Effect of Nitration Temperature and Time on Nitrocellulose Synthesized from Pineapple (*Ananas comosus*) Leaf Waste

Yelmida Azis*, *Anton* Algrinov, and *M Asyraf* Hazzamy

University of Riau, Chemical Engineering Department, Faculty of Engineering, 28295 Pekanbaru, Indonesia

Abstract. Nitrocellulose is widely used as a key energetic material due to its high combustion efficiency and strong ignition characteristics. This study investigates the effect of nitration temperature and reaction time on the synthesis of nitrocellulose derived from pineapple leaf waste, an abundant lignocellulosic by-product. Pineapple leaf cellulose was nitrated using 65% HNO_3 and 98% H_2SO_4 at temperatures of 5°C, 10°C, and 15°C with reaction times of 40, 50, and 60 min. The resulting nitrocellulose was washed and neutralized before undergoing combustion analysis and FTIR characterization. The combustion test showed that samples treated at 10°C and 15°C for 60 min exhibited the strongest flame intensity, indicating a higher degree of nitration. FTIR spectra confirmed the formation of nitro groups, as demonstrated by absorption peaks around ~1270 cm^{-1} and ~1645 cm^{-1} accompanied by a reduction in –OH intensity. The combined results confirm that both temperature and duration strongly influence the nitration process, with optimal conditions achieved at 60 min and moderate temperatures. Nitrocellulose synthesized under these conditions demonstrates properties consistent with potential applications in propellant formulations.

1 Introduction

The international community is currently competing to explore space. Elon Musk, CEO of SpaceX, is developing a spacecraft to carry humans and potentially build a civilization on Mars. In addition, countries are also racing to develop ballistic and intercontinental missiles. Meanwhile, as a developing nation, Indonesia is relatively new to the era of outer space flight. So far, Indonesia has developed microsatellites and solid-fuel rockets through the National Institute of Aeronautics and Space (LAPAN). However, rocket technology development has been slow due to limited funding, technology, and insufficient government attention. A key technological constraint is the difficulty in producing propellants that have strong, economical thrust [1].

* Corresponding author: yelmida.azis@lecturer.unri.ac.id

Rocket fuel or propellants are classified as single-base, double-base, or triple-base. In single-base propellants, the fuel and oxidizer are integrated within a single compound, such as nitrocellulose or nitroglycerin. Double-base propellants primarily consist of nitrocellulose and nitroglycerin, while the addition of nitroguanidine to a double-base formulation results in a triple-base propellant [2].

Nitrocellulose can be synthesized from both synthetic and natural cellulose sources [3]. Cellulose, the primary component of biomass, is widely available in Indonesia. For example, Padil [4] and Nuraini [5] utilized Riau palm oil frond waste to extract cellulose, which was found to contain 38.89% cellulose, 27.14% hemicellulose, and 19.87% lignin. Although this cellulose content is relatively low, delignification followed by bleaching can increase the cellulose concentration in palm oil frond waste to 92%.

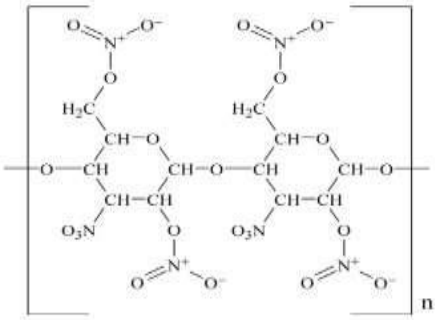

Fig. 1. Nitrocellulose structure

Pineapple leaves represent another abundant source of cellulose in Riau Province. The pineapple plant (*Ananas comosus*) is a short-stemmed, monocotyledonous species that grows in clumps. Its leaves, which are 55 to 75 cm long and possess upward-facing spines along the edges, cluster at the base of the stem and contain long fibers [7]. Despite their high cellulose content, pineapple leaves are typically regarded as agricultural waste since only the fruit is harvested. The chemical composition of pineapple leaf fiber includes cellulose, lignin, pectin, fats and waxes, ash, and various other organic substances [8], as shown in **Table 1**.

Table 1. Composition of Dried Pineapple Leaves

Chemical Composition	Pineapple Fiber (%)
Cellulose	69.5 – 71.5
Pentosa	17.0 – 17.8
Lignin	4.4 – 4.7
Fat and *Wax*	3.0 – 3.3
Pectin	1.0 – 1.2
Ash	0.71 – 0.87
Other substances (proteins, organic acids, etc.)	4.5 – 5.3

The usability and economic value of cellulose derived from pineapple leaf waste can be enhanced by employing it as a raw material for nitrocellulose production. Nitrocellulose is synthesized via a nitration reaction involving a mixture of nitric acid and sulfuric acid, with

the addition of water, with or without stirring. The reactant composition is regulated to yield a nitrocellulose product with a nitrogen content of 12.2%. The resulting nitrocellulose is stabilized using hot acid and subsequently treated with a hot, dilute sodium carbonate solution [8].

This research uses cellulose derived from pineapple leaf waste to produce nitrocellulose, a precursor for propellant manufacturing. The use of pineapple leaf waste will render pineapple leaves profitable, cost-effective, and eco-friendly.

2 Experimental Sections

2.1 Materials

Pineapple leaves served as the primary raw material of natural nitrocellulose. The materials used in the process were 65% HNO_3, 98% H_2SO_4, 2% H_2SO_4, 15% H_2O_2, 2 N KOH, $NaHCO_3$, and distilled water.

2.2 Materials Pre-treatment

In this process, the pineapple leaves, which had been cleaned of their spines, were ready for lignin and hemicellulose removal using a decorticator tool. The next step involved grinding the leaves using a blunt glass fragment to achieve the desired fiber results. The fiber was then washed thoroughly and dried in the sun for approximately 3 days.

2.3 The Cellulose Analysis

After the pre-treatment was completed and before the bleaching, the chemical composition of the pineapple leaf waste was analyzed to determine its chemical composition. The analysis performed consisted of a moisture content test (SNI 08-7070-2005) and an alpha-cellulose content test (SNI 0444-2009).

2.4 Bleaching Process

The bleaching process was carried out to increase the purity of the α-cellulose obtained and to remove unwanted dyes. H_2O_2 was chosen as the bleaching agent because the waste from the bleaching process was water (H_2O), which does not pollute the environment. Bleaching was performed once using 300 mL of a 15% H_2O_2 solution, with a reaction time of 30 minutes at 80°C. After the bleaching process was complete, an analysis was executed to determine the purity of α-cellulose (SNI 0444-2009).

2.5 Nitration Process

The nitration process was carried out using 65% HNO_3 and 98% H_2SO_4 titration reagents in a nitration reactor equipped with a cooling vessel, a condenser, a stirrer, a rubber stopper, and a stirring motor. The nitration conditions were 5, 10, and 15 °C, with reaction times of 40, 50, and 60 min. The ratio of pineapple leaves and nitric acid used was 1:12 with a constant stirring speed of 65 rpm. After nitration, the resulting nitrocellulose was washed and neutralized with $NaHCO_3$ (**Fig. 2**). The functional groups formed during the nitration process

were analyzed using FTIR, and the presence of $–NO_2$ absorption peaks was used to indicate the degree of nitration achieved.

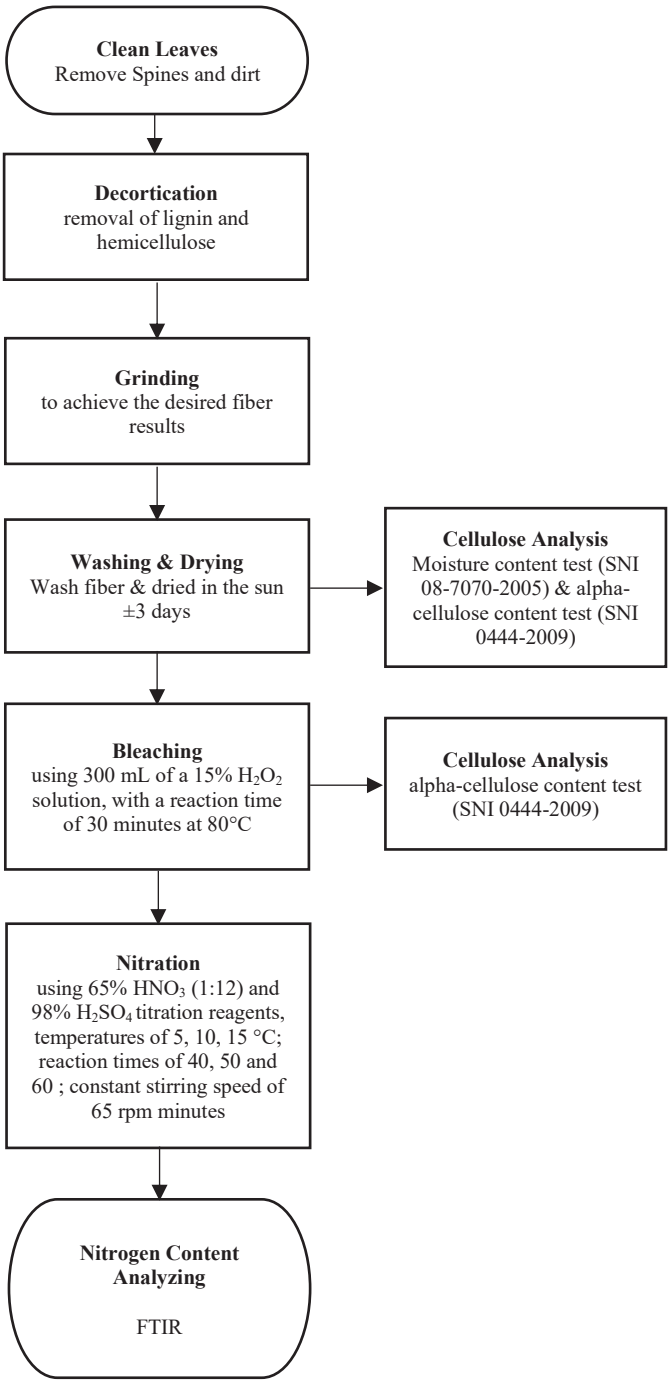

Fig. 2. Process flow diagram

3 Result and Discussion

3.1 Simple Analysis with Combustion Test

A preliminary indication of the nitrogen content in nitrocellulose can be obtained through a simple combustion test performed after the samples have been thoroughly dried. Although qualitative, this combustion assessment provides an initial visual indication of how effectively nitro groups have been incorporated into the cellulose structure during the nitration process. Nitrocellulose with a higher degree of nitration generally exhibits increased flammability, igniting more rapidly and producing a stronger, more sustained flame due to the presence of oxidizing nitrate groups within the polymer chain.

Fig. 3. Example of Combustion Test sample (60 min; 15°C)

In this study, small amounts of nitrocellulose produced under different temperature and reaction time conditions were ignited to observe variations in flame characteristics. The key parameters examined included the ease of ignition, flame size, burn continuity, and the amount of residue left after combustion. Samples that failed to ignite or produced weak flames typically indicated lower nitrogen substitution or incomplete nitration, whereas samples that generated larger, more intense flames suggested a higher degree of nitration, consistent with propellant-grade nitrocellulose.

Table 2. Nitrocellulose Combustion Test of Pineapple Leaf Waste

Time	Temperature		
	5 °C	10 °C	15 °C
40 min	Fire did not burn, burned, remained	Fire did not burn, burned, remained	Fire did not burn, burned, remained
50 min	Flame (small), burned, remained	Flame (small), burned, remained	Flame (moderate), burned, remained
60 min	Flame (small), burned, remained	Flame (large), burned, remained	Flame (large), burned, remained

The combustion patterns showed a clear trend with respect to both temperature and reaction time. For samples processed at 40 min across all temperatures (5, 10, and 15°C), ignition did not occur, and the cellulose structure remained largely intact after heating, indicating insufficient nitration at shorter durations. At 50 min, slight improvements were observed, with small flames appearing at all temperatures, although combustion remained weak and produced noticeable residues, indicating partial nitration.

The most significant enhancement occurred at 60 min. At this duration, samples synthesized at 10°C and 15°C produced strong, relatively large flames, as illustrated in Fig. 3. These findings suggest that at these temperatures, longer nitration times facilitate more effective substitution of hydroxyl groups by nitro groups, resulting in materials with higher combustibility. This qualitative outcome aligns with the FTIR results described in later sections, where distinct $–NO_2$ absorption peaks confirm successful nitration. Conversely, the sample treated at 5°C for 60 min showed only a small flame, implying that lower temperatures slow the nitration kinetics even with longer reaction times.

The combustion test results presented in **Table 2** demonstrate that both reaction temperature and time significantly influence the nitration degree of pineapple leaf cellulose. The strongest combustion—associated with the highest inferred nitrogen content—was consistently observed in samples treated at 10 and 15°C for 60 min, indicating that these conditions are the most favorable among the tested variations for producing propellant-grade nitrocellulose.

3.2 Fourier Transform Infra Red (FTIR)

Fourier Transform Infrared (FTIR) spectroscopy was used to determine the degree of substitution between hydroxyl (–OH) and nitro ($–NO_2$) groups, which indicates the success of the nitration reaction. According to Hartaya [9], cellulose exhibits a broad hydroxyl absorption band at 3200–3600 cm^{-1}, whereas nitrocellulose produces characteristic peaks at 1260–1390 cm^{-1} and 1560–1660 cm^{-1}. These wavenumber regions are therefore used as indicators of nitrocellulose formation.

Jamal et al. previously reported that nitrocellulose spectra generally show two strong $–NO_2$ absorption peaks accompanied by reduced –OH intensity as nitration progresses [10]. The same spectral trend was observed in the present study, although the data originate from experiments performed on pineapple leaf cellulose rather than from the cited reference.

Fig. **5-6** smoothly illustrates the evolution of the functional groups at different reaction conditions. At 5 °C, the $–NO_2$ peaks are present but relatively weak, as confirmed by higher transmittance values in **Table 3**. This implies incomplete nitration, particularly at 40 and 50 min. At 10 °C, the $–NO_2$ peaks intensify and show lower transmittance (**Table 4**), indicating a higher level of substitution. At 15 °C, a similar trend is observed (**Table 5**), particularly at 60 min, suggesting that moderate heating accelerates nitration efficiency.

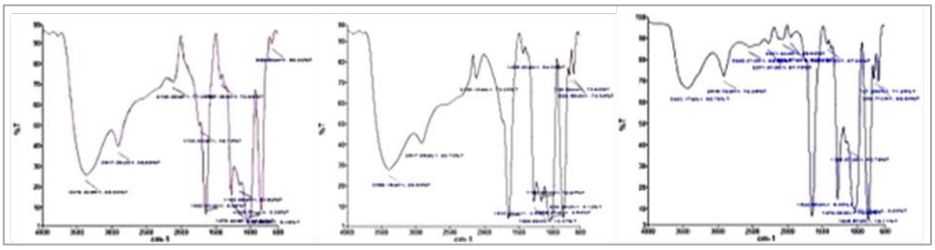

Fig. 4. FTIR spectrum of pineapple leaf nitrocellulose at 5 °C with reaction time of (a) 40 min; (b) 50 min; (c) 60 min.

Consistent with Rahmada et al. [11], lower transmittance signifies greater functional group concentration; therefore, the strongest nitration corresponds to the lowest transmittance values recorded for the –NO_2 peaks at 10 °C and 15 °C for 60 min. Across all spectra, the persistence of absorption near ~1270 cm^{-1} and ~1645 cm^{-1} aligns with the findings of Gismatulina et al. [12], confirming that nitrocellulose has been successfully formed.

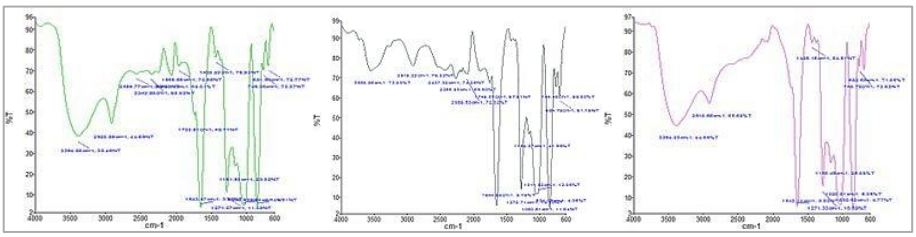

Fig. 5. FTIR spectrum of pineapple leaf nitrocellulose at 10 °C with reaction time of (a) 40 min; (b) 50 min; (c) 60 min.

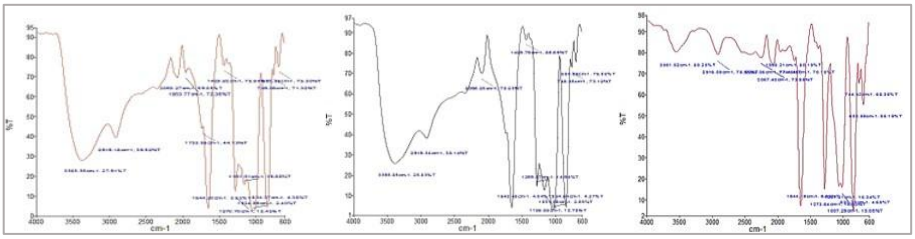

Fig. 6. FTIR spectrum of pineapple leaf nitrocellulose at 15 °C with reaction time of (a) 40 min; (b) 50 min; (c) 60 min.

Based on the relative reduction of –OH groups and the emergence of strong –NO_2 peaks, the nitrogen content can be inferred qualitatively. As suggested by Hartaya [9], full substitution of three hydroxyl sites yields ~12.73% nitrogen, which corresponds to the estimated value for the samples treated at 10 °C and 15 °C for 60 min. These findings match the combustion test in Section 3.1, which identified the same conditions as producing the highest burning intensity.

Table 3. Functional Groups –NO_2 and –OH in Pineapple Leaf Nitrocellulose (5 °C)

Functional Group	5 °C					
	40 min		50 min		60 min	
	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)
-NO_2	1272.44	15.63	1269.69	12.47	1272	14.80
	1642.50	6.45	1643.33	4.03	1642.66	6.36
-OH	3376.32	25.93	3398.19	28	3442.17	66.76

Table 4. Functional Groups –NO_2 and –OH in Pineapple Leaf Nitrocellulose (10 °C)

Functional Group	10 °C					
	40 min		50 min		60 min	
	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)
-NO_2	1271.07	11.38	1272.71	14.10	1271.33	15.59
	1643.47	3.85	1644.69	5.76	1643.14	5.32
-OH	3394.85	38.46	3558.86	73.83	3394.03	44.66

Table 5. Functional Groups $–NO_2$ and –OH in Pineapple Leaf Nitrocellulose (15 °C)

Functional Group	15 °C					
	40 min		50 min		60 min	
	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)	F (cm^{-1})	T (%)
$-NO_2$	1270.70	12.43	1273.64	14.93	1269.87	14.94
	1644.20	3.82	1644.19	6.90	1643.48	4.04
-OH	3383.35	27.91	3561.52	80.25	3393.85	25.83

Conclusions

Pineapple leaf waste, which contains 69.5–71.5% cellulose, was successfully converted into nitrocellulose through acid nitration. Variation in reaction temperature and time resulted in clear differences in nitration efficiency. FTIR characterization confirmed the formation of nitro groups at all tested conditions; however, the strongest absorption peaks and the lowest transmittance values, indicating the highest degree of substitution, were obtained at 10°C and 15°C for 60 min. Combustion observations supported these results, showing the most intense flame characteristics under the same conditions. In sum, the study demonstrates that both temperature and reaction time significantly influence the nitration process, and the nitrocellulose produced under optimal conditions exhibits properties suitable for potential propellant applications.

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