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Applications of nanocellulose and its composites in bio packaging-based starch

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A R T I C L E I N F O

Article history:
Available online 23 November 2022

Keywords:
Bio packaging
Biopolymer
Nanocellulose
Starch

A B S T R A C T

This mini-review discusses existing technology and future issues in applying nanocellulose as a starch-based packaging of food material. Biopolymers, mainly starch as packaging materials, are increasingly replacing petroleum plastics. This mini-review encompasses applying the commonly used nanocellulose starch-based bio packaging material, focusing on production processes, properties, and analysis of potential uses in starch-based bio packaging. The use of nanocellulose as an alternative material for starch-based bio packaging substitutes conventional polymers for food packaging and its entirely new properties and characterization. Microorganisms can produce cellulose biopolymers through the fermentation process of various biological resources (e.g., bacterial cellulose). Biomass can be produced directly from various plants (pineapple, water hyacinth, and others). Researchers are currently focused on reducing the problem of pollution due to conventional plastics produced from fossil fuels.

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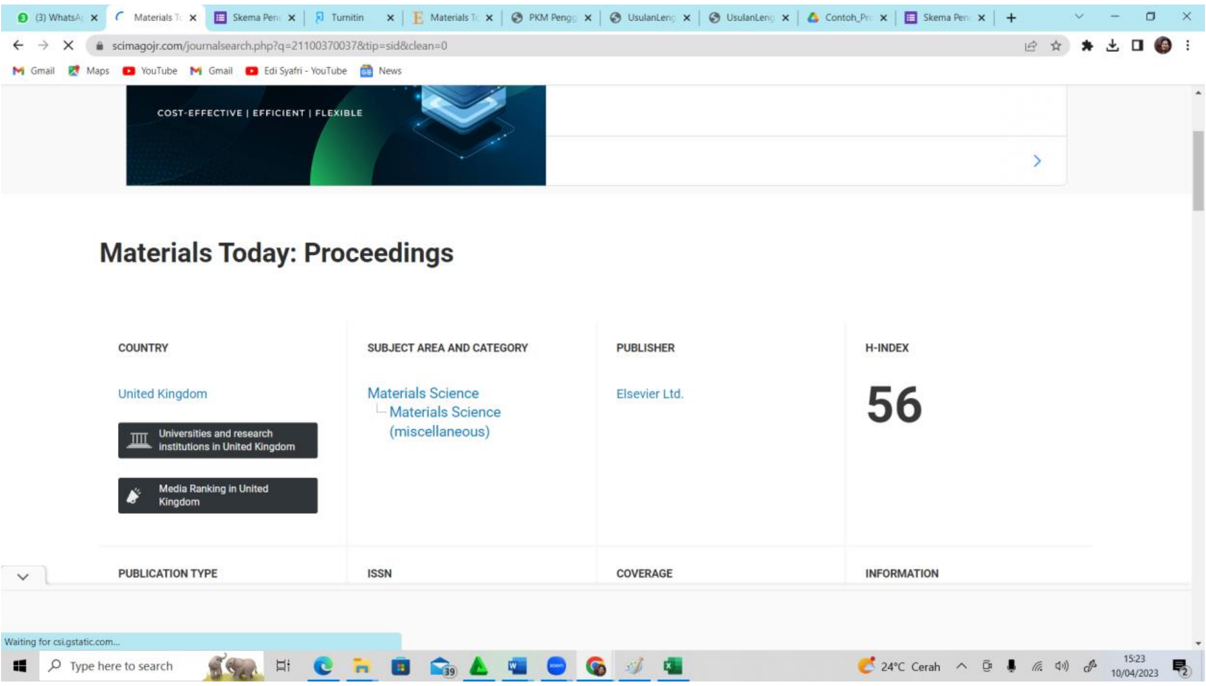

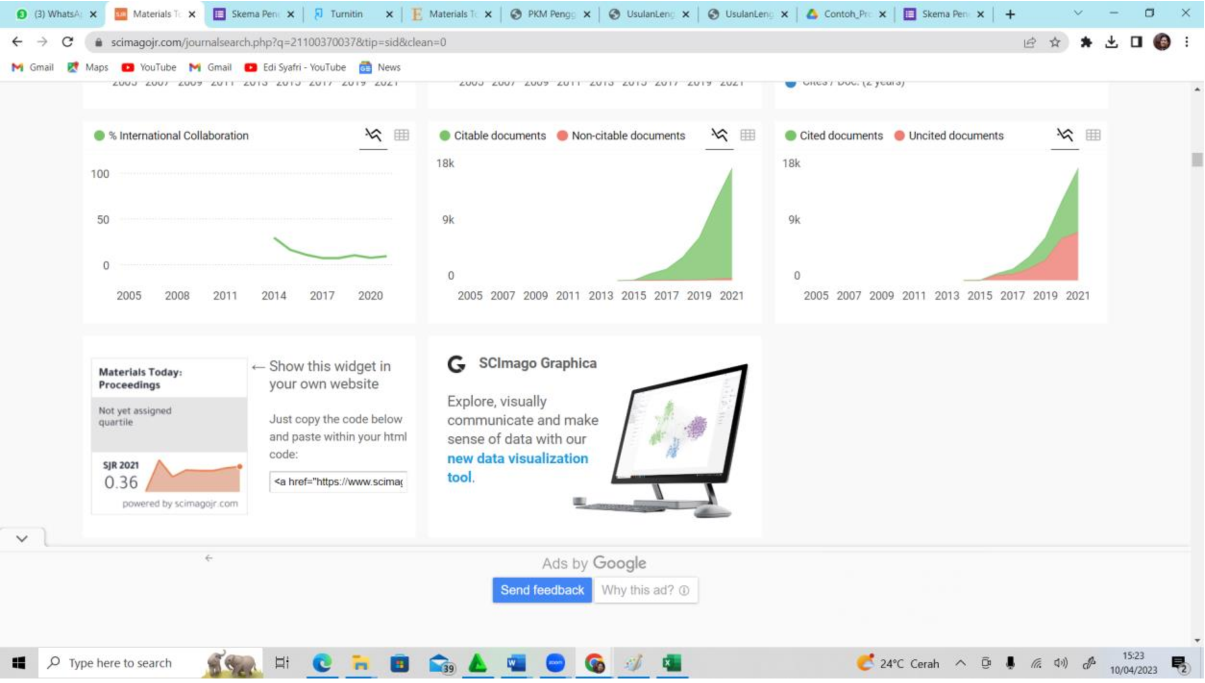

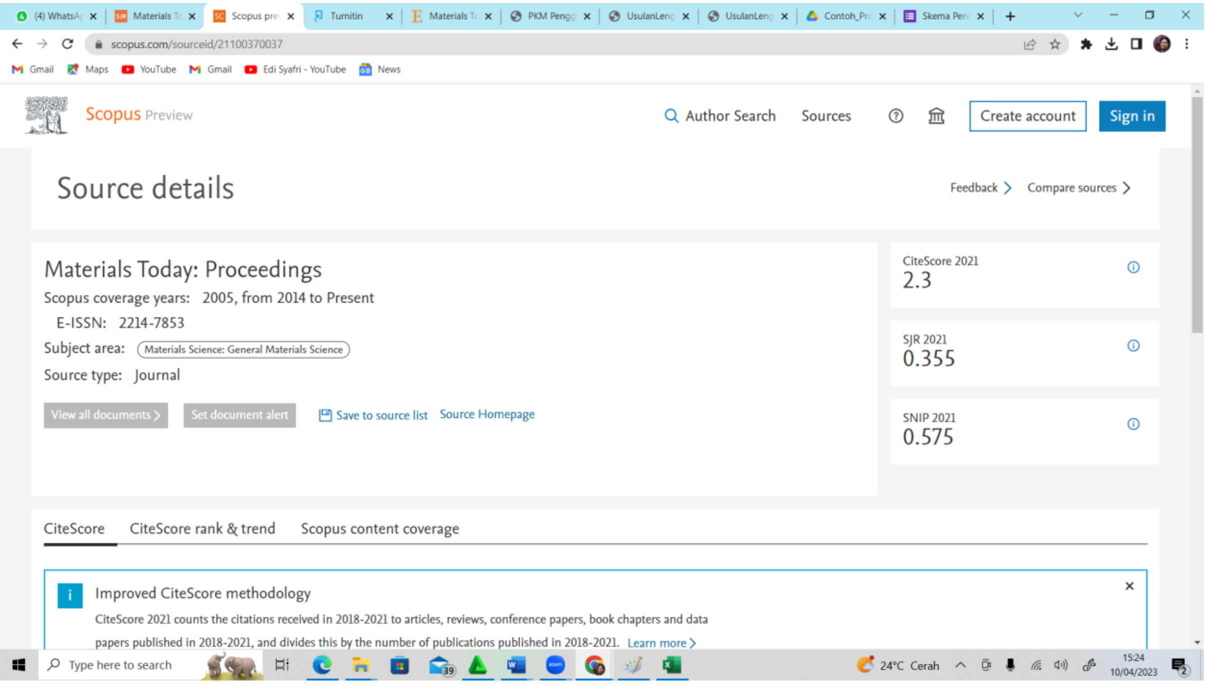

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1. Introduction

Conventional plastic polymers produced from fossil fuels have been used for food packaging. Plastic is utilized to assure the safety and integrity of packaged food items, from production, handling, and storage to final usage by consumers. In particular, plastics play an essential role in avoiding rapid deterioration in product safety and quality, which influences the overall usage of packaged foods during their lifespan and ultimately avoids product spoilage losses. The interconnections among food, packaging materials, and environmental conditions focus on food packaging. Conventional plastics as food packaging materials have caused environmental pollution on land and sea, showing synthetic plastic pollution. Therefore, using environmentally friendly materials such as starch-based bio packaging with nanocellulose filler is an alternative solution to replace synthetic plastic.

Starch is the most widely used renewable raw material for bio packaging. Starch consists of glucose, amylose, and amylopectin. Its chemical and physical characteristics are unique compared to all other carbohydrates. Sources of starch are obtained from whole grains, legumes, cereals, potatoes, and fruits [1]. Starch polymers are susceptible to moisture, with high water vapour permeability, and poor mechanical properties limiting the application of bio packaging. Several previous studies using starch as bio packaging are yam starch [2], cassava starch [3], sugar palm starch [4], corn starch [5], tapioca starch [6], sago starch [7], rice starch [8], and potato starch [9]. Starch derivatives are the most interesting and used in the food sector because they can be combined with various fillers, types, and amounts of plasticizers used during starch-based bio packaging. To obtain superior properties that determine the final product's physical, chemical, and thermal properties. Its main utilization in the food sector is inflexible and solid packaging (biofilm, packaging, bio-coating, lamination, and others). The properties of starch-based bio packaging generated are biodegradable and it has good properties as a barrier to oxygen, moisture, and amylose content is a strong restrictive factor, related to the mechanical qualities of the starch film [10]. In many cases, to modify the properties of starch-based films, fillers or reinforcing agents have been added to the starch-polymer matrix, such as microcrys-

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https://doi.org/10.1016/j.matpr.2022.11.138
2214-7853/
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talline cellulose (MC), fiber, nano-clay, carboxymethylcellulose (CMC), carbon nanotubes, and nanocellulose [11].

Of several cellulose derivatives that have been produced commercially, such as cellulose acetate, it is widely used for food packaging (baked foods and fresh products) [12]. The properties of cellulose acetate are low resistance to gas and moisture and use the plasticization process when used for film development. Several previous studies using cellulose in food packaging based on yam starch films with nanocellulose from pineapple leaves [2], cassava starch with oil palm mesocarp cellulose nanowhiskers [13], bacterial cellulose nanofibers/starch/chitosan for a food packaging alternative [14], and potato starch/cellulose for sustainable packaging [9]. The film-forming properties have many cellulose derivatives, so the cost is too high for wider industrial use. In order to produce lower-cost cellulose packaging materials, processing technology for the production of cellulose derivatives is required. Nanocellulose fiber (NCF) reinforced with starch through film casting technique processing has resulted in very successful bio packaging [15]. In this way, bio packaging improves its mechanical properties. In addition to this combination, starch mixed chitosan in the amount of 30 % significantly increased the mechanical properties of the bio packaging by 97.8 % [15]. Various types of starch reinforced with nanocellulose have been used in packaging applications for bread, vegetables, and meat products stored under standard conditions [16].

Bio packaging made of tapioca starch with bacterial cellulose nanofibers and filler chitosan as reinforcement has excellent mechanical properties, processability, water and gas vapour resistance, and thermal resistance [14]. Fully biodegradable polymers are not recyclable from currently applied technology used in recycling conventional petroleum-based polymers. Starch-based biopolymer with nanocellulose filler can be used for wrapping food products. In this way, non-biodegradable plastic materials will no longer be needed. Biopolymers provide attractive functionality while maintaining the environmentally friendly characteristics of the material. Chitosan [17], carrageenan [18], and starch [19] have become a commonly researched and applied material as a biomaterial for bio packaging. Previous studies have found that the use of starch-based nanocomposite films modified by nanocellulose and chitosan for food packaging applications showed significant performance improvements. Balakrishnan et al. (2017) wrote about making sustainable packaging based on potato starch with added pineapple leaf nanofiber reinforcement to obtain extraordinary results because the bio packaging has better barrier properties in UV resistance and higher transparency [9]. In addition, any given bio packaging material must fulfil requirements related to conventional packaging materials. It refers to the permeability properties (permeability to water vapour and gases, aroma substances, and light), mechanical, and optical properties (e.g., transparency) [20].

2. Starch-based bio packaging production process and its composites

Previous research conducted by Marichelvam et al. (2019) made alternative packaging materials from corn and rice starch [19]. In rice and cornstarch-based bio packaging, glycerol was used as a plasticizer due to its better mechanical properties and good water solubility [19]. The bio-plastics produced by the solution casting method were prepared according to the following procedure starch, glycerol, and citric acid were added to distilled water in various ratios. The mixture was stirred and heated on a hot plate under certain conditions [19]. Then it was poured into a Teflon-coated glass mold, leveled and naturally dried for 3–4 days. Saoza et al. (2020) reported different casting method process to get the bioplastic films where the films was casted in polystyrene (PS) plates and dried in an oven at certain temperature for 12 h [21]. Starch was dispersed in distilled water and continuously stirred to achieve starch gelatinization (see Fig. 1). Then, the mixture was added to the nanocrystalline glycerol cellulose, which is stirred and dried in the oven [21].

Abral et al. (2021) also reported a method of making the edible antimicrobial film using starch/chitosan gel mixed with bacterial cellulose suspension through a solution casting process by pouring the solution, which has been sonicated into a petri dish and dried in the oven [14]. The addition of chitosan in biocomposite was aimed to get transparent film and the sonication would produce the homogenous dispersion. Another study by Noorbakhsh et al. (2018) also used a certain amount of chitosan that was dissolved in acetic acid while stirring NCF-Thermoplastic starch (TPS) solution to produce a transparent solution [15]. They also employed the sonication procedure to get the homogenous solution. Solution casting seems to be typical method for preparing the films of NCF-TPS biocomposites [14,15,22].

Different kinds of molding substrate were used to cast the film including teflon, PS, common petri dish, and acrylic mold. The utilization depends on the process of gelatinization and plasticization, acidic acid and glycerol are the common solvent that acts as plasticizer to form the film during the drying process and the ratio of this solvent added in to the composite mixture is need to take into account [14,15,23].

3. Properties in bio packaging

The film from corn and rice starch with glycerol and citric acid can increase the tensile properties after rice starch [19]. Then, the water absorption and water solubility are also reduced [19]. Starch-based films with nanocellulose-stabilized pickering emulsions of ho wood (*Cinnamomum camphora*) exhibited strong chemical interactions, which significantly increased the mechanical resistance of the films [21]. The films exhibit high thermal stability due to strong molecular interactions between starch chains and pickering emulsions [21]. In addition, the film exhibits a lower rate of water vapour transmission. The starch-based film having good thermal stability is an indication that it can be safely applied in the food industry. Previous studies reported the highest tensile strength and high thermal resistance after nanofiber addition [14]. In addition, nanofibers also increase moisture resistance water barrier, increase in the content of nanocellulose and chitosan results in an increase in young's modulus, tensile strength, elongation at break, and transparency [15]. Characteristics of NCF in TPS film-based biocomposite showed high tensile strength; the percentage of elongation at break is reduced; reduction in water vapour and oxygen permeability compared to TPS control films [22]. As the reinforcement of the film, NCF provides strength and stiffness to the composite because of its strong interfacial adhesion and homogeneous dispersion with the TPS. Moreover, the crystallinity and nanometer size effect of the NCF play an important role to improve the mechanical behavior of the biocomposite film [22].

Bamboo, cotton linter, and sisal fibers are a source of nanocellulose used as reinforcement in corn starch films, with the schematic diagram of the development of the starch-based biocomposite films (see Fig. 2) [23]. The addition of nanocellulose did not affect the morphology and transparency of the three types of composite films. However, it improved the mechanical and barrier properties in certain ratio of the NCF and the TPS. The addition of more NCF to make the composite is not necessarily improve the mechanical characteristics even it can decrease the elongation break of the film [23]. TGA analysis was done by Q. Chen et al. (2019) suggest that the biggest ratio of NCF in TPS also decrease the thermal stability

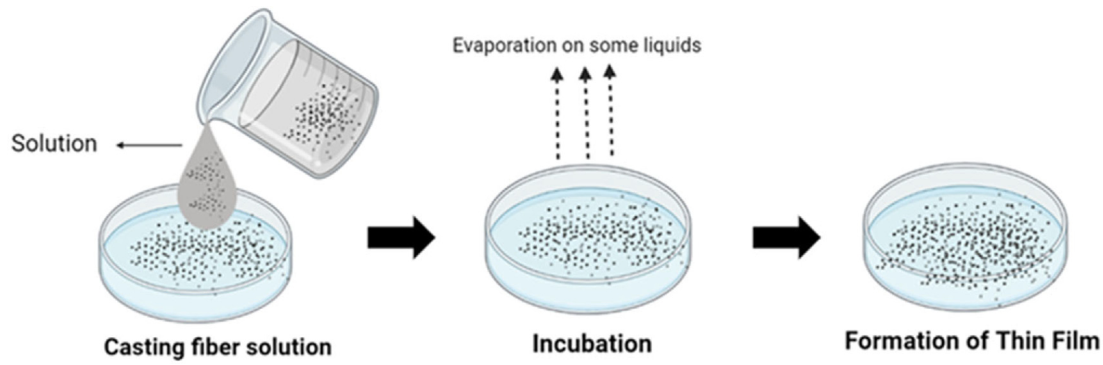
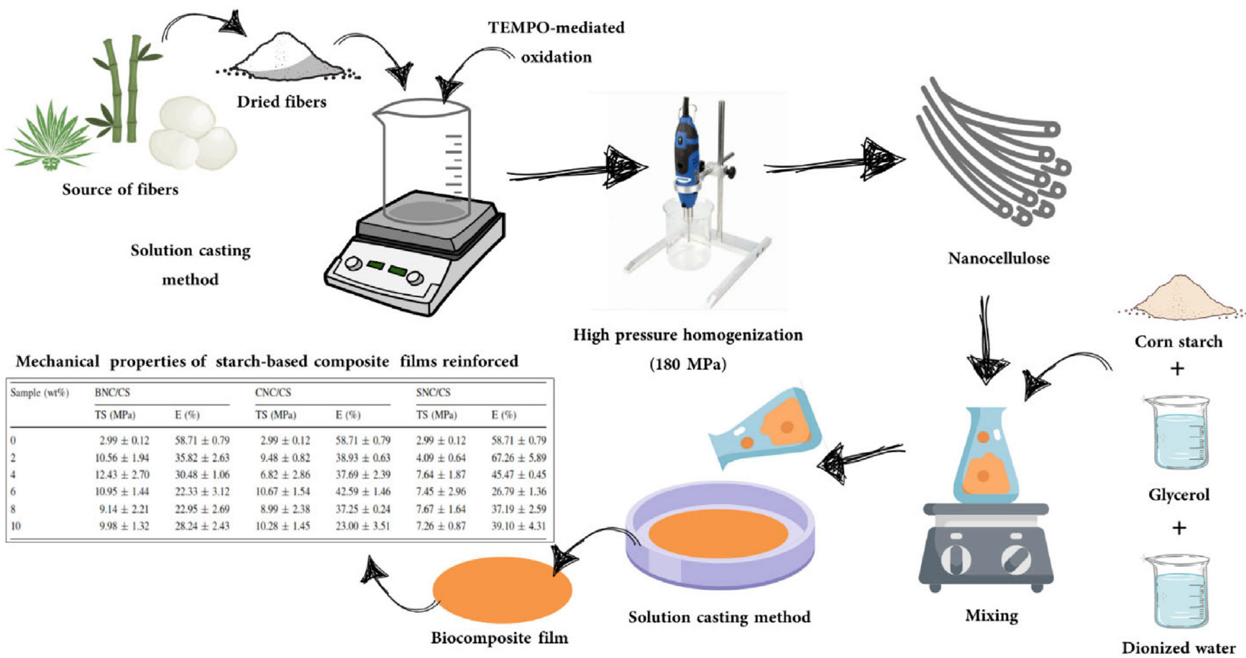


Fig. 1. Simple Illustration method of solution casting in manufacturing bioplastics.



Sample (wt%)	BNC/CS		CNC/CS		SNC/CS	
	TS (MPa)	E (%)	TS (MPa)	E (%)	TS (MPa)	E (%)
0	2.99 ± 0.12	58.71 ± 0.79	2.99 ± 0.12	58.71 ± 0.79	2.99 ± 0.12	58.71 ± 0.79
2	10.56 ± 1.94	35.82 ± 2.63	9.48 ± 0.82	38.93 ± 0.63	4.09 ± 0.64	67.26 ± 5.89
4	12.43 ± 2.70	30.48 ± 1.06	6.82 ± 2.86	37.69 ± 2.39	7.64 ± 1.87	45.47 ± 0.45
6	10.95 ± 1.44	22.33 ± 3.12	10.67 ± 1.54	42.59 ± 1.46	7.45 ± 2.96	26.79 ± 1.36
8	9.14 ± 2.21	22.95 ± 2.69	8.99 ± 2.38	37.25 ± 0.24	7.67 ± 1.64	37.19 ± 2.59
10	9.98 ± 1.32	28.24 ± 2.43	10.28 ± 1.45	23.00 ± 3.51	7.26 ± 0.87	39.10 ± 4.31

Fig. 2. Schematic diagram of the development of the starch-based biocomposite films [23].

of the bio composites. Savadeker et al. (2012) reported that melting temperature of NCF-TPS biocomposite assessed by the differential scanning calorimeter (DSC) shifted to the lower temperature if the ratio of the NCF in the biocomposite increased. In general, the mixed ratio is important to promote an effective utilization of NCF to achieve the best enhancement for TPS film.

4. Conclusions

In industrialized countries, biopolymer materials such as starch and nanocellulose largely replaced conventional polymers as food packaging materials, mainly natural, organic, and functional food packaging. Nanocellulose provides strength and stiffness to the composite because of its strong interfacial adhesion and homogeneous dispersion with thermoplastic starch for food packaging. Biopolymers were produced for various uses, ranging from food packaging to high technology. Despite biopolymers' benefits, many drawbacks limit the wide industrial use of these materials, especially in food packaging. It is generally due to higher efficiency and price when compared to the petroleum-based polymer. Biopolymers can be classified into various groups and quality categories according to the production method and applied in the food sector. Cellulose-based biopolymers occupy the most extensive industrial application in the food packaging sector. Only a few additional biobased polymers are used industrially in packaging traditional foods. Material technology improvements have been made in generating industrial solutions utilizing starch-based biomaterials for many types of food packaging. Accordingly, biopolymers have grown in importance in food applications. The leading cause is that bio packaging is made from renewable resources and reused via recycling. Researchers focusing on biopolymers have demonstrated that they have features suited for a greater variety of usage in the food sector to attain commercially acceptable prices. In the future, when comparing bio packaging with recycled petroleum plastics, packaging materials created from sustainable biological resources will soon have economic cost, and appropriate quality for food packaging application.

CRediT authorship contribution statement

Melbi Mahardika: Conceptualization, Writing – original draft, Writing – review & editing. **Devita Amelia:** Visualization, Writing – review & editing. **Azril:** Writing – review & editing. **Edi Syafri:** Supervision, Funding acquisition.

Data availability

No data was used for the research described in the article.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors are thankful to the Integrated Laboratory of Bioproducts (iLaB), National Research and Innovation Agency Republic of Indonesia for their kind support.

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