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Removal Ability of *Bacillus licheniformis* on Waxy Cuticle on Wheat Straw Surface

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Abstract: The outermost surface of wheat straw (WS) is covered with hydrophobic lipophilic extracts and silica, which affects follow-up processes such as impregnation pretreatment of pulping and papermaking. In this study, a strain named *Bacillus licheniformis* (*B. licheniformis*) was screened from the black liquor of papermaking, which was used to explore the effect of its treatment on the waxy cuticle of WS. Scanning electron microscope-energy dispersive spectroscopy (SEM-EDS) showed that the *B. licheniformis* had a certain destructive effect on the outer surface of WS and the content of Si on the outer surface decreased by 80%. The results of FTIR and X-ray photoelectron spectroscopy (XPS) displayed that the wax composition on the outer surface of WS decreased and the fiber structure inside appeared. The mechanical properties of paper demonstrated that the treated WS is still feasible in this field and the content of Si in the black liquor is reduced by 33%. Therefore, the WS treated by *B. licheniformis* can destroy the waxy cuticle on its outer surface and improve the wettability of WS. It provides a new idea to alleviate the "Si interference" problem of alkali recovery in WS traditional pulping and papermaking.

Keywords: *Bacillus licheniformis*; waxy cuticle; wheat straw; papermaking; black liquor; Si interference

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1. Introduction

With increasingly serious environmental pollution and the increase in fossil fuel consumption, alternative energy sources of renewable resources such as agricultural and forestry wastes have become an important development field of the global economy [1]. In China, the shortage of forest resources led to the high cost of papermaking from wood. Chemical pulp raw materials can be replaced by recycled wastepaper, but only low-grade paper can be produced [2]. In addition, the limited amount of recycled paper cannot guarantee the large demand of the paper industry. In view of the above situation, it is particularly important to obtain pulp from non-wood raw materials. Wheat straw (WS) is the second most abundant agricultural residue in the world [3]. WS is an important agricultural and forestry waste product and is a renewable fiber resource in China. The annual output of wheat will reach 22 million tons and the output of WS will reach 30 million tons at that time [4,5]. As an annual grass of gramineae, WS is mainly composed of cellulose, hemicellulose, and lignin and has strong environmental tolerance. In addition, it has the characteristics of high cellulose content, loose structure, thin cell wall, and so on [5–8]. WS can be used in many fields such as feed and compost and other fields including pulp and paper industry that can be utilized on a large scale due to its low cost and abundant reserves [9]. However, the outer surface cuticle of WS is covered with hydrophobic non-cellulose components, namely lipophilic extracts and silica. This organ has a certain degree of hardness and can protect the internal fibers of WS [10], but it also impedes the impregnation effect of the solution during the pre-treatment process. To improve the production level, lower equipment maintenance costs, and reduce the

incidence of defects, the extraction or removal of plant waxes is also considered as a possible preprocessing method for the pulp and paper industry [11]. The properties of WS can be improved when these hydrophobic components (such as waxes and silica) on WS surfaces are removed using oxidizer and alkali (H_2O_2 and NaOH). Although chemical methods can be used to remove these lipophiles, this can lead to secondary contamination and adversely affect fiber strength [12]. Therefore, biological treatment is highly desirable as an eco-friendly, efficient, and non-toxic keratinization method. The more common is biological enzyme treatment. At present, there are some enzymes used to treat lignocellulosic biomass, namely cellulase, hemicellulose, lignin-enzyme, lipase, etc. [13–16]; the pretreatment of biomass with different enzymes has different results. For example, the damage degree of lipase and cellulase on the outer surface of WS was different, it depends on the different action mechanism of the two enzymes [17,18]. However, there are still some deficiencies in the application of biological enzymes in the pretreatment of biomass. For example, the modification of fibers by enzymes needs further study and the high cost of enzyme production limits its application in papermaking industry to some extent [19]. Another biological treatment method, biological flora, usually plays an important role in the production of methane by the anaerobic fermentation of biomass, but this process needs to keep the balance of biota [20]. Furthermore, the cuticle of WS protects plants from drought, extreme temperature, ultraviolet radiation, chemical attack, mechanical damage, and pathogen/insect infestation while also providing mechanical support and acting as a barrier against organ fusion [21,22]. However, the existence of cuticles influenced the industrial utilization of WS, for example, it leads to the decrease in the efficiency of biomethane production by WS. Moreover, the Si of WS is mainly distributed in the outer surface cuticle in the form of SiO_2, resulting in high silicon content and high viscosity of black liquor in the pulp and paper industry, which is difficult to be applied in a conventional alkali recovery system [23]. Therefore, suitable strains are screened to achieve the destruction and removal of the outer surface of the WS cuticle from another perspective of the application of microbial agents in this paper [24]. The surface morphology, functional groups, basic elements, and other physical properties of WS before and after treatment were measured, which indicated that the surface barrier of WS was destroyed. The paper performance changes of WS under natural conditions and after treatment were compared to further verify the feasibility of *B. licheniformis* treatment in the pulp and paper industry.

2. Materials and Methods

2.1. Materials and Reagents

The papermaking black liquor comes from the State Key Laboratory of Qilu University of Technology. Wheat straw is acquired from a paper mill in Weifang City, Shandong Province. Experimental medium chemicals such as NaCl, Agar powder, Sodium hydroxide, tryptone, and yeast extract were purchased from Beijing Aobo Biotechnology Co., Ltd. (Beijing, China). All chemical reagents are analytical grade and no further purification is required. The WS with length of 3–5 cm, chemicals, and instruments were sterilized under 121 °C for 20 min using a vertical pressure steam sterilizer.

2.2. Test Scheme

2.2.1. Isolation and Identification of Strains

1. Screening and isolation of strains

The 100 µL of paper black liquor was dropped into 50 mL sterilized Luria-Bertani liquid medium and incubated overnight in a 37 °C 200 rpm shaker. After that, 100 µL bacterial solution was absorbed and diluted 1000 times step by step in a Luria-Bertani solid medium. The single colony was selected for enrichment culture after overnight in a biochemical incubator at 37 °C. Finally, the isolated colonies were cultured, preserved, and weighed for follow-up identification and experimental treatment.

2. Treatment of wheat straw using shaking flask fermentation of strain

For the shake flask containing the medium, 10 g sterilized dried WS was added. The medium formula is yeast extract 5 g/L, peptone 10 g/L, and sodium chloride 10 g/L, with a few drops of 2M NaOH solution to adjust the pH of the Luria-Bertani liquid medium to 7 ($\pm$0.2). The strain was not added in the control group. All the preserved strains were acquired for activation culture. Then, 50 μL of activated 24 h strain was inserted into the culture medium, respectively, in the experimental group and they were put together with the control group into a 37 °C 200 rpm shaker for 48 h. All medium elements were analyzed to determine the target strain. An X-MET8000 Expert X-ray fluorescence spectrometer (Oxford Company, UK) was used as the instrument: detection element range: 12 Mg–92 U, (Rh) anode target; testing voltage: 50 kV; current settings: 200 μA; testing time: 60 s; operating temperature: −25–50 °C; and method of measurement: fast screening- He 34 mm.

3. Identification of strain

The genomic DNA of the target bacteria was extracted using the Sangon Biotech Bacterial Genomic DNA Rapid Extraction Kit (universal) and it was used as a template to amplify by PCR with universal primers 27F (5′-AGAGTTTGATCCTGGCTCA-3′) and 1492R (5′-CTACGGCTACCTTGTTACGA-3′). The PCR products were detected using 1% agarose gel electrophoresis and recovered with an AxyPrep DNA gel recovery Kit. Finally, the products were sequenced using an ABI3730-XL. The Blast program from the NCBI website was used to compare the spliced sequence files with the data in NCBI's 16 s database to obtain the information of the species with the most similar sequences, which was the identification result.

2.2.2. Characterization of Basic Physical and Chemical Properties of WS

The WS samples under different treatment conditions (native, control, treated) were cut into 0.5 cm × 0.5 cm and fixed on the sample table. The scanning electron microscope images of the external surface of the WS were obtained using a scanning electron microscope (TM4000Plus, Hitachi, Tokyo, Japan, acceleration voltage 15 kV). The EDX spectra of Si content and distribution on the outer surface of WS were obtained by combining a Model 550I EDS Detector Power and a TM4000Plus scanning electron microscope.

A Fourier transform infrared spectroscopy (Bruker, Tensor 37, Karlsruhe, Germany) attached to an Attenuated Total Reflection (ATR) was used to record the spectral curves of different WS. An ATR probe was used to scan the outer surface of the wheat straw 32 times in the spectral range of 4000–700 cm^{-1} at a temperature of 25 °C and a resolution of 4 cm^{-1}. The external surface of the WS was irradiated by an ESCALAB 250 Xi X-ray photoelectron spectrometer using monochromatic Al Ka to obtain X-ray photoelectron spectroscopy (XPS) data.

The static water contact angle (CA) was completed by an LSA100 contact Angle instrument (LAUDA Scientific, Germany). During the measurement, 5 μL water droplets were hit on the surface of the WS and the CA was measured after 5 s. The CA was measured at least three times at different locations of each sample and the results were averaged.

2.2.3. Paper Forming Experiment of WS

The paper properties of WS under different conditions were analyzed and compared. The tensile index, tear index, and ring compression index of the paper were determined according to the international standards (ISO 1924-2-2008, ISO/DIS 12192, ISO 1974). In addition, the content of the Si elements in black liquor was determined using an Inductively Coupled Plasma Emission Spectrometer (ICP-OES) (Agilent 5110, RF power: 1.20 KW, plasma flow: 12.0 L/min, auxiliary flow: 1.50 L/min, nebulizer flow: 0.70 L/min, sample uptake delay: 15 s, instr stabilization delay: 15 s, replicate read times: 5 s, replicates: three times).

3. Results

3.1. Isolation, Screening, and Identification of Strains

After the WS was treated by seven isolated strains (Figure 1a, No. BL1-7) under experimental conditions, the XRF results showed that only the culture medium treated with a BL-03 strain contained the Si element (Figure 1b). As the Si layer in the wheat straw belongs to a part of the cuticle, BL-3 was used in subsequent experiments to verify its removal effect on the WS cuticle. The other elements displayed in the culture medium were all from the strain itself under control conditions. From the in-depth analysis of BL-3, it is seen as a thin rod under the microscope (Figure 1c). The bacterial colonies of BL-3 were round, with diameters from 2.0 to 5.0 mm and a thickness of 2.0 mm. The surface was white and transparent with irregular protruding and uneven edges (Figure 1d). Finally, the comparison between 16SrDNA gene sequence of BL-3 (NCBI Genbank accession number: OP782682) and the database in NCBI showed that the homology between BL-3 and *Bacillus licheniformis* was 99% (NCBI: NR_118996. 1). According to the results of the phylogenetic tree, the BL-3 strains and several strains of *Bacillus licheniformis* (*B. licheniformis*) were clustered into one branch (Figure 1e). Based on the phylogenetic analysis of the 16SrDNA gene sequence, combined with the experimental results of the morphological characteristics, the BL-3 strain can be identified as *Bacillus licheniformis* (*B. licheniformis*).

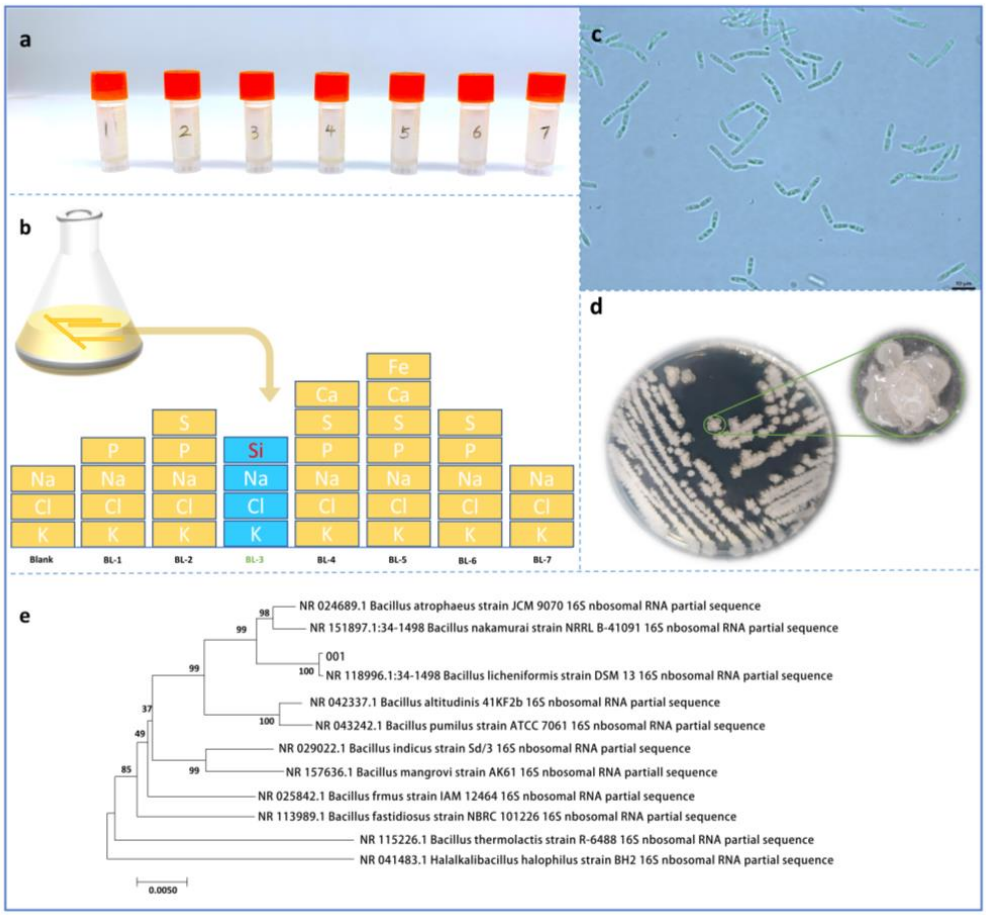

Figure 1. (**a**): Seven strains were screened from black liquor and preserved in glycerol; (**b**): Elements in the medium of wheat straw treated by different strains and the element Si was shown by BL-3. (**c**): Microscopic image of strain BL-3 shows its cell morphology. (**d**): Single colony morphology of strain BL-3 on plate medium. (**e**): The rooting phylogenetic tree based on the 16SrRNA sequence of BL-3 strain and related bacteria.

3.2. SEM-EDX

The SEM images of the WS raw material, the blank control group and the experimental group are shown in Figure 2. The outer surface of the untreated WS is smooth, with stomata and some horny bumps. The vascular bundles and some shallow furrows on the outer surface of the untreated WS are clearly shown. Some damage also occurred on the outer surface of the sample due to external forces, such as the loss of some horny bumps, which was caused by the collection or preparation of samples (Figure 2A). The outer surface of the native WS is nearly uniform, smooth and dense, which covered with a thicker waxy layer. Meanwhile, there is no new damage on the outer surface of the WS in the blank control group after the same time of treatment (Figure 2B). After treatment with *B. licheniformis* in the logarithmic growth stage, the outer surface of WS was no longer smooth and the damage degree was different from that caused by mechanical force. This is mainly manifested by the tearing of the previously flat outer surface (Figure 2C). The magnification continued to observe the external surface of the WS; it was clear that there was no significant difference between the external surface of native WS (Figure 2D) and the outer surface of WS treated with a medium for 48H (Figure 2E). However, the outer surface of the WS treated by *B. licheniformis* was severely damaged. The original dense surface was torn and some obvious dents were exposed on the new surface (Figure 2F) [25].

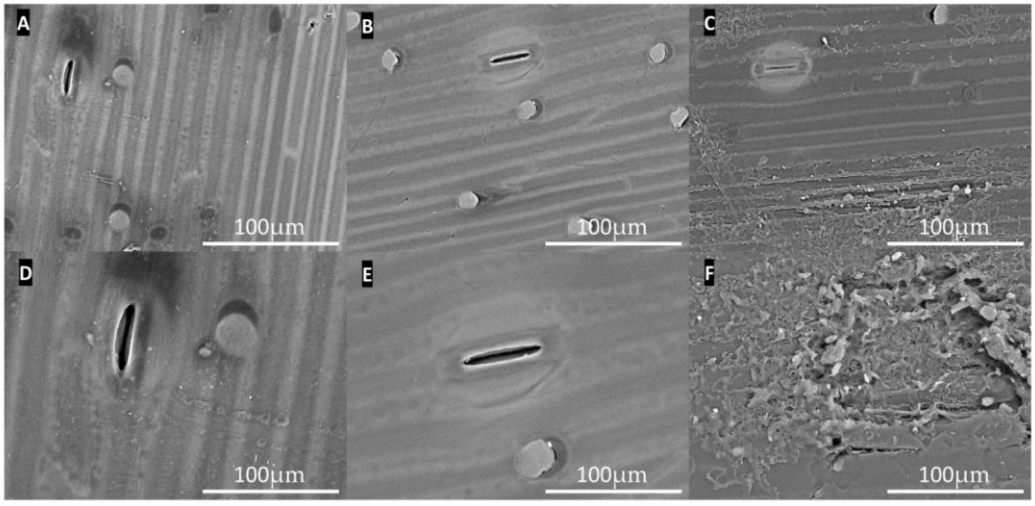

Figure 2. SEM images of WS under different processing conditions. (**A**,**D**): The outer surface is magnified by ×500 and ×1000 under natural conditions. (**B**,**E**): The outer surface is magnified by ×500 and ×1000 in control group. (**C**,**F**): The outer surface of WS treated 48 h by *B. licheniformis* is magnified by ×500 and ×1000.

As mentioned earlier in this paper, Si elements are mainly distributed on the outer surface of WS. In order to further analyze the effect of BL treatment on the outer surface of WS, the distribution of the Si element on the surface of WS was observed by a ×500 energy spectrometer. The results showed that the distribution of silicon in the outer surface was dense in the native WS (Figure 3a). The treatment of the medium did not affect the distribution of elements on the outer surface of WS (Figure 3b). The distribution of Si elements on the outer surface of WS became very sparse after *B. licheniformis* treatment (Figure 3c), indicating that the outer surface of WS was damaged after *B. licheniformis* treatment and the Si layer was affected. This result precisely explained the phenomenon of Si element in medium after the WS was treated by *B. licheniformis*.

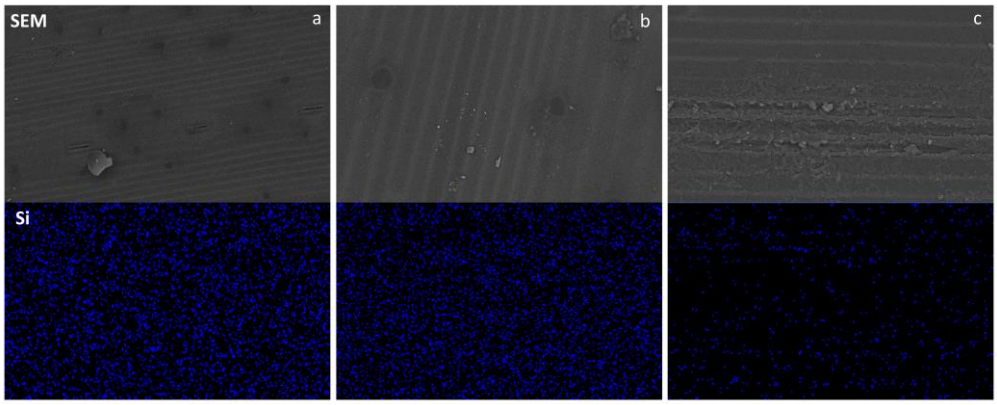

Figure 3. Distribution of Si element on the outer surface of wheat straw under different conditions. (**a**) WS without any treatment, (**b**) WS treated with LB medium for 48 h, (**c**) WS treated by *B. licheniformis*.

Figure 4 shows the analysis of total element content (C, O, Si) on the outer surface of WS magnified by 500 times. It can be seen from the table that after *B. licheniformis* treatment, the Si element content on the outer surface of the WS decreased by 80%. Combined with the above, the reason for this result may be that *B. licheniformis* destroys the outer surface of the WS and makes its Si layer fall off, thus reducing the silicon content of the outer surface of the WS. This is also the reason why *B. licheniformis* was selected to treat WS in the first place.

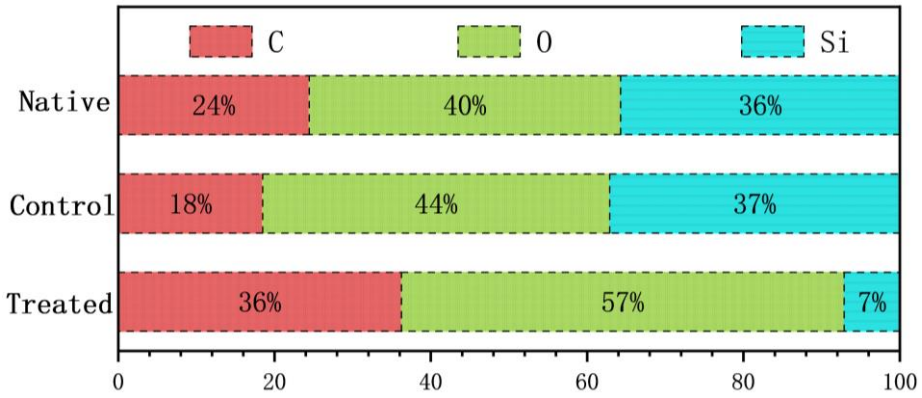

Figure 4. The contents of Si elements on the outer surface of WS under different conditions.

3.3. FTIR, XPS Spectra, and Contact Angle

The XPS spectra of WS under three conditions have obvious C1s, O1s, and Si2p peaks (Figure 5A). In addition, from the core energy spectra of C1s (Figure 5B,C), it can be seen that C1s can be accumulated into four Gaussian peaks, the values of which are 284.8 eV, 286.3 eV, 287.9 eV, and 289.0 eV, respectively, corresponding to C-C/C-H, C-O, C=O, and COOH groups [26]. The content of C-C/C-H groups on the WS surface is significantly higher than that of other three C-binding groups (Figure 5B,C). It is reported that C-C/C-H peak corresponds to aliphatic and aromatic carbon bonds, which come from carbon extracts [27]. It shows that the main substances on the outer surface of WS are long-chain aliphatic hydrocarbons with C-C/C-H chemical bond frames. The C1s (C-O) come from all wood compounds, especially cellulose and hemicellulose. The C1s (C=O) peak represents the bonding of carbon atoms in the ketone group or two oxygen atoms in cellulose and hemicellulose [28].

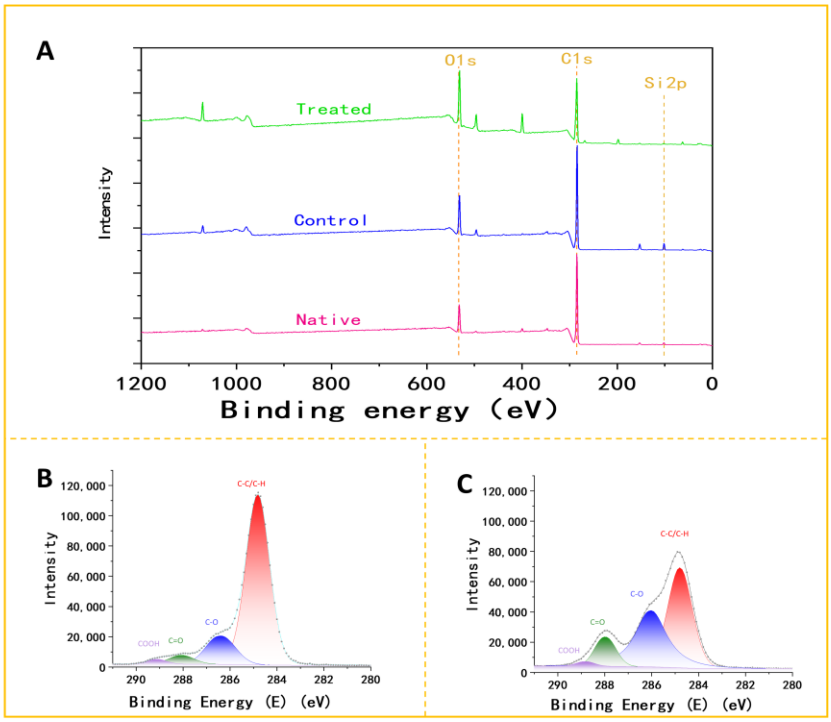

Figure 5. XPS spectrum of outer surface of WS under different treatment conditions, (**A**) full spectrum of XPS under three conditions; (**B**) the C1s spectrum of the untreated WS; (**C**) the C1s spectrum of WS after *B. licheniformis* treatment.

Through the analysis and calculation of XPS spectral data using Thermo Avantage, the outer surface atomic compositions of three kinds of WS are obtained and the oxygen-carbon ratio (O/C) is calculated (Table 1). It is reported that the theoretical O/C ratio of cellulose and hemicellulose is 0.83 and that of lignin is 0.33 [29]. The O/C of WS is only 0.20, which further confirms that the surface of WS is covered with a high layer of wax layers, showing strong non-polarity. In summary, with the treatment of WS by *B. licheniformis*, the peak values of C1s (C-C/C-H) groups decreased significantly, while the peak values of C1s (C-O) and C1s (C=O) groups increased somewhat (Figure 5B,C). The O/C ratio of the WS surface increased to 0.32, close to the theoretical ratio of lignin. The results showed that after *B. licheniformis* treatment, the aliphatic compounds (waxy layer) on the outer surface of WS were removed and the fibrous tissue inside was exposed.

Table 1. The outer surface of O/C ratio and C1s relative peak area for WS.

Sample	C1s (%)	O1s (%)	O/C	C1: C-C/C-H (%C1s)	C1: C-O (%C1s)	C3: C=O (%C1s)	C4: COOH (%C1s)
Native	82.94	17.06	0.21	74.56	22.03	1.84	1.57
Control	86.02	13.98	0.16	78.32	18.29	1.84	1.55
Treated	75.92	24.08	0.32	42.55	43.46	10.01	3.98

The change of the FTIR spectrum before and after WS treatment further confirmed the obvious structural difference between them. The typical FTIR spectrum of the sample before and after treatment is shown in Figure 6.

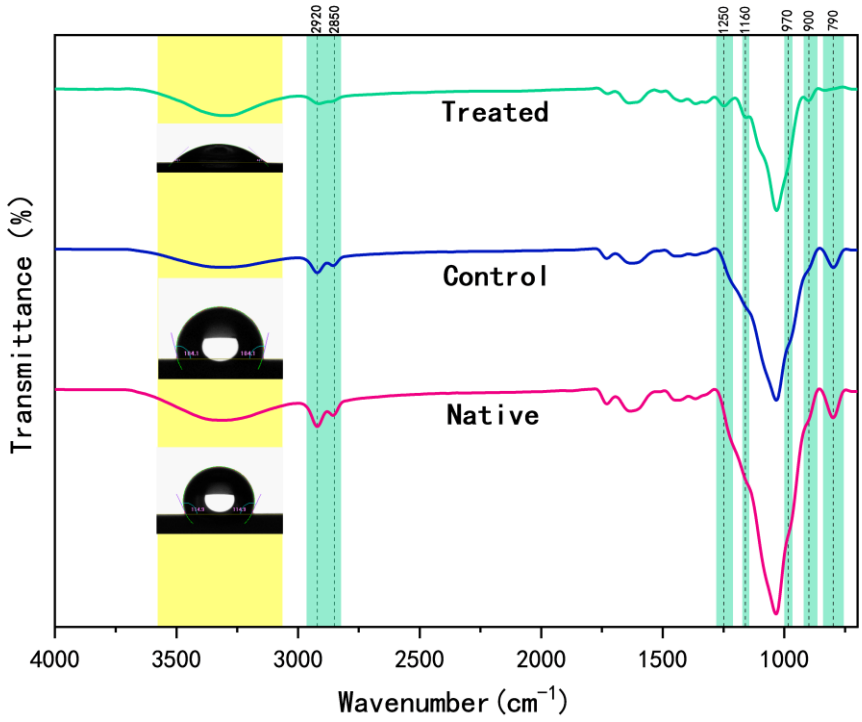

Figure 6. FTIR was used to measure the functional groups of WS outer surface and the average contact angle of WS outer surface under different conditions.

The intensities of all the peaks of WS components in the outer surface spectra of untreated and control are almost the same. These results showed that the medium hardly interfered with the evaluation of the effect of *B. licheniformis* on WS treatment. The infrared spectrum of the native and control WS in Figure 6 shows two strong and sharp peaks at 2920 cm^{-1} and 2850 cm^{-1}. These two peaks are classified, respectively, as asymmetrical and symmetrical extension of the CH_2 group, which includes the fatty fraction of most waxes, respectively [30]. However, the spectrogram treated by *B. licheniformis* showed that the sharp peak at 2920 cm^{-1} and 2850 cm^{-1} was significantly reduced and turned into a weak circular band (Treated). The formation of the remaining passivation peak may be caused by the CH_2 group in lignin and polysaccharide in WS [31]. In addition, a peak was observed at 790 cm^{-1} and 970 cm^{-1} in the WS outer surface spectra of untreated (Native) and control (Control), which is more likely to be attributed to the Si-C stretching vibration and Si-O stretching vibration [32,33]. This indicates that there is a portion of silicide in the outer surface of WS. The WS spectrum (Treated) shows that the peaks at these two places are reduced to corresponding weak shoulder peaks. This phenomenon is believed to be caused by the removal of part of the material containing silicon during *B. licheniformis* treatment, which can also be confirmed by the SEM-EDX analysis of the WS above. Further comparison of the spectral changes of WS before and after treatment showed that the intensity of several typical peaks of lignin and polysaccharide (cellulose and hemicellulose) increased significantly. The intensity of the sharp band at 900 cm^{-1}, which is characteristic of β-glycosidic bond linkages between the sugar units and is absent in lignin spectra, increased significantly after the *B. licheniformis* treatment [12]. In addition, WS spectra after treatment also showed peaks at 1250 cm^{-1} and 1160 cm^{-1}, with the former more likely to be attributed to C-O stretching of the partial acetyl groups in lignin and hemicellulose [34]. The latter is more likely due to C-O-C deformations in hemicellulose and cellulose or aromatic C-H deformations in clove and guaiac-based units in lignin [30,35]. Hydrogen-bonded hydroxyl groups at 3320 cm^{-1} became sharp in the spectra of WS after being treated. This indicated that there were much more hydroxyl groups in the WS outer surface treated than in the native one. This can also be verified by the contact angle. The contact angle of the surface of the WS can be used as an indicator of the bonding ability (wettability)

of the fiber [36] and the wettability of the substrate is good when the contact angle is less than 90° [10]. Therefore, Figure 6 also shows the changes of water contact angle on the outer surface of WS under various conditions. Due to the presence of Lipophilic extract such as hydrophobic waxes and SiO_2, the outer surface of native WS is quite hydrophobic, with a water contact angle of 104° (±12°). The water contact angle is still roughly within this range after being treated with solution of the medium. However, after *B. licheniformis* treatment, the water contact angle of the outer surface of WS decreased to about 52° (±10°). This shows that the outer surface of WS is hydrophilic at this time. The destruction of WS surface results in the change of wettability difference of external surface. The low surface energy provided by the waxy layer on the outer surface of WS caused its outer surface to be hydrophobic. *B. licheniformis* removed the lipophilic components (mainly waxy) and a part of silica layer on the surface of WS, which exposed lignin and carbohydrates linked to cellulose. Thus, the adsorption capacity of WS to water is enhanced. Improving the wetting agent properties of WS will promote its liquid impregnation pretreatment, thereby reducing drug consumption and processing time.

3.4. Paper Forming Experiment

The papermaking experiments of WS under various conditions were carried out. The results showed that there was no significant difference in the tensile index of any WS paper. The ring compression index of WS decreased and the tear index increased slightly (Figure 7A) after treatment. The ring compression strength of the paper is mainly related to the adhesion between fibers. The ring compressive strength of paper is mainly related to the adhesion between fibers. In particular, the fiber fines in the chemical pulp can strengthen the bonding between fibers in the process of paper fiber forming. The greater the fiber binding force, the closer the fiber binding, resulting in the increase in ring pressure strength [37]. After the treatment of WS by *B. licheniformis*, the effect of alkali on WS is improved. Thus, the content of fiber fines in the pulp is reduced and the void ratio between them is increased, ultimately leading to changes in the performance of the paper. In addition, the ICP-OES results show that the content of Si in the black liquor of WS after treatment is reduced by 30% and the remaining Si should come from a few Si layers connected to the fiber tissue of WS and the wax layer containing silicon that has not been removed. After *B. licheniformis* treatment, the WS still meets the requirements of paper and the Si content is decreased during the whole papermaking process. The silicon interference conditions that are detrimental to alkali recovery may be avoided and the cost of equipment maintenance is reduced.

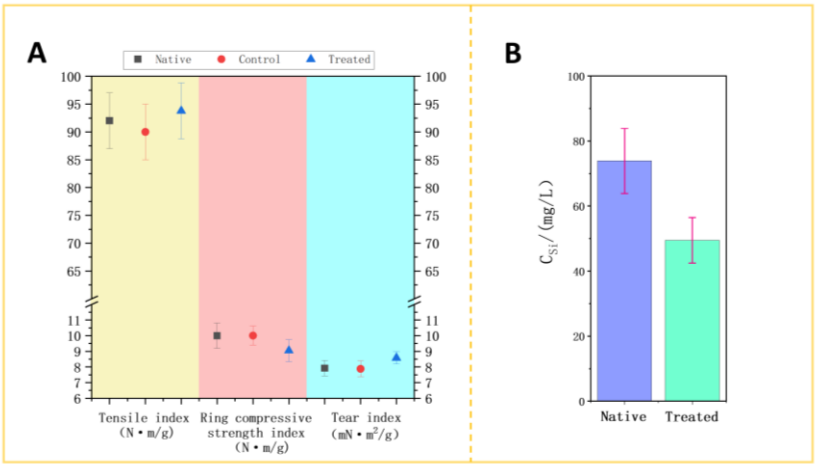

Figure 7. (**A**): The change of paper properties caused by the WS in different conditions. Native: the WS under natural conditions, Control: the WS treated by medium, Treated: the WS treated by *B. licheniformis*; (**B**): the content of Si in black liquor obtained by pulping different WS.

4. Conclusions

In this study, *B. licheniformis BL-3* was isolated from black liquor. This study verified the treatment effect of *Bacillus licheniformis* from papermaking black liquor on the outer surface cuticle of WS. The *B. licheniformis* could remove the hydrophobic waxy layer containing SiO_2 and reduce the Si content in the black liquor. In addition, the original hydrophobic barrier of treated WS was broken, increasing the concentration of -OH on the outer surface and enhancing the wettability of WS. The WS treated by *B. licheniformis* can still be used for pulp and paper and the properties of paper are still acceptable. In summary, this study provides a potential new biological bacterial treatment technology, which may provide new ideas for the follow-up study of WS and provide a basis for other uses.

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Conflicts of Interest: The authors declare that they have no known competing financial interest or personal relationships that could have appeared to influence the work reported in this paper.

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