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MORPHOLOGICAL AND THERMAL PROPERTIES STUDY OF PCL BIOCOMPOSITES WITH NANOCELLULOSE FILLER FROM RICE HUSK USING PCLacac COMPATIBILIZER AS MATERIAL CANDIDATE FOR BONE SCAFFOLDS

Yossie Juliastri Napitupulu[1], Muhammad Yusuf[1*], Eddiyanto[1], Innayah Wulandari[1], Winsyahputra Ritonga[2]
[1]Department of Chemistry, Faculty of Mathematics and Natural Sciences, Medan State University
[2]Department of Physics, Faculty of Mathematics and Natural Sciences, Medan State University
myusuf@unimed.ac.id

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ABSTRACT
Biocomposites from poly(ε-caprolactone) (PCL) matrix with nanocellulose filler are materials that have been developed for biomedical applications, such as scaffolds, tissue engineering, implants, and wound dressings. The PCL matrix is flexible and biocompatible. Meanwhile, the rice husk nanocellulose filler is environmentally friendly, renewable, and abundantly available in Indonesia. However, the differences in the hydrophobic properties of PCL and the hydrophilic properties of nanocellulose cause low interfacial adhesion, so a compatibilizer is needed to improve the compatibility of the two phases. This study aims to examine the effect of adding short-chain PCL (PCLacac) as a compatibilizer. The mixing of the PCL matrix, nanocellulose filler, and PCLacac compatibilizer used the solvent casting method. The characterization results using Scanning Electron Microscopy (SEM) showed that the addition of PCLacac can improve interfacial adhesion between the matrix and filler, characterized by a more even filler dispersion and reduced agglomeration. On the other hand, thermogravimetric analysis (TGA) analysis showed that the addition of 3% PCLacac resulted in an increase in decomposition temperature, which indicates an increase in thermal stability due to better interfacial interactions between the matrix and filler. Thus, the addition of PCLacac plays an effective role in improving the morphological structure and thermal resistance of PCL/rice husk nanocellulose biocomposite, so it has the potential to be developed as a biocomposite material in biomedical applications including bone scaffolds

Keywords: Biocomposite, Bone Scaffolds, Compatibilizer, PCL, Rice Husk Nanocellulose.

INTRODUCTION

In recent years, the development of biocomposites as environmentally friendly materials has been used as a substitute for synthetic materials with good mechanical properties, light weight, and the ability to degrade naturally (Diana et al., 2020). Biocomposites are composed of a matrix and filler mixed to improve material properties (Nurfajriani et al., 2026). One of the widely used matrices is a polymer such as poly(ε-caprolactone) (PCL), which is biodegradable, biocompatible, and has the potential for widespread application, especially in the biomedical field, such as scaffolds, tissue engineering, implants, and wound dressings.

On the other hand, the filler used can be derived from cellulose. Cellulose sources are obtained from biomass such as empty oil palm fruit bunches, bamboo fiber, sugar cane fiber, straw, and coconut fiber. However, the cellulose used is generally still in the form of micrometer-sized fibers that easily agglomerate. Therefore, the particle size of cellulose needs to be reduced to nanocellulose to improve its dispersion and performance in composite materials. Furthermore, nanocellulose has a larger surface area than cellulose, allowing for better interfacial interactions with the polymer matrix. Consequently, the filler can be dispersed more homogeneously, potentially producing biocomposites with a more uniform morphology and improved thermal stability (Celebi et al., 2022) (Siqueira et al., 2013).

However, one of the challenges in producing PCL- and nanocellulose-based biocomposites is the poor compatibility between the relatively hydrophobic matrix and the hydrophilic nanocellulose filler (Kurniawan et al., 2026). This difference in properties can lead to filler agglomeration and poor interfacial adhesion, thus reducing the morphology and thermal stability of the biocomposite. Therefore, the addition of a compatibilizer capable of enhancing the interaction between the two components is necessary.

In previous research, the research team successfully synthesized short-chain PCL using bis(acetylacetonato)Zr complex as a catalyst (PCLacac) (Yusuf et al., 2023) (Yusuf et al., 2022). PCLacac was used as a compatibilizer because it has a similar structure to the PCL matrix, thus bridging the interaction between the hydroxyl groups in nanocellulose and the carbonyl groups in PCL through the formation of hydrogen bonds (Kurniawan et al., 2026). This interaction is expected to increase the dispersion of nanocellulose in the matrix, resulting in a more homogeneous biocomposite structure.

The success of adding a compatibilizer to a biocomposite is indicated through morphological and thermal characterization. Morphological characterization using Scanning Electron Microscopy (SEM) aims to observe the distribution of nanocellulose, the level of homogeneity, and the quality of interfacial adhesion between the filler and the PCL matrix. Meanwhile, characterization using Thermogravimetric Analysis (TGA) is also carried out. (TGA) aims to evaluate the thermal stability of materials based on degradation patterns and mass changes due to increasing temperature. The combination of these two characterization techniques provides complementary information regarding the relationship between microstructure and thermal resistance of biocomposites.

Based on the description, this study focuses on the development of PCL biocomposites with nanocellulose filler from rice husk using PCLacac compatibilizer as material candidate for bone scaffolds. The addition of a compatibilizer can improve the compatibility and interfacial adhesion between the matrix and filler. Through this approach, the resulting biocomposite is expected to have better filler dispersion and thermal properties.

RESEARCH METHOD

Method

The instruments used in this study were SEM (Zeiss EVO MA10) and TGA (Shimadzu DTG-60). The materials needed for the isolation of rice husk cellulose consisted of rice husk waste biomass obtained from City X. nitric acid (HNO_3) (Sygma Aldrich), Sodium Nitrite (NaNO2) (Sygma Aldrich), sodium chlorite ($NaClO_2$) (Sygma Aldrich), sodium sulfite (Na_2SO_3) (Sygma Aldrich), sodium hydroxide (NaOH) (Sygma Aldrich), and concentrated sulfuric acid (H_2SO_4) (Sygma Aldrich). Meanwhile, the materials for making biocomposites include long-chain PCL (CAPA 6800) and short-chain PCLacac which are the results of synthesis using bis(acac)$_2$Zr catalyst, and chloroform ($CHCl_3$) (Sygma Aldrich).

Isolation of Nanocellulose from Rice Husk

The process of isolating rice husk cellulose begins with washing, drying, and refining, as well as sieving to obtain rice husk powder with a uniform size. Next, a chemical process is carried out through several stages. The first stage uses a mixture of 3.5% HNO_3 solution and 10 mg $NaNO_2$ while heating. The second stage uses a mixture of 2% NaOH solution and 2% Na_2SO_3 , then heated. The third stage uses a 1.75% NaOCl solution and heated, at this stage it is repeated until a white residue is produced. The fourth stage uses a 17.5% NaOH solution and heated again. From these four stages, alpha (α)-cellulose will be produced. which is hydrolyzed using a 64% sulfuric acid solution at a temperature of 45 °C. Then, it is centrifuged at a speed of 1100 rpm to separate the hydrolysis precipitate. The precipitate is then dried in an oven to obtain nanocellulose (Napitupulu et al., 2026).

Biocomposite Manufacturing

The PCL/PCLacac biocomposite film with nanocellulose filler was prepared using a solvent casting technique. PCL was dissolved in chloroform and short-chain PCLacac compatibilizer was added. After that, nanocellulose filler was added . The mixture was refluxed for 1 hour while stirring at 300 rpm. The resulting solution was then poured into an ASTM D638 mold to form a biocomposite film. Variations in the addition of PCLacac to the PCL matrix were 0% and 3%. Meanwhile, rice husk nanocellulose was added at 1% of the total material. The resulting biocomposite was then tested for its thermal morphology using SEM and TGA (Kurniawan et al., 2026)

RESULT AND DISCUSSION

SEM Analysis of Nanocellulose

Morphological analysis of nanocellulose from rice husk was performed using Scanning Electron Microscopy (SEM). This characterization aims to observe the morphology, size and structure of the particles, as well as changes in the morphology of the cellulose fibers after the hydrolysis process. In addition, this characterization aims to compare the biocomposite without the addition of 0% PCLacac as a compatibilizer with the biocomposite with the addition of 3% PCLacac. The following is an image of the SEM morphology.

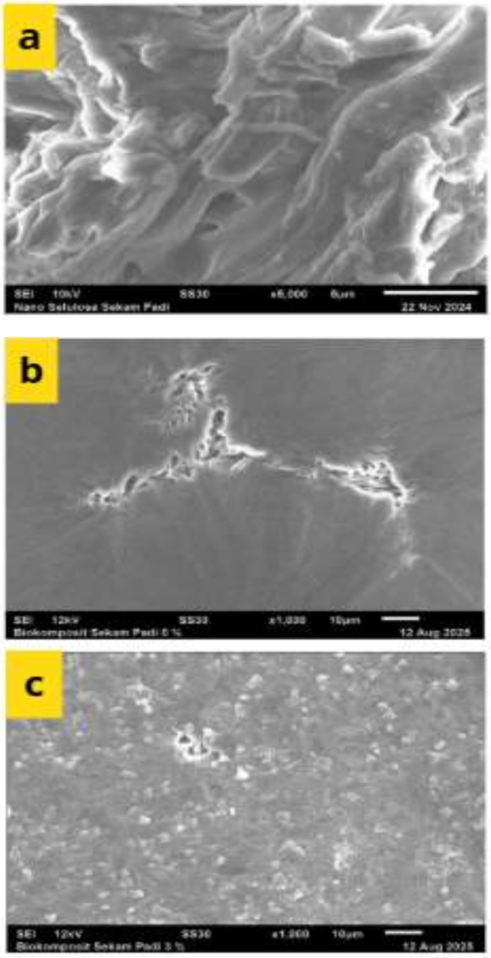

Figure 1. SEM morphology of a). Rice Husk Nanocellulose, b). Biocomposite with 0% PCLacac, c). Biocomposite with 3% PCLacac

Figure **1a** shows the morphology of nanocellulose particles isolated from rice husks (Napitupulu et al., 2026). The fiber structure obtained from the rice husk nanocellulose appears to be irregularly distributed and interconnected. This indicates that crystalline and amorphous cellulose have separated (Gea et al., 2020) (Ningtyas et al., 2020). Furthermore, the morphology of particle distribution in biocomposites with 0% PCLacac composition and 3% PCLacac composition is shown in Figure **1b** and **1c**. Figure **1b** shows a relatively smooth surface, but there are cracks and voids in the center. These cracks indicate a failure of the interface between the matrix and the filler. Consequently, the presence of voids indicates that the mixing process of the filler in the matrix has not taken place perfectly. Meanwhile, Figure **1c** shows a rougher surface, but the particles are distributed throughout the surface. The particle distribution appears more even, although there are still voids in the center. This indicates that there is interfacial contact between the filler and the matrix caused by the addition of 3% PCLacac compatibilizer.

TGA Analysis Biocomposite

Thermogravimetric analysis (TGA) was carried out to evaluate the changes in mass and decomposition temperature of the biocomposites during heating, so that their thermal stability against increasing temperature can be observed.

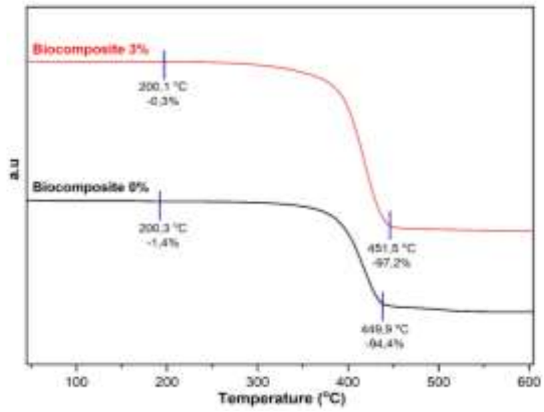

Figure 2. TGA analysis of PCL/PCLacac/Rice husk nanocellulose biocomposite

Figure 2 shows the TGA curves of biocomposites with PCLacac compatibilizer compositions of 0% and 3%. Both biocomposites underwent thermal degradation stages that occurred in the temperature ranges of 0–200 °C and 200–450 °C. The first stage showed a mass reduction of 0.3% and 1.4%, respectively, which was associated with the evaporation of water and volatile compounds, such as residual solvents that had not completely evaporated during the synthesis and molding processes of the biocomposites (Saputra et al., 2025).

Furthermore, the second stage saw mass reductions of 94.4% and 97.2%, respectively, associated with primary thermal degradation. This stage involves the cleavage of intermolecular interactions and the cleavage of the polymer chains making up the biocomposite, namely PCL, PCLacac, and nanocellulose, accompanied by the depolymerization of PCL into its monomers (Hejna & Kosmela, 2020).

The addition of 3% PCLacac resulted in a slight improvement in the thermal stability of the biocomposites, which was reflected by the increase in the decomposition temperature from 449.9 °C to 451.5 °C. This improvement is due to stronger interchain interactions promoted by the compatibilizer. Furthermore, PCLacac can act as a protective layer (compatibilizing layer) that inhibits oxygen diffusion during heating.

CONCLUSION AND SUGGESTION

The addition of the PCLacac compatibilizer was found to enhance the morphological characteristics and thermal stability of the PCL biocomposites with nanocellulose filler from rice husk. SEM observations show that the addition of PCLacac produces a more even dispersion of nanocellulose filler in the PCL matrix. Meanwhile, the results of TGA analysis show that the biocomposite with the addition of 3% PCLacac has a slightly higher decomposition temperature than the biocomposite without a compatibilizer. This increase indicates that PCLacac plays an effective role as a compatibilizer in strengthening the interaction between the carbonyl groups of PCL and the hydroxyl groups of nanocellulose, resulting in a biocomposite with better morphology and higher thermal resistance. Thus, this biocomposite is suitable for use as material candidate for bone scaffolds.

In future research, the use of an internal mixer is recommended to achieve more homogeneous mixing of the filler, matrix, and compatibilizer, as well as a more uniform filler dispersion. This is expected to enhance the interfacial interactions between the components, resulting in biocomposites with improved mechanical properties and thermal stability.

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