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Natural Fibers from Vietnam Coffee Grounds as a Potential Reinforcement for Biocomposite Materials

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ABSTRACT

Coffee grounds were a common agricultural byproduct available in large quantities in many countries, containing a relatively high cellulose content of approximately 26.6–31.3%. Extracting cellulose fibers from coffee grounds was therefore both economically and environmentally significant. This study aimed to extract cellulose fibers from coffee grounds using a non-toxic and cost-efficient alkaline hydrogen peroxide extraction method. Scanning electron microscopy (SEM) analysis results indicated that, after being dried, the cellulose fibers tended to aggregate into bundles, with no individual fibers observed. The extraction yield was found to be 63.24%. Fourier transform infrared (FTIR) spectroscopy analysis revealed absorption peaks at wavenumbers corresponding to O-H, C-H, and C-O-C group vibrations, characteristic of the chemical structure of cellulose. The crystallinity index determined by X-ray diffraction (XRD) technique of the extracted cellulose fibers was 38.9%, higher than that of the raw coffee grounds. Thermogravimetric analysis (TGA) result indicated that the thermal stability of the obtained cellulose fibers was relatively lower than that of the coffee grounds. The obtained cellulose fibers have potential application as a reinforcing agent for biocomposite materials.

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1. Introduction

Agriculture plays a crucial role in Vietnam's economy, with coffee being one of its key agricultural export products. According to the Ministry of Agriculture and Rural Development and VICOFA (Vietnam Coffee and Cocoa Association), Vietnam's coffee production for the 2023-2024 crop year is approximately 1.47 million tons, with about 1.43 million tons for export [1, 2]. On average, each cup of coffee requires 20 - 25 g of coffee beans. After brewing, coffee grounds account for approximately 40-50% of the initial weight of the coffee beans [3]. Currently, most coffee grounds are either used for low-value purposes or discarded into the environment, leading to water and soil pollution and disrupting soil microorganisms [3, 4]. Utilizing this abundant agricultural waste to produce high-value materials having potential applications in various fields holds significant scientific and practical importance. It helps address the environmental pollution caused by coffee grounds while also offering economic benefits.

The coffee roasting process typically occurs at temperatures ranging from 180 – 240 °C, and when the coffee is brewed (with hot water at around 90 °C), neither the coffee beans nor the cellulose in the coffee grounds undergo thermal degradation [5]. Coffee grounds contain components such as: moisture content below 10%, approximately 7.83% inorganic matter, about 2.87% fats and oils, around 18.07% fiber, 17.32-18.29% lignin, 20.92% hemicellulose, and 26.60-31.26% cellulose [4-6]. Depending on the soil conditions and the type of coffee, the chemical composition of coffee grounds can vary in their proportions.

Cellulose is the most abundant natural materials, with an annual production of 10^{11}–10^{12} tons/year [7]. It is a polysaccharide with the molecular formula $(C_6H_{10}O_5)_n$, composed of numerous β-glucose units

linked by β-1,4-glycosidic bonds, forming unbranched chains. As an inexpensive, biodegradable, and renewable polymer, cellulose is fibrous, durable, insoluble in water, and being a supporter for the structural integrity of plant cell walls. It has a semi-crystalline structure with notable properties, including high crystallinity, large surface area, non-toxicity, high mechanical strength, hydrophilicity, low density, flexibility, and the ability to form networks [4]. However, it is incompatible with hydrophobic polymers and has poor water resistance [5]. Cellulose fibers act as reinforcing agents effectively enhancing mechanical properties of biocomposite materials due to their strong hydrogen bonding network within the polymer matrix [8, 9]. With the good distribution, dispersion, and interfacial adhesion with polar natural polymer matrix, cellulose fibers can act as a nucleating agent enhancing the crystallinity of polymer matrix leading to the improved mechanical properties of biocomposite materials based on biodegradable polymers.

Methods used for extraction of cellulose fibers from biomass materials typically involve a combination of chemical and mechanical treatments to isolate cellulose from lignocellulosic materials by eliminating the lignin and hemicellulose, two other components found in coffee grounds [9]. Chemical treatments are used to remove lignin and hemicellulose, improving the efficiency of cellulose fibers isolation. Common chemical treatments include: alkaline treatment (using alkaline solutions like NaOH or KOH to hydrolyze and break down lignin and hemicellulose structures, thereby releasing cellulose), bleaching treatment (using oxidizing agents such as NaOCl, H_2O_2 to remove residual lignin after alkaline treatment to obtain cellulose fibers with lighter color), and acid hydrolysis (using strong acids like H_2SO_4 or HCl to hydrolyze amorphous region of cellulose resulting in cellulose nanocrystals) [5, 9]. Mechanical treatments are then applied to further separate cellulose bundles into individual fibers after chemical treatment [5, 9]. Common mechanical treatments include: ball milling (applying high-impact forces between balls to separate cellulose bundles), ultrasonic treatment (using ultrasonic waves to break hydrogen bonds between cellulose fibers), and steam explosion (using high-pressure steam to disrupt cellulose structure) [5, 9]. In addition, enzymatic treatment (using enzymes to degrade lignin and hemicellulose, releasing cellulose) and ionic liquid treatment (using ionic liquids to dissolve cellulose and separate it from lignin and hemicellulose) were also used to extract cellulose fibers from biomass materials [9]. In this study, the simple, non-toxic alkaline hydrogen peroxide treatment was used to extract cellulose fibers from agricultural waste coffee grounds. The obtained cellulose fibers have potential application as a reinforcing agent to improve the mechanical properties of biocomposite materials based on natural polymers.

2. Materials and Methods

2.1. Materials

Coffee grounds were collected from coffee shops in Rach Gia city, Kien Giang province, Vietnam. Sodium hydroxide (NaOH), hydrogen peroxide (H_2O_2), and cellulose filter paper (Whatman®, Grade 1) were purchased from Sigma-Aldrich.

2.2. Extraction of cellulose fibers from coffee grounds

The extraction of cellulose fibers from coffee grounds was carried out by alkaline hydrogen peroxide treatment. After collection, the coffee grounds were sieved and dried at 70 °C for 24 hours to remove moisture. Next, a mixture of coffee grounds and 11% (v/v) H_2O_2 solution with a ratio of 1:10 (w/w) was conducted. 8 wt% NaOH solution was then gradually added to the mixture to adjust the pH to 11. Subsequently, the mixture was stirred continuously for 4 hours at 80 °C. Finally, the mixture was filtered to obtain the solid portion (cellulose fibers), while the filtrate containing hemicellulose, lignin, and non-cellulosic components eliminated during the extraction reaction was discarded. To remove residual chemicals during filtration process, the solid was then centrifugated and repeatedly washed with distilled water until the pH reached 6 - 7. The extracted cellulose fibers were then freeze dried. The steps of extraction process of cellulose fibers from coffee grounds are illustrated in Figure 1.

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Figure 1. *Extraction process of cellulose fibers from coffee grounds.*

2.3. Characterization

2.3.1. Extraction yield of cellulose fibers

The extraction yield of cellulose fibers was calculated using Eq. (1) below [10]:

$$\text{H (\%)} = \frac{m_1}{m_0} \times 100 \quad (1)$$

Where H (%) is the extraction yield, m_1 is the weight of the final dried extracted cellulose fibers (g), and m_0 is the initial weight of raw coffee grounds (g).

2.3.2. Scanning electron microscopy

The surface morphologies of coffee grounds and obtained cellulose fibers were observed using a scanning electron microscope (FESEM model S-4800 HITACHI, Japan) at the R&D Center, SHTP – Ho Chi Minh City High-Tech Park, with an accelerating voltage of 10 kV. The samples for observation were prepared by mounting on a metal frame with carbon tape. Before measurement, all samples were coated with a Pt layer to ensure electrical conductivity.

2.3.3. Fourier transform infrared spectroscopy (FTIR)

FTIR spectroscopy was employed to investigate the chemical structure of the coffee grounds and cellulose fibers. Measurements were conducted using a NICOLET 6700-Thermo (USA) spectrometer at the University of Finance - Marketing in Ho Chi Minh City. Samples were analyzed over a wavenumber range of 4000 to 500 cm^{-1}, with a resolution of 4 cm^{-1} and 64 scans per sample.

2.3.4. X-ray diffraction analysis (XRD)

The crystalline structure of coffee grounds and obtained cellulose fibers was determined using an EMPYREAN X-ray diffractometer from PANalytical (Netherlands) at the University of Finance - Marketing in Ho Chi Minh City, Vietnam. Samples were analyzed over an angular range of 5–80°, utilizing a CuKα X-ray source (α = 1.54056 Å) at 40 kV and 45 mA. The crystallinity index was calculated using an empirical Eq. (2) [11]:

$$CI\ (\%) = \frac{I_{200} - I_{am}}{I_{200}} \times 100 \quad (2)$$

Where *CI* (%) is the crystallinity index, I_{200} is the maximum diffraction intensity of the (200) plane at a 2θ angle between 20° and 23°, and I_{am} is the minimum diffraction intensity representing the amorphous region of cellulose, which is taken at a 2θ angle between 18° and 20°.

2.3.5. Thermogravimetric analysis

The TGA analysis of coffee grounds and extracted cellulose fibers was measured using a TGA-DSC thermal gravimetric analyzer (STA PT 1600, Linseis, Germany) at the Vietnam National University Ho

Chi Minh City University of Science, Ho Chi Minh City, Vietnam. All samples were heated from 30 °C to 600 °C at a heating rate of 10 °C per minute with an Argon gas flow of 10 mL/min.

3. Results and Discussion

3.1. The extraction yield of cellulose fibers from coffee grounds

The extraction yield of cellulose fibers from coffee grounds using the alkaline hydrogen peroxide treatment was 63.24%, calculated using Eq. 1.

3.2. Surface morphologies of coffee grounds and cellulose fibers

As observed in Figure 2, compared to the coffee grounds with black color, the extracted cellulose fibers appeared light brown, suggesting that the alkaline hydrogen peroxide treatment effectively removed the amorphous impurities and non-cellulosic components such as hemicellulose and lignin from the raw coffee grounds. The surface morphologies with various magnifications of coffee grounds (a,b) and cellulose fibers (c,d) were shown in Figure 3. Figure 3a indicates that the untreated coffee grounds had a porous, heterogeneous surface structure, layered and covered with organic compounds such as lignin, hemicellulose, and other impurities.

After chemical treatment, the morphological changes were evident in the coffee grounds. The treated sample surface appeared smoother and more homogeneous, resulting from the removal of non-cellulosic components, as well as partial removal of hemicellulose and lignin from the raw coffee grounds [12]. The purification process also revealed distinct fiber bundles. However, after drying process, the cellulose fibers exhibited a tendency to accumulate into larger bundles, likely due to the formation of hydrogen bonds between hydroxyl groups on adjacent cellulose molecules.

Figure 2. *(a) The coffee grounds and (b) the extracted cellulose fibers.*

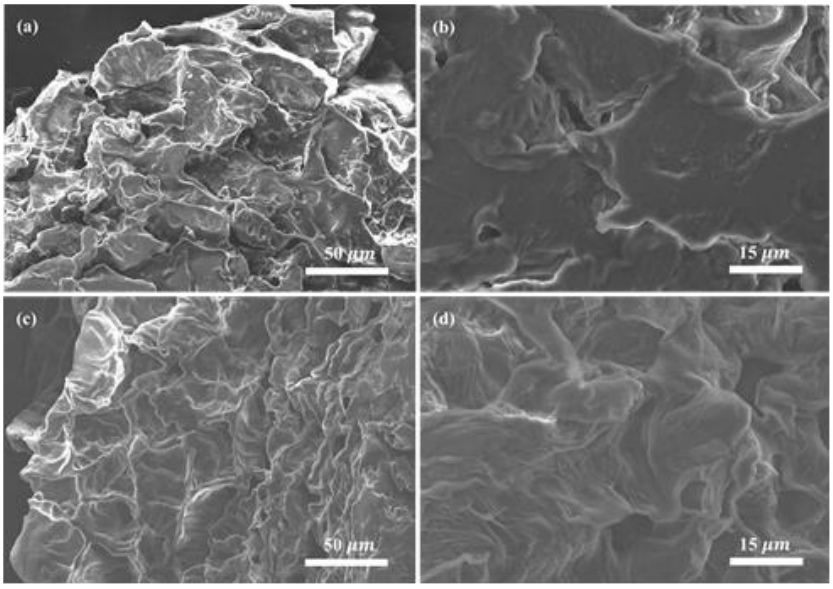

Figure 3. *SEM images of coffee grounds (a, b) and cellulose fibers (c,d) at different magnifications.*

3.3. Fourier transform infrared spectroscopy (FTIR)

The chemical structure of coffee grounds and obtained cellulose fibers was analyzed using FTIR spectroscopy. Figure 4 presented that cellulose fibers and coffee grounds display similar characteristic absorption peaks.

The FTIR spectrum of coffee grounds showed absorption peaks associated with the vibrations of functional grounds in the chemical structure of cellulose, lignin, and hemicellulose [13]. In the absorption range of 3500–3260 cm^{-1}, with a peak at 3429 cm^{-1}, there was a stretching vibration of hydroxyl (O-H) groups in cellulose molecules. Stretching vibrations of the (C-H) bond were observed at the 2919 cm^{-1} peak, corresponding to the alkyl groups of cellulose. Additionally, the peak at 2848 cm^{-1} reflected the asymmetric stretching vibration of (C-H) bonds in methyl and methylene groups in the chemical structure of lignin and hemicellulose. The acetyl and ester groups, associated with the stretching vibration of the (C=O) bond in hemicellulose, pectin, or carboxylic acid groups of ferulic and p-coumaric acids in lignin, were identified at the 1741 cm^{-1} peak. Absorption peaks observed in the range of 1639–1650 cm^{-1} were attributed to the stretching vibration of hydroxyl (O-H) groups from absorbed water. The stretching vibration of (C=C) bonds, corresponding to aromatic rings in lignin, appeared at 1537 cm^{-1}. Meanwhile, the peak around 1380 cm^{-1} corresponded to (C-H) stretching vibrations in cellulose structure. The absorption at approximately 1037 cm^{-1} was identified as the pyranose ring stretching vibration (C-O-C) of cellulose. Finally, a small peak around 897 cm^{-1} corresponded to the bending vibration of the (C-H) bond and the stretching vibration of β-glycosidic bonds in cellulose. These results were consistent with previously reported study, as published by Sataporn Malarat et al. [5].

Compared to the FTIR spectrum of untreated coffee grounds, the FTIR spectrum of cellulose fibers extracted by alkaline hydrogen peroxide treatment presented the similar characteristic absorption peaks. These results demonstrated that upon chemical treatment, the chemical structure of cellulose was intact. However, the FTIR spectrum of the cellulose fibers showed absorption peaks at 1741 cm^{-1} and 1537 cm^{-1}, corresponding to the vibrations of (C=O) and (C=C) bonds, respectively, with reduced absorption intensity compared to those in the FTIR spectrum of raw coffee grounds. This decreased absorption intensity was likely attributed to the partial removal of hemicellulose, lignin, and pectin components in the coffee grounds after the alkaline hydrogen peroxide treatment [14].

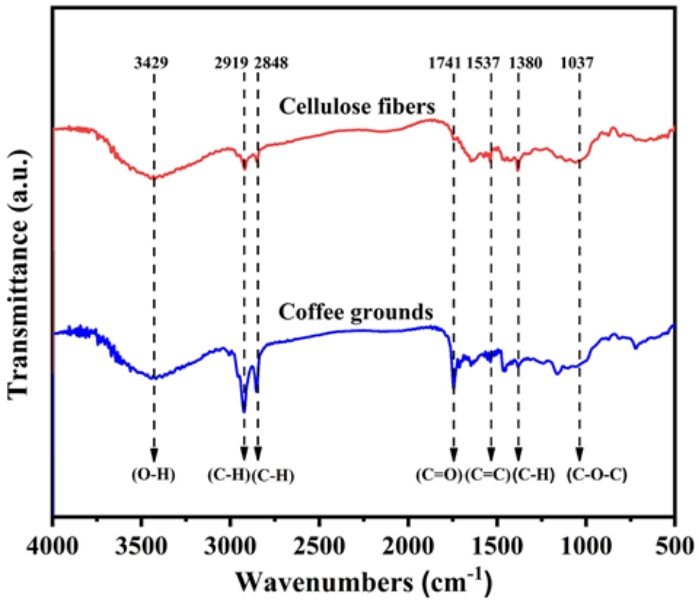

Figure 4. *FTIR spectra of coffee grounds and cellulose fibers.*

3.4. X-ray diffraction analysis (XRD)

The XRD pattern of cellulose fibers showed both crystalline and amorphous regions, while hemicellulose and lignin exhibited an amorphous structure. The XRD patterns of coffee grounds and cellulose fibers were presented in Figure 5.

Figure 5 displayed that XRD patterns of both coffee grounds and extracted cellulose fibers showed characteristic diffraction peaks at 2θ angles of approximately 16.1°, 20.3°, and 34.8°. These peaks corresponded to the lattice planes (110) (peak 1), (200) (peak 2), and (004) peak 3, indicating the cellulose I crystalline structure [15]. Additionally, the XRD patterns revealed that the diffraction intensity of the extracted cellulose fibers was higher than that of the raw coffee grounds. This was attributed to the efficient removal of hemicellulose and lignin, which enhanced the formation of hydrogen bonds and increased the diffraction intensity of the extracted cellulose fibers. This result is consistent with the FTIR result above. The chemical treatment process helped remove some amorphous impurities from the raw coffee grounds, resulting in the disappearance of certain undefined diffraction peaks in the XRD pattern of obtained cellulose fibers (Figure 5).

The crystallinity index of the obtained cellulose fibers and raw coffee grounds was 38.9% and 23.3%, respectively. The increased crystallinity index of the cellulose fibers demonstrates the efficacy of removal of amorphous non-cellulosic components from the raw coffee grounds upon alkaline hydrogen peroxide treatment. These results were consistent with previous study reported by Yucheng Liu and et al. [16].

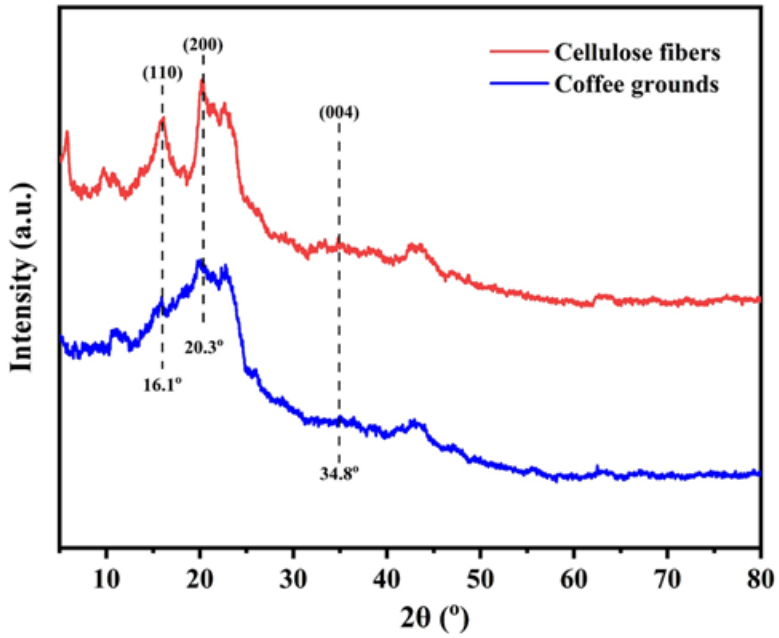

Figure 5. *XRD patterns of coffee grounds and cellulose fibers.*

3.5. Thermal stability

The TGA and DTG curves of coffee grounds and cellulose fibers were presented in Figure 6. The thermal decomposition of the samples showed three main stages of mass loss: (I) below 150 °C, (II) between 150 and 380 °C, and (III) from 380 to 600 °C. Table 1 summarizes the mass loss of both samples. In Stage I, from 30 to 150 °C, both samples exhibited a slight mass loss of less than 11%, attributed to the removal of water vapor and moisture [17]. Stage II, the primary decomposition stage, showed significant mass loss in the temperature range of 150 – 380 °C, with mass loss of 39.7% for coffee grounds and 52.3% for cellulose fibers. The thermal decomposition of hemicellulose began at 150 °C and continued up to 350 °C, while cellulose primarily decomposed within the range of 275 – 350 °C. In contrast, lignin degradation occurred over a broad temperature range of 250 – 500 °C [18]. As seen in Figure 6, the extracted cellulose fibers exhibited lower thermal stability than the raw coffee grounds, likely due to the effective partial removal of hemicellulose and lignin after chemical treatment. In addition, the reduced thermal stability of cellulose fibers may be attributed to the smaller size of

cellulose fibers compared to the raw coffee grounds, which generated a higher surface area that accelerates heat transfer and degradation rate [19]. The final decomposition stage, carbonization, occurred between 380 and 600 °C, with the residual mass after this stage being 38.4% for coffee grounds and 26.5% for cellulose fibers.

These results indicate that the alkaline hydrogen peroxide chemical treatment partially removed lignin from the coffee grounds structure, resulting in cellulose fibers with lower thermal stability.

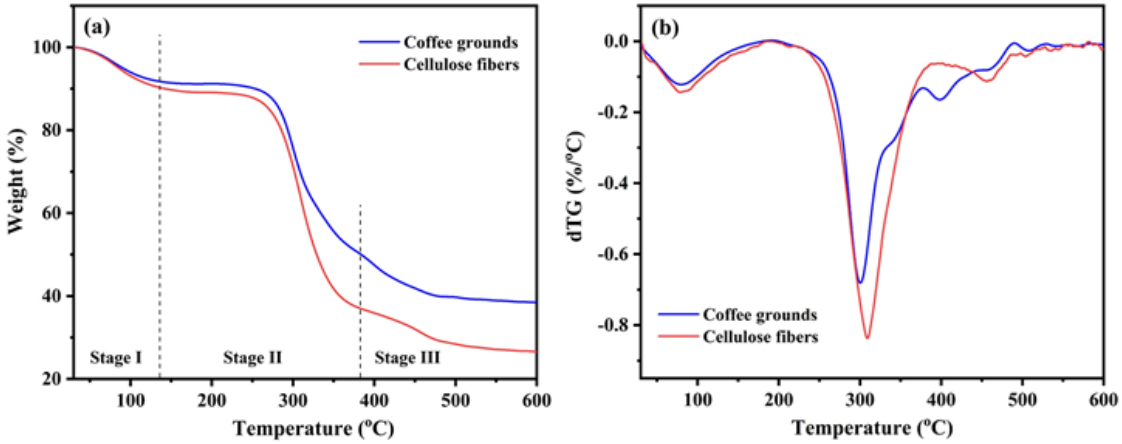

Figure 6. *TGA plots of coffee grounds and cellulose fibers.*

Table 1. *Decomposition temperatures of coffee grounds and cellulose fibers.*

Sample	T_{max} (°C)	Weight loss (%)	Residue (%) at 600 °C
Coffee grounds	300	39.7	38.4
Cellulose fibers	309	52.3	26.5

4. Conclusions

In this work, the cellulose fibers were successfully extracted from coffee grounds, yielding 63.24% using a simple, non-toxic alkaline hydrogen peroxide chemical treatment procedure. The chemical structure of cellulose was intact after chemical treatment. The produced cellulose fibers had a bundle shape and a smoother, more homogeneous surface structure than raw coffee grounds. The physicochemical characteristics of the analyzed product showed that its chemical structure was cellulose with a crystalline structure of cellulose I. The crystallinity index of cellulose fibers (38,9%) was found to be higher than that of coffee grounds (23,3%). With an enhanced crystallinity index and purity, the produced cellulose fibers have the potential to be used as a reinforcement to improve the mechanical properties of biocomposite materials based on natural polymers.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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