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Valorization of Textile Waste into Cellulose Citronellal Fiber Using Natural H-Zeolite Catalyst: Towards a Circular Bioeconomy Approach

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ABSTRACT

Mosquito-borne diseases such as malaria, dengue, chikungunya, and Zika remain major global health threats, while the rapid growth of the textile industry generates large volumes of waste largely disposed of in landfills due to limited recycling pathways. This study explores the valorization of cotton-based textile waste into functional biomaterials within a circular bioeconomy framework, using Indonesia as a case study due to its abundant textile waste, citronella biomass, and natural zeolite deposits. Cellulose was extracted from textile waste through acid hydrolysis, citronellal was isolated from citronella oil by fractional distillation, and natural zeolite was activated through acid treatment, ion exchange, and calcination to produce an H-Zeolite catalyst. The recovered cellulose was subsequently reacted with citronellal through a condensation-impregnation process using the H-Zeolite catalyst to synthesize cellulose-citronellal fibers. All materials were characterized using FTIR, SEM, and XRD. Acid activation and ion exchange significantly increased zeolite acidity to 1.382 g NH3/g sample, indicating the formation of Brønsted acid sites suitable for catalytic reactions, while the zeolite crystalline framework remained stable after activation. Cellulose recovered from textile waste exhibited morphological and functional group characteristics comparable to commercial cellulose. FTIR analysis indicated the incorporation of citronellal into the cellulose matrix, suggesting the formation of cellulose-citronellal fibers with potential bioactive properties. However, FTIR evidence alone cannot confirm covalent bond formation, and further analyses including solid-state NMR, TGA, and direct bioassay testing are recommended to fully validate functional performance. Overall, this study demonstrates a potential pathway for converting textile waste into higher-value bio-based materials using renewable biomass and a natural mineral catalyst, offering a circular bioeconomy model relevant for tropical countries facing similar biomass availability and vector-borne disease challenges.

1. INTRODUCTION

Mosquito-borne diseases such as dengue fever, malaria, and chikungunya remain a global health threat, with millions of cases occurring annually. Climate change is expanding the geographic distribution of mosquito vectors due to rising temperatures and changing rainfall patterns, making previously risk-free areas increasingly vulnerable (Fariz et al., 2026; Siniscalchi et al., 2026). This situation demands the development of more innovative, sustainable, and environmentally friendly vector control strategies, especially in tropical countries with high disease prevalence like Indonesia (Abbasi, 2025; Baitharu et al., 2021).

Essential oils from plant families such as Cymbopogon spp., Ocimum spp., and Eucalyptus spp. are widely recognized for their mosquito repellent properties. Active components with notable repellent activity include citronellol, citronellal, alpha-pinene, camphor, limonene, and thymol (Abdelgaleil et al., 2024; Nerio et al., 2010). Citronellal in particular has been studied as an insect repellent candidate, although its direct topical application provides only short-lived protection (Alighiri et al., 2018; Iovinella et al., 2022; Pujiastuti et al., 2017). Incorporating citronellal into functional textile materials offers a more practical and sustained approach to personal protection. Developing such functional textiles, however, requires simultaneous consideration of raw material sustainability and textile waste management, as the textile industry generates substantial amounts of solid residue (Hasan et al., 2025; Riza et al., 2021). Most textile waste is still disposed of in landfills due to high recycling costs, creating significant environmental burdens (Singh et al., 2025).

Globally, textile waste is estimated to exceed 92 million tons per year (Ghosh et al., 2025). At the national level, data from the Ministry of Environment (2025) recorded total waste generation in Indonesia at 25,141,493.03 tons, with fabric waste contributing approximately 2.46% of that volume. Although relatively modest in percentage, the absolute quantity remains substantial and largely underutilized. Textile recycling continues to be constrained by technological limitations and the economic burden of material separation, with most existing pathways downcycling waste into non-textile products such as composite materials. Cotton-based textile waste, by contrast, represents a promising feedstock due to its comparatively high cellulose purity relative to lignocellulosic sources such as wood, enabling more efficient and cost-effective cellulose recovery.

Cellulose recovered from cotton textile waste can be chemically modified into a range of functional derivatives. Post-consumer cotton textiles have been converted into wrinkle-resistant fabrics via esterification using polycarboxylic acids (Olivito et al., 2021), while surface functionalization followed by mechanical treatment has yielded cellulose citrate nanocrystals (CitCNCs) with tunable surface chemistry and favorable mechanical properties (Ma et al., 2023). Cellulose recovery yields for such applications have been reported between 65% and 88%, with degrees of substitution suitable for downstream processing into sodium carboxymethylcellulose (DS 0.27 to 0.61) and cellulose acetate (DS 2.59) (Ma et al., 2023). Beyond conventional derivatives, aromatic and functional textiles have also been developed through encapsulation approaches. Beta-cyclodextrin complexation has been employed to delay essential oil release and improve stability in textile applications (Costa et al., 2022), while microcapsule systems prepared via coacervation using gelatin and sodium sulfate, hardened with formaldehyde, have been optimized through response surface methodology to enhance encapsulation efficiency (Solomon et al., 2012). These diverse modification pathways confirm the versatility of textile-derived cellulose as a platform for producing value-added functional materials.

In the context of sustainable development, the circular bioeconomy framework provides a compelling rationale for integrating biomass valorization with waste reduction. Circular economy principles emphasize product design, business model innovation, and lifecycle management to minimize waste and increase resource efficiency (Awan and Sroufe, 2022; Mestre and Cooper, 2017). Circular bioeconomy extends this further by incorporating biological resources through biotechnology and biorefinery approaches, encompassing bioenergy, bioplastics, and bio-based chemicals, while also addressing ecosystem and environmental considerations (Chikhalet et al., 2026; Kumari et al., 2026; Manhongo and Mbohwa, 2026; Venkatesh, 2022). Within the textile sector, most existing studies address the circular economy solely through mechanical or chemical fiber recycling without explicitly engaging with bioeconomic dimensions (Ghosh et al., 2025; Patti et al., 2020). The present study goes further by simultaneously valorizing two biomass streams, namely cellulose from cotton-based textile waste and citronellal from citronella oil, to produce a citronellal-modified cellulose fiber as a bio-based material with potential applications in vector-control-related protective textiles.

The condensation-impregnation of citronellal within the cellulose matrix requires a catalyst capable of activating the carbonyl groups of citronellal and the hydroxyl groups of cellulose. Natural zeolite modified into H-Zeolite through acid treatment and ion exchange provides a relatively inexpensive and accessible solid acid catalyst suitable for this purpose. The citronellal-modified cellulose fiber developed in this study represents a preliminary model for utilizing essential oil-derived compounds in sustainable cellulose-based biomaterials. The present study primarily focuses on synthesis and physicochemical characterization, while further investigations on release behavior, material stability, and direct bioassay testing remain subjects for future work.

Based on this background, the study aims to: (1) evaluate the yield and characteristics of cellulose extracted from cotton-based textile waste, (2) examine the role of natural H-Zeolite catalyst in the condensation-impregnation of citronellal into the cellulose matrix, and (3) contextualize the overall process within a circular bioeconomy framework for textile waste valorization.

2. MATERIALS AND METHODS

2.1. Research Design

This experimental research was conducted in two main stages. The first stage was the recycling of textile waste into cellulose and the isolation of citronellal from citronella oil. The second stage was the preparation of natural H-Zeolite catalyst and the synthesis of cellulose-citronellal fibers. In addition to laboratory experiments, this study also examined the conceptual framework of the circular bioeconomy in the context of the valorization of textile waste as a secondary biomass source for the development of cellulose-based functional materials. This study used Indonesia as a case study, considering the availability of textile biomass, citronella oil production, and the existence of local natural zeolite resources such as Malang zeolite from East Java Province, Indonesia. The conceptual framework of circular bioeconomy in this study was supported by a study of relevant literature.

2.2. Extraction of Cellulose from Textile Waste

Preparation of cellulose citrate from fabric adapted from Ruiz-Caldas et al., (2023). Cellulose extraction from fabric waste was carried out through acid hydrolysis. Small pieces of textile fabric (<1 cm^2) were placed in a round-bottom flask containing an 85% aqueous citric acid solution by weight. The ratio of textile to pure citric acid was maintained at 1:20 ($g\ g^{-1}$).

The mixture was stirred at 300 rpm at 100°C. After the citric acid was completely dissolved, the reaction was maintained for seven hours. The reaction was then stopped by a fivefold dilution with DI water. The mixture was then filtered using a 5 mm pore size polyethersulfone (PES) membrane to separate the citric acid solution from the solid fraction containing carboxylated cotton fibers and residual acid. The citric acid was recovered through evaporation and crystallization. The remaining citric acid is then rinsed from the solids by slowly adding DI water to the filter until the filtrate conductivity drops below 5 $mS\ cm^{-1}$. Synthetic fibers or other fibers that remain intact during the process are separated through filtration and referred to as residue.

2.3. Isolation of Citronellal from Citronella Oil

Preparation steps for isolating citronellal from lemongrass oil, adapted from (Cahyono et al., 2010). The citronellal content in *Cymbopogon winterianus* lemongrass oil was analyzed using GC-MS. Isolation of citronellal from lemongrass oil containing more than 35% citronellal was carried out using fractional distillation at low pressure. The fraction with the highest citronellal content was then separated and continued with the re-distillation process. The results of the redistillation were then analyzed using GC.

2.4. Preparation of Natural H-Zeolite Catalyst

Acid treatment was performed to control the acidity and selectivity of zeolite. Natural zeolite chunks were ground using a mortar grinder to obtain homogeneous granules that passed a 100-mesh sieve. The sieved granules were oven-dried to remove moisture. A total of 300 g of dried 100-mesh natural zeolite was immersed in

350 mL of 1% hydrofluoric acid solution for 20 minutes. The sample was then washed until neutral and dried. Next, 30 g of the dried sample was immersed in 125 mL of 6 N HCl solution for 30 minutes at 50°C. The sample was washed until neutral and dried at 120°C. This activation product was designated Za.

A total of 20 g of Za was then added to 250 mL of 3 M ammonium chloride solution. The mixture was stirred for 24 hours at room temperature, then filtered and washed with running water until free of chloride ions (tested using $AgNO_3$ solution). The resulting solid was heated at 120°C until dry. The dried solid was then calcined at 450 °C for 1 hour using a Cahyono et al., (2010) modified calcinator to remove NH_3 gas. The final product of this treatment is referred to as H-Za.

2.5. Synthesis of Cellulose-Citronellal Fiber

The synthesis of cellulose-citronellal fibers was carried out in a reflux reactor equipped with a magnetic stirrer and an inert N_2 gas flow. A total of 1 gram of natural H-Zeolite catalyst, 3 grams of cellulose, and 30 mL of citronellal in ethyl acetate solvent were loaded into a 100 mL reactor flask with a total reaction volume of 50 mL. The mixture was stirred at reflux temperature. Samples were taken at reaction times of 3 hours, 6 hours, and 9 hours. The catalyst was separated from the sample by centrifugation and then extracted using n-hexane. The obtained solids were analyzed using FTIR.

3. RESULTS OR RESULTS AND DISCUSSIONS

3.1. Preparation and Characterization of Natural H-Zeolite Catalyst

3.1.1. Determination of Acidity Degree

The acidity level of the catalyst is an important parameter in determining the catalytic activity in the condensation reaction between citronellal and cellulose. In this system, acid sites play a role in activating the carbonyl groups of citronellal and facilitating interactions with the hydroxyl groups in the cellulose polymer chain. Therefore, increasing the number of acid sites on the catalyst surface is expected to increase the efficiency of the condensation reaction that occurs during the cellulose-citronellal fiber formation stage. The acidity degree was determined using a gravimetric method based on ammonia adsorption. The measurement results are shown in Table 1.

Table 1: Determination of Acidity Degree

Sample Code	Mass of Ammonia (g)	Acidity Degree (g)
HF Washing	0.009	1.040
NH_4Cl Washing	0.007	0.820

The results in Table 1 show that acid treatment using hydrofluoric acid increased the acidity level of the catalyst compared to the original zeolite. This increase occurred due to the release of impurities in the form of free SiO_2 and metal oxides from the zeolite surface, thereby increasing the number of available acid sites in the aluminosilicate structure. It should be noted that the acidity values reported in Table 1 are expressed in grams of adsorbed ammonia per gram of sample, as measured by the gravimetric method. Future studies are recommended to report acidity in standardized units (mmol NH_3 g^{-1}) obtained through temperature-programmed desorption (NH_3-TPD) to enable more rigorous inter-study comparisons.

In the next stage, ion exchange using NH_4Cl solution caused a temporary decrease in the acidity level. This occurred because NH_4^+ ions replaced metal cations such as Na^+ and Ca^{2+} in the zeolite framework. After calcination, the NH_4^+ ions decomposed into NH3 gas, forming new Brønsted acid sites in the zeolite framework. This transformation produced a natural H-Zeolite catalyst with the highest acidity level of 1.382. The increase in acidity degree after acid treatment and NH_4^+ ion exchange followed by calcination is in line with various previous studies which reported that acid activation and H-zeolite formation can increase the concentration of Brønsted sites and improve pore accessibility in natural zeolites (Muhammad et al., 2024; Tanirbergenova et al., 2025).

Compared with several other natural zeolite activation studies, the increase in acidity obtained in this study is in a comparable range to mordenite and clinoptilolite type zeolites that have been activated through a combination of acid treatment and ion exchange (Stoyanov et al., 2024). This indicates that the activation method used in this study is effective in producing heterogeneous catalysts with relatively high acid properties. Mechanistically, the Brønsted acid sites in H-Zeolite function as protonation centers that can activate the aldehyde carbonyl group in citronellal. This protonation increases the electrophilicity of the carbonyl carbon, thus facilitating the nucleophilic attack of the hydroxyl group on the cellulose chain, which ultimately facilitates the formation of new bonds in the condensation process. However, it should be noted that the exact nature of the interaction, whether covalent bond formation or surface adsorption, cannot be ascertained from acidity data alone and requires further physicochemical analysis such as solid-state NMR or thermogravimetric analysis. Therefore, increasing the acidity of the catalyst is an important parameter in supporting the citronellal–cellulose interaction during fiber production.

3.1.2. Functional Group Analysis using FTIR

FTIR analysis was performed to identify changes in functional groups in the zeolite structure after chemical treatment. The FTIR spectra of natural zeolite, HF-treated zeolite, and NH_4Cl-treated zeolite are shown in Fig 1.

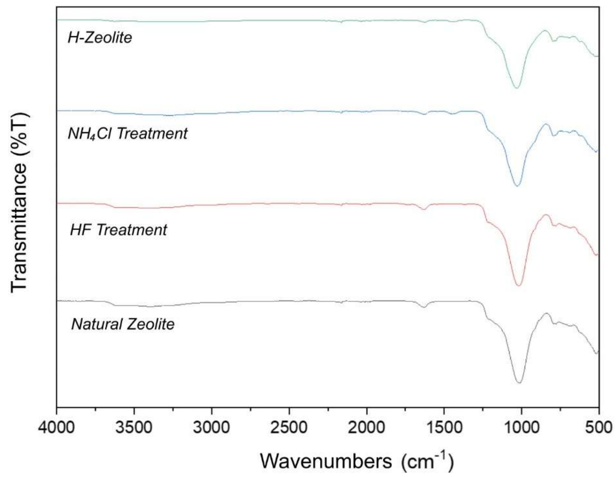


Fig. 1: Functional Groups (a) Natural Zeolite, (b) Zeolite Resulting from Reaction with HF, (c) Zeolite Resulting from Reaction with NH4Cl (d) Natural Zeolite

The main absorption band at wavenumbers around 1000–1100 cm^{-1} is associated with Si–O–Si stretching vibrations in the zeolite aluminosilicate framework. This band is a general characteristic of zeolite structures and has been widely reported in FTIR characterizations of natural and modified zeolites (Adila et al., 2024; Gebretsadik and Gebremedhin, 2020). After acid activation, the band appears sharper, indicating increased regularity of the silica-alumina structure. Furthermore, the appearance of additional bands in the wavenumber region associated with –OH groups (silanol and bridging hydroxyl) indicates the formation of new acid sites on the catalyst surface.

These –OH groups are generally associated with the presence of Brønsted acid sites in the zeolite structure and an increase in their intensity is often observed after acid activation or ion exchange processes in various zeolite types such as clinoptilolite, mordenite, and zeolite X (Adila et al., 2024; Hieu et al., 2022). These spectral changes indicate that the chemical treatment carried out does not cause damage to the zeolite aluminosilicate framework, but only modifies the surface and increases the number of catalytic active sites. The stability of this structure is important because the catalytic activity of zeolites is highly dependent on the integrity of their crystal framework.

3.1.3. Structural Analysis using XRD

XRD analysis was performed to evaluate whether the chemical activation process causes changes in the crystal structure of the zeolite. XRD diffractograms of natural zeolite and H-Zeolite are shown in Fig 2. The diffraction patterns show that the characteristic peaks of the zeolite are maintained after the activation process. This indicates that the acid treatment and ion exchange treatments do not damage the primary crystalline structure of the zeolite. It is acknowledged that the XRD peaks in this study have not been fully indexed to specific zeolite phases (e.g., clinoptilolite or mordenite), and quantitative parameters such as crystallinity percentage, Si/Al ratio, and unit cell parameters were not determined. These data would strengthen the characterization and are recommended for future work.

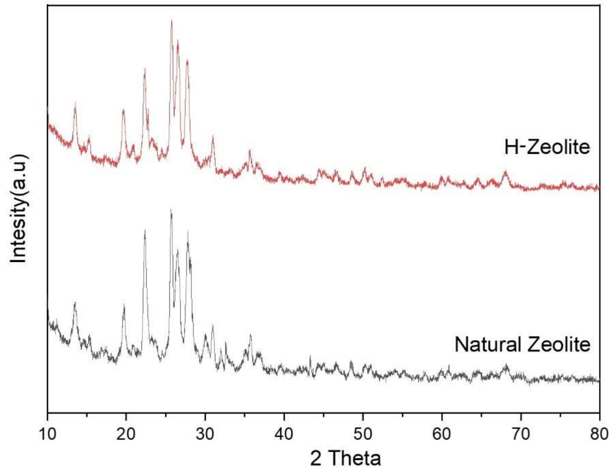
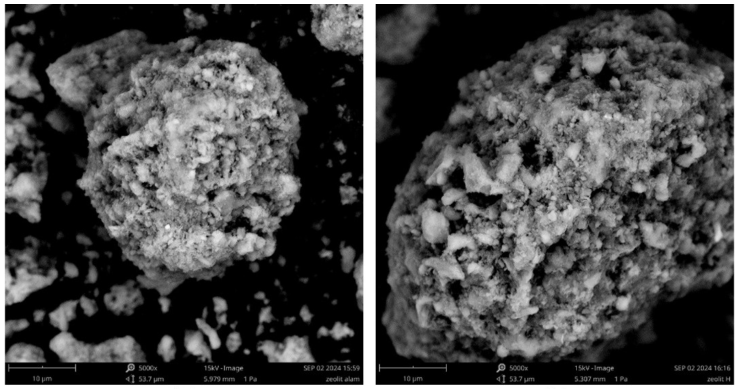


Fig. 2: XRD Results of Natural Zeolite and H-Zeolite

This phenomenon has been reported in various studies showing that moderate acid treatment of natural zeolites can increase porosity and acidity without destroying the main crystalline framework, which usually retains the typical diffraction pattern of mordenite or clinoptilolite (Muhammad et al., 2024; Stoyanov et al., 2024; Tanirbergenova et al., 2025). Compared with other activation methods such as extreme dealumination or high-concentration strong acid treatment, the activation method used in this study tends to be more selective in removing impurities without destroying the zeolite framework structure. The stability of this crystal structure is very important because the catalytic activity of zeolites is strongly influenced by the pore distribution and the stability of their aluminosilicate structure. However, the present study does not include quantitative measurements of surface area (BET), pore volume, or Si/Al ratio, which would be necessary to fully substantiate the relationship between structural changes and catalytic performance. These limitations are acknowledged, and such measurements are recommended in follow-up studies.

3.1.4. Morphological Analysis using SEM

Morphological analysis using SEM showed that the surface structure of the zeolite underwent a textural change after activation. Natural zeolite appears relatively dense with limited pore distribution. After activation to H-Zeolite, the surface exhibits a rougher texture with more pronounced porosity (Fig 3).



(a) (b)

Fig. 3: Morphology (a) Natural Zeolite, (b) Zeolite-H

These morphological changes indicate that the activation process successfully opened some of the zeolite pores, thereby increasing the surface area available for catalytic processes. Similar textural changes have also been observed in various natural zeolite activation studies, where acid treatment can remove mineral impurities and metal oxides from the zeolite surface, resulting in a more porous structure and increasing the accessibility of catalytic sites (Side et al., 2023; Tanirbergenova et al., 2025). This increase in porosity is important because it can enhance the diffusion of reactant molecules such as citronellal into the catalyst's pore structure during the reaction. However, it should be noted that the current SEM analysis is qualitative in nature. Quantitative surface characterization data, namely BET surface area, pore volume, and average pore diameter, were not measured in this study, and their absence limits the extent to which claims of increased porosity can be substantiated. Such data are recommended for future characterization to provide a more complete picture of the activation outcome.

3.2. Cellulose Recovery from Textile Waste

3.2.1. Morphological Analysis using SEM

Morphological analysis using SEM was performed to compare the structure of cellulose obtained from textile waste with commercial standard cellulose (Merck cellulose). The results showed that the acid hydrolysis process successfully separated the cellulose component from the textile matrix. The extracted cellulose fibers had a morphology similar to standard cellulose, although the resulting particle size was still relatively larger (Fig 4).

(a) (b)

Fig. 4: Morphology (a) Results of Cotton Fabric Decellulose, (b) Merck Cellulose

These findings align with previous studies showing that chemical treatment of cellulose extracted from cotton textile waste can produce morphologies very similar to commercial cellulose (Maciel et al., 2019; Rana et al., 2023). The observed differences in particle size are likely due to varying degrees of fiber degradation during the hydrolysis process. In some studies, more aggressive chemical treatments or the use of additional mechanochemical processes such as mechanical fibrillation can produce smaller cellulose particles. Nevertheless, the similarity in morphology suggests that cellulose from textile waste has great potential as a raw material for the development of cellulose-based materials.

3.2.2. Functional Group Analysis using FTIR

The FTIR spectrum of the extracted cellulose showed significant similarity to the spectrum of standard cellulose. Absorption bands associated with vibrations of the –OH group, C–H stretching, and C–O–C bonds, typical of polysaccharide structures, were clearly detected. The presence of these bands indicates that the basic structure of the cellulose was maintained during the extraction process (Fig 5).

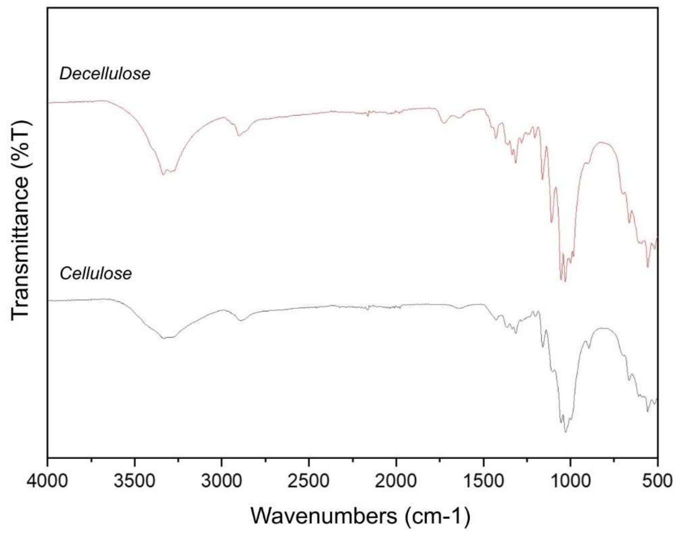


Fig. 5: FTIR spectra of standard cellulose and cellulose extracted from acid treatment of textile waste

These results are consistent with previous studies that reported that cellulose obtained from cotton textile waste exhibited FTIR characteristics very similar to standard cellulose, with the characteristic –OH, C–H, and C–O–C bands still clearly detected (Maciel et al., 2019; Rana et al., 2023). Thus, cellulose obtained from textile waste can be considered to have sufficient structural quality for use in further chemical modification processes.

3.2.3. Formation of Cellulose-Citronellal Fibers

The synthesis of cellulose-citronellal fiber was carried out through a condensation reaction between citronellal and cellulose with the aid of a natural H-Zeolite catalyst. This process aims to incorporate citronellal into the cellulose matrix, producing a functional fiber material with potential bioactive properties. It is important to emphasize that in this study, the nature of the citronellal–cellulose interaction — whether it involves covalent bond formation or physical adsorption — could not be definitively established based solely on FTIR analysis. A more precise characterization using complementary techniques such as solid-state NMR, TGA, or GC-MS of wash extracts would be required to distinguish between chemical bonding and physical incorporation.

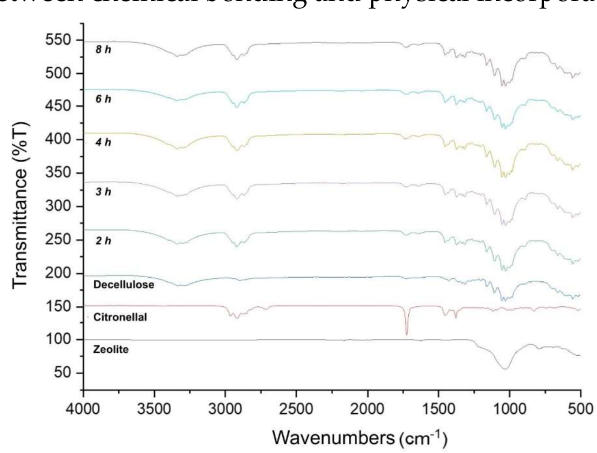


Fig. 6: FTIR spectra of cellulose-citronellal fiber synthesis products with variations in reaction time

Mechanistically, the Brønsted acid sites in H-Zeolite can activate the carbonyl group of aldehydes through protonation. This activation increases the electrophilic nature of the carbonyl carbon, facilitating interaction with the hydroxyl group in cellulose. This mechanism aligns with various reports on the role of zeolite catalysts in facilitating condensation reactions or acetalization between aldehydes and compounds containing hydroxyl groups (Hieu et al., 2022; Muhammad et al., 2024).

FTIR analysis of the reaction products revealed changes in absorption band intensity at several characteristic wavenumbers (Fig 6). At various reaction times of 3, 6, and 9 hours, the intensity of the absorption band at approximately 3000 cm^{-1} gradually increased with increasing reaction time. This band corresponds to the C–H stretching vibrations of the alkyl groups within the citronellal molecules. Furthermore, an increase in intensity was also observed in the 1300–1400 cm^{-1} wavenumber region, which corresponds to the CH deformation vibrations of the alkyl groups.

The increase in intensity in these two spectral regions indicates that the citronellal molecules were successfully integrated into the cellulose matrix. This analytical approach has also been widely used in polymer modification studies, where an increase in the aliphatic C–H band in the FTIR spectrum is considered an indication of successful binding of organic groups to the solid matrix (Adila et al., 2024; Gabrienko et al., 2018). Overall, these results suggest that the natural H-Zeolite catalyst facilitated the incorporation of citronellal into the cellulose matrix, as indicated by the observed changes in FTIR spectral intensity. However, several important limitations must be acknowledged. First, the observed increase in C–H stretching intensity does not alone confirm the formation of a covalent chemical bond between citronellal and cellulose; this spectral change may also be attributable to physical adsorption of citronellal onto or within the cellulose matrix. Distinguishing between these two mechanisms would require additional analyses such as solid-state $^{1}H/^{13}C$ NMR, TGA (thermogravimetric analysis) to assess the thermal stability of loading, or GC-MS analysis of wash extracts to evaluate release behavior. Second, this study did not include reaction yield, citronellal loading efficiency, or washing stability data, which limits the assessment of practical feasibility. Third, no control experiment without a catalyst was conducted, making it difficult to attribute the observed spectral changes specifically to the catalytic role of H-Zeolite. These limitations are acknowledged, and future studies incorporating these analyses are encouraged to provide more conclusive evidence of the material's chemical structure and functional performance.

3.3. Circular Bioeconomy Perspective of Textile Waste Valorization

The results of this study indicate that cellulose obtained from cotton-based textile waste can be used as a raw material for the synthesis of functional materials through a condensation reaction with citronellal using a natural H-Zeolite catalyst. These findings have important implications for textile waste management and the development of biomass-based materials within a circular bioeconomy framework.

Globally, textile waste continues to increase in line with the growth of the fashion industry and clothing consumption. It is estimated that more than 92 million tons of textile waste are generated annually worldwide (Ghosh et al., 2025). However, the utilization rate remains very low. In the United States, approximately 16.9 million tons of textile waste were recorded in 2017, but only approximately 15% was recycled or donated (Wagaw and Babu, 2023). A similar situation also occurs in Europe, where only approximately 15–20% of textile waste is collected for reuse or recycling, while most of the remainder is landfilled or incinerated (Sandin and Peters, 2018). In fact, globally, it is estimated that around 75% of textile waste still ends up in landfills (Juanga-Labayen et al., 2022). Therefore, various studies in the United States and Europe conclude that only around 15% of global textile waste is actually recycled into new products, and only a small portion is processed through fiber-to-fiber recycling.

This trend is also reflected at the national level. According to data from the Ministry of Environment (2025), total waste generation in Indonesia in 2023 reached 23,878,638.66 tons, with fabric waste contributing approximately 2.52% of the total waste generation. In 2024, total waste generation increased to 36,009,662.6 tons, with textile waste contributing approximately 2.55%. Meanwhile, in 2025, waste generation was recorded at 25,141,493.03 tons, with fabric waste contributing approximately 2.46% of the total national waste generation. Although the percentage is relatively small, the absolute volume of textile waste produced remains significant and most of it has not been optimally utilized.

From a circular bioeconomy perspective, textile waste actually holds significant potential as a secondary biomass source. One of the main components of textile waste is cellulose-rich cotton fiber. An estimated 11.6 million tons of cotton waste are generated annually in the global textile industry (Johnson et al., 2020). Cotton fiber contains high-purity cellulose, making it highly potential for conversion into biomass-based materials with higher added value (Homem and Amorim, 2020). Therefore, recovering cellulose from textile waste through chemical recycling can be seen as a promising strategy for textile waste valorization.

Beyond its potential as a biomass source, textile waste also holds significant economic value. It is estimated that globally, unutilized clothing waste has an economic value of approximately USD 400–500 billion per year (Ghosh et al., 2025; Wagaw and Babu, 2023). However, most literature indicates that textile recycling systems still face economic challenges, primarily due to the relatively high costs of collection and processing compared to virgin fiber production. Recycled textile fibers often command slightly higher prices than virgin fibers due to logistics and material recovery costs (Boschmeier et al., 2024; Wagner and Heinzel, 2020). Nevertheless, in the European market, recycled textile fibers still have an economic value of around 2–3 euros/kg, indicating that textile waste can be viewed as an economic resource within a circular economy (Renewable Matter, 2025).

In this study, a valorization approach was employed by utilizing cellulose from textile waste as a raw material for the synthesis of new materials through a condensation reaction with citronellal. The integration of cellulose and active compounds from citronella oil resulted in a biomass-based material with potential applications as a mosquito repellent fiber. Citronella itself is a regionally relevant biomass commodity, as Indonesia is known as a major producer of citronella oil in the growing global essential oil market (Ahmad et al., 2025).

In addition to biomass availability, the successful development of valorization technology also depends on the availability of economical and sustainable catalysts. This study used a natural H-Zeolite catalyst derived from natural zeolite. Globally, natural zeolite production is estimated at around 1.1 million tons per year, with industrial prices ranging from USD 564–935 per ton depending on the country (Research and Markets, 2026). In Indonesia, zeolite is an abundant industrial mineral.

Natural zeolite from Malang, particularly from the South Malang region, primarily contains mordenite and has been widely used in various applications such as bioethanol purification adsorbents, catalysts, filtration media, and soil conditioners (Apandi, 2002; Kurniawan et al., 2019). Natural zeolite is relatively affordable, ranging from IDR 5,000 to IDR 70,000 per kg (approximately USD 0.30–4.25 per kg), making it highly potential for development as a catalyst in biomass-based chemical processes (Ismuyanto et al., 2025). Furthermore, the market value of zeolite in Indonesia is projected to increase from approximately USD 98 million in 2024 to USD 154 million in 2032, demonstrating the growing role of this mineral in various industrial sectors.

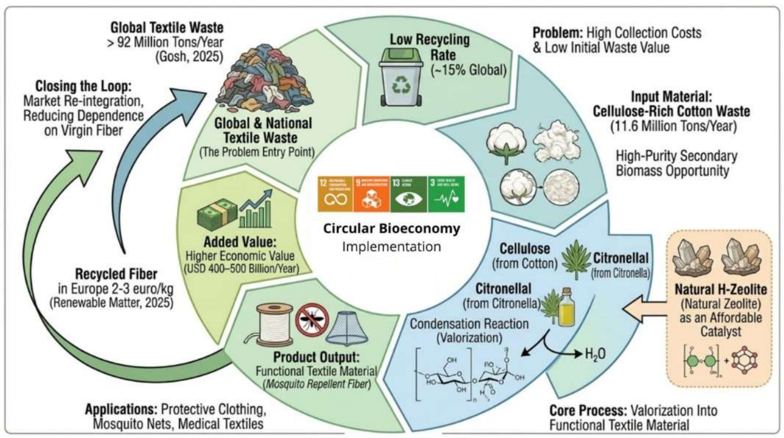


Fig. 7: Circular bioeconomic logic diagram on textile waste fabric valorization

Considering the availability of textile biomass, the economic potential of waste, and the availability of local mineral-based catalysts, the approach developed in this study can be seen as an example of implementing a circular bioeconomy in Indonesia and perhaps applicable to tropical countries with similar challenges. Textile waste is no longer treated as the final residue in a linear production system, but as a source of secondary raw materials that can be converted into functional materials with higher added value. The logic diagram for applying the circular bioeconomy to textile waste in this study is presented in Fig 8.

This approach directly supports SDG 12 (Responsible Consumption and Production) through efforts to reuse waste materials and SDG 9 (Industry, Innovation, and Infrastructure) through the development of waste-based functional biomaterials. Indirectly, this approach also has the potential to contribute to SDG 13 (Climate Action) and SDG 3 (Good Health and Well-being) through biomass utilization and the development of textile materials with potential mosquito repellent activity.

Although this study demonstrates the potential of utilizing textile waste as a source of cellulose for the synthesis of functional materials, several limitations still need to be considered. First, the material characterization conducted in this study is still limited to chemical structure and morphology analysis using FTIR, SEM, and XRD. Evaluation of the physical and mechanical properties of cellulose-citronellal fibers, such as tensile strength, thermal stability, and resistance to environmental degradation, has not been comprehensively analyzed. These parameters are important for assessing the material's suitability for industrial-scale textile applications. Second, the mosquito repellent activity testing in this study is still preliminary. A more comprehensive evaluation is needed to assess the protective effectiveness under real-life conditions. Important parameters such as the duration of protection, the release kinetics of citronellal from the cellulose matrix, and the stability of the active compound against washing processes and environmental exposure have not been analyzed in depth. Third, this study has not yet quantitatively evaluated the sustainability aspects of the process. Although the approach used in this study utilizes biomass and a natural mineral-based heterogeneous catalyst, a quantitative process sustainability analysis has not been conducted. Previous studies have shown that the wet processing stages in the textile industry, particularly pretreatment, dyeing, and finishing, are dominant contributors to various environmental impact categories, such as global warming potential, aquatic ecotoxicity, and water consumption (Catarino et al., 2025; Li et al., 2024). In fact, in some studies, the dyeing process unit was reported to contribute the largest contribution to almost all environmental impact indicators at the midpoint impact level (Gonzalez et al., 2023). Therefore, a quantitative evaluation of the technology developed in this research is essential to comprehensively assess its potential environmental benefits.

The approach commonly used in these studies is a Life Cycle Assessment (LCA) with cradle-to-gate or gate-to-gate system boundaries, which evaluates various indicators such as carbon footprint, energy consumption, water use, and potential toxicity in the textile production system (Imran et al., 2023). Therefore, integrating LCA analysis, evaluating process energy consumption, and estimating potential greenhouse gas emission reductions is a crucial research agenda for further research to assess the contribution of this textile waste valorization technology to a more sustainable textile production system. Furthermore, the scale-up process has not been evaluated in this study. The cellulose extraction, citronellal isolation, and material synthesis processes are still being conducted at the laboratory scale, requiring further studies on process efficiency, energy requirements, and production costs at an industrial scale.

4. CONCLUSIONS

This study demonstrates the feasibility of converting cotton-based textile waste into functional biomaterials through a catalytic modification process within a circular bioeconomy framework. Cellulose recovered from textile waste through acid hydrolysis exhibited structural and morphological characteristics comparable to commercial cellulose, confirming that textile waste can serve as a viable secondary biomass resource. Natural H-Zeolite catalyst prepared through acid activation, ion exchange, and calcination showed significantly increased acidity while maintaining zeolite crystalline integrity. The Brønsted acid sites present on the H-Zeolite catalyst facilitated the condensation-impregnation process between citronellal and cellulose, and FTIR analysis indicated the incorporation of citronellal into the cellulose matrix, suggesting the formation of cellulose-citronellal fibers with potential bioactive properties.

Beyond laboratory-scale synthesis, this study highlights the relevance of the proposed approach within a circular bioeconomy context. By integrating cellulose from textile waste and citronellal from plant-based essential oils, the pathway demonstrates a strategy for valorizing textile residues into bio-based functional materials with higher added value. Using Indonesia as a case study, with its available textile waste, citronella biomass, and natural zeolite deposits, the approach is particularly applicable to tropical countries facing similar resource conditions and vector-borne disease challenges.

Several limitations remain to be addressed. The current work focuses primarily on chemical and structural characterization, and future research should investigate the mechanical properties, durability, wash stability, and citronellal release kinetics of the synthesized fibers. Quantitative sustainability assessments, including Life Cycle Assessment, energy consumption analysis, and greenhouse gas emission estimation, are also recommended to evaluate the environmental benefits and scalability of this valorization approach.

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