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# Optimization of high pressure homogenization parameters for the isolation of cellulosic nanofibers using response surface methodology

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A B S T R A C T

Response surface methodology (RSM) was used to determine the effects of high pressure homogenization (HPH) parameters (pressure and number of cycles) on the isolated yield, crystallinity, and diameter of kenaf bast cellulose nanofibers (CNF). Central composite design of experiments was utilized to determine the optimal pressure and number of cycles of HPS for the highest CNF yield, crystallinity, and lowest CNF diameter. The linear terms for the pressure and homogenization cycles had significant effects on the CNF yield, crystallinity, and diameter, whereas the interaction between the pressure and homogenization cycles had a significant effect on the CNF crystallinity. The optimized experimental conditions for the HPH process were a homogenization pressure of 56 MPa, 44 P homogenization cycles, and a 0.1 wt% fiber suspension concentration. Under these conditions, the isolated kenaf bast CNF yield was 89.9% with 56.5% CNF crystallinity and a CNF diameter of 8 nm.



## 1. Introduction

Interest in nanocellulose, in either whisker or fiber form, has motivated scientists to produce new materials with promising and unique properties (Eyholzer et al., 2010). In recent years, the application of nanocellulosic fibers has been extended to various fields owing to their potential advantages over commercial fibers, including sustainability, inexpensive price, flexibility and stiffness as well as their unique mechanical, thermal, and electrical properties (Kalia et al., 2011; Salajkova et al., 2013). Kenaf, as a source of natural fibers, is an economically viable and ecologically friendly cellulosic source. The kenaf bast fibers possess attractive mechanical and thermal properties for use in polymer composites (Abdul Khalil et al., 2012; Abdul Khalil et al., 2014). Studies have been conducted on the isolation of cellulose nanofibers (CNF) from various cellulosic sources using mechanical, chemical, or the combination mechanical and chemical (chemo-mechanical) methods (Fatah et al., 2014). However, it has been reported that high pressure homogenization (HPH) is the most straightforward method to isolate CNF (Besbes et al., 2011a; Jonoobi et al., 2009, 2012). During the homogenization process for CNF isolation from various cellulosic sources, the fiber suspension passes through the very narrow orifice of a homogenizer, wherein a pressure drops facilities nanofibrillation. Repeating this process can increase the degree of fibrillation. The collision of particles, magnitude of the pressure drops, turbulent flow, and high shear forces might also influence the size of the obtained fibers (Kalia et al., 2014). In fact, the extent of fibrillation is highly dependent on the pressure and the number of homogenization cycles.

The concentration of the fiber suspension plays a significant role during the fibrillation process using a homogenizer. Owing to the narrow size of the HPH nozzles, fiber suspension over a certain concentration might clog the nozzle during fibrillation. In order to avoid clogging, an appropriate concentration of the fiber suspension should be used. For instance, Özgür Seydibeyoğlu and Oksman, (2008) studied the effect of the concentration of a hardwood suspension (0.025–0.2%) on CNF preparation using HPH. The study found that slurry concentrations over 0.025% clogged the homogenizer. However, the appropriate fiber suspension concentration

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may differ with the type of fiber and their processing pretreatments.

Numerous studies have been conducted on the generation of CNF using a homogenizer from kraft pulp (Nakagaito and Yano, 2005), hardwood and softwood (Stelte and Sanadi, 2009), eucalyptus (Besbes et al., 2011b), alfa, eucalyptus, pine (Besbes et al., 2011a), banana peel (Pelissari et al., 2014), etc., Studies have reported that the number of cycles and the pressure drop in the homogenization process potentially influence the degree of nanofibrillation (Besbes et al., 2011a,b; Qua et al., 2011). Despite the abundance of published research examining the effect of the homogenization cycle on the properties of the obtained nanofibers, there is limited information available in the literature on the impact of the process parameters on the production of kenaf bast CNF using HPH. Jonoobi et al. (2009) isolated kenaf bast CNF using HPH by passing the fiber suspension through the homogenizer at a constant pressure of 50 MPa for 40 cycles. The study reported that the isolated kenaf bast CNF displayed superior thermal stability with high crystallinity (Jonoobi et al., 2009).

The isolation process, processing parameters, and cellulosic fiber source play important roles in facilitating nanofibrillation, as well as influencing the properties and reinforcing potential of the fiber. The degree of nanofibrillation mainly depends on the pressure and number of passing cycles in the HPH process. Thus, optimization of the process parameters for the isolation of kenaf bast CNF is crucial to obtain the maximum yield and good physical and thermal properties, as well as to improve the applicability of these fibers as a reinforcement material in various polymer matrixes.

As many variables influence the CNF isolation processes, a quantitative assessment of their efficiency is required. However, the conventional approach utilized in the investigation of process variables is based on examining one variable at a time. This method may potentially miss some important aspects that contribute to the response, which usually involves interactions between or among the variables that are being investigated. Response surface methodology (RSM) is an effective statistical technique to study any process behavior to estimate the effects of several variables simultaneously (Hossain et al., 2015). RSM is a collection of mathematical and statistical techniques used for modeling and analysis of the influence of several input variables on the response of interest, with the objective of optimizing the response (Hossain et al., 2012). Therefore, RSM was utilized in the present study to optimize the effect of homogenization pressure and cycles on the isolated yield, as well as morphological, physical, and thermal properties, of the resultant kenaf bast CNF.

## 2. Material and methods

### 2.1. Materials

Six month old cultivated kenaf plants were supplied by the National Kenaf and Tobacco Board, Malaysia. The kenaf bast fiber was extracted from the stem. Sodium hydroxide beads (99%) (NaOH; Fine Chemical, Germany), anthraquinone (98%) (AQ; Merck, Malaysia), hydrogen peroxide (32%) ($H_2O_2$; OReC, Asia), magnesium sulphite (97%) ($MgSO_4$; R & M Chemical, UK), and hydrochloric acid (37%) (OReC, Asia) were used to treat the fibers.

### 2.2. Preparation of kenaf bast CNF

Cellulose was extracted from the kenaf bast fibers using soda-AQ pulping and a subsequent alkaline-peroxide bleaching procedure. The kenaf fibers were manually cut into 2–3 cm lengths. The pulping was performed following optimal experimental conditions (i.e., 200 g of fiber, 19.4% NaOH, a cooking liquor to kenaf fiber ratio of 7:1, and 0.1% AQ at 160 °C for 2 h) as reported by Ang et al. (2010), in a batch digester (model: Buatan Malaysia, RB Supply Enterprise, Malaysia). . The pulp was then rinsed with water to remove the chemicals used in the digester. The yield of pulping process was about 56%. Subsequently, a bleaching treatment was performed to eliminate any residual lignin using 3% $H_2O_2$, 3% NaOH, and 0.5% $MgSO_4$ at 80 °C for 2 h (Ashori et al., 2006). Finally, 10 g of the bleached kenaf bast fibers was hydrolyzed using 1.5 M HCl at 80 °C for 2 h. The hydrolyzed fibers were then rinsed in distilled water to neutralize the pH, filtered using a membrane filter (Nylafo membrane disc filter 0.2 μm, Pall Corporation, Malaysia), and then dried in oven at 60 °C for 24 h. A HCl hydrolysis was implemented in this study as a pretreatment prior to the homogenization process in order to reduce the size of fibers for preventing of clogging the homogenization's nozzle, facilitating disintegration, removing amorphous sections from fibers and for disintegrating CNF from the kenaf bast cell wall (Pan et al., 2013).

The nanofibrillation of the kenaf bast fibers was carried out by passing a suspension of the fibers with a concentration of 0.05, 0.1, and 0.2% through a high pressure homogenizer (Model: HPH 2000/4, IKA, Germany). The optimal suspension concentration was determined based on the ease of fibrillation. The optimal fiber suspension concentration was utilized to isolate CNF from the kenaf bast fibers in all further studies.

### 2.3. Design of experiments

RSM was applied to determine the optimum experimental conditions to obtain the maximum response for yield (%), crystallinity (%), and fiber diameter (nm). A central composite design was utilized to determine the optimal homogenization pressure and number of cycles. The homogenization pressure ($X_1$) and cycles ($X_2$) were the independent variables and the variables were coded according to the following equation:

$$X = \frac{x - [x_{\max} + x_{\min}]/2}{[x_{\max} - x_{\min}]/2} \quad (1)$$

where $x$ is the natural variable, $X$ is the coded variable, $x_{\max}$ is the high level of the natural variable, and $x_{\min}$ is the low level of the natural variable. The low, intermediate, and high levels of each variable were designated as −1, 0, and +1, respectively, as shown in Table 1. The nanofibrillation behavior of kenaf bast in HPH can be explained by the following second-order polynomial equation:

$$U = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{12} X_1 X_2 + \epsilon \quad (2)$$

where $U$ is the response as a function of yield (%), crystallinity (%), and diameter (nm), $\beta_0$ is a constant term, $\beta_1$ and $\beta_2$ are the coefficients of linear effects, $\beta_{11}$ and $\beta_{22}$ are the coefficients of quadratic effects, and $\beta_{12}$ is the coefficient of interaction effects. MINITAB software (ver.16.2.1) was used to analysis the regression model and fit the second-order polynomial equation to the model. In order to minimize the influence of unexplained variability, the experiments were randomized. For each response function, analysis of variance (ANOVA) was conducted to estimate the significance of the terms. In addition, the adequacy of the model was determined by lack of fit, coefficient of determination ($R^2$), and adjusted coefficient of determination ($R^2_{adj}$). The three-dimensional graphical

**Table 1**
The coded and uncoded levels of the independent variables.

| Factor | Symbol | Level | | |
|---|---|---|---|---|
| | | Low (−1) | Intermediate (0) | High (+1) |
| Pressure, MPa | $X_1$ | 20 | 35 | 50 |
| Cycles, P | $X_2$ | 20 | 30 | 40 |