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Low-Voltage SEM of Natural Plant Fibers: Microstructure Properties (Surface and Cross-Section) and their Link to the Tensile Properties

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Abstract

In this study, the microstructure of different natural plant fibers (flax, jute, ramie, and sisal fibers) were characterized by using low-voltage Scanning Electron Microscopy (LV-SEM). The LV-SEM observations indicated that jute and sisal fibers exhibit less variation in terms of the fiber cross-sectional area, internal lumen shape and size, and cell wall thickness in comparison to flax and ramie fibers. We find that this is also reflected in the tensile properties of the fibers. The tensile properties of single ramie fibers and their fracture behavior was investigated in detail. The stress-strain behavior showed two distinctive regimes. For linear curves, the tensile strength varies from 648-1086 MPa whereas nonlinear curves result in much lower values (177-452) MPa. This variation was linked to differences in the microstructure of the fibers. The LV-SEM of the tensile fracture surfaces of ramie fibers revealed details on the cell wall structure and its fracture behavior under tensile load. Moreover, the SEM images confirm that the collapse of the primary cell wall generally leads to a non-linear stress-strain curve for single ramie fibers.

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Keywords: Plant fibers; Fiber's microstructure; Scanning electron microscopy; Tensile properties

1. Introduction

Natural plant fibers such as jute, flax, hemp, sisal, and ramie fibers are increasingly being used as reinforcements in polymer matrix composites [1], due to their wide availability, low cost, eco-friendliness, low density, and high

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specific mechanical properties [2, 3]. Moreover, the high density, non-recyclability, health hazards, high energy consumption and environmental issues of synthetic fibers [4, 5] have motivated many researchers to investigate sustainable materials. Natural plant fibers represent a renewable resource and form an interesting class of reinforcing materials, which can be used as a replacement for synthetic fibers in polymer composites [5].

However, the mechanical properties of natural plant fibers typically vary over a large range [6-9], partly due to variability typical for biological samples, but also due to errors arising from testing techniques [3, 6]. Natural plant fibers have a complex structure and organization, which can be considered as a natural composite material consisting of cellulose fibrils embedded in an amorphous matrix of hemicellulose and lignin [2, 3, 6, 10]. The single plant fibers have a typical structure that consists of a central lumen surrounded by a thick cell wall [2]. The fiber cell wall is formed from two main layers: a very thin primary cell wall (70-110 nm) [11], and a secondary cell wall, which is made up of three sub-layers (S1, S2, and S3) [7]. S2 is the thickest layer (3-13 μm thick) [11] in the cell wall and represents the most important layer which determines the mechanical properties of the fibers [12, 13]. Each layer of the fiber cell wall is composed mainly of cellulose, hemicellulose and lignin [7, 14]. The long chain cellulose molecules, often referred to as microfibrils, are organized in a crystalline network [7]. The typical diameter of these microfibrils are in the range of 10-30 nm and they are made up of 30-100 of cellulose molecules in the form of an extended chain. The microfibrils greatly contribute to the mechanical strength of the fibers [15, 16], and thus the complex structure of natural plant fibers can highly influence the fiber properties.

Fiber singular testing techniques also can influence the apparent fiber properties. The reported tensile properties of single fibers are highly effected by the cross sectional area [6, 8, 9]. Some of the variation in tensile properties commonly reported in the literature are likely due to inaccurate cross-sectional area measurements of single fibers. The assumption that natural plant fibers have a uniform and circular cross sectional area is the most common method employed in the literature to calculate the cross sectional area of single fibers [6, 7, 17-19]. Most natural plant fibers exhibit considerable deviation from circularity in their cross sectional area [7], therefore, the conventional method which is based on the fiber diameter measurement may not be a suitable method to calculate the cross sectional area. For example, Thomason and Carruthers [8], found that the average values of the cross sectional area obtained from the fiber diameter measurements of flax and sisal fibers are almost double that of the actual cross sectional area, leading to the underestimation of tensile properties by up to 60% . Hu et al. [20] also reported that the conventional method of cross sectional area measurements leads to inaccurate results with high standard deviations.

The review of the literature shows that fiber microstructure and also the fiber testing techniques can highly influence the fiber properties. Therefore, this study aims to investigate the microstructure of different natural plant fibers (flax, jute, ramie, and sisal fibers) by using low voltage scanning electron microscopy (LV-SEM). The link between fiber microstructure and property variations of these fibers was also studied. The tensile testing of single ramie fibers followed by SEM observations of the fractured fibers indicated details on the fiber microstructure and mechanical properties of the cell wall. The true cross sectional area of single ramie fibers was measured by analysis of LV-SEM images using image J software. The obtained tensile strength results were compared to other reported results assuming circularity, in terms of standard deviations.

2. Materials and experimental procedure

Flax, Jute, ramie, and sisal fibers were used in this study. The as received fibers were cryofractured in order to investigate the fiber cross section structure. An LV-SEM (Nova Nano SEM 450) was used to observe the fiber microstructure. Natural plant fibers are not conductive materials, therefore, the observations were performed using a low accelerating voltage (1 kV) to avoid fiber charging. The images were collected using a through lens detector (TLD) at 4.5 mm working distance with a beam deceleration of 2000 V.

A Zwick Roll tensile testing machine with a 500 N load cell was used to test the tensile strength of single ramie fibers. The test was carried out at a constant crosshead displacement rate of 40 %/min. In accordance with the ASTM D 3822-01 standard the single ramie fiber was mounted on the paper card with a 5 mm gauge length by using cyanoacrylate glue. The fibers were carefully glued in the exact center of the card as shown in Fig. 1. Thereafter, samples were loaded into the testing machine and just before starting the test, the supporting side of the card was carefully cut. The tests were carried out at room temperature (22 ± 3 °C). During the test the force-strain

values were recorded and these values were used to measure the fiber tensile strength properties. Only samples which broke in the middle of their gauge length were used to calculate the tensile strength, whereas the fibers which broke near to the glue clamp or card frame were not used in the calculations.

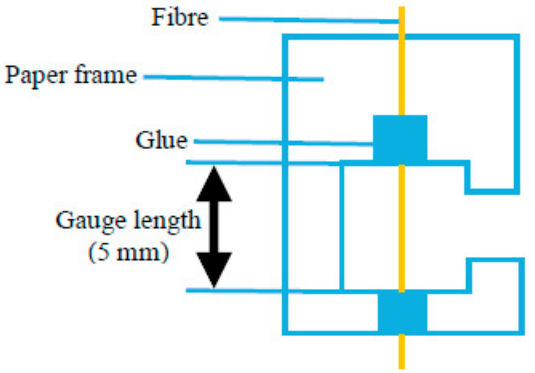

Fig. 1. Schematic representation of the paper frame for the single fiber tensile testing.

After testing, the cross sectional area can be estimated from the fractured sample due to the elastic behavior of the plant fibers. LV-SEM was used to observe the cross section of the fractured fibers, as shown in Fig. 2. Fractured fibers with a flat and clear cross section were selected for the cross section area calculations. The SEM images were used to calculate the actual cross sectional area of the fractured fibers by using image J software. The hollow structure (lumen) can be clearly seen as demonstrated in the example in Fig. 2. This area was found to be about 10% of the total cross sectional area and was excluded from the total area.

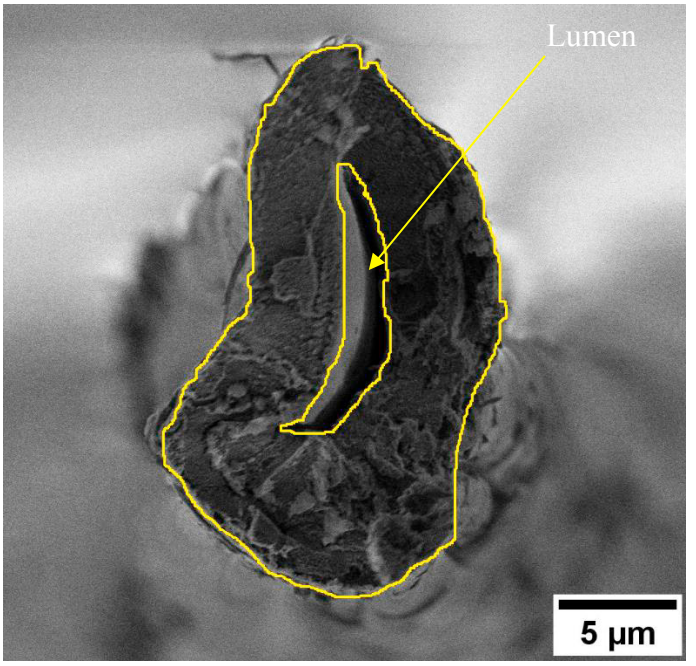

Fig. 2. Cross-sectional area determination of single ramie fiber using image J software.

3. Results and Discussion

3.1. Microstructure comparison of different fibers

The SEM images of the cross-sections of flax, jute, ramie, and sisal fibers are shown in Fig. 3. It can be seen that jute and sisal fibers are in the form of a bundle, that the single fibers are combined by means of middle lamella (ML). The flax fibers are in a partly separated bundle exposing the single fibers, and the ramie is in the form of a single fiber. All fibers show similar structures, comprising a lumen in the center which is surrounded by several cell walls. We observe that the cross-section shape, cell wall thickness, internal lumen size and shape vary substantially for the different materials. For instance, our flax and ramie fibers show irregular internal lumen shape and more polygonal fiber cross-section. In contrast, jute and sisal fibers show similar structure in terms of cross-section shape and the internal lumen shape and size are almost circular. Analysis of the tensile strength data in the literature [21] shows that jute and sisal fibers have smaller variations in reported tensile strength values than flax and ramie fibers (Fig. 4), which we believe could be due to the greater variation in fiber microstructure of the flax and ramie fibers, as indicated by SEM observations. In addition, the higher density and strength of flax fibers reported in the literature [21], could partly be explained due to the smaller lumen and dense cell wall structure compared to other fibers.

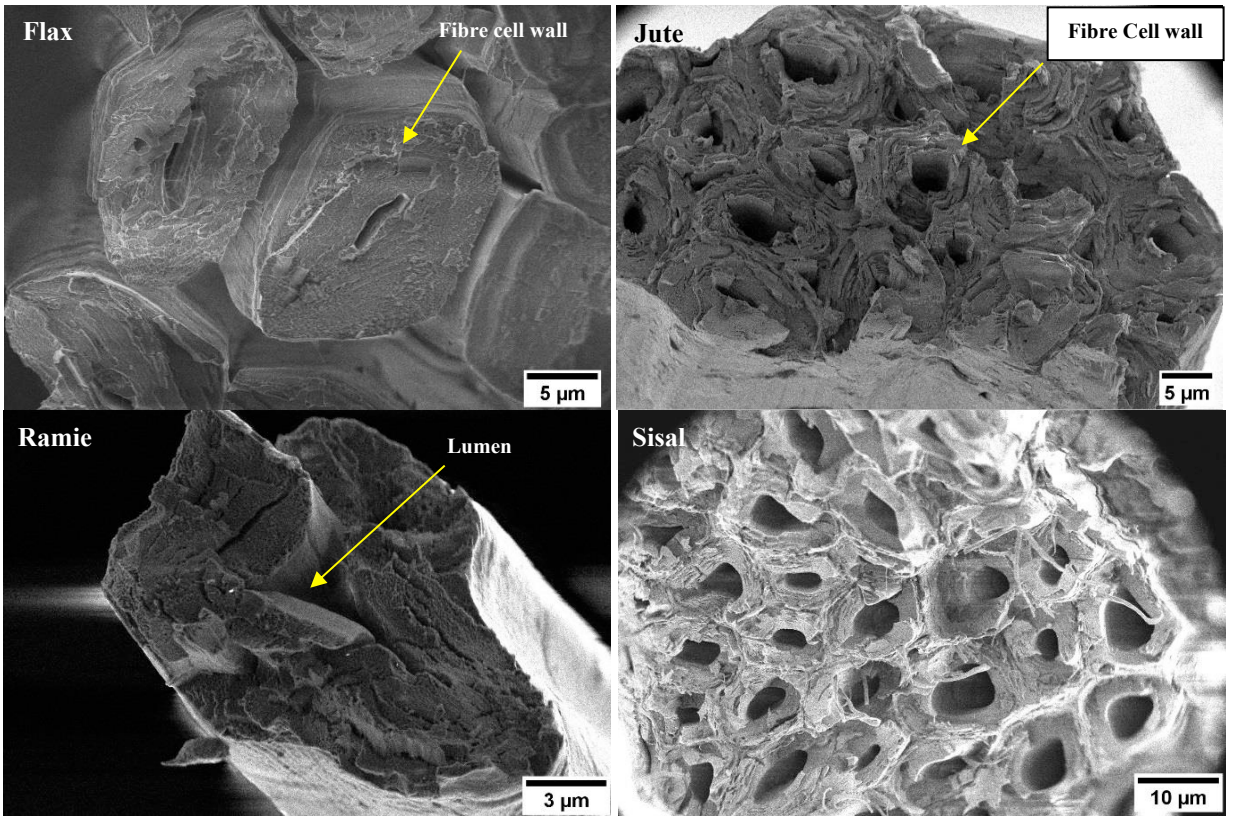

Fig. 3. SEM images of the cross section of flax, ramie, jute, and sisal fibers.

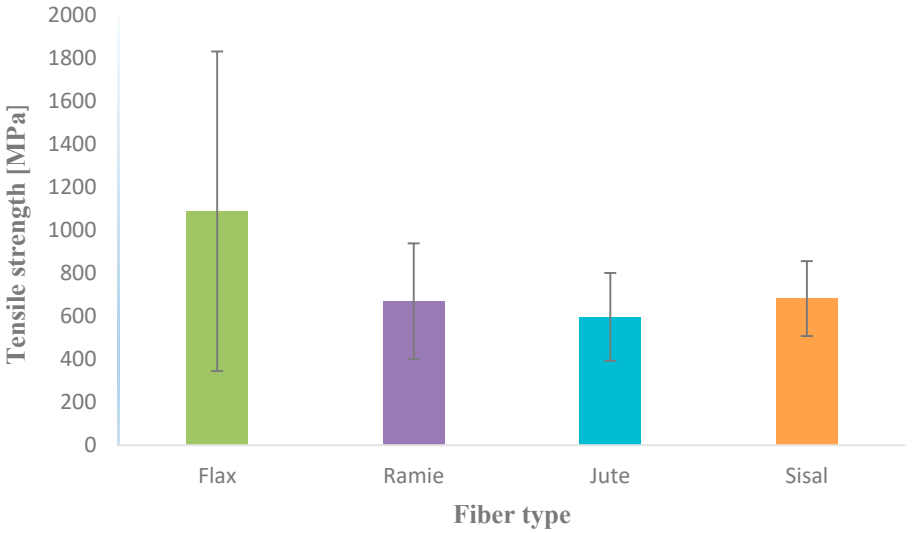

Fig. 4. Bars represent the relative variation, and error bars represent the range of the tensile strength values of flax, ramie, jute, and sisal fibers based on data reported in [21].

3.2. Microstructure of ramie fibers visible on tensile fracture surfaces

30 single ramie fibers were investigated after tensile fracture. The LV-SEM observations of the cell wall structure of ramie fibers followed by image J analysis showed that the thickness of the primary wall was approximately 100 nm, as shown in Fig. 5a, b. This is in the range of primary wall thickness reported for hemp fibers [11]. It can also be seen from Fig. 5a, that the secondary cell wall main consists of two phases: (1) bright nanoscale features (~ 30-70 nm), and (2) the dark phase in between. Based on literature reports that fiber cell wall can be considered as a natural composite [22], the bright nanostructures features would be expected to be crystalline cellulose microfibrils embedded in non-crystalline regions of hemicellulose and lignin (Fig. 5a). The observed cracks between the layers of the cell wall (Fig. 5a), are possibly due to the interlaminar stress which formed during the fracture process.

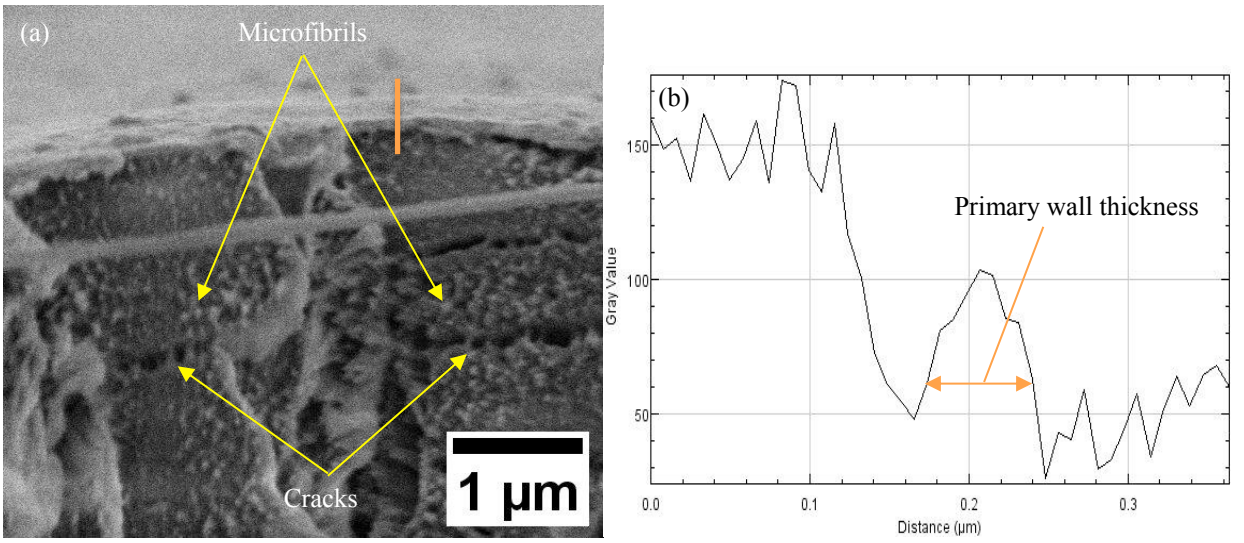

Fig. 5. (a) SEM image of the cell wall of ramie fiber, (b) the line profile of the orange vertical line in Fig. 5a.

3.3. Fracture behavior of single ramie fibers under tensile load

The typical stress – strain behavior of a single ramie fiber in the tensile test is demonstrated in Fig. 6a. It can be seen that the curve is almost linearly elastic without any signs of plastic deformation. Similar behavior and shape of plots were found for plant fibers reported in other works [6, 23, 24]. The LV-SEM observations of the fractured fibers showed a very flat and clear fracture end (Fig. 7a). However, some of the tested fibers showed two regions (non-linear) of the stress-strain curve (Fig. 6b). Mukherje [25] reported that the initial region of the stress-strain curve of sisal fibers is mostly due to the collapses of the weak primary cell wall and decohesion between fiber cells. The SEM observations in this work clearly show the collapse of the primary cell wall, as illustrated in Fig. 7b. Such fibers were found to have lower strength values than those with a linear stress-strain curve, which is possibly due to the defects and cracks that already present in the cell wall as well as the weak bond between the primary and secondary wall. On the other hand, some other tested fibers showed that different fiber cells have fractured in different planes (Fig. 8). According to Silva [26], this behavior of fiber cells is probably due to the variability in the fiber cell strength and also due to cell wall flaws. Such fibers showed a linear stress-strain curve. These variations in the fiber cell wall fracture can strongly influence the fiber properties and therefore large scatter was found in the values of the tensile strength of ramie fibers in this study. However, the standard deviation of the tensile strength values obtained from linear curves only in this study was (872 ± 190) MPa considerably lower than that reported elsewhere [27] as (936 ± 320) MPa. We believe this is due to the more accurate method used to calculate the cross sectional area of single fibers.

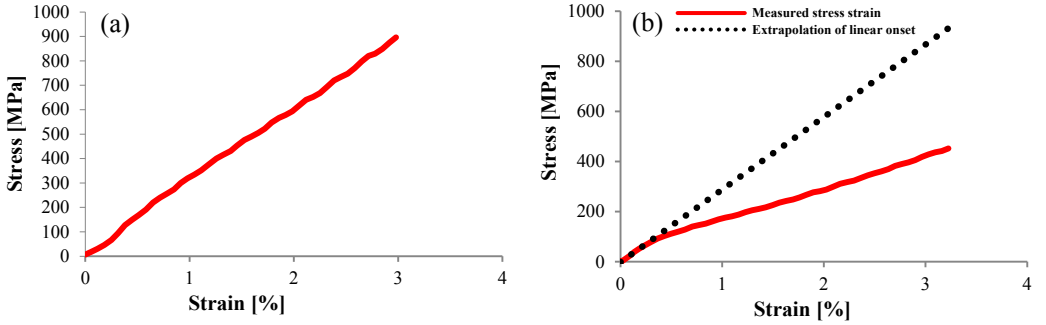

Fig. 6. The stress-strain curve of the single ramie fiber (a) example of linear curve, (b) example of non-linear curve, the extrapolation of the linear onset results in curve very similar to (a), the beginning of the deviation from this curve might indicate primary wall failure.

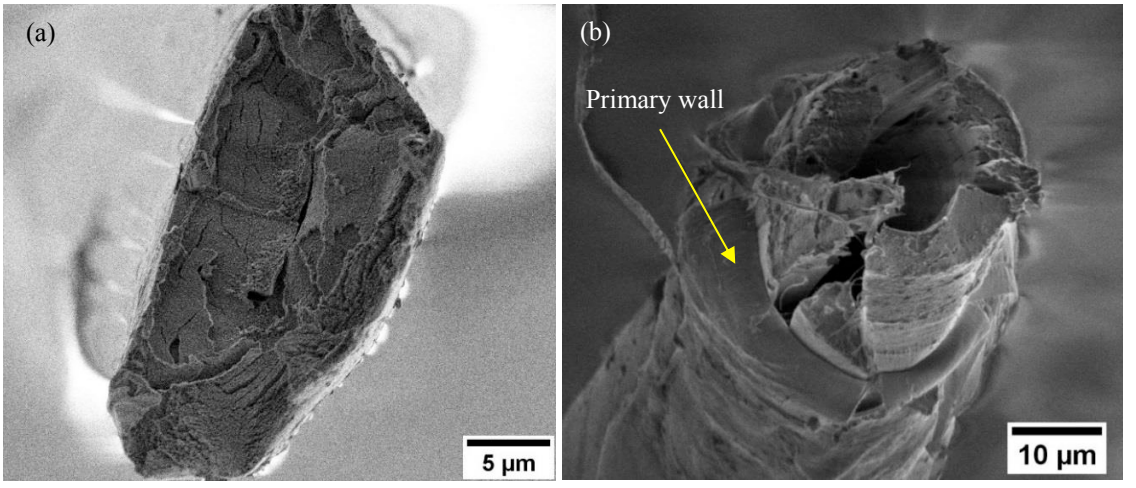

Fig. 7. SEM images of the fractured surface of single ramie fibers under tensile load (a) fractured in a flat surface, (b) showing the collapse and retraction of the weak primary wall and subsequent protrusion of the secondary wall.

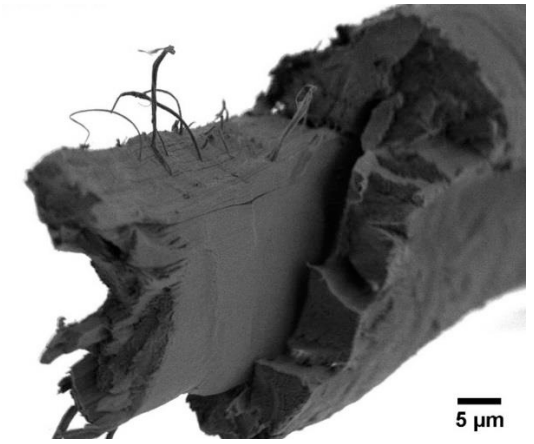

Fig. 8. SEM image of the fractured surface of single ramie fiber under tensile load showing different fiber cells have fractured in different planes.

4. Conclusions

LV-SEM observations in this work showed that the jute and sisal fibers exhibit less variation in their microstructure than those of flax and ramie fibers. This can be reflected on their mechanical properties. Detailed investigation for example of ramie fiber shows that some variation is due to differences in fracture behavior (e.g. collapse of primary wall and variability in cell wall strength), but further variation might be due to the methods of cross-sectional area measurement. The latter can be minimized through the use of our LV-SEM technique. Hence, LV-SEM is powerful tool for the understanding of the fracture behavior of plant fibers.

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