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# Study of contents ratio of cellulose, hemicellulose and lignin on the mechanical properties of sisal fibers reinforced polylactic acid (PLA) composites

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**Abstract.** Untreated sisal fibers (USFs) with the highest tensile strength and alkali-treated SFs (ASFs) with the maximum interfacial strength with polylactic acid (PLA) matrix were mixed to make hybrid SFs (HSFs) according to different mass ratios and then compounded with PLA to prepare PLA/HSF composites. It was found that PLA/HSFs had the best mechanical property when the mass ratio of USFs and ASFs was 4:6. and the content of cellulose, hemicellulose and lignin at this moment were 71.4%、16.6% and 12.0% respectively.

## 1. Introduction

The interest of using natural fibers as reinforcement in polymer composites has increased in recent years due to their lightweight, non-toxicity, low cost, and biodegradable properties. <sup>[1-2]</sup> Natural fibers consist of cellulose, hemicellulose and lignin. They can be considered as composites of hollow cellulose fibrils held together by a lignin and hemicellulose matrix. <sup>[4]</sup> Lignin can be considered as a structural supporting material in natural fibers. In our previous work, SFs were alkali treated with different time to prepare ASF samples. The changes of chemical compositions of SFs were investigated, and their influences on the microstructure, tensile strengths of SFs and the interfacial strength of SFs and PLA were studied. It was found that USF had the best tensile strength and ASF1h (be alkali-treated for 1h) had the maximum interfacial strength with the PLA matrix. It could also be observed that suitable treatment intensity can reserve appropriate content of lignin and hemicelluloses, which had some positive effect on the performance of SF and SF-reinforced PLA composites. <sup>[6]</sup> Therefore, USFs was mixed with ASF1h according to different mass ratios to make HSFs in this work. Then different HSFs were compounded with PLA to make PLA/HSF composites. The aim is to obtain the PLA/HSFs composite with the best mechanical properties, the optimal hybrid ratio of USFs and ASF1h and the contents of cellulose, hemicellulose and lignin at this time.

## 2. Experiment

### 2.1. Materials

PLA polymer 3051D, Nature Works™, was provided by Shenzhen Bright China Industrial Company (China). SF bundles (GB/T 15031-94) were provided by Guangxi Sisal Company of Guangxi Province, China. 10% w/v aqueous solution of NaOH was commercial product.



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## 2.2. Preparation of USFs and ASF1h

Firstly, SF bundles were cut short and submerged into 10% w/v aqueous solution of NaOH for 1h. Then alkali-treated SFs (ASFs) were washed with distilled water and dried in the open air for 48h. Finally, the USFs and ASF1h were dried in an oven at 80°C for 4h.

## 2.3. Preparation of PLA/HSF composites

Firstly, PLA, ASFs and USFs were dried at 80°C for 4h. Then, USFs were manually mixed with ASF1h to fabricate eleven kinds of HSFs, which mass ratios of USF and ASF1h were listed in Table 1. After that, PLA were melted and compounded with different HSFs respectively to make into eleven kinds of PLA/HSF composites according to a mass ratio of 4: 1 using a two-roll plastic mill. At last, they were compressed into sheets with a thickness of 1 mm or 4 mm by a hot press and cut into standard samples (GB/T 1039-1992) for mechanical tests. Five specimens were used for each test.

**Table 1.** Ratios of USFs and ASFs of PLA/HSFs composites

	C1	C2	C3	C4	C5	C6	C7	C8	C9	C10	C11
Mass ratio of USF to ASF1h	0:10	1:9	2:8	3:7	4:6	5:5	6:4	7:3	8:2	9:1	10:0

## 2.4. Scanning electron microscopy (SEM)

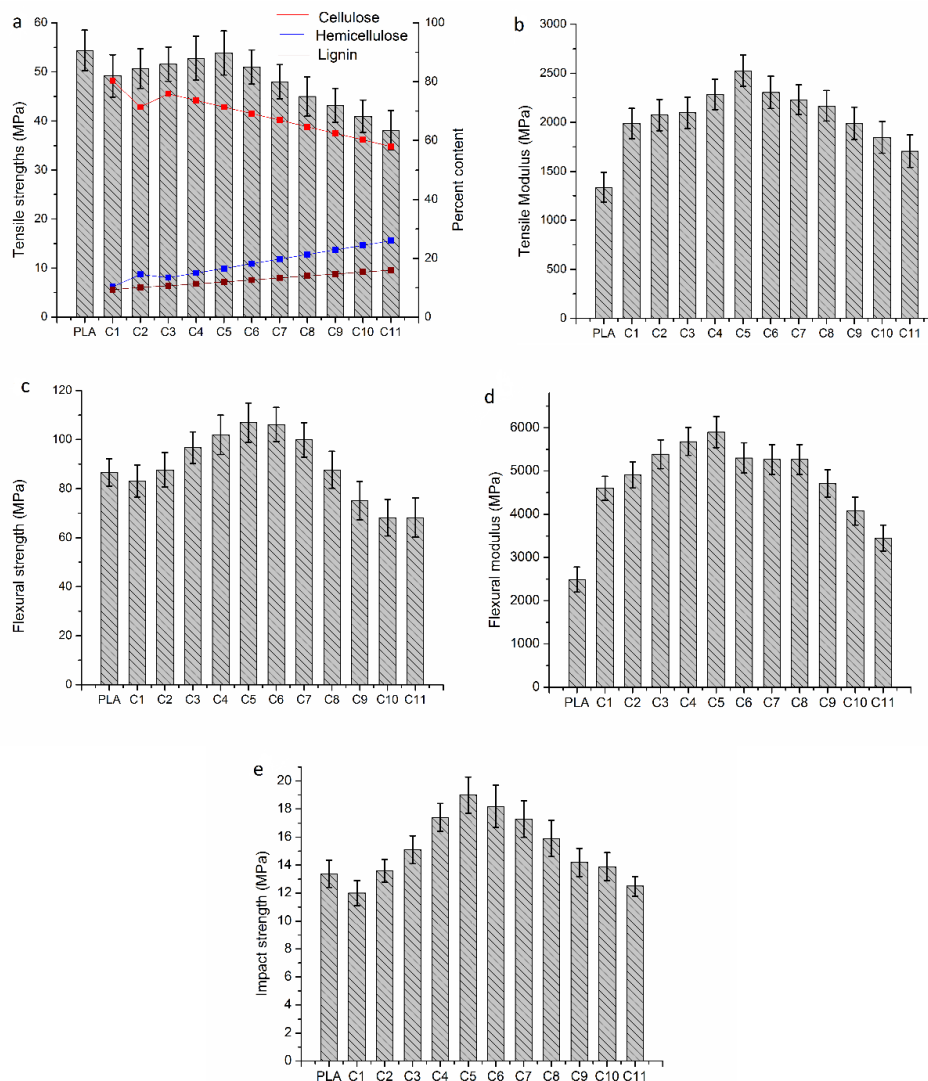
The specimens were fractured in liquid nitrogen to obtain unaffected fractured surfaces. The fractured surfaces of the samples were then sprayed with gold powder to prepare for scanning electron microscopic (SEM) observation by a SEM (S-3700N, Hitachi, Tokyo, Japan).

## 3. Results and discussion

### 3.1. Mechanical property of SFs reinforced PLA composites with different SFs

In our previous work, it can be found that the tensile strength of USF was the highest while the interfacial bonding between ASF1h and PLA was the strongest. It was generally considered that the mechanical properties of fiber-reinforced composites primarily depends on three factors: (1) the strength and modulus of the fibers, (2) the strength and toughness of the matrix, and (3) the effectiveness of fiber–matrix bonding in transferring stress across the interface. <sup>[5]</sup> If the processing parameters are the same, the performance of PLA as a matrix is constant. Therefore, the mechanical properties of PLA composites mainly depend on the properties of fibers and the interface of fiber and matrix. Therefore, USF was mixed with SF with ASF1h according to different mass ratios to make HSFs. The aim is to make cellulose, hemicellulose and lignin play their roles adequately and thus prepare the PLA/HSFs composites with the best mechanical properties.

Figure 1 showed the mechanical properties of PLA, PLA/USFs, PLA/ASFs and PLA/HSF composites. It was found that the mechanical properties of PLA/USFs composite and PLA/ASFs composite were worse than those of pure PLA, which was mainly because of the bad interfacial compatibility of PLA and USFs and the poor mechanical property of ASFs.



**Figure 1.** Mechanical properties of PLA and PLA composites

However, the addition of HSFs could improve the flexural and impact properties of PLA effectively. For example, the flexural strength, flexural modulus and impact strength of C5 composite were 23.7, 137.3 and 41.7% higher than those of pure PLA. Comparing PLA/HSFs with PLA/USFs and PLA/ASFs, it can be observed that the strength and Young's modulus of PLA/HSFs were better than those of PLA/USFs and PLA/ASFs except for C9 and C10 composites. For example, the tensile strength and modulus of C5 were 53.9MPa and 2526.7MPa, which were 41.5% and 48.1% higher than those of PLA/USFs, and 10.0% and 27.1% higher than those of PLA/ASFs. The impact strength of C5 was 19 KJ/m<sup>2</sup>, which was 48% and 58.3% higher than that of PLA/USFs and PLA/ASFs.

Therefore, it can be concluded that HSFs could strengthen PLA matrix more effectively than USFs and ASFs. There may be the following several reasons. Firstly, the interface bonding of PLA and ASFs was better and the mechanical property of USFs was higher. When USFs and ASFs were compounded, they could play their synergistic effect and strengthened PLA matrix effectively. Secondly, appropriate contents of hemicellulose and lignin are good for SFs and the interface bonding of PLA and SFs, which had been proved by above experiment. Thirdly, hemicellulose and lignin can

make SFs well disperse in the polymer matrix and hence enhance the dispersion uniformity of HSFs<sup>[6]</sup> At last, some substance existing in the USFs can induce the formation of  $\beta$ -crystal, which had been proved in our previous work.<sup>[7]</sup>  $\beta$ -crystal higher impact strength and better thermal properties than  $\alpha$ -crystal, which may especially explain why PLA/HSFs and PLA/USFs had better impact strength than PLA/ASFs.

### 3.2. Influence of hybrid ratios of USF and ASF on the strength of PLA/SF composites

The mechanical properties of PLA/HSF composites changed with the hybrid ratios of USFs and ASF1h. When the mass ratio of USFs and ASF1h was 0:10 and 1:9, the mechanical properties of PLA/HSFs composites were relatively worse, which was mainly due to the higher content of ASF1h. As the content of USF increasing, the mechanical properties of PLA/HSFs composites became better and better. When the mass ratio of USFs and ASF1h was 4:6, the mechanical properties of PLA/HSFs composite was the best and the content of cellulose, hemicellulose and lignin at this moment were 71.4%、16.6% and 12.0% respectively. However, with the content of USFs further increasing, the mechanical properties of PLA/HSFs composites reduced. Especially with the USFs content increasing, the reduction amplitude in mechanical properties increased. It presented that the interfacial strength was important for the mechanical properties of fiber reinforced PLA composite. When the interface bonding is too weak, it is possibly difficult for SFs to play their reinforcing roles.

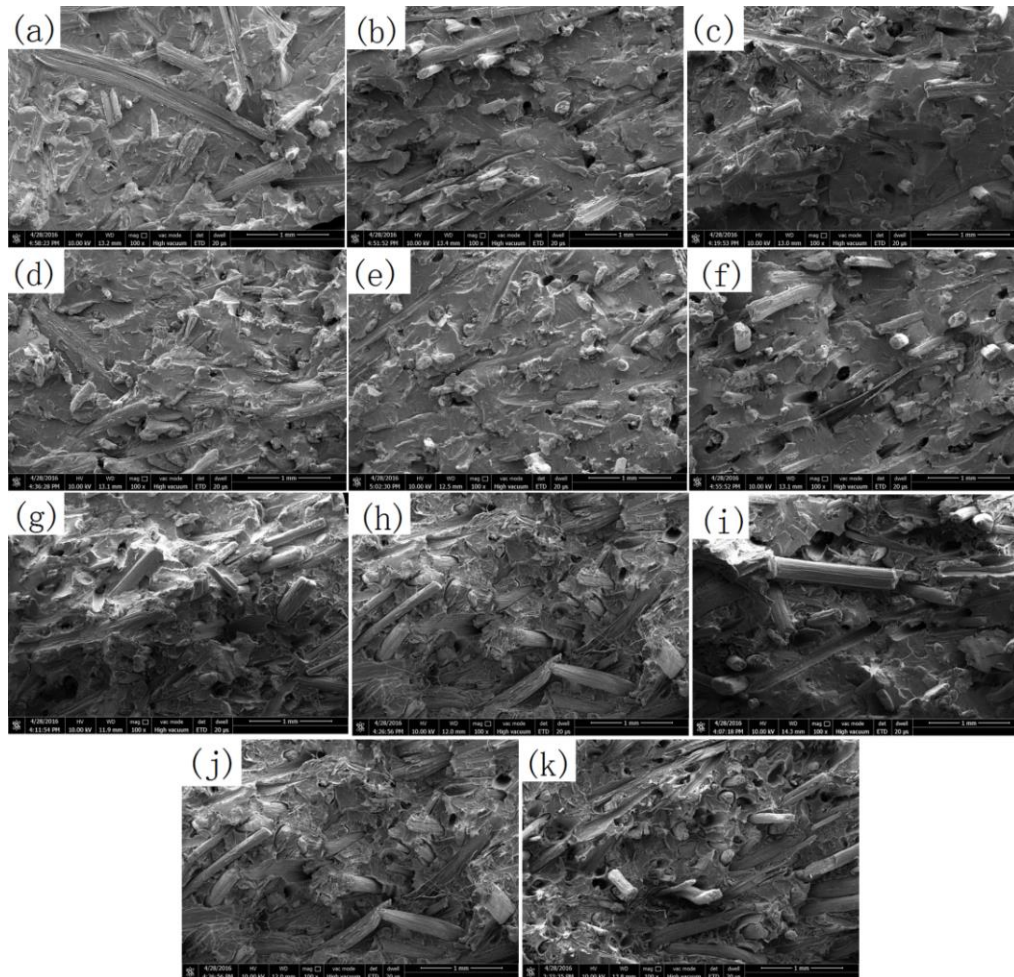
From the above investigation and analysis, it can be found there existed doubts about the pretreatment of natural fibers. Although pretreatment promoted the interface bonding of fibers and matrix, it also caused the loss of the mechanical properties of fibers. By mixing untreated fibers with pretreated fibers to prepare hybrid fibers and fill them in the polymer matrix, the contradiction can be preferably solved and then the reinforcement of fibers for matrix can be achieved effectively. Therefore, different pretreatments can be employed to prepare natural fibers with different surface morphology, compositions and properties. If these natural fibers with different properties are filled in the polymer matrix, composites with excellent properties will perhaps obtained through synergistic effect of different natural fibers.

### 3.3. SEM analysis

Figure 2 was the SEM graphs of the impact sections of eleven PLA composites obtained under magnification of 100X. It can be found from Figure 2(a) that the impact section of PLA/ASF composite was smooth and fewer cavities were observed, which showed better interface bonding of PLA matrix and ASF.

Meanwhile, many torn SFs were visible in Figure 2(a). It was mainly because that the strength of SF declined obviously after alkali treatment and better bonding between ASF and matrix made SFs absorb more impact energy. It was also found in the following Figure 2(b-k) that pulled-out SFs became more and more with the USF content increasing. This phenomenon is more obvious in Figure 2(g-k). For example, in the 5\*4 mm<sup>2</sup> range, the number of holes in Figure 2(a) is 10 and those in Figure 2(g) and Figure 2(k) reached 26 and 32, respectively. Those presented that the interface bonding became weaker with the USF content increasing. Moreover, it can be observed that lengths of pulled-out SFs in Figure 2(f-k) were longer than those in Figure 2(a-e) obviously. In combination with experiment conditions, the length of SFs was 1cm in the beginning, and became shorter and shorter after alkali treatment and processing. It is mainly because alkali treatment removed some compositions, the

diameter and length of sisal fiber were greatly reduced to about 30% of its origin. The fibers also became brittle. Then in the process of mixing with the matrix, the fibers broke and get shorter after turning and twisting. Thus, it may be concluded that a large number of SFs are pulled out directly and didn't bear external force when composites suffering from external force. These stated that fibers can't play an effective role as reinforcement if the interface bonding of fiber and matrix is too weak.



**Figure 2.** SEM graphs of eleven PLA composites. (a): C1; (b): C2; (c): C3; (d): C4; (e): C5; (f): C6; (g): C7; (h): C8; (i): C9; (j): C10; (k): C11.

#### 4. Conclusions

It was found the reinforced effect of HSFs for PLA composites is better than that of USFs and ASFs in this work. The mechanical properties of PLA/HSFs were better than those of PLA/USFs and PLA/ASFs in this work. PLA/HSFs composite could reach the best mechanical properties when the hybridizing ratio of USFs and ASF1h was 4: 6, and the content of cellulose, hemicellulose and lignin at this moment were 71.4%、16.6% and 12.0% respectively. Based on the above analysis, it can be found the reason why treated fibers are difficult to strengthen polymer composites effectively. Though various pretreatments improve the compatibility between fiber and matrix, they can also decline the mechanical properties of fibers themselves. By using hybrid fibers, the high strength of untreated fibers and the good compatibility of treated fibers can play a synergistic role to strength the polymer

matrix effectively. Furthermore, we can use different pretreatments to obtain fibers with different morphology, composition and properties. Then these fibers can be mixed and filled into polymer matrix to prepare composites with high performance.

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