Journal of Fiber Bioengineering and Informatics 16:1 (2023) 89–101
Cite as: S. Shao et al.
doi:10.3993/jfbim02201

Extraction of Cellulose Nanofibrils (CNFs) from Pomelo Peel via Hydrothermal Auxiliary Alkali Treatment Combined with Ball-milling

Shuai Shao [a], Xiao-Yun Wang [a], Run-Tao Xia [a], Ju Huang [a], Zhi-Qin Peng [b,*]

[a] *College of Materials Science and Engineering, Zhejiang Sci-Tech University, Hangzhou, China*

[b] *National Engineering Lab for Textile Fiber Materials and Processing Technology (Zhejiang), Zhejiang Sci-Tech University, Hangzhou, 310018, China*

Abstract

The pomelo peel, a by-product of pomelo processing, is predominantly discarded. To fully exploit the pomelo peel resources and contribute to environmental improvement and resource recycling, this study reports a green method for extracting cellulose nanofibrils (CNFs) from pomelo peel through hydrothermal-assisted alkali treatment. Lemon juice is employed for further ball milling treatment to purify the CNFs. The chemical structure, crystallinity, thermal stability, and microstructure of pomelo peel powder and CNFs are analyzed. The study results demonstrate that the combination of alkali treatment, hydrothermal processing, bleaching, and ball milling progressively eliminates hemicellulose, lignin, and other impurities from the pomelo peel, yielding high-quality CNFs. The CNFs retain the natural cellulose structure, with enhanced thermal stability and crystallinity. The diameters of the CNFs range from 2-30 nm. This method employs lower NaOH concentrations than previous studies, resulting in CNFs with higher crystallinity and smaller diameters. This study provides a promising method for enhancing the value-added of pomelo peel and increasing the volume of CNF production.

Keywords: Pomelo peel; Cellulose nanofibers; Hydrothermal; Extraction

1 Introduction

In recent years, the continual advancement of science and technology has dramatically improved pomelo output. However, the disposal of pomelo peel as a byproduct has resulted in the wastage of resources and environmental contamination [1]. To address this issue, researchers have conducted numerous studies on the byproducts of pomelo peel, including the extraction of limonin analogs,

*Corresponding author.
Email addresses: 2935659129@qq.com (Shuai Shao), 3293134784@qq.com (Xiao-Yun Wang), pengqiao6858@126.com (Zhi-Qin Peng).

1940–8676 / Copyright © 2023 Textile Bioengineering and Informatics Society
Mar. 2023

flavonoids, dietary fiber, pectin and pigment [2]. However, the cellulose nanofibrils (CNFs) extraction from pomelo peel has received relatively lesser attention. Cellulose is a linear polymer comprising D-glucose as the basic unit and it is connected by β-1,4' glycosidic bonds [3]. As cellulose is a component of plant cell walls, it exists widely in agricultural wastes. Obtaining cellulose from agricultural waste will be a meaningful way to transform waste into wealth. CNFs are widely used in various fields, such as papermaking [4], chlorine adsorption [5], food [6], conductive composite materials [7]. and more, which is due to their unique interwoven network structure, better biocompatibility, excellent mechanical properties, and larger specific surface area. In addition, due to their high dielectric constant, excellent puncture resistance, superior chemical stability, strong thermal stability, etc. CNFs are also employed in the field of battery diaphragm [8].

CNFs can be obtained through physical, chemical, and biological methods. Biological methods involve processes such as enzymatic hydrolysis [9] and bacterial synthesis [10]. Chemical methods include acid hydrolysis [11], alkali treatment [12, 13], 2, 2, 6, 6-tetramethylpiperidine oxide (TEMPO) oxidation [14], deep eutectic solvent [15], and ionic liquid [16], etc. Physical methods include micro-jet [17], high-pressure homogenization [18, 19], and ball milling [20, 21], etc. However, each method has its limitations and drawbacks. Acid hydrolysis requires a high concentration of acid solution, which is difficult to recycle and can cause equipment corrosion. Biological methods produce pure CNFs but are expensive, time-consuming, and difficult to control. The TEMPO method is expensive and involves toxic reagents [22]. The ionic liquid method is highly selective but difficult to recover. Alkali treatment requires a high concentration of NaOH [23, 24], leading to excessive alkali consumption and equipment corrosion. The steam explosion method can strip cellulose fibers and loosen the cellulose cell wall structure, facilitating nanofibrillation. However, this method produces non-uniform fiber sizes, requiring reagent waste, equipment corrosion, and high energy consumption. Overall, each method has its advantages and disadvantages, and further research is needed to develop more efficient and sustainable approaches for obtaining CNFs.

The hydrothermal method is commonly used to synthesise inorganic compounds due to its ability to create high pressure and tight seal. However, there has been little research on its applicability in cellulose extraction. In reality, during the hydrothermal process, water vapor permeate the biomass raw material and degrade the hemicellulose in the cell wall of the biomass raw material. At the same time, cellulose is minimally affected [25]. Ball milling is a mechanical-chemical process that utilizes mechanical energy to induce chemical and structural changes in materials [26]. Under the strong compression and shear force of ball milling, the connection among hemicellulose, lignin, and cellulose in the cell wall is damaged to a certain extent. Additionally, the structure of the pomelo peel fiber can be opened, allowing NaOH reagents to enter into the fiber and remove hemicellulose and lignin. Moreover, by controlling the parameters of the ball mill, the amorphous area of the cellulose can be destroyed, leaving the crystalline region intact. As a result, ball milling as a pretreatment method can reduce the concentration of NaOH reagents required and the size of the CNFs. Most of the fiber bonding materials are removed, and the cellulose is effectively refined, making it easier to obtain nano-sized cellulose fibers.

In this study, we extract CNFs from pomelo peel by hydrothermal auxiliary alkali treatment combined with ball-milling without using any chemical buffer to further reduced the use of chemical reagents and simplifies the extraction process. This study presented a more sustainable and effective extraction process for obtaining cellulose nanofibers from pomelo peel with higher crystallinity and smaller diameter.