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Effects of Fiber Dimension and Its Distribution on the Properties of Lyocell and Ramie Fibers Reinforced Polylactide Composites

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Abstract: The regenerated cellulose fiber (Lyocell) and natural cellulose fiber (ramie) with different initial lengths were used to reinforce polylactide (PLA) by melt compounding and injection molding. The fiber dimension and its distribution in composites after injection molding were analyzed by high-resolution Fiber Quality Analyzer (FQA). Moreover, the contents of small fibers with length < 0.2 mm were also provided by FQA. Then the influences of fiber dimension and its distribution on the mechanical properties of the composites were investigated. The results showed that fiber length was reduced during compounding and injection molding processes, and the longer the initial fiber length, the more severe the length declined. Compared with ramie fiber, Lyocell fiber showed a longer fiber length, smaller diameter and narrower width distribution in composite after injection molding. Additionally, the content of small fiber played an important role in the analysis of the relation between the fiber dimension and mechanical properties of composites owing to its smaller aspect ratio. The different mechanical characteristics of the PLA composites were attributed predominantly to the fiber aspect ratio and the small fiber content. By comparison, Lyocell fiber exhibited better reinforcement for PLA due to the higher aspect ratio and lower small fiber content. The tensile strength, tensile modulus and impact strength of Lyocell/PLA composite were increased by 8.61 %, 75.43 % and 26.9 %, respectively.

Keywords: Polylactide composites, Lyocell fiber, Ramie fiber, Fiber dimension, Distribution

Introduction

Recently, the biodegradable polymers are increasingly gaining importance as the promising substitute materials for petro-based polymers [1]. Polylactide (PLA) is considered as one of the most promising biodegradable polymers and has been widely used as biomedical and packaging materials. Although PLA is a relatively stiff polymer with good tensile strength, its low impact strength and slow crystallization rate prevent its spread applications. In order to solve these problems, some fibers such as glass fiber, carbon fiber and cellulose fibers have been used to reinforce PLA to accelerate the crystallization rate and improve the mechanical properties of PLA in recent years [2-5].

Compared to the glass fiber and carbon fiber, cellulose fibers offer a series of advantages, such as renewability and biodegradability, lower density and cost, and less abrasion to the processing equipment [6]. In addition, some natural cellulose fibers also have comparable specific mechanical properties as traditional glass fibers [7]. Therefore, many natural plant cellulose fibers, such as bamboo [8], flax [9], jute [10], hemp [11], ramie [12] and cotton fibers [13], were used as the reinforcing fiber to improve the properties of PLA and decrease the cost of resultant material [14,15].

Among the natural plant cellulose fibers, ramie has much longer fiber length and higher tensile modulus than jute, hemp and flax, and its specific tensile modulus is even approximate to that of glass fiber [5]. Xu *et al.* [16] found that the tensile strength and tensile modulus of injection molded ramie/PLA composites were increased by 39.2 % and 102.8 %, respectively, without sacrifice of toughness. Chen *et al.* [17] found that the addition of ramie fiber accelerated the crystallization rate of PLA, and the tensile modulus and flexural modulus of 24 wt% ramie/PLA composite were increased by 54.9 % and 45.7 %, respectively.

In addition to the natural plant cellulose fibers, the regenerated cellulose fiber is another attractive reinforcing fiber for PLA. Lyocell fiber is a new kind of regenerated cellulose fiber with good mechanical properties, and it has lower variability in the surface constitution and roughness in comparison to natural cellulose fibers. Therefore, Lyocell has the potential to be an ideal reinforcing fiber for PLA and can offer the composite high impact strength due to its higher elongation, compared with the natural cellulose fibers such as flax, kenaf and hemp [18,19]. Baghaei *et al.* [11] found that the impact strength of 30 wt% Lyocell/PLA composite was increased by 126 % compared with pure PLA. An overview of mechanical properties and application fields of natural and regenerated cellulose fibers-reinforced PLA composites was well concluded by Graupner *et al.* [18]. The results showed that Lyocell/PLA composite had high tensile strength, tensile modulus and impact strength,

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and exhibited the best comprehensive characteristics than PLA composites reinforced with cotton, kenaf and hemp.

Gunning *et al.* [20] investigated the effect of twin-screw extrusion on the final length of Lyocell fibers in PLA-based composites prepared by injection molding. It was found Lyocell fibers showed slightly decrease in average fiber length after extrusion and injection molding due to the intensive shear stresses during the process. However, the relationships between the fiber dimension & distribution and the mechanical properties of the cellulose fibers/PLA composites need to be further investigated.

In this paper, the typical natural cellulose fiber (ramie) and the regenerated cellulose fiber (Lyocell) were used to reinforce PLA by melt compounding and injection molding. The fiber dimension and its distribution in PLA composites after compounding and injection molding were analyzed by high-resolution Fiber Quality Analyzer (FQA). Then the influences of fiber dimension and its distribution on the mechanical properties of the composites were investigated.

Experimental

Materials

PLA chips used in this work were provided by Zhejiang Haizheng Biomaterials Co., Ltd., China. Its melt flow index was 16.3 g/10 min (190 °C/2.16 kg), and the glass transition temperature and melting temperature were 58.0 °C and 159.3 °C, respectively.

Lyocell filaments (™Newcell) were supplied by Akzo Nobel Co., Germany. Ramie fiber (degumm and refined ramie fiber) was purchased from Hunan Dongting Hemp Industry Co., Ltd., China. The average diameter of Lyocell fiber and ramie fiber were ~18 µm and ~30 µm. The tensile strength of Lyocell fiber and ramie fiber were 3.28 cN/dtex and 5.95 cN/dtex, and the elongation of Lyocell fiber and ramie fiber were 7.18 % and 3.71 %, respectively.

Preparation of Cellulose Fiber Reinforced PLA Composites

The fibers were cut into different initial lengths (1 mm, 5 mm, 10 mm, 15 mm and 20 mm), and then the fibers and PLA chips were dried in a vacuum oven at 60 °C for 12 h before compounding processing. Then 15 wt% reinforcing fibers were compounded with PLA matrix in a torque rheometer (CTR-100, Shanghai Changkai Mechanical & Electrical Technology Co., Ltd., China) with a rotating rate of 50 rpm at 180 °C for 5 minutes for uniform mixing of fiber and PLA matrix. The resultant compounds were cut into small granules and then injection molded using an injection-molding machine (SZ-5-C, Shanghai Dehong Rubber & Plastics Machinery Co., Ltd., China). The injection temperature and mold temperature were 180-190 °C and 50 °C, respectively. The injection pressure was 0.5 MPa, and the dwell time was 8 s. Moreover, the specimen of pure PLA was also prepared as reference materials.

Measurements of Fiber Dimension and Its Distribution

In order to study the fiber dimension and its distribution in the PLA composites, the fiber must be extracted from the composites. Firstly, the composite standard test bar was soaked in chloroform and stirred with a magnetic bar at 2000 rpm and 25 °C for 24 h to dissolve PLA matrix. Then the mixed suspension was filtrated to obtain the extracted fibers. The procedure was repeated three times. At last, the extracted fibers were dried in a vacuum oven at room temperature for 48 h.

The fiber dimension was analyzed using the high-resolution Fiber Quality Analyzer (FQA, LDA02, OpTest Equipment Inc., Canada). It is a special testing equipment for fiber quality, more than 10000 fibers were measured at a time within several minutes, and some information of the fibers including the fiber length, fiber width, and small fiber (with length < 0.2 mm) content can be calculated automatically and accurately. Briefly, the fiber suspension by using deionized water as diluent was transported through a sheath flow cell where the fibers were oriented and positioned. The images of the fibers were detected by a built-in CCD numerical camera, and the data (including the fiber length, fiber diameter, and small fiber content) of the fibers was measured by circular polarized light. Each fiber can be identified by the real-time two-dimensional image analysis software of the FQA from the image signal, and then its morphological parameters can be measured according to the birefringence of cellulose. The introduction of the FQA and experimental operations were described by Robertson [21] and Olson [22]. All samples were run in triplicate, and each FQA measurement collected the data of fibers up to ten thousand. The FQA measurements can provide the results of number-average fiber length (L_n, $Ln=\Sigma n_i L_i/\Sigma n_i$), weight-average fiber length (L_w, $Lw=\Sigma n_i L_i^2/\Sigma n_i L_i$), the number-average small fiber content (F_n, %), weight-average small fiber content (F_w, %) and the average fiber diameter [23].

Scanning Electron Microscope

The surfaces of initial cellulose fibers and the extracted fibers from PLA-composites were observed by using a scanning electron microscope (SEM) (JSM-5600LVJEOL Co., Ltd., Japan).

Measurements for Mechanical Properties of the Composites

The tensile properties of cellulose fiber reinforced PLA composites were tested with a universal testing machine (Instron 5969, Instron Corporation, USA) according to ASTM D638-03. The notched izod impact strengths of the composites were determined using a XJJUD-50Q Izod Pendulum impact tester (Chengde COTS Scientific Instruments, Co. Ltd., China) according to ASTM D256-10.

For above-mentioned mechanical properties tests, five replicates were evaluated for each sample.

Results and Discussion

Comparison of Fiber Dimension and Its Distribution of Two Cellulose Fibers in PLA Composites

Figure 1 shows the average fiber lengths of Lyocell and ramie in composites after compounding and injection molding (with an initial length of 5 mm). Wherein, C and I mean the compounding process and injection molding process, respectively. C-L_n and C-L_w mean number-average fiber length and weight-average fiber length in the cellulose fiber/PLA mixture after compounding, respectively. In the same way, I-L_n and I-L_w mean number-average fiber length and weight-average fiber length in the fiber/PLA composite after injection molding, respectively. The results showed that the average fiber lengths (L_n and L_w) of Lyocell and ramie fibers were shortened drastically after compounding, and they were further reduced after injection molding. Moreover, the length of ramie fiber was shorter than that of Lyocell fiber not only during compounding process but also during injection molding. The possible reason maybe that ramie fiber is more rigid and easily be cut off when it is subjected to shear stress. It is also worth noting that the aspect ratio of Lyocell and ramie fibers with the same initial fiber length was different due to the different diameter. However, the fiber length also decreased while the diameter changed during the compounding and injection molding process. Therefore, the final size of the fibers in the composite is the key to the reinforcement.

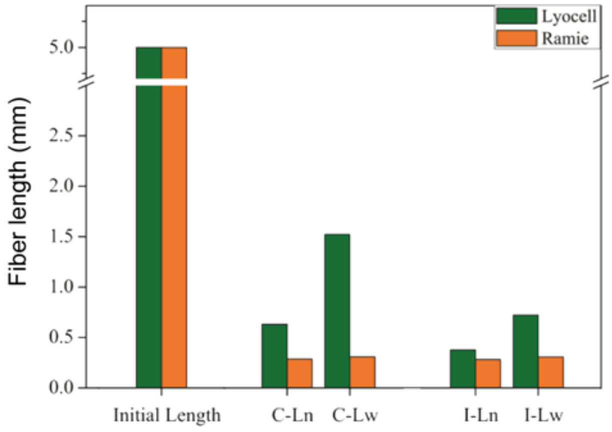

Figure 1. Average fiber lengths of Lyocell and ramie in PLA-composites after compounding and injection molding (with an initial length of 5 mm).

Figure 2 shows the distributions of fiber length (L_n) and diameter of Lyocell and ramie in composites after injection molding. It can be seen that the fiber length distribution of Lyocell fiber (L5) was wider than that of ramie (R5), and

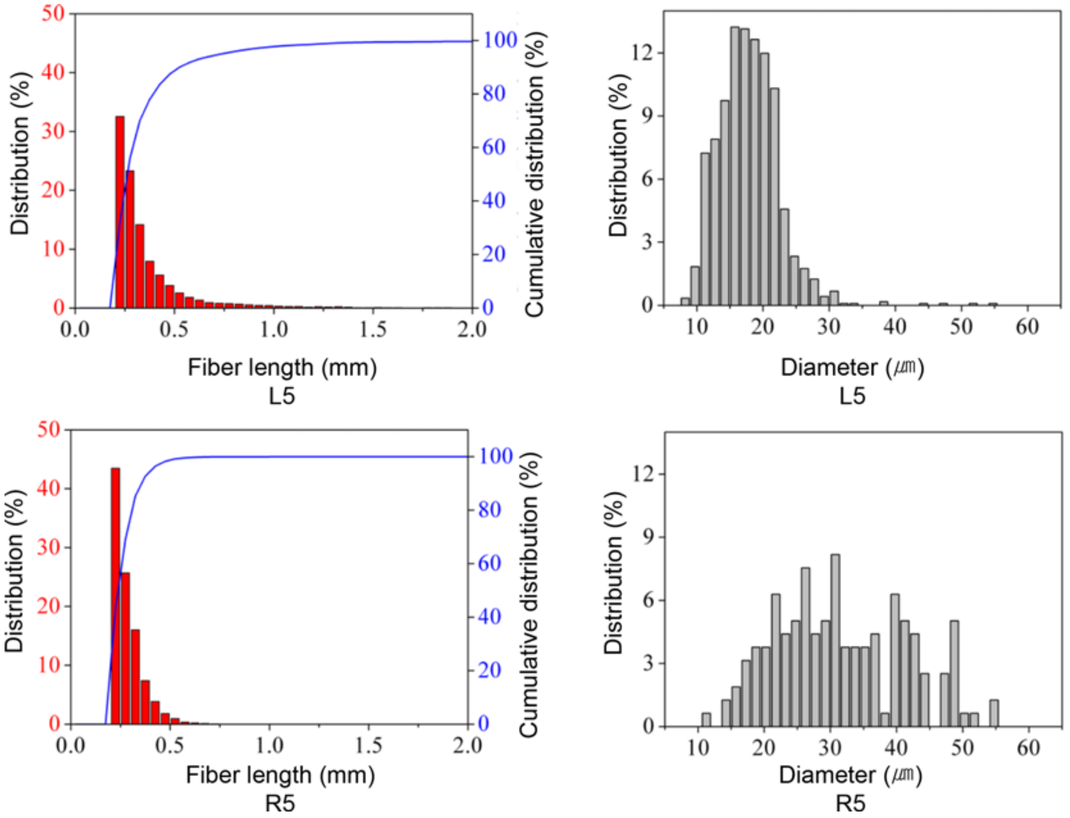

Figure 2. Distributions of fiber length (L_n) and diameter of Lyocell and ramie in composites after injection molding (with an initial length of 5 mm).

Table 1. Fiber dimensions of two types of fibers extracted from PLA composites reinforced with different initial lengths fibers

Fibers with varying initial length*	Average fiber length (mm)		Average fiber diameter (μm)	Aspect ratio of fiber		Content of small fiber (%)	
	L_n	L_w	D	L_n/D	L_w/D	F_n	F_w
L1	0.363	0.690	18.75	19.36	36.80	21.90	11.15
L5	0.377	0.722	15.75	23.94	45.84	24.82	13.15
L10	0.385	0.779	17.25	22.32	45.16	29.76	14.88
L15	0.381	0.839	18.75	20.32	44.75	33.28	17.85
R1	0.256	0.282	25.50	10.04	11.06	55.97	37.92
R5	0.282	0.308	26.25	10.74	11.73	67.25	47.81
R10	0.306	0.404	21.75	14.07	18.57	63.95	41.44
R15	0.356	0.524	23.25	15.31	22.54	57.78	34.70

*L and R mean the Lyocell and ramie fiber, respectively. L1 and R1 mean the Lyocell and ramie fiber samples with an initial fiber length of 1 mm, respectively. Others can be done as the same manner.

many longer Lyocell fibers were retained in composites after injection molding, which is consistent with the results of Figure 1. Moreover, it can be found that Lyocell fiber showed a smaller diameter and narrower width distribution than ramie. The longer average fiber length and smaller diameter of Lyocell fiber resulted in a larger aspect ratio than ramie fiber. Thus, the Lyocell fibers may be more conductive to reinforce PLA, compared with ramie fibers.

Table 1 shows the fiber dimensions of two types of fibers extracted from the PLA composites reinforced with different initial lengths fibers. It was found from Table 1 that the longer the initial fiber length, the more severely the fiber length declined after processing, i.e. the longer initial fiber was easier to suffer more damage during the process. Moreover, the average fiber lengths (L_n and L_w) of these two reinforcing fibers were both increased slightly when their initial lengths increased from 1 mm to 15 mm. Most importantly, the average length of Lyocell fiber was longer than that of the corresponding ramie fiber, regardless of the initial length of the fiber.

Moreover, it can be found from Table 1 that the average fiber diameters of two types of fibers extracted from PLA composites varied in different degrees with different initial fiber lengths, and Lyocell fibers showed the smaller average fiber diameters than those of ramie fibers under the same conditions. Consequently, the larger aspect ratios were obtained for Lyocell fibers under the same initial fiber lengths.

In addition, Table 1 also provided the content of small fiber, F_n and F_w. The small fiber refers to small particle with length < 0.2 mm. The content of small fiber is an important factor in the analysis of the relationship between the fiber dimension and mechanical properties of composites due to its smaller fiber aspect ratio. It can be found obviously that both F_n and F_w of Lyocell fibers were less than those of ramie fibers in all varying initial length cases, which confirms further that ramie fiber was more susceptible to break during the process. Moreover, the small fiber content of Lyocell fibers increased with the increase of the initial fiber length, whereas the ramie fibers showed a highest small fiber content when the initial fiber length of ramie was 5 mm. The reason for this difference was that ramie fiber, as a natural cellulose fiber, was not uniform in the diameter and the fiber strength, compared to the high uniformity of Lyocell fiber. Therefore, the small fiber content of ramie fiber varied from Lyocell fiber. It was noted that the F_n of Lyocell fibers was only 20-30 %, while the F_n of ramie fibers was more than 50 %. Such high level of small fiber content will probably affect the performance of the ramie

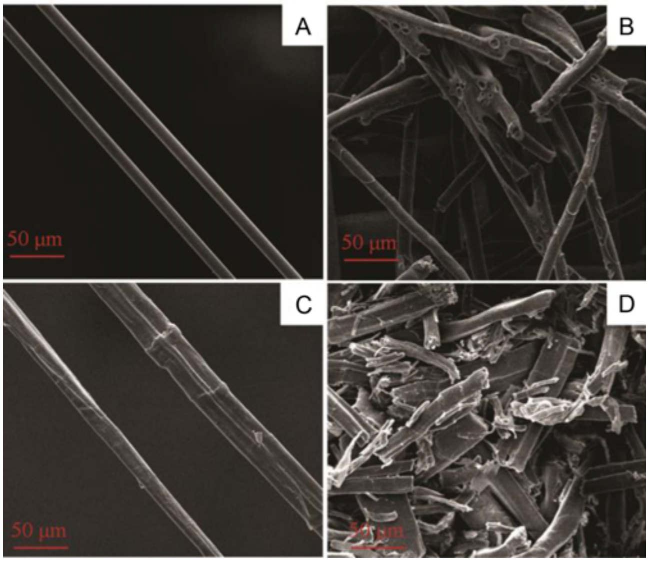

Figure 3. SEM images of the initial fibers and extracted fibers (initial length 5 mm) (A) initial Lyocell fiber, (B) the extracted Lyocell fiber from the Lyocell/PLA composite, (C) initial ramie fiber, and (D) the extracted ramie fiber from the ramie/PLA composite.

fiber reinforced PLA composites.

Figure 3 shows SEM images of the initial fibers and the extracted fibers from PLA composites. It can be found that the initial ramie fibers were nonuniform not only in length, but also in fiber diameter, compared with Lyocell fibers. On the whole, it seemed to be more “fragmented” for the exacted ramie fibers, which resulted in the higher content of small fiber. By contrast, the extracted Lyocell fibers showed the longer length and a wider distribution of lengths, therefore the small fiber content was lower for Lyocell fibers. This was coincided with the results in Table 1.

The Effect of Fiber Dimension and Its Distribution on the Mechanical Properties of Cellulose Fiber Reinforced PLA Composites

Figure 4 shows SEM images of fractured surfaces of Lyocell/PLA composite and ramie/PLA composite. As can be seen in Figure 4(a), a part of the Lyocell fibers were pulled out from the PLA matrix, and the corresponding holes can be visible in the fractured surface of composite. Furthermore, it can be seen that the surfaces of the pulled out fibers were clean, which indicated that there was no good adhesion between the Lyocell fibers and the PLA matrix. By contrast, the better interfacial adhesion can be found between ramie fibers and PLA matrix, and ramie/PLA composite showed shorter fiber pull-outs than Lyocell/PLA composites. The similar results can also be found in the literature for the PLA/Cordenka and PLA/flax composites [24].

Figure 5 shows the tensile properties of PLA composites reinforced by Lyocell and ramie fibers with varying initial fiber lengths. Obviously, all reinforcing fibers exhibited the enhancement on the tensile strength and tensile modulus of PLA matrix. Although the mechanical properties of Lyocell fibers were lower than those of ramie fibers and the weak interfacial adhesion was found for Lyocell/PLA composite, the Lyocell/PLA composites showed the greater increase in tensile strength and tensile modulus than the ramie/PLA composites, which may be related to the Lyocell fiber dimension and its distribution in the composites. When the initial fiber length was 5 mm, the tensile strength and tensile modulus of Lyocell/PLA composites were increased by 5.42 % and 8.65 % than ramie/PLA composites, respectively.

Some studies suggested that fiber length of reinforcing fiber is an important factor in the improvement of tensile properties of their composites [25]. It was considered that longer fiber had beneficial effect on tensile properties of composites, because the fibers with lengths longer than critical fiber length would have a larger contribution to the composite strength. Moreover, the small fiber content played an important role in the mechanical properties of composites due to its smaller aspect ratio. The higher small fiber content

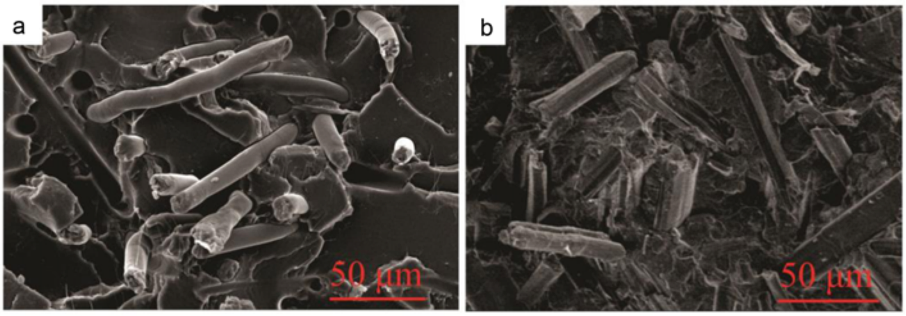

Figure 4. SEM images of fractured surfaces of Lyocell/PLA composite (a) and ramie/PLA composite (b).

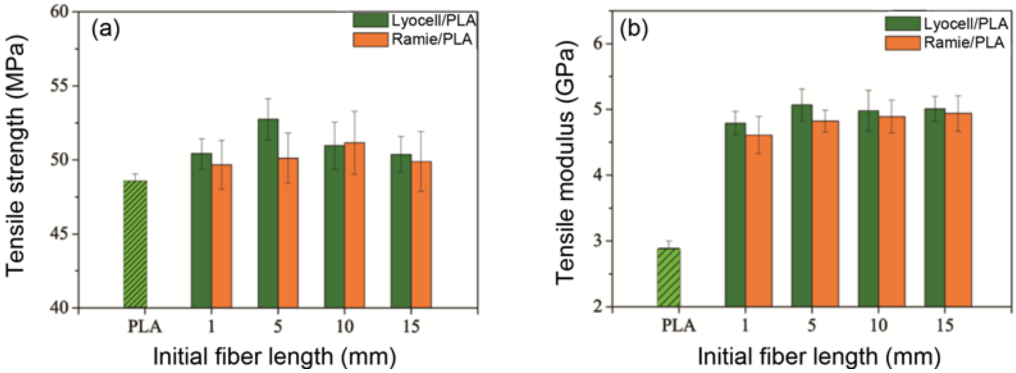

Figure 5. Tensile strength (a) and tensile modulus (b) of PLA composites reinforced by Lyocell and ramie fibers with varying initial fiber lengths.

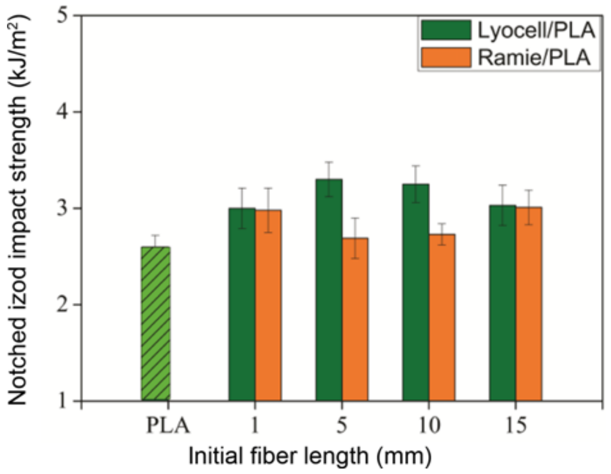

Figure 6. Impact strength of PLA composites reinforced by Lyocell and ramie fibers with varying initial fiber lengths.

in the composites, the worse mechanical properties of composites showed. The results of Table 1 showed that Lyocell fibers in the composites exhibited longer average fiber length and lower small fiber content than ramie fiber, which resulted in better tensile properties of Lyocell/PLA composites. In addition, when the initial length of Lyocell fiber was 5 mm, Lyocell/PLA composites showed the best tensile properties due to the highest aspect ratio of fiber (Table 1). For the fiber reinforced PLA composites, the larger fiber aspect ratio means the higher specific surface area of fiber, which resulted in better interfacial bond between the fibers and matrix, and then the better fiber reinforcement effect could be obtained.

Figure 6 showed the impact strength of PLA composites reinforced by ramie and Lyocell fibers with varying initial fiber lengths. It can be seen that Lyocell/PLA composite showed higher impact strength than ramie/PLA composite in the cases of the initial fiber length of 5 mm and 10 mm. Whereas if the initial fiber length was 1 mm or 15 mm, Lyocell/PLA composite and ramie/PLA composite showed the similar impact strength. The impact strength of composite was affected by many factors. It was reported that an higher elongation of reinforcing fibers can lead to a better improvement in the impact strength of the composites [26]. Compared with the elongation of ramie (3.6-3.8 %) [27], Lyocell fibers have higher elongation at 8-10 % [28]. It is one of the reasons of higher impact strength of Lyocell/PLA composite with the initial fiber length of 5 mm and 10 mm.

Furthermore, the aspect ratio of reinforcing fiber and small fiber content are important factors for the mechanical properties of fiber reinforced polymer composites. In this work, the aspect ratio of Lyocell fiber was higher than that of ramie fiber in the composites although the initial length of the two fibers was equivalent. Consequently, Lyocell fiber exhibited the better reinforcement on the tensile strength and impact strength for PLA composites. Compared with ramie fiber, Lyocell fiber had the lower content of small fiber (20-30 %, see Table 1) and achieved the greatest improvement on the mechanical properties of PLA composites especially in the impact strength. However, the high content of small fiber of ramie fiber (up to 50 %) was not conductive to the improvement in the impact strength of PLA composites because small fiber with lower aspect ratio will result in a decline of the stress transference and energy absorption. Too much small fiber may cause structure defect and decrease the mechanical properties of composite. Therefore, the larger aspect ratio and lower small fiber content for Lyocell fiber led to the better impact strength of Lyocell/PLA composites with the initial fiber length of 5 mm and 10 mm.

In addition, the various initial fiber lengths of reinforcing fibers also showed different effect on the impact strength of PLA composites. The impact strengths of Lyocell/PLA composites increased firstly and then decreased with the increasing initial length of Lyocell fiber. The highest impact strength for the Lyocell/PLA composite was found at the initial fiber length of 5 mm. However, the impact strength of ramie/PLA composites decreased firstly and then increased with the increasing initial fiber length. This tendency was consistent with the changes of small fiber content of ramie with the different initial fiber lengths (Table 1). The highest small fiber content for ramie occurred in the initial fiber length of 5mm, which resulted in the lowest impact strength of ramie/PLA composites.

In summary, Lyocell fiber exhibited the better improvement in tensile and impact properties of PLA composites compared with the ramie fiber, and the different mechanical characteristics of the PLA composites were attributed predominantly to the fiber aspect ratio and the content of small fiber in the composites. Moreover, it should be noted that the neat rami and Lyocell fibers are very variable in average diameter, and it is difficult to keep the same aspect ratio and fiber diameter of Lyocell and ramie. Therefore, the mechanical properties of composites can be changed to some extent if the fiber diameters were changed.

Conclusion

In this work, natural ramie fiber and regenerated Lyocell fibers were used to reinforce PLA materials, the fiber length was both reduced during compounding and injection molding processes. Lyocell fiber showed a longer fiber length, smaller diameter and narrower width distribution in composites, compared with ramie fiber, which resulted in a higher aspect ratio for Lyocell fiber. Moreover, the longer the initial fiber length, the more severe the length declined, regardless of the fiber type. For the same kind of fiber, the final fiber length increased slightly with the increasing of the initial fiber length.

In addition, the small fiber content of the reinforcing fiber was an important parameter in the analysis of the relationship between the fiber dimension and mechanical properties of composites. The small fiber showed negative effect on the mechanical properties of composites owing to its smaller fiber aspect ratio. The higher content of small fiber of ramie reduced the tensile and impact properties of composites. By comparison, Lyocell fiber exhibited better reinforcement for PLA due to the larger aspect ratio and lower small fiber content.

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