

## Mechanical Fibrillation Capability of Kraft Pulp for Obtaining Cellulose Nanofibers

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### Abstract

Cellulose nanofibers (CNFs), which exist in the plant cell wall, are packed densely with the hemicellulose and lignin matrix to support the mechanical strength of the plant growth. Due to CNF's structural characteristics, it owns excellent features of mechanical strength, lightweight, high specific surface area, and low coefficient of thermal expansion. These outstanding features made CNFs potential raw materials in various areas and were appealing to use as reinforcement in nanocomposites. The CNF fabrication is processed by chemical and mechanical methods, including isolating cellulose from raw material and disintegration cellulose into nanoscale fibers, CNFs. In order to upscale and promote the practical usage of CNFs, cost-effective and efficient methods are needed.

In order to obtain high aspect ratio fibers that induce greater mechanical strength, the mechanical disintegration methods were focused for interest. In previous studies, the dried commercial pulp could be disintegrated with a grinder and homogenizer with extensive energy input. Later, it was reported that uniform CNFs could easily be obtained from the never-dried pulp with one grinder treatment pass. The easiness in mechanical fibrillation of pulps was thought to be attributed to the never-dried pulps in yet hornified and yet formed irreversible hydrogen bonds during the drying process; hence the pulp could be easily disintegrated. However, it was found that the disintegration of commercial pulp is hard to process with excessive mechanical treatment, even in its never-dried state. It was then considered that if it was the manufacturing process of the raw material pulp for making cellulose nanofibers affects the nanofibrillation process. Therefore, the motivation of the current study is to understand whether the pulp-making process influences never-dried pulp fibrillation.

To identify the issue of why the never-dried state pulp is still hard to fibrillate into uniform CNFs, (1) the most common pulp-making process, kraft cooking, was used as the method for preparing the never-dried pulp, (2) the study hopes to find out the key to prepared pulp that is suitable to prepared CNFs with ease. Kraft cooking was chosen as the study object since it is the world's dominant pulping method, and its uptake is the most abundant pulp raw material, kraft pulp. This study aims to optimize kraft cooking for fabricating never-dried pulp, which is feasible for mechanical fibrillation at ease.

In Chapter 2, the effect of kraft cooking on the never-dried pulp nanofibrillation process was conducted by comparing the nanofibrillation capability between ambient-made wise-based pulp and the kraft cooking processed kraft-based pulp. The difference in the nanofibrillation shows that the kraft-based pulp has less fibrillation tendency. Thus, kraft cooking hinders the nanofibrillation process; even the never-dried pulp was adopted.

In Chapter 3, the wise-based pulp, which is more easily fibrillated, was prepared to identify the influence of high temperature and alkaline kraft cooking conditions on the pulp nanofibrillation. The results prove that the agglomerations of CNFs happened during the cooking conditions of kraft cooking and are confirmed with the simulation of CNFs at high temperatures and alkaline autoclaved conditions.

In Chapter 4, three stages of experiments were conducted to adjust the kraft cooking conditions for better pulp nanofibrillation capabilities. The temperature variation under similar energy input was examined for their influence on nanofibrillation capability. Later on, the different cooking energies with milder temperatures and the hydrothermal cooking with milder temperature was examined subsequently. The limitation to modified kraft cooking for making pulp with better fibrillation capability of wise-based pulp is hard was confirmed.

The main findings of this study are summarized as follows: (1) The kraft cooking will hinder the pulp nanofibrillation process even in its never-dried state. (2) The kraft cooking conditions, high temperature, and high pressure were determined to deteriorate the nanofibrillation capability of the never-dried pulps. Also, the direct agglomeration of cellulose nanofibers under kraft cooking conditions is proved. (3) The nanofibrillation of the pulp will not be affected by the temperatures variation under similar energy input (4) The matrix of hemicellulose and lignin remains during the pulp cooking did not stand as a blocking role to prevent the aggregations among cellulose microfibrils. The agglomerations between CNFs happen even if a matrix exists during the pulping process. (5) The kraft cooking process is necessary and prominent in the paper and pulp industry, but it was not suitable for producing pulp for making CNFs. The development of the pulp-making process oriented for CNF fabrication is needed in the future.