EFFECT OF ALKALINE TREATMENT UNDER TENSILE STRESS ON MECHANICAL PROPERTIES OF NATURAL FIBRE REINFORCED PLA COMPOSITES

SEIJI MITSUBAYASHI & KENICHI TAKEMURA
Department of Mechanical Engineering, Kanagawa University, Japan

ABSTRACT

Recently, the development of green composites (GCs), which are 100% biomass-derived composites made of biodegradable resin reinforced with natural fibres, has attracted much attention. To improve the mechanical properties of GCs, it is necessary to enhance the adhesion between the fibre/resin interface, and alkaline treatment and cyclic tensile loading of natural fibres are considered to be effective methods. However, the optimal conditions of alkaline treatment and tensile loading for maximizing the mechanical properties of GCs have not been clarified. Therefore, in this study, to investigate the effect of alkaline treatment under tensile stress on the mechanical properties of continuous natural fibre reinforced PLA composites, continuous natural fibre reinforced PLA composites were prepared by simultaneously applying alkaline treatment and tensile loading to the continuous natural fibres and their mechanical properties were evaluated. The results showed that the mechanical properties were further improved by applying tensile load during alkaline treatment. The tensile strength of the composites approached a certain value when the tensile load during alkaline treatment exceeded a certain percentage. In conclusion, the increase in tensile strength with increasing tensile load during alkaline treatment was attributed to the decrease in yarn twist angle, suggesting that it is not necessary to apply a load exceeding a certain percentage of the breaking load during alkaline treatment.

Keywords: green composites, natural fibre, alkaline treatment, tensile stress.

1 INTRODUCTION

Recently, the development of green composites (GCs), which are 100% biomass-derived, has been attracting significant attention [1]–[3]. GC is a composite that uses biodegradable resin as the matrix and natural fibres as reinforcement. However, compared to carbon fibre reinforced plastics (CFRPs) and grass fibre reinforced plastics (GFRPs), GC has the disadvantage of weak adhesion at the fibre/resin interface [4], [5]. Therefore, improving interfacial adhesion is a crucial challenge.

Fibre surface treatment is one of the effective methods to enhance interfacial adhesion. Surface treatment techniques can be broadly classified into physical and chemical methods. A physical approach involves adding nanofillers such as carbon nanotubes (CNTs) [6] and cellulose nanofibres (CNFs) [7] to fibres or resins. However, due to their microscopic size, achieving uniform dispersion of nanofillers is challenging [8]. Non-uniform dispersion can cause localized fractures and deformations [9]. Additionally, nanofillers are expensive, which may lead to increased material costs [10]. On the other hand, chemical treatments [11] include alkaline treatment [12] and silane coupling treatment [13]. Silane coupling treatment utilizes silane compounds to form strong chemical bonds between materials, thereby improving interfacial adhesion. While this method significantly enhances adhesion, its high cost and the need for specialized equipment limit its applicability. In contrast, alkaline treatment is a method that removes lignin and hemicellulose from the fibre surface using alkaline chemicals such as sodium hydroxide. This approach has the advantage of being more cost-effective than physical treatments and silane coupling treatment, as the chemicals used are relatively

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inexpensive. Therefore, alkaline treatment is particularly effective when improving interfacial adhesion while keeping costs low.

Numerous studies have been conducted to enhance the mechanical properties of GCs by applying alkaline treatment to natural fibres [14]–[16]. Matsuo et al. have demonstrated that applying cyclic tensile loads to natural fibres during alkaline treatment improves the mechanical properties of green composites (GC) [17]. Additionally, it is well known that longer fibre length generally enhances the mechanical properties of fibre reinforced plastics (FRP). Therefore, to improve the mechanical properties of GC, it is crucial to perform continuous alkali treatment on natural fibres while applying tensile loads.

Typically, natural fibres are spun into single or plied yarns before being distributed in the market [18]. However, alkali treatment of these yarns is often performed in a discontinuous manner, resulting in low productivity. To address this issue, the authors utilized a system that enables alkaline treatment while continuously applying tensile load to natural fibres, aiming to improve both the mechanical properties and production efficiency of natural fibres and green composites. Nevertheless, the effect of tensile stress variations during alkaline treatment on the mechanical properties of natural fibre-reinforced PLA composites remains insufficiently understood.

This study investigates the mechanical properties of natural fibres and green composites subjected to continuous alkaline treatment under different tensile stresses. Tensile testing was conducted to compare their mechanical performance and analyse the key factors influencing these variations.

2 EXPERIMENTAL METHOD

2.1 Materials

In this study, 20-count twisted ramie yarn (Tosco Co., Ltd., Tokyo, Japan) was used as reinforcement. It was a single-ply and right-handed twill (S-twist) [19]. In this context, the term ‘count’ refers to the metric count (Nm), which can be converted to tex, the mass per kilometre of twisted yarn, using eqn (1):

$$\text{tex} = \frac{1000}{\text{Nm}}, \tag{1}$$

where tex and Nm are the linear density in tex and the metric count, respectively. Therefore, the count of twisted yarns used in this study was equivalent to 50 tex. Moreover, the diameter of ramie yarn, which was observed by zoom-type stereo microscope (SZX7, Olympus Corporation, Japan), was approximately 0.3 mm.

PLA in pellet form (Terramac TE-2000, Unitika Ltd., Osaka, Japan) was used for matrix. It had a specific gravity of 1.25, a melting temperature of 170°C and a melt flow rate of 10 g/10 min (190°C, 21.18 N) [20].

In addition, sodium hydroxide (NaOH) and acetic acid (both purchased from Fujifilm Wako Pure Chemical Corporation, Osaka, Japan) were used for alkaline treatment and subsequent neutralization.

2.2 Fabrication process of specimens

2.2.1 Fabrication process of ramie yarn reinforced PLA composites

Fig. 1 shows the apparatus used for producing ramie yarn reinforced PLA filament. Alkaline treatment and compositing with PLA were conducted using this setup while feeding twisted yarns at a constant speed. The yarns were unwound for transport (part no. 1 in the Fig. 1), and rollers (part no. 2) were rotated to nip the twisted yarn. These rollers were powered by a motor, and the speed of the yarn conveyance was regulated by adjusting the rotation speed of the rollers. Tension was applied to the yarns by creating a difference in the rotational speeds of the front and rear rollers, and the tension on the yarns was measured (part no. 3). Containers for both alkaline solution and solution for neutralization (part no. 4) were set up, and alkaline treatment and neutralization were performed during transporting. In this study, to prevent hydrolysis of PLA resin, the alkaline-treated yarns were rinsed in water and thoroughly dried in a vacuum dryer at 130°C for 4 hours. The dried yarns were then conveyed into the crosshead die (part no. 7) and composited with PLA supplied from a single-shaft extruder (part no. 8). After composition, the ramie twisted yarn reinforced PLA composite was air-cooled and finally wound using a winding device (part no. 9).

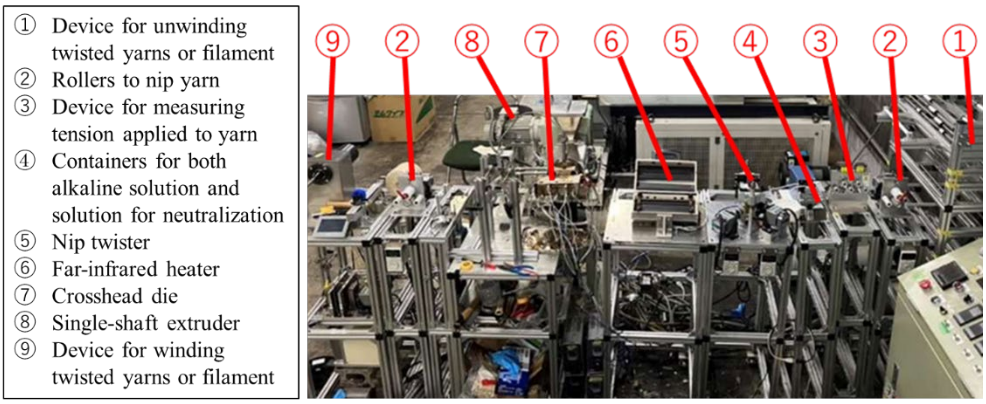

Figure 1: Equipment for the alkali treatment of ramie yarn and fabrication of ramie yarn reinforced composites.

2.2.2 Process of alkaline treatment to ramie yarn

In this study, aqueous sodium hydroxide and acetic acid solutions were used for alkaline and neutralization treatments, respectively. Fig. 2 shows the details of the container for alkaline treatment and neutralization treatment. These containers were placed at position no. 4 in Fig. 1. Inside these containers, twisted yarns were hooked to grooved bearings to reduce the frictional force acting on the twisted yarns. In addition, the length of the path that the twisted yarns travelled through the containers and the treatment time were ensured for the treatment. Here, the path length of twisted yarn passing through each container was 600 mm. When the transport speed of the twisted yarns is 18.3 mm/s, the time required for treatment is equivalent to 33 seconds. The containers were used so that the time required for alkaline treatment and neutralization treatment were equal. In this study, tension was applied to the twisted yarns during alkaline treatment at a concentration of 10 wt.% of sodium hydroxide solution. Here, the tension applied was set at 0%, 20%, 40%, 60% and 80% of the breaking load of the twisted yarns.

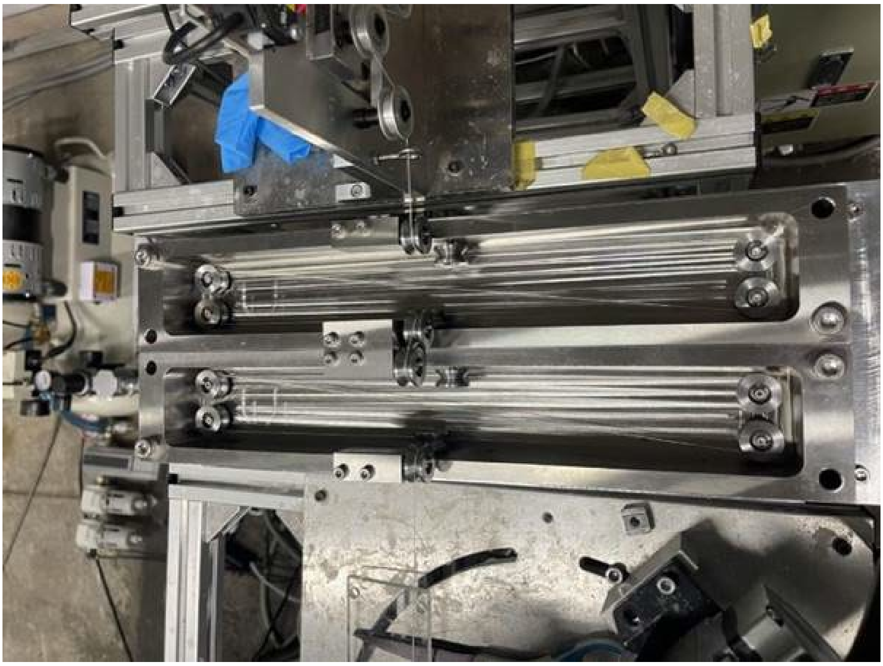

Figure 2: Containers for alkali treatment and neutralization.

2.2.3 Fabrication process of ramie yarn reinforced PLA composites

After alkaline treatment of the twisted yarns, ramie/PLA composites (filaments) were fabricated using the filament fabrication equipment shown in Fig. 1. The filaments were prepared by adjusting the filament diameter to 1.6 mm so that the volume content of twisted ramie yarn in the filaments was approximately 3%.

2.3 Evaluation method of tensile properties

2.3.1 Tensile test of ramie yarn

In this study, specimens were prepared by gluing alkaline-treated ramie yarns under various conditions to a U-shaped backing paper with a distance of 10 mm between the points and then cutting the backing paper with scissors. The test specimens were mounted on a universal testing machine (Tensilon, RTC-1250A, A&D Company, Limited, Tokyo, Japan) and static tensile tests were performed. The number of specimens was set to 10 or more, and the tensile speed of the specimens was set to 1.0 mm/min.

When calculating the cross-sectional area, the shape of the twisted yarn was considered to be elliptical because of the flattened shape of the fibre. When measuring dimensions, the diameters of the major and minor axes were measured by image analysis. Eqn (2) was used to calculate the cross-sectional area.

$$A = \frac{\pi ab}{4}, \tag{2}$$

where A, a, and b are the cross-sectional area, diameter of the major axis, and diameter of the minor axis of the twisted yarn, respectively.

2.3.2 Tensile test of ramie yarn reinforced PLA composites

Static tensile tests were performed on ramie/PLA composites with filament shape. Here, the length of the specimen was 150 mm, the distance between the points was 50 mm, the number of tests was 5, and the tensile speed of the specimen was 1.0 mm/min. A micrometre was used to measure the diameter.

2.4 Observation method of ramie yarn reinforced PLA composites

To investigate the factors that cause changes in the tensile strength of twisted yarns treated with alkaline under tensile stress and filaments formed using these yarns, the condition of the filaments was observed using zoom-type stereo microscope (SZX7, Olympus Corporation, Japan). The observation results were photographed using a C-mount camera for microscopy, and the twist angle of the ramie yarn inside the filament was measured. Image processing software (ImageJ) was used to measure the twist angle. Here, the number of twist angle measurements was set to 5 or more.

3 RESULTS AND DISCUSSION

3.1 Effect of alkaline treatment under tensile stress on tensile properties of ramie yarn

Fig. 3 shows the tensile strength and Young's modulus of alkali-treated ramie yarn under tensile stress. As a result of the tensile load, an increase in tensile strength and a decrease in Young's modulus were observed. Additionally, the tensile strength and Young's modulus remained largely consistent regardless of the magnitude of the tensile load applied during alkaline treatment. This suggests that the mechanical properties are independent of the tensile load during alkaline treatment.

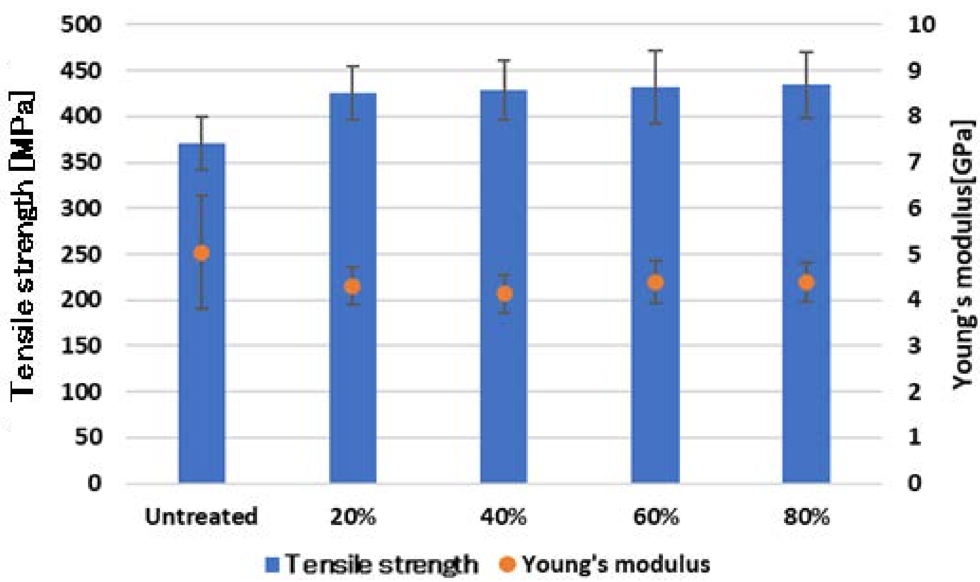

Figure 3: Effect of alkaline treatment under tensile stress on tensile properties of ramie yarn.

3.2 Effect of alkaline treatment under tensile stress on tensile properties of ramie yarn reinforced PLA composites

Fig. 4 shows the relationship between tensile stress and filament tensile strength during alkaline treatment. The maximum tensile strength was obtained when alkaline treatment was performed while applying a load of 40% of the breaking load. This result indicates that there is no advantage to alkaline treatment while applying a tensile load of 40% or greater.

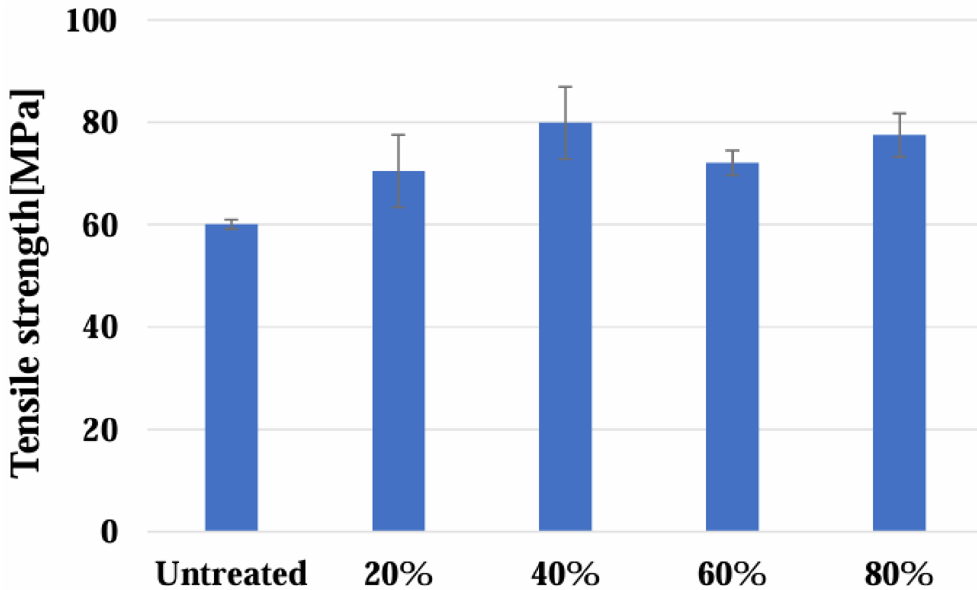

Figure 4: Effect of alkaline treatment under tensile stress on tensile strength of ramie yarn reinforced PLA composites.

3.3 Effect of alkaline treatment under tensile stress on tensile properties of ramie yarn reinforced PLA composites

Fig. 5 shows the results of optical microscope observations of filaments obtained by alkaline treatment with a 10 wt.% NaOH solution while applying tensile loads of 20%, 40%, 60% and 80% relative to the tensile strength of untreated ramie yarn. Fig. 6 presents the calculated twist angles of ramie twisted yarn subjected to alkaline treatment under a 10 wt.% NaOH solution while applying loads corresponding to 0%, 20%, 40%, 60% and 80% of the breaking load of the twisted yarn.

In all treatment conditions, the twist angle tended to decrease compared to the untreated samples. Furthermore, when alkaline treatment was performed under a load exceeding 40% of the breaking load, the twist angles were found to be approximately constant. This reduction of twist angle is considered a key factor contributing to the improvement in the strength of both twisted yarn and filaments.

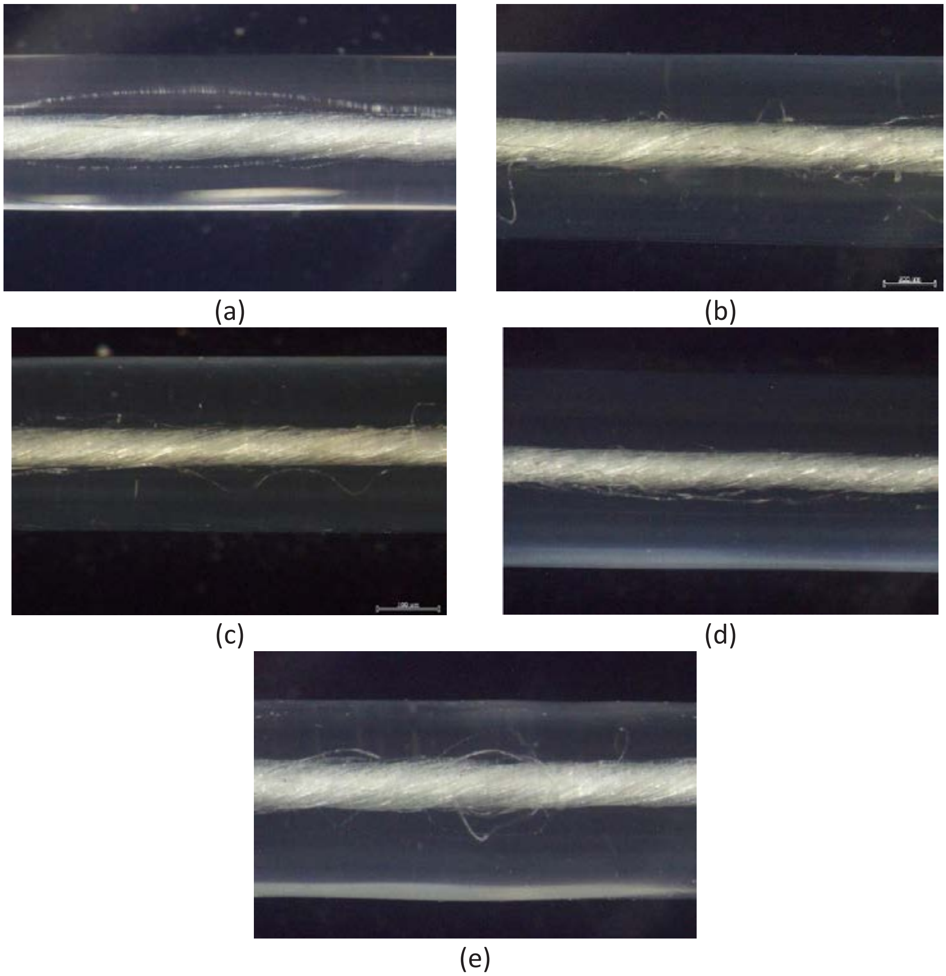

Figure 5: Filaments prepared by alkaline treatment under tensile load. (a) 0% of breaking load; (b) 20% of breaking load; (c) 40% of breaking load; (d) 60% of breaking load; and (e) 80% of breaking load.

4 CONCLUSION

The purpose of this study was to clarify the effects of changes in alkaline treatment concentration, treatment time, and tensile load on the mechanical properties of ramie/PLA composites. As a result, the following findings were obtained.

1. Measurements of the tensile strength of ramie twisted yarn showed that the tensile strength increased and Young's modulus decreased due to the applied tensile load. Additionally, the tensile strength and Young's modulus remained largely consistent regardless of the magnitude of the tensile load applied during alkaline treatment. This suggests that the mechanical properties are independent of the tensile load during alkaline treatment.

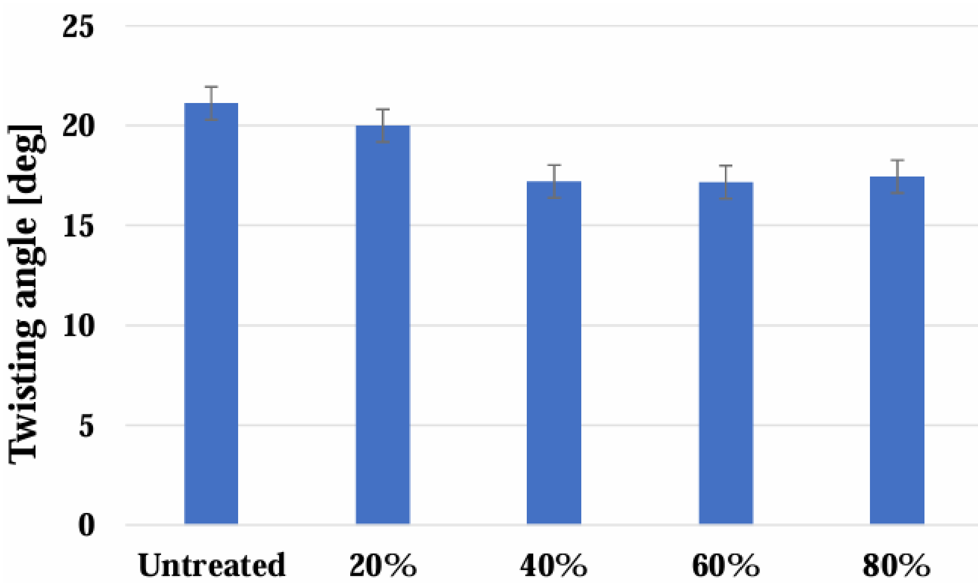

Figure 6: Relationship between tensile stress during alkali treatment and twist angle of twisted yarns.

2. The maximum tensile strength of alkali-treated ramie/PLA composites was obtained when alkaline treatment was performed while applying a load of 40% of the breaking load. This result indicates that there is no advantage to alkaline treatment while applying a tensile load of 40% or greater.
3. Tensile loading has a significant effect on the tensile strength of ramie/PLA composites. The continuous processing equipment made it possible to reduce the twist angle of the yarn. The variation of the twist angle became small when the tensile load exceeded 40%, and it was found that higher tensile loads were not necessary to increase the strength of the yarn.

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