids and fiber pullouts phenomena. But the fiber surface was coated by polymer improved the bonding properties avoided fiber damage during processing. Zhu et al. [63] reported that the nano attapulgite (ATP) solutions with different contents (0.5, 1, 1.5, 2%) coating the rice straw(RS) surface formed the new intermediate layer between the RS and PLA, which had a positive effect on the mechanical performance of the RS/PLA composite. The tensile strength of MRS/PLA composites (30% MRS, 2% ATP) was increased by 19% compared to the uncoated polymer one. The highest tensile modulus had occurred at 1.5% ATP, which was 5.9% higher than the uncoated composites. Qin et al. [64] reported that the poly butyl acrylate(BA) coating the RS surface (10% RS) can improve the mechanical strength of RS/PLA composite. The tensile strength of RS/PLA composite was increased at first and then decreased with the increasement of BA content. When the content of BA accounted 20% of RS, the tensile strength of RS/PLA composite was reached the maximum value (30 MPa), which was 25% higher than that of the uncoated composites.

Although the mechanical properties of RR/PLA composites had improved by coating polymer, interface performance was not improved. The reason was no direct effect on the properties of interface connection. Generally, the interface

Fig. 1 Fracture surface of RR/PLA composite: **a** 20% RS [61], **b** 20% RH [62], **c** 20% RH [65], **d** 20% RH [66]
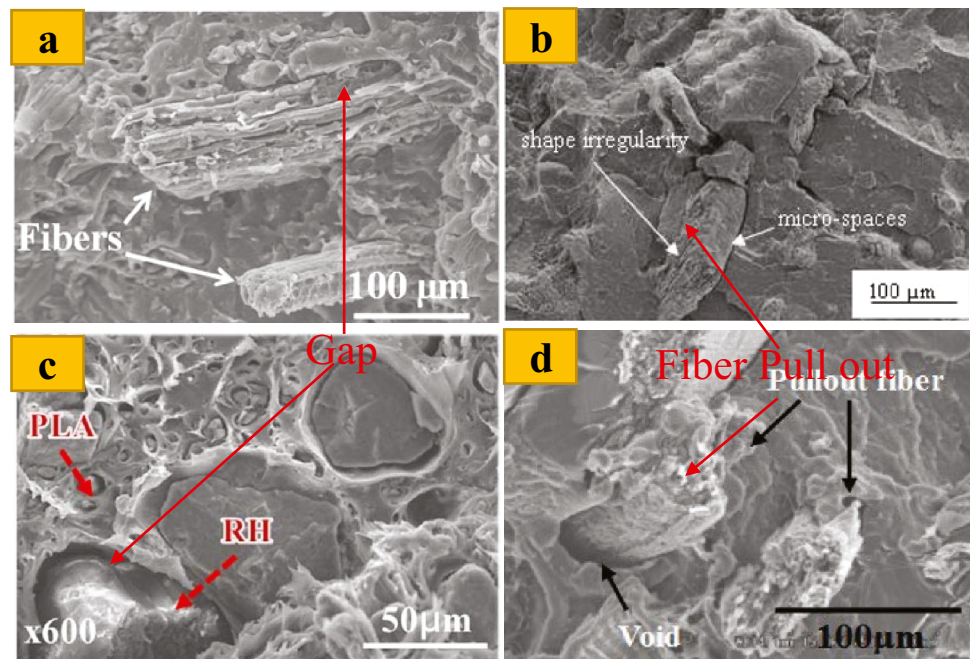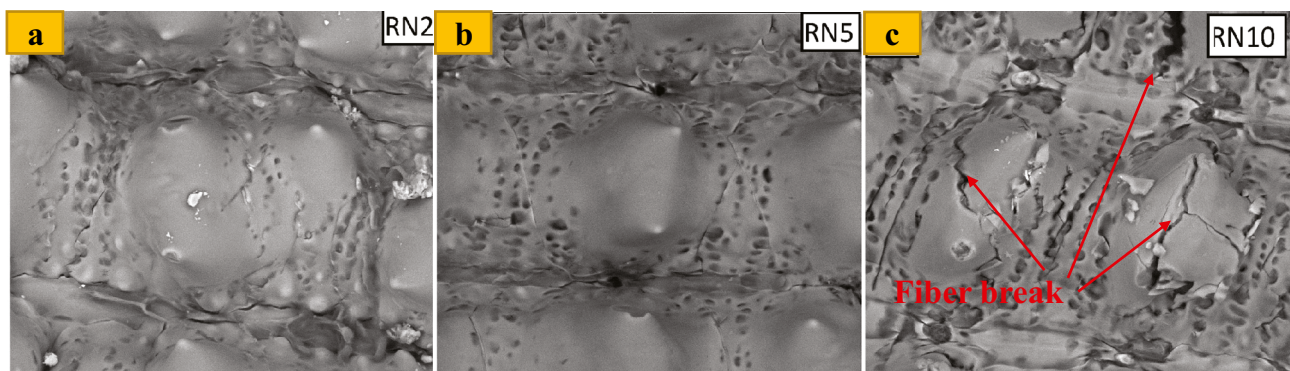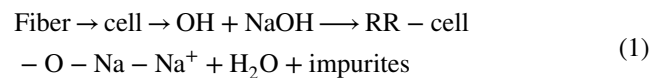

Table 1 Husks compositions (%) as a function of the NaOH concentration for an alkaline treatment duration of 24 h [68]

Husk composition	R	RN 2	RN 5	RN 10
Wax	2.2 ± 0.9	0.8 ± 0.4	0.7 ± 0.6	0.5 ± 0.4
Linin	37.1 ± 2.7	38.2 ± 1.4	39.9 ± 0.4	27.4 ± 1.3
Holocellulose	46.3 ± 1.4	49.8 ± 0.8	52.0 ± 0.2	65.1 ± 0.6
Inorganic(ash)	14.3 ± 0.2	11.2 ± 0.2	7.5 ± 0.1	6.9 ± 0.0

performance of RR and PLA can be improved by various chemical and physical treatments [67]. Alkali-treated was the simplest and cheapest method in natural fiber surface treatment. The proportion of cellulose was increased by alkali treatment due to the removal of some non-cellulose components. Tran et al. [66] studied the effect on the rice husk (RH) treated by different alkali concentration(2%, 5%, 10%). It was confirmed that the alkali-treated removed part of the lignin and hemicellulose in RH, and removed a large amount of pectin, wax oil and other substances (Table 1). It is clear from Fig. 2 that 5% NAOH.
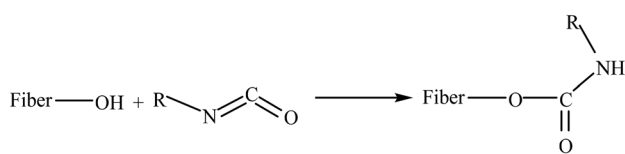
treated had the best fiber surface and 10% NAOH had broken part of fiber. Alkali would react with the hydroxyl groups in the surface of cellulose, which reduced the content of hydroxyl groups and increased the performance of interface. The formula of NaOH reaction was shown in (Eq. 1). Yu et al. [68] reported that the water absorption of RS/PLA composites was significantly reduced by alkali-treated. Compared with the untreated composites, the water absorption was reduced by 27.7%. The mechanical performance test results showed that the tensile strength, tensile modulus, flexural strength, and flexural modulus were increased by 31.19%, 16.48%, 18.75%, and 25.27%, respectively. The surface of fiber treated by alkali was easier to react with other modifiers. Therefore, alkali treatment was generally used a pretreatment method in fiber treatment process. In addition, the pretreatment methods were applied to fiber, also including microwave treatment and ultrasonic treatment.

$$\text{Fiber} \rightarrow \text{cell} \rightarrow \text{OH} + \text{NaOH} \longrightarrow \text{RR} - \text{cell}$$
$$- \text{O} - \text{Na} - \text{Na}^+ + H_2O + \text{impurites} \tag{1}$$

Fig. 2 Variation of surface morphology of the RH treated by NaOH: **a–c** NaOH concentration of 2%, 5% and 10%, respectively [68]

Fig. 3 The reaction principle of KH 570 connecting fiber and PLA [72]

Fig. 4 The reaction principle of MAPP connecting fiber

Fig. 5 The reaction mechanism of isocyanate treated fiber

Surface modification

Silane, a multifunctional molecule, was used as a coupling agent modified fiber surface, which formed chemical connection by the siloxy group and the alkyl group [69]. The process that silane coupling agents modified fibers involved several stages of hydrolysis, condensation, and bonding [37, 70–72]. The silanol was formed by the hydrolysis of siloxy groups. Its one end can react with the hydroxyl group of cellulose during the condensation process (which also enhanced the hydrophobicity), and its other end was reacted with the PLA matrix to form a bond (Fig. 3). Tran et al. [66] reported that the rice husk (RH) modified by silane had a positive effect on the properties of RH/PLA composites. The tensile strength and flexural strength increased by 6.6% and 10.9%, respectively, which was attributed to the improvement of interface properties. The second function of silane penetrate into the pores of fiber surface and act as a surface protective layer. Chen et al. [72] studied the effects of silane coupling agents (APS, KH 560, KH 570 and KH 590) on the interface properties of wheat straw (WS)/PLA composites. Compared with the unmodified composites, the tensile strength and flexural strength of WS/PLA composite treated by KH 570 were increased by 43.29% and 40%, respectively. The SEM showed that the interface properties of the WS and PLA had been significantly improved.

Maleic anhydride coupling agents were commonly used on the surface of fiber and matrix provided an effective interaction. Maleic anhydride, one end reacted with the hydroxyl group of cellulose reducing the hydrophilicity, and other end can also form stable carbon–carbon bonds with PLA during the modification process, which provided a chemical connection that was similar to the effect of silane between RR and PLA. Hamdan et al. [33] reported that the mechanical properties of RH/PLA composite modified by the toluene solution of MAPP and MAPE had been significantly improved. The MAPP concentration was 6%, the tensile strength reached the peak (20.9 MPa), which was 39.3% higher than the unmodified one. When the MAPE concentration was 6%, the maximum bending strength was 49 MPa, which was 48.5% higher than the unmodified one. The values of tensile modulus and flexural modulus was made a little change after composite modified. The reaction mechanism of maleic anhydride polypropylene (MAPP) was shown in Fig. 4.

The reaction mechanism of isocyanate modified fiber was that the functional group of isocyanate ($-N=C=O$) reacted with the hydroxyl groups of cellulose and lignin to form urethane bond, which provided a strong interfacial bonding. In addition, the isocyanate can also react with water on the fiber surface to form urea that can further reacted with the hydroxyl groups of cellulose. The secondary reaction provided a higher hydrophobicity and better interfacial bonding. The reaction formula between the fiber and isocyanate coupling agent was shown in Fig. 5. Tsou et al. [73] reported that the mechanical properties of RH/PLA composites modified by isocyanate (MDI) had a significant positive effect. The tensile strength of RH/PLA composite (with 20% RH content) was increased by 55.7% (from 32.5 MPa to 50.6 MPa). The water absorption of composite had been clearly reduced, and the printing test suggested that the RH/PLA composite (with 20% RH content) filament with the solubilizer MDI can be used in the 3D printing application.

Acrylic acid had two functional groups: carbon—carbon double bond and carboxyl group. Under the induction of benzoyl peroxide (BPO), the carbon–carbon double bond of acrylic acid broke and grafted to the PLA chain [74]. The carboxyl group of acrylic acid reacted with the hydroxyl group in the fiber to form stable ester bonds. This provided more pathways for the free radicals of the active cellulose to enter the polymer. Wu and Tsou [65] used acrylic acid (AA) to graft polylactic acid (PLA-g-AA) and coupling agent to treat rice husk (TRH) to improve the properties of PLA/RH composites. Compared with PLA/RH, the tensile strength of PLA-g-AA /TRH (30% TRH) composites was increased by 112.5%. In this study, the AA was successfully grafted onto PLA by BPO as initiator. The esterification reaction occurred between PLA-G-AA and TRH, and the interface between the two was stronger. The reaction formula was shown in Fig. 6.

Other modification methods such as the ester compounds also have a strong improvement in compatibility between fiber and polymer. Zhao et al. [75] studied that the effect of

**Fig. 6** The reaction formula of AA connecting TRH and PLA
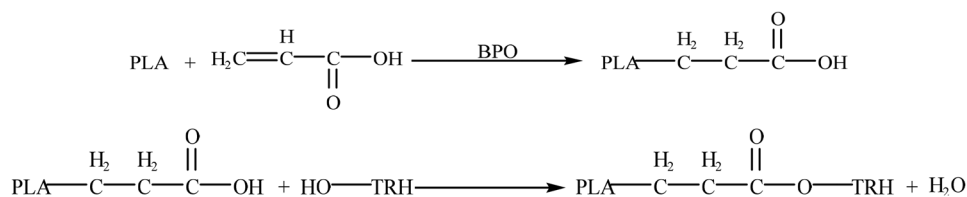
the methyl methacrylate (MMA) modified rice straw fiber (RSF) on the properties of RSF/PLA composites. Compared with the unmodified one, the tensile strength (RSF content of 30%, 0.02% MMA) increased by 53.6%. Wu et al. [61] studied the effect of glycidyl methacrylate (GMA) on RS reinforced PLA composites. The tensile strength of composite (30% RS) increased by 31.4%. It was a high molecular polymer that the polyethylene glycol (PEG) possessed excellent dispersibility and adhesion was used as a plasticizer in the preparation of RR and PLA composite with the effect of improving performance. Zubir et al. [76] reported the effect of the polyethylene glycol (PEG) on the adhesion of RS and PLA/PHBV. As a results, the tensile strength of the composites was increased by 14.6%. As shown in Fig. 7, the RS/PLA composites had good interface compatibility and fiber breaks were found at the section after chemical treatment, indicating to play role in enhancing PLA.

Some researchers had extracted the cellulose from RS and RH to enhance PLA composites. Battegazzore et al. [77] extracted cellulose from RH and fabricated cellulose/PLA composite by melt blending. From the SEM image, the cellulose particle was uniformly distributed and had excellent adhesion in the PLA matrix. The storage modulus of the cellulose/PLA (30% cellulose) increased by 41%. In subsequent work, they extracted the silica from RH to prepare silica/PLA composites [78], and the mechanical properties were comparable to the industrial silica/PLA composites [79]. It can be seen that the cellulose extracted from RR had similar properties to commercial cellulose, and the RR had excellent advantage for enhancing biopolymers.

This part reviewed the main literature in term of RR/PLA composites and introduced the mechanism of surface treatment and its effect on mechanical properties. Many modification methods, including alkali treatment, silane coupling agent, crosslinking agent treatment, GMA, AA, MDI, MAPP, MAPE,

Fig. 7 Surface morphology of chemical treatment of RR/PLA composites: **a** 1% RS and alkali + ultrasound, **b** 20% RH and 2 wt% MAPP [33], **c** 20% RH and AA [65], d 30% RH and MDI [73], **e** 20% RS and GMA [61], **f** 30% RS and 0.02% MMA [75]
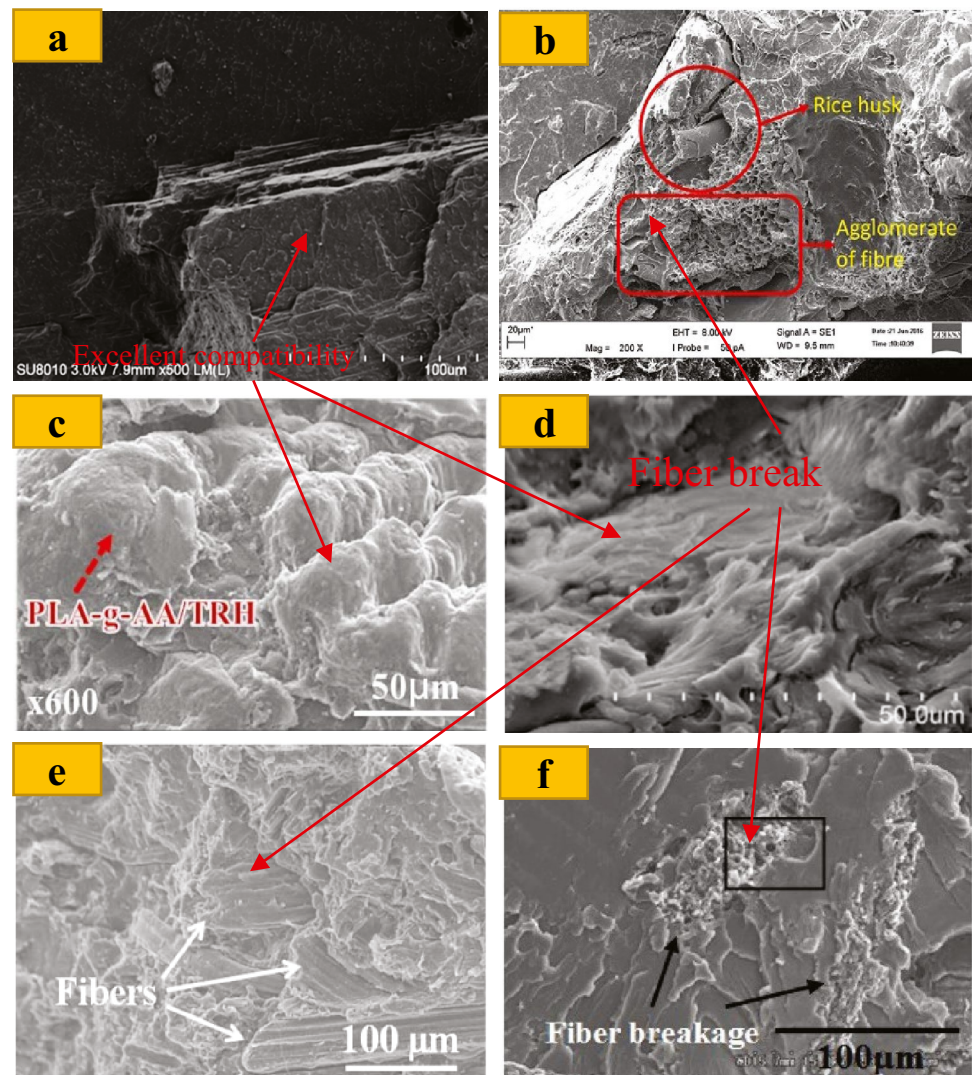
MMA, ATP covering, microwave carbonization (HC), were applied in the RR surface for adjustment the interface connection [59]. It is apparent from the images (Fig. 1) that the fractured surface of untreated RR/PLA composite has large number of fiber pullouts and void, which indicates the poor adhesion between matrix and fiber. On the other hand, fracture surface of silane and MAPP modified RR/PLA composites exhibits fiber breakage (Figs. 2 and 7) rather than fiber pull out, which proves the improvement in the interfacial bonding. Due to this phenomenon, better stress transfer will lead to improved mechanical properties [80]. Furthermore, Table 2 provides a list of mechanical properties of RR/PLA composite that employ various treatment method. The tensile strength of RH/PLA composites treated with MAPP and PEG was the lowest (about 25 MPa). The tensile strength of RH/PLA composites treated with silane, MMA, and ATP was the highest (about 60 MPa). That is mostly improved by about 40% compared to unmodified composites. It can be concluded that surface modification has been excellent effect on improving mechanical properties.

Mechanical properties of GF/PLA composites

Glass fiber (GF), with high strength and low cost, is often used to prepare high performance composite. It has many applications in the aerospace field, but a few applications in the construction and automotive fields [84–86]. This section summarized recent literature on the mechanical properties of GF/PLA composites.

Methods with untreated materials

The performance of GF/PLA composite was largely depended on the content, aspect ratio and dispersion of GF [87]. Different from natural fibers, the increasement of mechanical properties of GF/PLA composite were positively correlated with GF content [86]. Varsavas et al. [88, 89] reported the mechanical properties of GF/PLA composites was increased with the increasement of GF content. Compared with pure PLA, the tensile strength, flexural strength, tensile modulus and flexural modulus of GF/PLA composites were increased by 31.9%, 20.7%, 47.7% and 100%, respectively. Huda et al. [90] prepared GF/PLA composite (30 wt% GF) by twin-screw extruder and injection molding machine. Compared with the neat PLA, the tensile strength, flexural strength, elastic modulus, flexural modulus and notched impact strength of GF/PLA composite were increased by 28%, 10%, 145%, 152%, and 53%, respectively. The mechanical properties of PLA composites were obviously improved by GF, and some performance was enhanced double time. On the same aspect with nature fiber, the interface properties of GF and PLA was greatly affected by dispersibility and compatibility. The literature showed that many fiber pull-outs and voids were discovered at the interface between the GF and PLA (Fig. 8). The reason was that the interface performance of GF in the PLA matrix was inferior. It is necessary to improve the interface performance of GF and PLA by surface modification.

Surface modification

A number of researchers adopted the modification measures of natural fiber to modify GF surface for improvement the interface performance. This method made the stress smoothly transferred from the matrix to the fiber. The versatility of silane coupling agent can also serve as a medium for inorganic (GF) and organic (PLA) [92]. It was different from other coupling agents that the silane was improved the

Table 2 Mechanical properties of RR/PLA composites

Author	TS/MPa	TM/GPa	FS/MPa	FM /GPa	Mark	Year
Yussuf et al. [62]	-	-	85	4	-	2010
Nyambo et al. [81]	56	4.6	83	5.8	30% straw	2011
Zhao et al. [75]	63	-	-	-	0.02% MMA	2011
Qin et al. [64]	30	-	-	-	5% RSF	2011
Wu et al. [61]	51.8	-	-	-	GMA、TEOS	2013
Kellersztein et al. [82]	36.7	4.1	75.4	4.47	20%	2015
Zubir et al. [76]	27	-	-	-	RS 25%, PEG	2017
Chaboki et al. [34]	28.39	-	-	-	RS 10%	2019
Zhu et al. [63]	63	2.86	-	5.46	1.5% ATP	2019
Hamdan et al. [33]	22	1.4	48	3	MAPP 20% RH	2019
Wu et al. [61]	53	-	-	-	(AA) grafting	2019
Tsou et al. [73]	50	-	-	-	MDI 20% RH	2020
Chen et al. [72]	38	-	75	-	30% WS	2021
Nizamuddin et al. [83]	36.17	6.182	-	-	20% HC	2021
Yu et al. [68]	59	0.55	92	3.25	A + U	2021

Fig. 8 Surface morphology of GF/PLA composites: **a** 20% GF [87], **b** 30 GF [90], **c** 15% GF [88], **d** 20% GF [88]
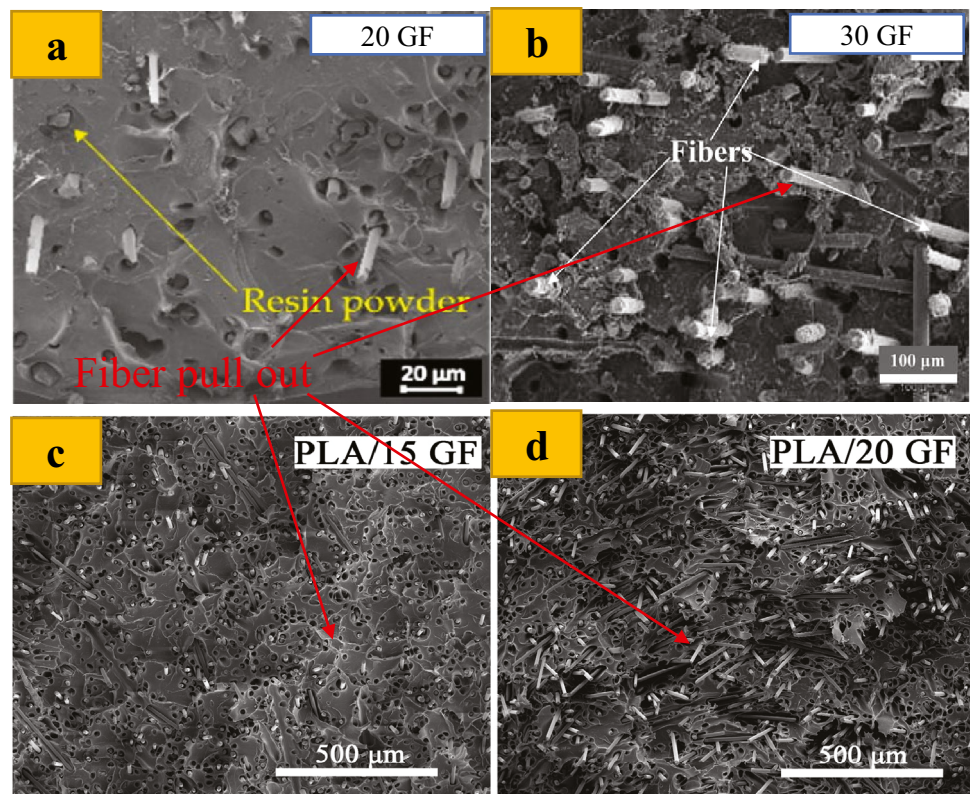

mechanical properties of the composites by increasing the length of the fiber. Jing et al. [91] reported that the GF modified by silane was seem coated a protective film that prevented fiber breakage and kept the fiber length closest to the original (Fig. 9). Compared with the unmodified composites, the tensile strength was increased by 52.9%. Wang et al. [93] reported that the GF modified by silane greatly improved the mechanical properties of PLA composites. Compared with pure PLA, the tensile strength, flexural strength, tensile modulus and flexural modulus of GF/PLA composites (20% m-GF) increased by 90.6%, 42.2%, 137.5 and 103.3%, respectively. It was attributed to form the chemical connection by the reaction of the silane coupling agent. One end of the silane had three siloxy groups that can be hydrolyzed

to form silanol, and the silanol reacts with the surface of the inorganic material. The other end of silane had the long alkyl chain that can react with the polymer. The reaction mechanism of silane coupling agent action with GF and PLA matrix was shown in Figs. 10 and 11.

Graphene oxide (GO), which was the closest precursor to graphene, had a large number of epoxy, hydroxyl, carbonyl and carboxyl functional groups that was easy to disperse in water and polar organic solvents [94]. It was reported in the literature that the GO can be coated on glass fiber by electrostatic assembly technology, as the modifier of fiber surface like silane coupling agent [95]. Jing et al. [91] studied the effect of GF modified by graphene oxide (GO) on the mechanical properties of GF/PLA composites. The

Fig. 9 Schematic diagram of GF enhancement mechanism for PLA composites [91]
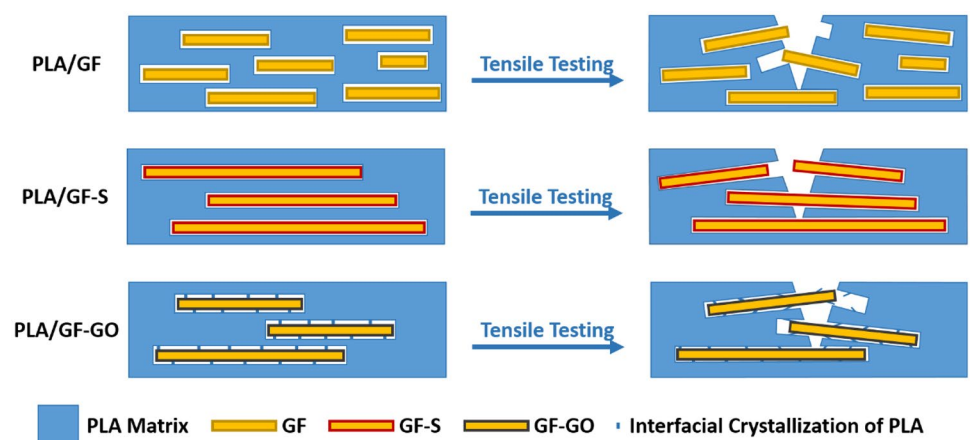
Fig. 10 Silane coupling agent hydrolysis
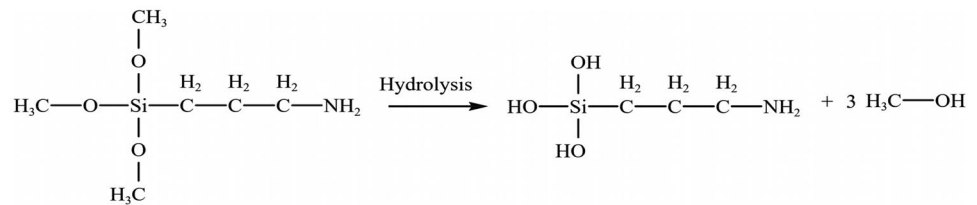
results showed that the GO can greatly promote the nucleation of composites, which improved the modulus of GF/PLA composite. The author conducted an in-depth analysis of the reinforcement mechanism and fracture evolution process of the graphene oxide modified GF. The interface adhesion between GF and PLA treated by graphene oxide was significantly improved (Fig. 9). Among them, improving compatibility and adhesion was the most fundamental factor for the improvement of the mechanical properties of GF/PLA composites. The chain extender was used for the functionalization to restore the molecular weight of GF/PLA composites and improved the crystallization performance of PLA. Akindoyo et al. [96] reported that the GF modified by impact modifier (BS) and chain extender (CESA) improved the interfacial properties, and the impact strength of the composites was increased by 21.4%. Wang et al. [93] reported that the crystallinity of GF/PLA composites was increased after heat treatment. It can be inferred that the heat treatment was an effective method to improve the mechanical properties of semi-crystalline polymers. Goh et al. [97] used the fused filament manufacturing (FFF) technology to manufacture the thermoplastic composites reinforced by the continuous carbon fiber and glass fiber. Compared with the short glass fiber, the tensile strength and Young's modulus of the continuous GF composites sample was 450 MPa and 7.20 GPa, respectively. It made thermoplastic high-performance structural parts possible by the development of continuous GF compound extrusion technology.

Others

Some researchers have experimented with other aspects, such as blend PLA with other polymers. Lu et al. [98]

took the solubilizer PTW (the content is 5%) for increasement the compatibility of PLA and HDPE added different GF contents (5, 10, 20, 30 wt%) to enhance the PLA/HDPE matrix. The SEM observed that the compatibility of PLA and HDPE was significantly improved. The PTW could effectively reduce the contact angle of PLA/HDPE mixture, which improved the compatibility of composites. When the addition of GF was 30 wt%, the tensile strength of the GF/PLA composite was increased by 75%, and the impact strength and elongation at break of GF/PLA composite were reduced by 57% and 83%, respectively. Lin et al. [99] employed different GF contents (10, 20, 30 wt%) to reinforce the PLA/PC (70/30) matrix. The tensile strength, flexural strength and notched Izod impact strength of composites were separately increased by 84%, 73%, and 200%. Hazer and Aytac [100] conducted a similar research to study the mechanical properties of the PLA/PC (50/50) blend reinforced by GF. The tensile strength of GF/PLA/PC (30% GF) composites was 93.3% higher than that of PLA/PC. In another study [52], they used different mixed of carbon fiber (CF) and GF to reinforce PLA/PC(50/50) blend. The SEM observation found that there was a better interface adhesion between the GF/CF and PLA/PC matrix. With the synthetic fiber admixture increasement, the tensile strength of composites was increased. Among the PLA/PC blend with 15 wt% GF and 15 wt% CF, the tensile strength was the highest at 115.23 MPa, which was 102.2% higher than that without addition. The mixture using of CF-GF can reduce the economic cost. Varsavas and Kaynak [88, 89] reported that the GF and thermoplastic polyurethane elastomer (TPU) mixed to enhance PLA. The SEM observation revealed that the dispersion of TPU in the PLA matrix was inferior.

Fig. 11 Grafting with GF and PLA reaction
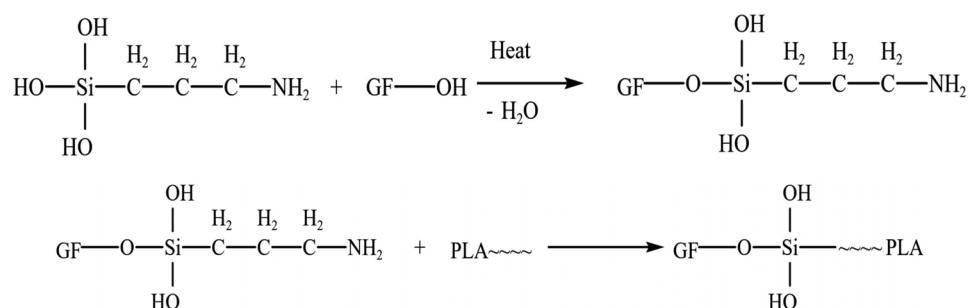
Compared with the TPU/PLA composites (10% TPU), the tensile strength, flexural strength, tensile modulus, and flexural modulus of GF/TPU/PLA composites (15% GF, 10% TPU) were increased by 20.9%, 12.1%, 52.6%, 54.7% respectively. Rafiquzzaman et al. [50] processed the jute/GF reinforced epoxy resin composites by manual layout, and evaluate its tensile, bending and impact properties. The mechanical properties have been greatly improved due to addition GF to the polymer.

Most current studies showed that the high strength GF had greatly improved the mechanical performance of PLA composites. In general, in all GF/PLA composites, it is noted that the mechanical properties of the composites increased linearly up to a certain stage and drops afterwards. The critical point is the optimum fiber content. In view of the poor interface performance of GF and PLA, researchers had conducted a number of modification studies on GF/PLA composites, such as silane coupling agents, graphene oxide, impact modifiers and chain extenders. As shown in Fig. 12, these modifications improved the interface properties of the GF and PLA, when the mechanical properties and reliability of the GF/PLA composite had been greatly improved after chemical modification. The mechanical properties of GF/PLA composites from the above studies were shown in Table 3. It could draw a conclusion that GF/PLA composite with the optimum content and the most effective modification would achieve the highest performance.

Thermal properties of RR/PLA and GF/PLA

The thermal characteristics of RR/PLA composites and GF/PLA composites are highly influenced by the characteristics of the RR and GF [34, 75, 102]. When RR/PLA composites and GF/PLA composites are subjected to heat, they are likely to experience prominent alterations in their physical and chemical properties [103]. The RR/PLA and GF/PLA will undergo processes such as evaporation, sublimation, water absorption, etc., which is a function of both temperature and time [104, 105]. As a consequence, there will be variations in mechanical and thermal properties. In RR/PLA composite, the reduction in the thermal stability may be predominant due to the degradation of natural fiber [106]. In GF/PLA composite, the slower reduction in the thermal stability may be occur because of glass fiber for high temperature resistance. The thermal degradation of natural fibers generally happens at 200–210 °C, and the thermal degradation of PLA begins at around 300 °C, whereas the thermal distortion of GF takes place above 500 °C.

The thermal stability of composites is studied through thermogravimetric analysis (TGA) [107]. During TGA, the mass of the sample is observed as the temperature of the sample is increased [108]. Chougan et al. [109] used TGA to evaluate the thermal degradation of wheat straw/PLA composites. The TGA curve showed that there were three distinct stages in the degradation (Fig. 13). In the first

Fig. 12 Surface morphology of chemical treated GF/PLA composites: **a** 20% GF and silane coupling agent [93], **b** 30% GF and S [91], **c** 30% GF and GO [91], **d** 10% GF and BS + CEAS [96]
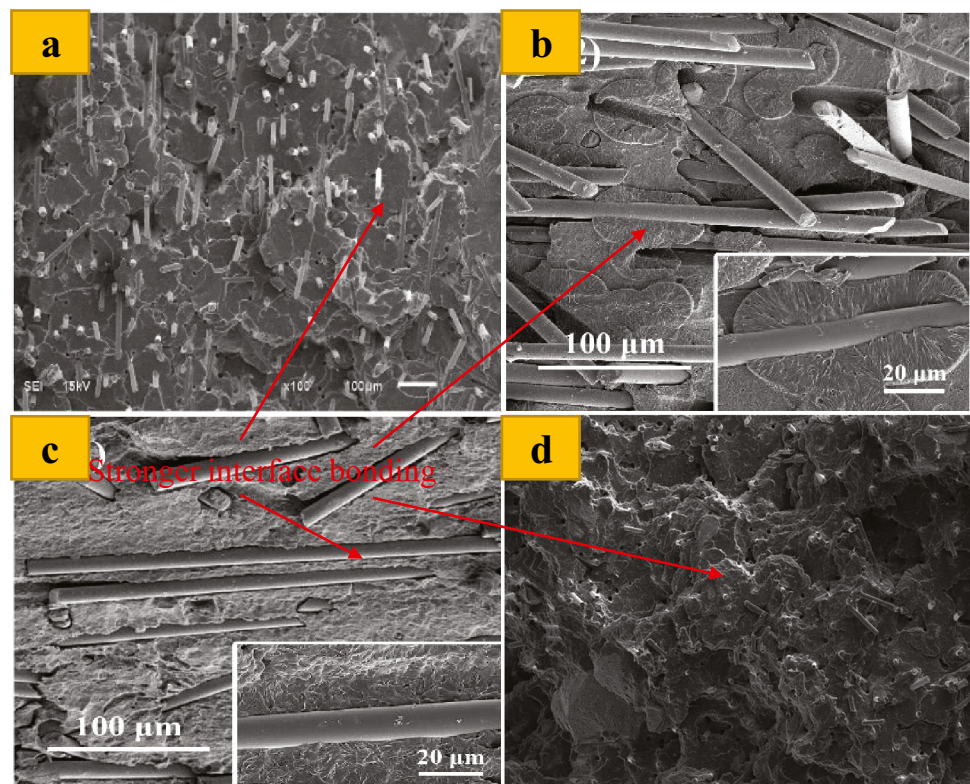
Table 3 Mechanical strength of GF/PLA composites

Author	TS	TM	FS	FM	Remark	Year
Huda et al. [90]	80.2 MPa	6.7 GPa	108.9 MPa	8.2 GPa	30% GF	2010
Lin et al. [99]	124.7 MPa	-	173.2 MPa	7.3 GPa	30% GF, PLA/PC	2011
Wang et al. [101]	118 MPa	9.98 GPa	168 MPa	11.6 GPa	20% GF, HOT	2014
Hazer and Aytac [100]	108 MPa	-	-	-	30% GF	2019
Jing et al. [91]	140 MPa	5.8 GPa	-	-	30%GF, GO/S	2019
Akindoyo et al. [96]	80.4 MPa	6.03 GPa	123 MPa	5.89 GPa	10% GF	2019
Lu et al. [98]	67 MPa	-	-	-	30% GF	2020
Hazer and Aytac [100]	108 MPa	-	-	-	30% GF	2021
Wang et al. [93]	80 MPa	8 GPa	123 MPa	6.2 GPa	20% GF, SILANE	2021

stage (100~150 °C), a weight loss of 5%-10% can be seen in the composites. Here, the weight loss happens due to the evaporation of moisture that is trapped within the fiber. The second stage of degradation (250~350 °C) corresponds to the decomposition of hemi-cellulose and cellulose. Simultaneously, PLA degradation also happens in this stage [110]. Out of the three most important constituents of fiber: cellulose, hemi-cellulose and lignin, hemi-cellulose has the lowest thermal stability and it was the first compound to degrade among the three. It is followed by cellulose degradation while degradation of lignin takes place at a higher temperature. The third stage (above 350 °C) of degradation is attributed to the decomposition of lignin and other non-cellulosic molecules. Tran et al. [66] reported that the thermal degradation of PLA and it composites was a single-step

degradation mechanism that occurs at around 360 °C. The degradation temperature of RH/PLA composites modified by alkali and silane was 354–357 °C, and second only to pure PLA, significantly higher than untreated composites (340 °C). It showed that the thermal decomposition temperature of RR/PLA composites will be improved after surface modification.

The thermal degradation of GF/PLA composites can be divided into two section due to glass fiber for high temperature resistance. The initial weight loss is the evaporation of moisture, and then the degradation of PLA. Wang et al. [101] reported that the thermal degradation temperature of pure PLA and PLA/GF composites was all almost 330 °C. For an increase content GF, the thermal degradation temperature of GF/PLA composites tends to shift slowly to high

Fig. 13 TGA curves of untreated RH/PLA, KH 550 treated RH/PLA
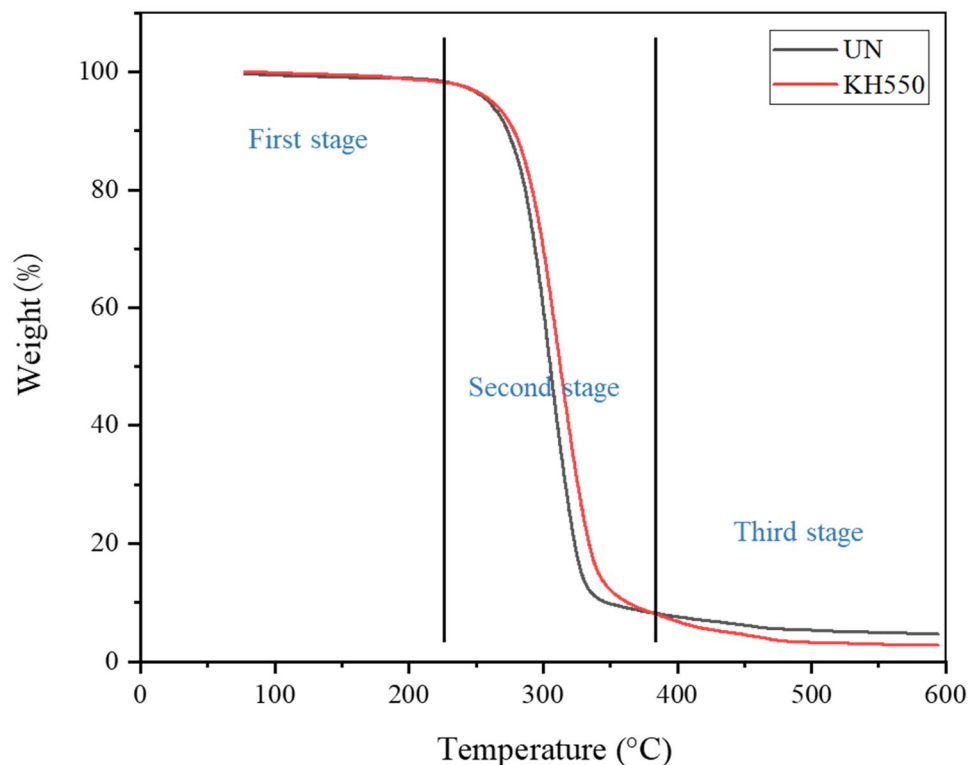
temperature. The study had shown that the thermal degradation properties of GF/PLA composites can be adjusted by GF surface modification. Akindoyo et al. [96] reported that the initial degradation temperature of PLA composites and the temperature of materials degraded by 50% was increased by Jing et al. [91] reported that the GF treated by graphene oxide (GO) promoted the properties of PLA nucleation, which enhanced the thermal performance. Wang et al. [101] and Deng et al. [111] also had similar reported.

RR had little effect on the T_g and T_m of PLA composites, but it significantly improved the crystallinity of PLA composites, the reason was that the fiber act a role in promoting nucleation (Qin et al. [64], from 10.7 to 16.6%). Battegazzore et al. [77] reported that cellulose extracted from RS can reinforce crystallinity of PLA composite. The thermal properties of RR/PLA composites were changed on account of the different treatment in RR surface. In lijun qin's work, the RS/PLA composite was treated by PBA. The crystallinity of composites treated by PBA was less than that of pure PLA, as the PBA prevented crystallization of RS/PLA composites. Nizamuddin et al. [83] suggested that the melting point of composites had little change with carbonization RS, and the agglomeration nature of carbonization RS prevented the free movement of polymer chains, which reduced the crystallinity of PLA composites. Wu et al. [61] reported that RS reduced the melting point of PLA composites. The reason was that the distance between the PLA chains is magnify during the grafting process, as the crystallization of polymer became more difficult. Zhu et al. [63] reported that ATP-coated RS lead an increase in heat resistance. But they have not studied the crystallinity of composites. The modification methods in the above-mentioned documents all have a certain effect on the crystallinity, but failed to improve the comprehensive thermal performance of RR/PLA and GF/PLA

composite. The influence of other modifiers on the thermal properties of RR/PLA composites needs to be studied in depth.

The T_g, T_m and X_c of GF/PLA composites showed different results for RR/PLA composites [112, 113]. Jing et al. [91] reported that GF treated by graphene oxide promoted the nucleation process of PLA, which can greatly improve the crystallinity of PLA composites. Wang et al. [101] reported that the heat treatment greatly improved the crystallinity of GF/PLA composites. The heat resistance of GF/PLA composites was significantly improved in heat treatment induced crystallization. Akindoyo et al. [96] reported that chain extenders and impact mixed modifiers improved the crystallinity of GF/PLA composites. The T_g, T_c, and T_m of GF/PLA composites almost had little change, which indicated the presence of GF had little effect on the mobility of PLA chain. As the GF size was too large, it affects the fluidity of the polymer segment. The thermal properties of RR/PLA and GF/PLA composites were shown in Table 4. It showed that RR and GF can improve the crystallinity of PLA composites, especially cellulose and graphene oxide. Neither RR nor artificial fibers (GF) had much influence on the Tg, Tc, and Tm of PLA composites.

Degradability of RR/PLA and GF/PLA

Fiber composites (RR/PLA, GF/PLA), the marvel materials of the present world are inevitable in various applications as a result of their mechanical performance and cost feasibility [117–119]. The degradation mechanism of RR/PLA and GF/PLA composites has been studied and reported by very few researchers though experimentation by nature-degradation, exposure to different environmental

Table 4 Thermal properties of RR/PLA and GF/PLA

Samples	$T_g(^\circ C)$	$T_{cc}(^\circ C)$	$T_{m1}(^\circ C)$	$T_{m2}(^\circ C)$	ΔH_m (J/g)	$X_c(\%)$	Treat	Ref
PLA	58.8	100.4	144.1	150.6	27	10.7	-	Qin et al. [64]
PLA/RS	58.8	96.7	143.4	151.5	28.6	16.6	-	Lijun qin
PLA/RS	59.4	99.9	144.1	151.7	14.5	9.8	PBA	Lijun qin
PLA/cellulose	61	124	-	153	21.7	29	From RS	Battegazzore et al. [77]
PLA/HC	-	-	-	165.3	14.2	17.3	RS(HC)	Nizamuddin et al. [83]
PLA/RS	67	-	-	153.2	-	-	GMA	Wu et al. [61]
PLA/RH	66	-	-	169	77	-	Water T	Arjmandi et al. [114]
PLA/RH	57	-	-	160	52	-	Alkaline T	Arjmandi
PLA/PHBV/RS	-	-	-	166.2	-	42.6	-	Zubir et al. [76]
PLA/RH	66.1	-	-	153.1	-	-	AA	Wu and Tsou [65]
PLA/GF	57	-	-	170	41.7	44.5	-	Huda et al. [115]
PLA/GF	61.45	113	150.1	156.7	25.8	32.5	Hot	Wang et al. [101]
PLA/GF	61.9	112.3	162.8	168.8	55.37	51.29	GO	Yin and Bao [116]
PLA/GF	61.9	-	-	154.9	-	40.4	GBC	Akindoyo et al. [96]
PLA/PC/GF	64.3	-	-	150.7	5.9	14.9	-	Hazer and Aytac [100]

conditions, as well as attack from fungi and microbial [120]. These tests were conducted to study their resistance against biodegradation to ensure long durability of composites [121, 122]. The weight reduction in composites after degradation is noted to comprehend the degradation mechanism.

The degradation of RR/PLA composites potentially occurred on biological, chemical, thermal and water environments. The soil burial experiment was the simplest method to simulate the natural degradation of composite. The degradation of cellulose in the soil mainly included microbial degradation and hydrolysis [123]. Yussuf et al. [62] reported that the RH/PLA composites had stronger degradation properties than pure polylactic acid in soil burial experiments. There was slow weight loss on the 10th and 30th days. On the 90th day, the weight loss of RH/PLA composite was twice that of PLA (Fig. 14). The same result appeared in the experiment of Guo et al. [124]. During soil burial, wood fiber/PLA composite was more prone to degradation than pure PLA. Gunti et al. [125] reported that the weight loss percentage of composite increased linearly with the number of days buried in the soil. In addition, the degradation rate in the enzymatic environment was significantly higher than that of soil burying. Wu et al. [61] reported that the PCL/RSF composites had stronger biodegradability than that of pure PCL. The biodegradation rate increased with the increasement of RSF content. It can save cost of time that the experiments of accelerated degradation were implemented under certain conditions (constant heat, constant humidity, UV aging). Kyutoku et al. [36] conducted the experiment

of accelerated degradation on cellulose/PLA composites under the constant temperature and humidity conditions. The results showed that the storage modulus decreased by 50% over time. During the experiment of accelerated degradation of constant temperature and humidity, hydrolysis in PLA composite was occurred, but adding modifier can inhibit the hydrolysis of PLA composites [126].

During the degradation process (chemical degradation, thermal degradation) of GF/PLA composites, because the ingredient of GF was unaltered, it prevented the degradation of mechanical properties of composite. Varsavas and Kaynak [89] studied natural weathering degradation performance of GF/PLA composite. The exposure of pure PLA and GF/PLA composite (with 15% GF) was tested by the accelerated weathering conditions of ultraviolet light irradiation and water circulation. The results showed that the mechanical property of PLA was reduced by 92%, the mechanical property of GF/PLA was only reduced by 34%.

The above documents showed that the RR/PLA composites had strongly degradation performance in the natural environment, which can disappear completely after a certain period of time. The degradation rate can be adjusted by adding different fiber and modifying agent. In addition, RR/PLA composites was manifested in a lower initial degradation temperature and a faster degradation rate, which showed lower thermal degradation performance than pure PLA. The heat resistance of GF/PLA composite was higher than that of pure PLA slightly and the thermal degradation resistance of GF/PLA was improved by surface modification of GF. There were not many studies on reporting the degradation performance of RR/PLA composites, especially in

Fig. 14 Relationship between weight loss and compost time (PLA, RH/PLA) [62]
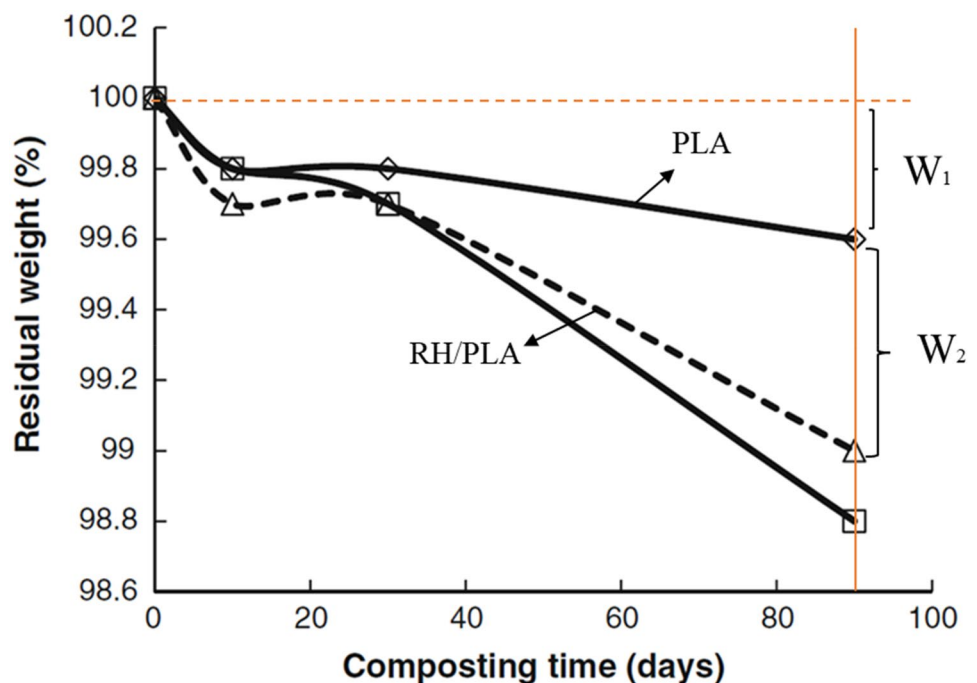
hydrolysis and UV degradation. As GF kept not degradable, it was conducive to be used in the long-term durability of composites. The degradation performance of PLA composites needs researchers to study deeply.

Conclusion

Petroleum polymers had been fully used in industry and produced a large number of non-degradable materials, and resulted in multiple environmental issues. Alternatives for petroleum polymers are attracting the attention of the research community. Concerning this purpose, green composites is regarded as the new technology and opportunity. Due to its excellent processability and natural biodegradability, PLA was a suitable matrix for natural fiber composites. Many techniques to improve the performance of PLA composites are constantly evolving. There is a clear difference between the strength of natural fiber composites and synthetic fiber composites. This article reported the research progress of mechanical properties, thermal properties, and biodegradation properties of RR/PLA and GF/ PLA composites.

For the first, it was information on the mechanical properties of PLA composite. The defects in RR and GF reinforced PLA composites were improved with many methods of surface treatment. Also, the mechanical properties of RR/ PLA and GF/PLA composite was improved under surface modifying. We therefore introduced the surface modified methods of RR and GF, and summarized the enhancement mechanism of fiber surface modified. The results showed that the tensile strength and bending strength of RR/PLA composite was about 50 MPa and 80 MPa, respectively. The mechanical performance of GF/PLA composites was highly superior than that of RR/PLA composite. The tensile strength and bending strength of GF/PLA composites can reach 120 MPa and 170 MPa, respectively.

For the second, the thermal properties and degradation properties of RR/PLA composites and GF/PLA composites was analyzed. Thermal performance of RR/PLA composite was worse than that of pure PLA, whereas the thermal stability of GF/PLA was slightly better than that of pure PLA. The TGA weight loss process of RR/PLA composites can be divided into three stages. In the first stage, the moisture in the fiber evaporates; the second stage was the degradation of cellulose and hemicellulose; and the third stage was the degradation of non-fiber components. During the TGA (room temperature-600 °C) test process of GF/PLA composites, the weight of GF was hardly changed. Biodegradability analysis showed that RR/PLA composites had preeminent biodegradability, and the biodegradability increased with the increasement of RR content. The aging of GF/PLA composite was merely the degradation of PLA components, so

the decline of mechanical properties of GF/PLA composite was weakened. Also, the modification of both RR/PLA and GF/PLA may also lead to increased values of the thermal properties and degradation properties.

Compared to GF/PLA composites, the mechanical performance of RR/PLA composites was lower. However, the combination of RR and GF creates the possibility for the functionalization of fiber composites. In summary, the research of fiber reinforced PLA composites was a dynamic process that needs to delve into multiple performance features. It was necessary to develop models for the performance prediction of RR/PLA and GF/PLA composites and to explore the degradation laws under various conditions. This review will help readers to understand the performance of mechanical, thermal and degradation of RR/PLA and GF/ PLA. The future composites will possess higher mechanical properties, more durability and versatility. The development of fiber composites in automotive, transportation, aerospace and other manufacturing industries should be concerned, which will help maintained a sustainable ecological environment.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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