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Flax/Polylactic Acid (PLA) Core/Sheath Structural Composite Yarn Fabricated by Spindle Braiding Technique

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Abstract: The increasing demand for sustainable and environmentally friendly materials has driven research towards the development of green composites. In this work, the flax/polylactic acid (PLA) braided yarns were fabricated by braiding PLA filaments with 4 to 24 spindles on flax yarns. After curing at different temperatures (180 °C and 190 °C), the core/sheath structural flax/PLA composite yarns were manufactured. According to the results of the tensile test, the flax/PLA composite yarn with 4-spindle PLA yarns as a sheath layer and at a curing temperature of 180 °C reached the maximum elastic modulus of about (5.79 ± 0.65) GPa and the maximum tensile strength of about (162.17 ± 18.18) MPa. This flax/PLA composite yarn with good mechanical properties would be suitable for green composites in the automobile manufacturing industry and building materials.

Key words: flax; polylactic acid (PLA); braiding; composite yarn; tensile property

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0 Introduction

In recent years, there is a growing global emphasis on sustainable practices and environmentally friendly alternatives across various industries. This paradigm shift has prompted significant research and innovation in the development of materials that not only meet the performance requirements of modern applications but also adhere to ecological and ethical considerations. Among these materials, green composites have emerged as promising avenues for creating environmentally sustainable and high-performance materials^[1].

Green composites, also known as bio-composites or eco-composites, refer to composites composed of natural

fibers or reinforcements embedded within a biodegradable or recyclable matrix^[2]. These composites aim to combine the mechanical properties of traditional composites with the environmental benefits of using renewable resources. Commonly employed natural fibers include ramie^[3], jute, flax, hemp^[4], bamboo and kenaf, while biodegradable matrices often include polymers like polylactic acid (PLA), starch-based polymers and other bio-based resins^[5-7].

The development of green composites has become an important field of study and innovation in response to the growing need for sustainable materials^[8]. The combination of flax and PLA is driven by their complementary abilities to produce a biobased yarn with improved environmentally favorable properties^[9-11]. Flax fibers, derived from the flax plant, have notable tensile strengths and low environmental impact^[12-13]. PLA, a biopolymer, is derived from renewable resources with biodegradability and versatility^[13]. Current research focuses on understanding mechanical, thermal and morphological properties of flax/PLA composite yarns, including the effects of factors such as the fiber content, the fiber orientation and processing conditions on the final properties of the composite yarns^[14]. Gobi et al.^[15] indicated that the structure of flax/PLA composite yarns had the direct influence on the properties of the composites. For the braiding structure, the number of spindles is a crucial variable in the fabrication process. Different numbers of spindles can influence the orientation, the structure and mechanical properties of the yarn. Exploring various spindle braiding techniques provides valuable insights into optimizing the fabrication process and achieving the desired characteristics for green composite applications^[16-19].

This research aims to develop a flax/PLA biobased yarn with tailored mechanical and thermal properties suitable for green composites. It also investigates the

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influence of spindle numbers on the final yarn composition and performance. The developed flax/PLA biobased yarn might have the potential to find applications in industries such as automotive industry, construction and textiles, offering a sustainable alternative to other traditional materials without compromising on performance.

1 Materials and Methods

1.1 Materials

Flax yarns with a linear density of 200 tex purchased from Zhejiang Kingdom Linen Co., Ltd., (Zhejiang, China) were used as core yarns. PLA filaments with a linear density of 1 350 tex purchased from Smartwin International Group Co., Ltd., (Hongkong, China) were used as sheath yarns.

1.2 Preparation of flax/PLA braided yarns and composite yarns

PLA filaments underwent a twisting process and double ply, resulting in a final linear density of 2 700 tex. The twisting process was completed on the

spinning frame (Digital Spinning Testing Machine-DHU-X1, Shanghai, China). The twist factor of PLA filaments was 300 twists per meter. The machine speed was 6 500 r/min during the twisting process. Three braiding machines with 8 spindles, 12 spindles and 24 spindles (Shanghai Xianghai Strap Weaving Machinery Co., Ltd., Shanghai, China) were used to prepare five types of braided yarns with different spindle numbers: 4-spindle braided yarns, 6-spindle braided yarns, 8-spindle braided yarns, 12-spindle braided yarns and 24-spindle braided yarns.

Figure 1 (a) presents the braiding process of the flax/PLA braided yarns. Figure 1 (b) presents the curing process of flax/PLA braided yarns to manufacture flax/PLA composite yarns. These braided yarns were cured in an industrial oven (Shanghai Jinghong Experimental Equipment Co., Ltd., Shanghai, China) for 15 min and at the curing temperature of 180 °C and 190 °C, respectively^[20]. The flax/PLA braided yarn sample was fixed on metal bars, and then it was put in the curing oven for curing. Figure 1 (c) presents the flax/PLA composite yarn after the curing process.

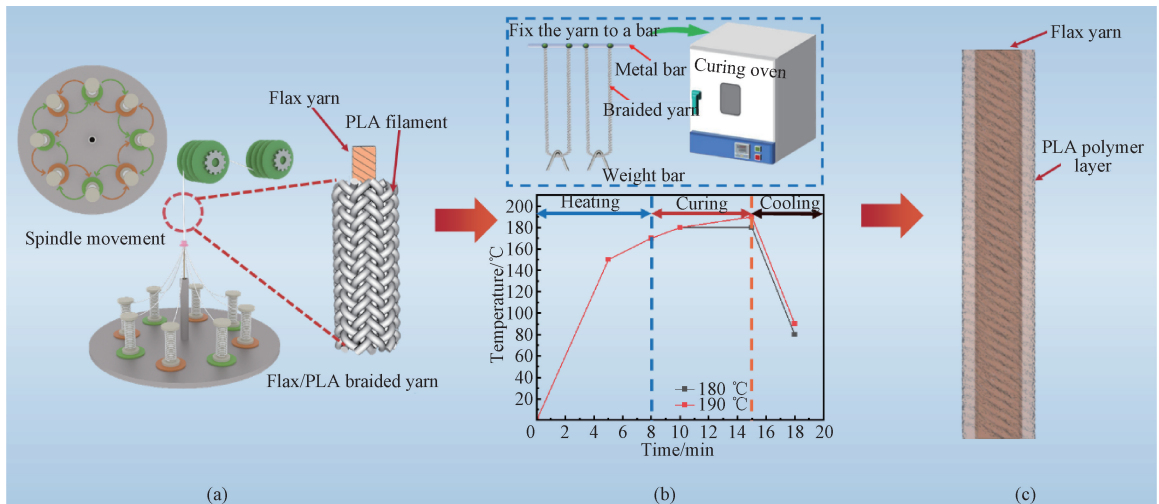
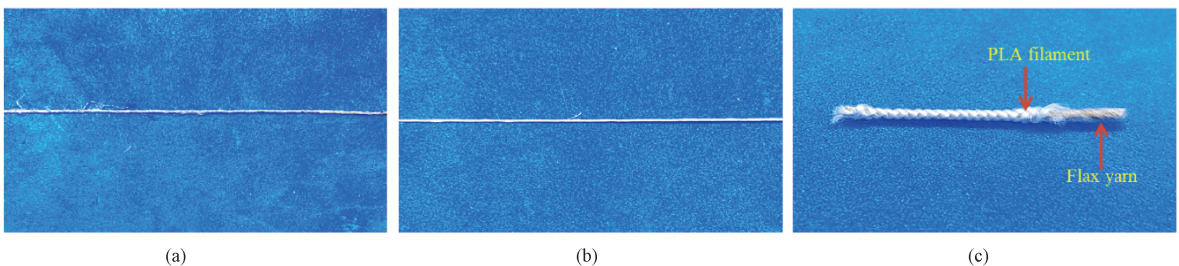


Fig. 1 Manufacturing process of flax/PLA composite yarn: (a) braiding process; (b) curing process; (c) structure of flax/PLA composite yarn

1.3 Yarn specification of flax/PLA braided yarns

The braiding angles of 4-spindle braided yarns, 6-spindle braided yarns, 8-spindle braided yarns, 12-spindle braided yarns and 24-spindle braided yarns were similar, around 45°. The morphologies of the flax/

PLA braided yarns are shown in Figs. 2 (a) – 2 (g). Mass fractions of flax and PLA, and the linear densities of flax/PLA braided yarns and composite yarns are shown in Figs. 2 (h) and 2 (i), respectively.



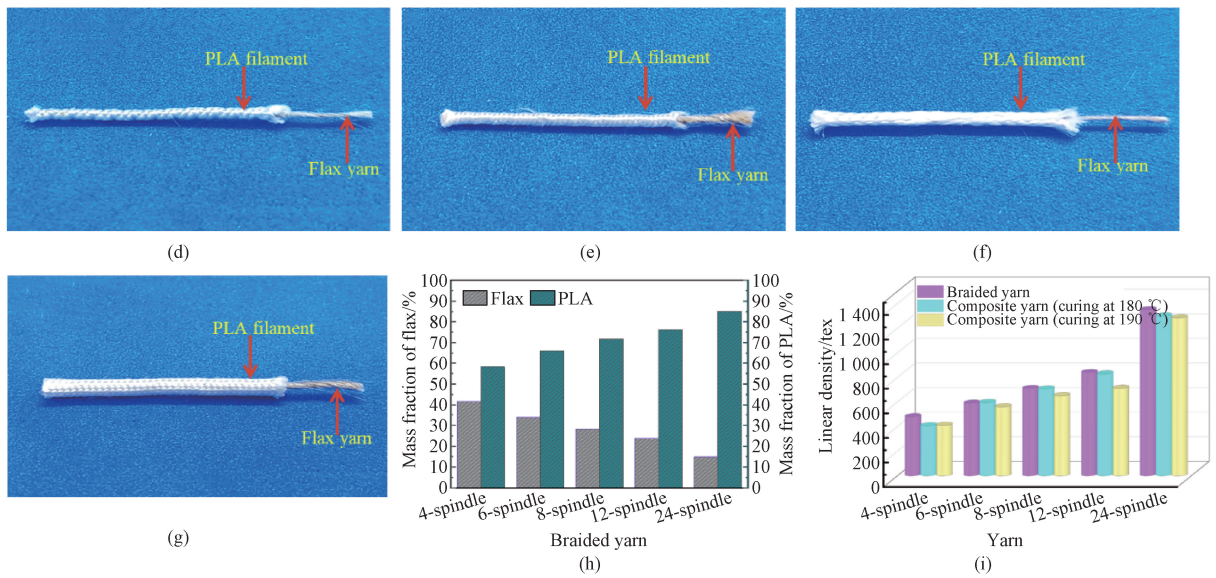


Fig. 2 Yarn morphologies and yarn specification: (a) flax yarn morphology; (b) PLA filament morphology; (c) 4-spindle braided yarn morphology; (d) 6-spindle braided yarn morphology; (e) 8-spindle braided yarn morphology; (f) 12-spindle braided yarn morphology; (g) 24-spindle braided yarn morphology; (h) mass fractions of flax and PLA; (i) linear densities of flax/PLA braided yarns and composite yarns

1.4 Thermal property test

The TGA 4000 thermogravimetric analyzer (PerkinElmer Enterprise Management Shanghai Co., Ltd., China) with a heating rate of 15 °C/min^[21] was used to test the thermal property of flax yarns and all braided and composite yarns (4-spindle composite yarns, 6-spindle composite yarns, 8-spindle composite yarns, 12-spindle composite yarns and 24-spindle composite yarns). The temperature range was 30–600 °C^[21].

1.5 Scanning electron microscope (SEM) test

The SEM (Hitachi SU8010, Japan) was used to study surface and cross-section morphologies of the flax/PLA braided yarns and composite yarns. Gold sputtering was applied to flax/PLA braided yarn and composite yarn specimens.

1.6 Tensile property test

The electronic single yarn strength meter (Laizhou Electron Instrument Co., Ltd., China) was used to test the tensile properties of the flax/PLA braided yarns and composite yarns. The tests were conducted according to the standard ASTM D 6775–13 (2017). The effective gauge length was 250 mm and the crosshead speed was 1000 mm/min. Each sample was tested five times, and the average result was taken^[22-25].

1.7 Flexural property test

Due to the complexity of the interwoven and the spiral structure of the braided yarns, many difficulties are brought to the theoretical research. In order to compare the flexural performance of different braided yarns and composite yarns, the cantilever beam method was used. If a braided yarn with a certain length is fixed on the bracket and is bent due to its own weight, the flexural

stiffness is^[22]

$$B = ql^4 / (3f), \quad (1)$$

where B is the flexural stiffness ($\text{cN} \cdot \text{cm}^2$); q is the uniform load generated by the self-weight of the braided yarn (cN/cm); l is the length of the braided yarn (cm); f is the deflection at both ends of the braided yarn (cm).

One end of the flax/PLA braided yarn was fixed on a vertical bracket with adhesive and the horizontal extension length was 25 cm. During the test, the yarn was kept horizontal with the help of the bracket, then the bracket was quickly removed, and the braided yarn sample drooped naturally due to its own weight. f was measured by the image software after standing for 30 min and the braided yarn was weighted on the electronic balance.

2 Results and Discussion

2.1 Morphologies and diameters of flax/PLA braided yarns and composite yarns

SEM images of yarns are shown in Figs. 3(a)–3(f). In Fig. 3(f), the flax yarn surface is not so smooth with fibers pulling out. In Fig. 3(a), the flax yarn is fully covered by PLA filaments, after curing at 180 °C, PLA filaments melt and the smooth surface is observed. The diameters of all flax/PLA braided yarns decrease after curing. For the 4-spindle braided yarn, the diameter decreases 26.47% and 26.57% after curing at 180 °C and 190 °C, respectively. For the 6-spindle braided yarn, the diameter decreases 34.15% and 34.67% after curing at 180 °C and 190 °C, respectively. For the 8-spindle braided yarn, the diameter decreases 25.72% and 14.99% after curing at 180 °C and 190 °C, respectively. For the 12-

spindle braided yarn, the diameter decreases 27.92% and 30.45% after curing at 180 °C and 190 °C, respectively. For the 24-spindle braided yarn, the diameter decreases

46.95% and 43.31% after curing at 180 °C and 190 °C, respectively. Notably, the 24-spindle braided yarn experiences the most significant diameter decrease.

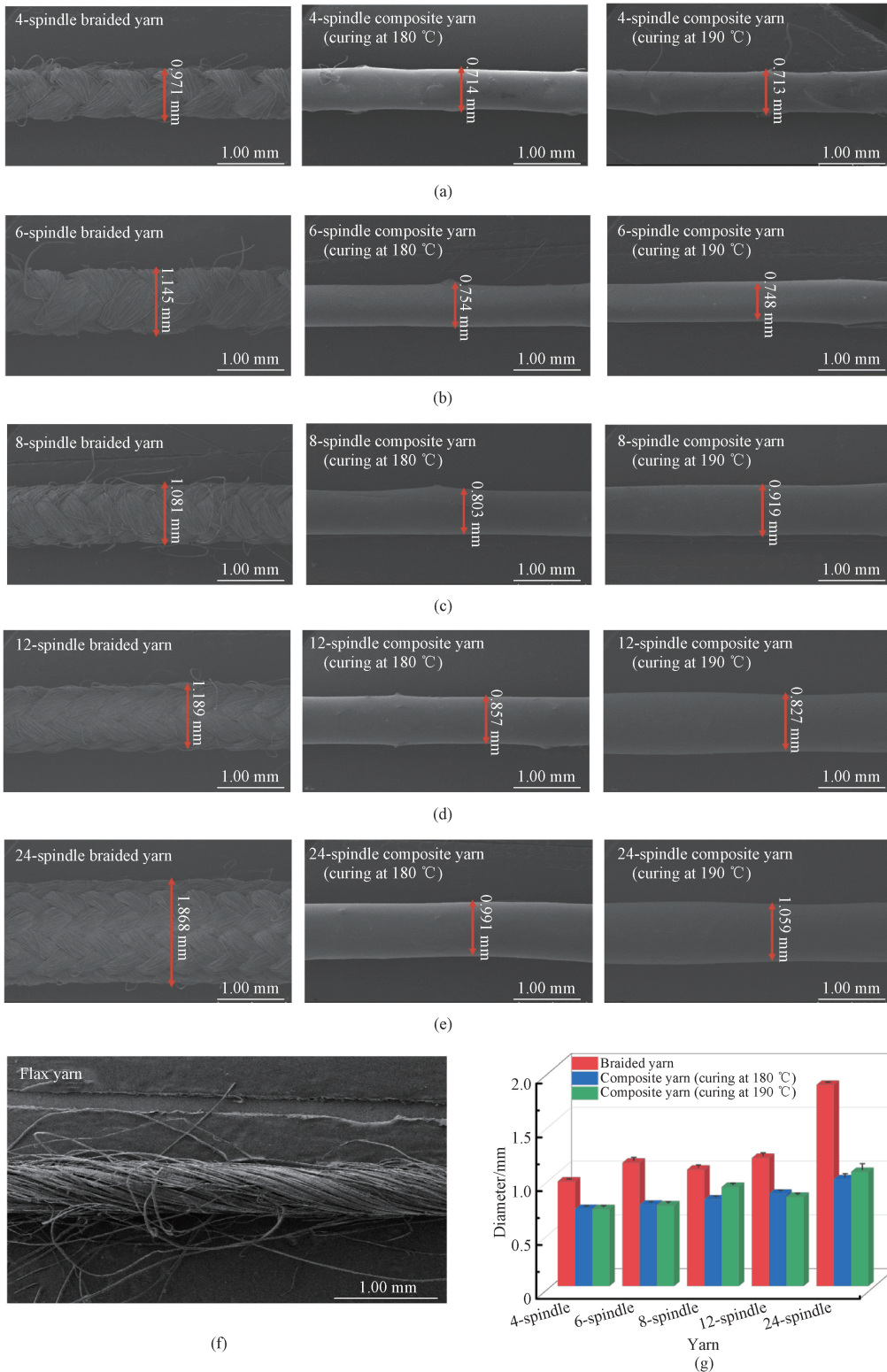


Fig. 3 Surface morphologies and diameters of braided yarns and composite yarns; (a) 4-spindle yarn; (b) 6-spindle yarn; (c) 8-spindle yarn; (d) 12-spindle yarn; (e) 24-spindle yarn; (f) flax yarn; (g) diameter histogram

PLA, being a thermoplastic polymer, undergoes thermal shrinkage when subjected to the elevated temperature during the curing process. As PLA undergoes thermal shrinkage, the yarns compress in sizes, contributing to the overall decrease in the diameter from 1.868 mm to 0.991 mm. The more PLA mass fraction (85.19%) in the braided yarn, the more thermal shrinkage. Natural fibers like flax fibers can undergo changes in their structure and properties during the curing process. Flax fibers may undergo activation and subsequent relaxation when exposed to heat. This activation and relaxation process can lead to a reorganization of the flax fibers, contributing to a reduction in the yarn diameter. The specific curing temperature and time can significantly influence the extent of shrinkage. The curing process aims to set the structure of the composite, and as a consequence, it may cause the materials to compact and contract. A densely braided structure may experience more compacting during curing, resulting in a more substantial decrease in the diameter from 1.868 mm to 0.991 mm.

2.2 Thermal properties of flax/PLA composite yarns

As shown in Fig. 4, the sample mass is slightly reduced due to the removal of water in the low temperature range. The initial thermal degradation of all samples occurs between 260 °C and 275 °C. This degradation can be attributed to the thermal degradation of pectin and hemicellulose of flax fibers. This initial degradation phase signifies the start of degradation of organic components within the flax yarns, leading to a loss of structural integrity and masses. The second major degradation occurs at around 300–400 °C, and PLA, cellulose and hemicellulose are rapidly degraded. Above 405–410 °C, the mass loss rate is low again, and the thermal degradation is mainly attributed to lignin degradation and carbonization of pyrolysis products. When the temperature reaches more than 500 °C, the curves of all samples tend to be gentle. When the temperature exceeds 400 °C, PLA is completely degraded and the lignin of flax fibers is gradually degraded. Therefore, the final residual mass rate of composite yarns gradually decreases with the increase of spindle numbers in the Fig. 4. At 500 °C, the residual mass rate of 24-spindle composite yarns is the least.

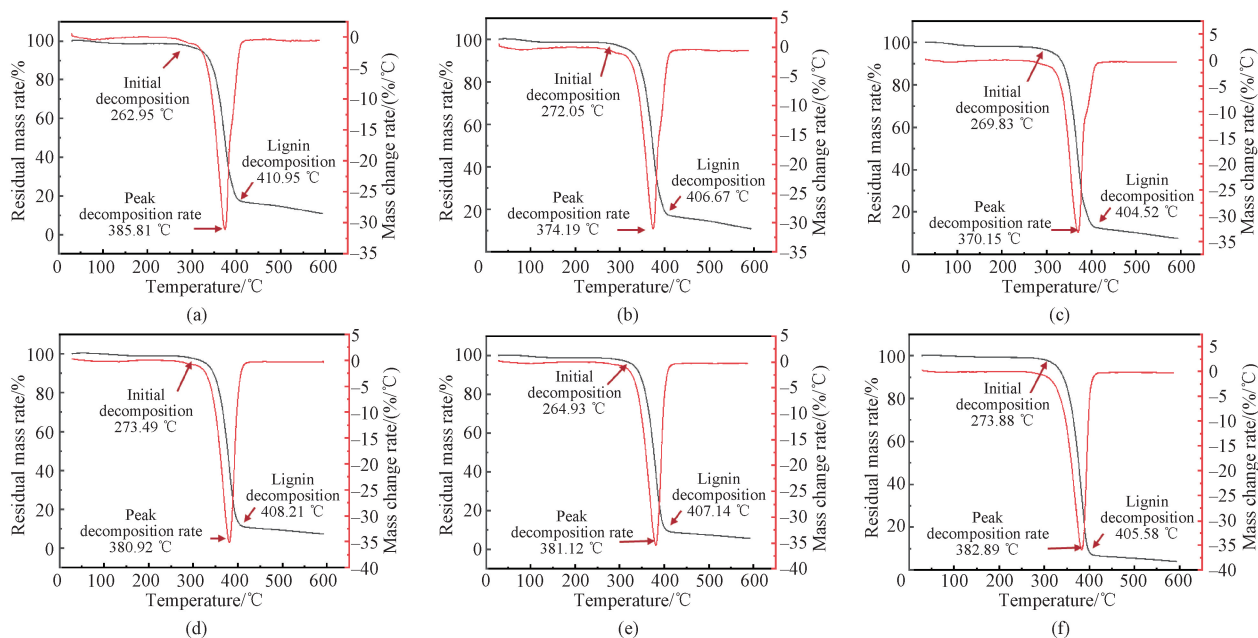


Fig. 4 Thermal analyses of flax yarn and flax/PLA composite yarns curing at 180 °C: (a) flax yarn; (b) 4-spindle composite yarn; (c) 6-spindle composite yarn; (d) 8-spindle composite yarn; (e) 12-spindle composite yarn; (f) 24-spindle composite yarn

2.3 SEM analyses of flax/PLA composite yarns

Figure 5 shows the cross section of flax/PLA composite yarns. In Fig. 5(a), there is no obvious void inside the flax/PLA composite yarns, and there is less PLA in the outer surface. In Fig. 5(b), there is more void and pores between flax fibers and PLA matrices, and there is more PLA in the outer surface. It is demonstrated that the porosities inside the flax/PLA composite yarn is more due to the increase of the PLA mass fraction and the

curing temperature. It should be noted that the 24-spindle composite yarn has a heterogeneous flax fiber contribution as well as rather large pores and empty area. Poor adhesion between flax fibers and PLA matrices as well as the increasing PLA content on the outside surface is shown in Fig. 5 (b). The voids between flax fibers and PLA matrices are obvious defects that will directly influence the mechanical properties of the flax/PLA composite yarns.

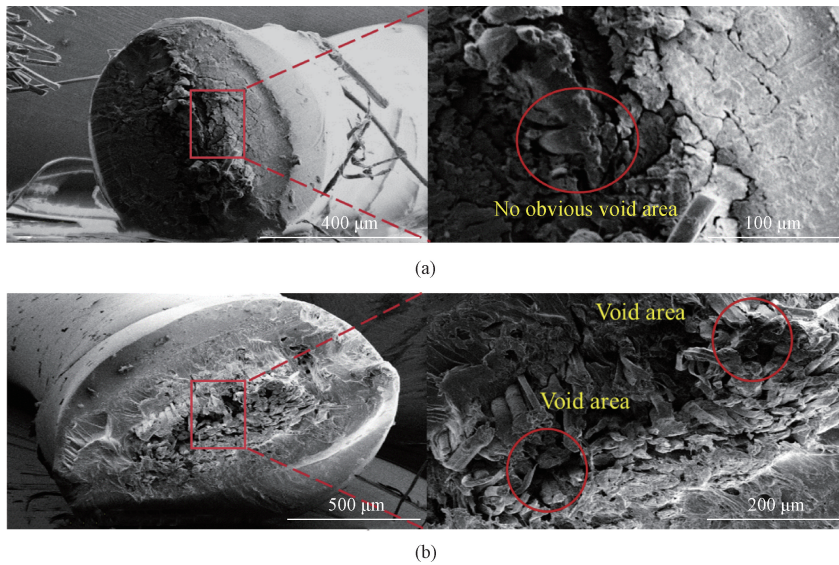


Fig. 5 Cross section of flax/PLA composite yarns: (a) 4-spindle yarn (curing at 180 °C) (b) 24-spindle yarn (curing at 190 °C)

2.4 Mechanical properties of flax/PLA braided yarns and composite yarns

Figure 6 shows the stress-strain curves of flax/PLA braided yarns and composite yarns. Before curing, the stress-strain curves of flax/PLA braided yarns exhibit a zigzag pattern upon breakage. The flax yarns fracture first,

and then the PLA filaments break with more strain. Because of the elastic behavior of PLA filaments, the flax/PLA braided yarns exhibit a zigzag pattern rise with the strain up to fracture. After curing, the stress-strain curves of flax/PLA composite yarns rise with the strain up to fracture, and then exhibit a sharp decline.

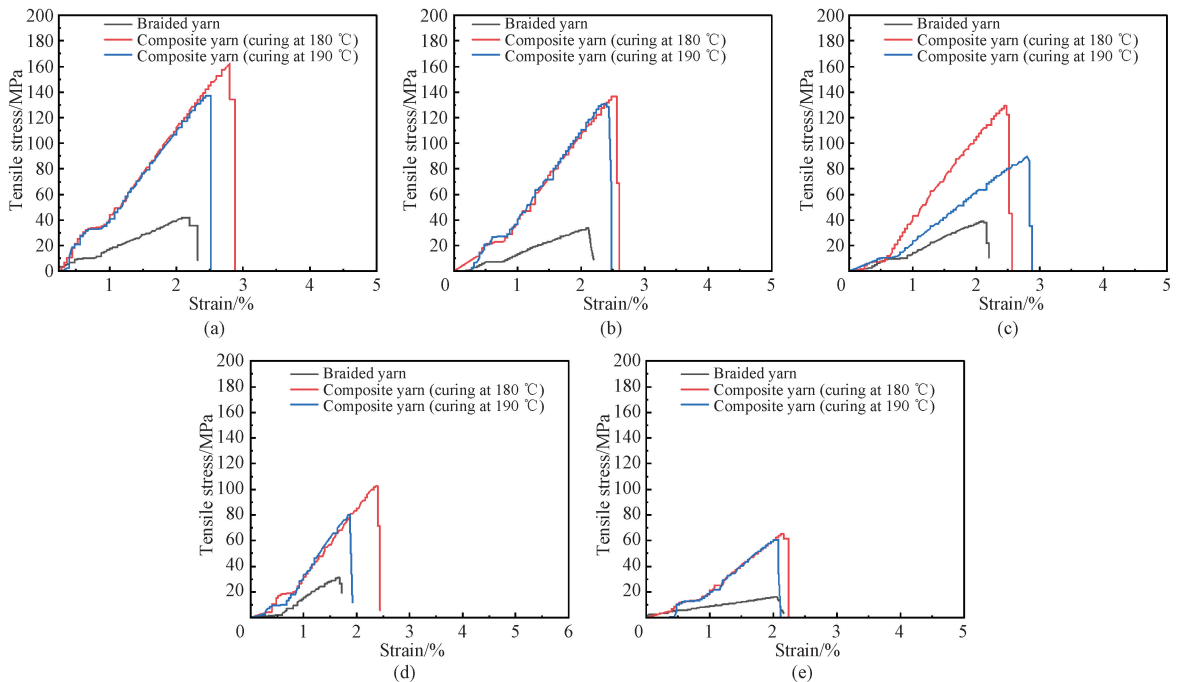


Fig. 6 Tensile stress-strain curves of flax/PLA braided yarns and composite yarns: (a) 4-spindle yarn; (b) 6-spindle yarn; (c) 8-spindle yarn; (d) 12-spindle yarn; (e) 24-spindle yarn

The 4-spindle composite yarns likely result in better fiber alignment and orientation of the flax fibers within the composite structure. Proper alignment of fibers along the load direction enhances load transfer and improves the tensile strength. The curing at 180 °C might have

promoted effective bonding between flax fibers and PLA matrices. A strong interfacial bond is crucial for efficient load transfer between the fiber reinforcement and the matrix, leading to the enhanced tensile strength. The specific curing temperature of 180 °C might have

provided optimal conditions for the curing reaction, resulting in a well-consolidated composite with improved tensile properties. Variations in the curing temperature can significantly impact the degree of polymer cross-linking and matrix-fiber adhesion, influencing the tensile strength of the composite. The chosen curing temperature and the properties of the PLA matrices may have contributed to favorable interactions with the flax fibers. PLA is known for its compatibility with natural fibers like flax, and the selected processing conditions might have facilitated a strong bond between the fibers and the matrices, enhancing the tensile strength. The 4-spindle composite yarn could have minimum defects and voids

within the composite structure. Uniform distribution of fibers and reduced porosity contribute to improved tensile strengths.

Figure 7 shows the elastic modulus, the tensile strength, the tensile strain at breakage and the flexural stiffness of all braided yarns and composite yarns. The elastic moduli of 4-spindle composite yarns and 6-spindle composite yarns are similar and high. The highest tensile strength of braided yarns is observed on 12-spindle yarns and the highest tensile strength of composite yarns is observed on 4-spindle yarns cured at 180 °C. The tensile strength of composite yarns decreases with the increase of the mass fraction of PLA.

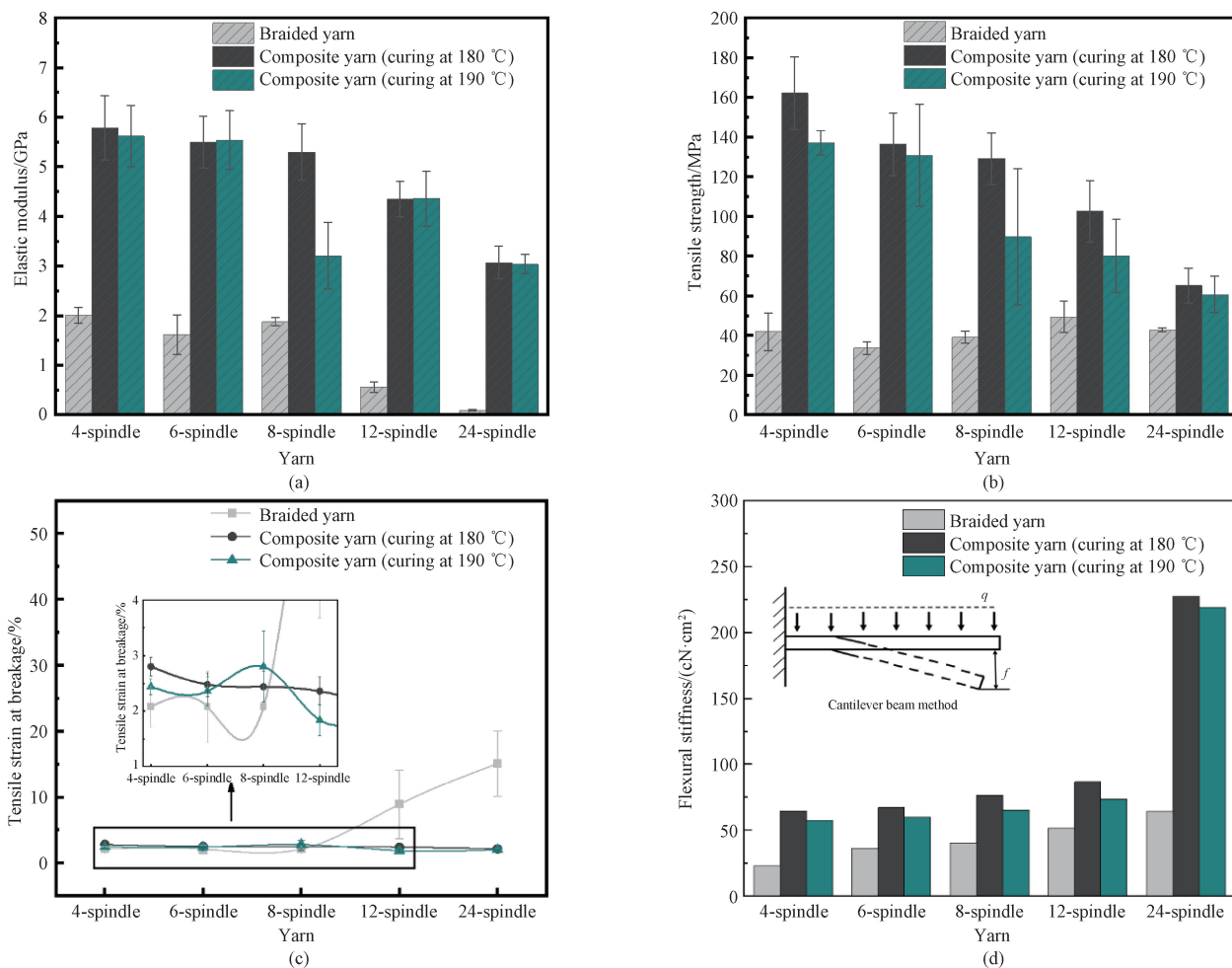


Fig. 7 Mechanical properties of flax/PLA braided yarns and composite yarns; (a) elastic modulus; (b) tensile strength; (c) tensile strain at breakage; (d) flexural stiffness

The 4-spindle composite yarn has a high tensile strength and a high stiffness due to its balanced structure, providing a foundation for improvement during the curing process. Curing involves molecular changes and increased intermolecular forces, leading to enhanced tensile properties. After curing, the 4-spindle composite yarn may experience more efficient molecular alignment and cross-linking between flax fibers and PLA matrices. This optimal arrangement allows for a more effective

utilization of the curing process to enhance tensile properties. The curing process promotes stronger interfacial bonding between flax fibers and PLA matrices, resulting in a more densely packed and organized structure. This contributes to a higher elastic modulus ((5.79±0.65) GPa). The enhanced molecular structure also contributes to the increased tensile strength. The alignment of molecules and the formation of additional cross-links during the curing process strengthen the yarn,

making it more resistant to deformation under tension. The curing conditions, such as a temperature of 180 °C and curing time of 15 min, may have been particularly well-suited for the 4-spindle yarn, leading to optimal improvements in its tensile properties.

The 24-spindle braided yarn has the highest tensile strain ($(15.04\% \pm 4.96\%)$) and the lowest elastic modulus, indicating a highly flexible structure. The 24-spindle yarn may not benefit as much from the curing process compared to other yarns with low initial flexibility. The inherent flexibility of the 24-spindle yarn structure, even after curing, can limit the development of a highly organized and tightly packed molecular arrangement, which results in a structure with the low stiffness and elastic modulus. The curing process may not induce sufficient molecular alignment and cross-linking due to its more PLA mass fraction in the 24-spindle yarn.

The flexural behavior of the braided yarns is affected by the braided structure, the arrangement of the spindles and the interaction between the fibers. The flexural stiffness of yarns is shown in Fig. 7(d). The highest flexural stiffness ($227.62 \text{ cN} \cdot \text{cm}^2$) is seen in the 24-spindle composite yarn. The larger the flexural stiffness, the stiffer and less easy to bend the composite yarn. With more spindles, the number of interweaving points per unit length increases, the sheath yarn forms a tighter weaving structure, and the increase of the contact pressure at the interweaving points of the adjacent spindle improves the friction between the spindles and prevents the fibers from flexure. The high degree of yarn buckling leads to an increase in the mass of the yarn per unit length (4-spindle yarn), and the positive contact pressure between the core yarn and the sheath yarn increases under the same flexural deformation. Yarns braided with less spindles are easy to deform under the extrusion than that braided with more spindles.

3 Conclusions

The mechanical properties of flax/PLA braided yarns were investigated to create green composites using a braiding technique. By the SEM analysis, the thermal analysis and the tensile property test, the effects of PLA mass fractions, curing conditions and flax contents on the mechanical properties of flax/PLA braided yarns and composite yarns were examined. The tensile strength of the flax/PLA composite yarns increased with an increase in the PLA mass fraction. The maximum tensile strength was achieved at a PLA mass fraction of 58.33%. Notably, the maximum tensile strength of the flax/PLA braided yarns was achieved under specific curing conditions of 180 °C for 15 min. The curing temperature significantly influenced the tensile strength of the composite yarns. The developed composite yarns hold promise for various applications, including automotive body parts, building materials and aerospace manufacturing, and would contribute to reducing

environmental impact while maintaining high-performance standards.

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编织工艺制备亚麻/聚乳酸芯鞘结构复合材料纱线

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摘要: 可持续和环保材料的需求日益增长, 推动了绿色复合材料的研究发展。该文以 4~24 锭的聚乳酸 (polylactic acid, PLA) 长丝为鞘, 亚麻纱线为芯, 编织成亚麻/PLA 芯鞘纱线。经不同温度 (180 °C 和 190 °C) 熔融固化后, 制得亚麻/PLA 复合材料纱线。拉伸试验结果表明, 在 180 °C 的固化温度下, 4 锭亚麻/PLA 复合材料纱的弹性模量为 (5.79±0.65) GPa, 拉伸强度为 (162.17±18.18) MPa。该亚麻/PLA 复合材料纱线具有良好的力学性能, 适用于汽车制造业和建筑材料的绿色复合材料。

关键词: 亚麻; 聚乳酸 (PLA); 编织; 复合材料纱; 拉伸性能