A mini-review of bio-scrubber derived from bacterial cellulose impregnated by flavonoid of moringa leaves

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Abstract. Scrubber is widely used in various products, such as cosmetics, facial cleansers, and soaps. The use of scrubber releasing 209.7 trillion microplastics would harm the environment due to lack of process in treatment facilities. Efforts to substitute plastic-based scrubbers with more environmentally friendly materials need to be made. In previous studies, substitution scrubber with grape seeds has been done but has a low viscosity. This problem may be solved by using bacterial cellulose (BC) in the manufacture of bio-scrubbers. Several methods are currently being investigated to produce bacterial cellulose microparticles, such as mechanical methods using high-pressure homogenizer (HPH), acid hydrolysis, microbial hydrolysis, hydrogel fiber cultivation, microfluidic process, and ultrasonication. This review recommends the manufacture of bacterial cellulose microparticles by ultrasonication method. The recommendation is based on the literature study that has been carried out. The ultrasonication method has more advantages than other methods. It does not use solvents that pollute the environment and increasing the number of bacterial cellulose microparticles. The synthesis of bio-scrubber from bacterial cellulose ends with the drying process of bacterial cellulose microparticles. This review recommends the ambient pressure drying method. The ambient pressure drying method can produce bio-scrubber with high crystallinity, high mechanical properties, and transparency.

1. Introduction
Plastic waste can be decomposed into microplastics by environmental factors, such as exposure to sunlight, wind, and water [1]. Microplastics have been classified as contaminants by Scotland’s Centre of Expertise for Waters due to its small size, which is difficult to remove from the environment [2,3]. Microplastics can come from cosmetic products, such as facial scrubbers. The use of scrubbers could release 209.7 trillion microplastics into the environment each year [4]. Waste treatment facilities cannot process microplastics in scrubber products, so it is toxic to the environment [5]. Plastic scrubbers can absorb chemical pollutants in aquatic habitats and can be transferred into the bodies of animals that consume them. It can lead to diseases such as cellular necrosis, inflammation, and tissue laceration [6]. Therefore, some countries have banned the use of microplastics in cosmetic products, including Canada, the UK, the USA, and the Netherlands [5].

However, the use of scrubbers is indispensable for exfoliating dead epidermis cells, producing clean skin, accelerating cell renewal, scar removal, stain removal, and acne treatment [7]. Therefore scrubbers are increasingly produced in various products, such as cosmetics, facial cleansers, and soaps [8]. It is necessary to make substitution efforts of plastic-based scrubbers with more environmentally friendly materials to avoid the negative impacts caused by the use of scrubbers. Scrubber made from
environmentally friendly materials is one of the green products innovations besides biodegradable films [9] that can save the environment.

Scrubber ingredients include olive seeds, apricot seeds, grape seeds, jojoba oil granules, and cellulose [7]. Cellulose is a potential ingredient because of its abundant quantity in nature [10]. Cellulose can be extracted from biomass such as wood [11], bamboo [12–17], cotton straw fibers [18], some species of algae [11], tapioca by-products (onggok) [19], sago pulp [20], and rice straw [21]. Cellulose sourced from biomass is bound to hemicellulose, lignin, and other impurities. It takes a lot of treatment (chemical and physical) to separate cellulose from other components. These various chemical and physical treatments can cause environmental pollution. To overcome this problem, bacterial cellulose (BC) can be used as a source of cellulose. Bacterial cellulose can produce biodegradable, high purity, high flexibility, high crystallinity cellulose and requires only simple treatment [22,23].

The manufacture of bacterial cellulose (BC) microparticles for various applications has been carried out through various methods. These methods include mechanical method using High-pressure homogenizer (HPH), hydrolysis with acids, microbial hydrolysis, cultivation of hydrogel fibers, microfluidic processes, and ultrasonication. Mechanical method using High-pressure homogenizer (HPH) produces blocking on homogenizer so the intact fibers can be formed [24]. Hydrolysis with acids can cause environmental pollution due to the use of strong acids [37]. Microbial hydrolysis produces only low yields [25]. Hydrogel fiber cultivation requires complex processes and a long time [26–28]. In comparison, microfluidic processes produce microparticles bacterial cellulose with hollow morphology. Therefore, it cannot be used as a scrubber [29]. The mechanical method by ultrasonication can produce microparticle bacterial cellulose which is more transparent than microparticle bacterial cellulose without ultrasonication [30]. Hence, ultrasonication methods are more suitable for making bio-scrubbers from bacterial cellulose.

The last stage in the manufacture of bio-scrubber from bacterial cellulose is the drying process. The drying process can be done by various methods. The spray-dried method produces microparticles bacterial cellulose with an amorphous shape [31]. Another method is ultrasonication with freeze-drying which produces microparticles bacterial cellulose with low mechanical properties and high porosity [32]. Therefore, this paper recommends the ambient pressure drying (APD) drying method. The microparticles bacterial cellulose produced through the ambient pressure drying (APD) method has higher crystallinity, better mechanical properties, and transparent [33].

To improve the performance of bio-scrubber bacterial cellulose (BC) is required the addition of antibacterial agents, such as alginate, chitosan [34], Chlorella sp. acid. [35], and flavonoids [27]. Flavonoids have significant potential as antibacterial because this compound is widely distributed in parts of plants, so it is easy to find [36]. One plant that has the potential as a source of flavonoids is moringa leaves. Moringa leaves have a fairly high flavonoid content of 5.53% [37]. This study uses flavonoids from moringa leaves as an antibacterial that will be interpreted into bio-scrubber bacterial cellulose. The final product in the form of bio-scrubber bacterial cellulose is expected to have environmentally friendly properties and antibacterial capabilities.

2. Materials and method

This review article is built upon reputable international journals on the Science Direct, SpringerLink, and Scopus databases. Journal searches are based on several relevant keywords. Keywords used in literature searches include scrubber, bacterial cellulose, flavonoid, ambient pressure drying, and moringa leaves. The literature obtained is 111 journals.

3. Result and discussion

3.1. Scrubber

In 1977 a chemical engineering engineer and professor of the Norwegian Institute of Technology (NIT) from Norway, John Ugelstad, invented a manufacturing method of monodisperse polymer particles without utilizing weightless space patented in 1979 [38–40]. This discovery is known by the particle name Ugelstad, but now we often know it by the name of Microbeads or scrubber [41]. Scrubber is a
round or amorphous particle measuring 0.5 μm to 500 μm commonly used in personal care and cosmetic products such as facial scrubs, soaps, toothpaste, and other products as exfoliating agents [42,43].

Scrubber is a small piece of plastic commonly used in personal care products because of its shape that can be used as a skin scrub. Scrubbers feature a large surface area per volume. This combined with uniformity of size and shape provides excellent accessibility and kinetics of fast liquid phase reactions, and fast and efficient binding. Scrubber is applied as a substitute for natural exfoliating agents such as almonds, oatmeal, apricot seeds, peanut skin, and pumice stones. Scrubber is usually made from polyethylene (PE), polymethylmethacrylate (PMMA), and polypropylene (PPE) which are cheaper to produce than previous natural exfoliating agents that make scrubbers rapidly develop and be applied by the industry in almost all cosmetic products today [44].

Scrubber has properties that are difficult to decompose. During application on the body, scrubber is designed to be instantly lost when rinsed with water. Its small shape makes scrubber often unfiltered in water treatment systems until it is eventually dumped into aquatic ecosystems and pollutes the environment. It was estimated that 8 trillion scrubbers had been dumped into the aquatic environment every day through wastewater treatment plants [6]. Scrubber may undergo physical modification after being incorporated into the water stream, and they can absorb or absorb other existing contaminants. They may float or settle in time depending on density and physical metamorphosis [45]. Scrubbers can float or sink according to their density and physique. This trait is dangerous because if the scrubber floats, it can contact with birds or aquatic species. While if it is drowned, it can contaminate the soil because it is difficult to decompose.

Due to the many negative impacts provided by scrubbers, there have been many moves made by both the government and non-government companies such as Unilever, The Body Shop, IKEA, Target Corporation, L’Oreal, Colgate/Palmolive, Procter &Gamble, and Johnson &Johnson in the form of campaigns and the realization of scrubber use tires in their products [6]. Other ingredients such as sugar, coffee, glycolic acid, etc., are used instead of plastic scrubbers in their products, as shown in table 1. However, many of these replacement ingredients have drawbacks such as availability issues, high prices, and potential hazards that are not much different from plastic scrubbers [46,47]. Some researchers have researched the utilization of various ingredients to overcome this problem, such as the manufacture of polyvinyl-alcohol scrubbers combined with calcium alginate, alginate, and calcium sulfate [48-50]. Nonetheless, it has not been perfectly decomposed. Moreover, other raw materials can be used to manufacture scrubbers, such as silica and cellulose [51,52]. Cellulose scrubbers also can be made from biomass such as wood, bagasse, cotton, rice straw, fruit, and bacterial cellulose (BC) [53-58].

BC has advantages among other cellulose in mechanical strength, crystallinity, nanofibril structure, purity, and biocompatibility [59]. BC has the potential to be used in biomedical [59], audio membrane [60], electronic paper [61], and biocomposites [22,62]. With this many potentials, it does not close the possibility of using BC as an environmentally friendly scrubber. BC scrubber can replace plastic scrubber that prevents environmental brightening of water and soil.

3.2. Production of Bacterial Cellulose Microparticles

Bacterial cellulose (BC) can be produced by Acetobacter xylinum [23], Acanthamoeba, Achromobacter, and Zoogloea [66]. Bacterial cellulose has many advantages, including good mechanical strength, high crystallinity, high purity, biocompatible, biodegradable, and it can form hydrogels with better material properties [67]. Bacterial cellulose has ultra-fine fibrils and it has superior properties compared to plant cellulose [68]. Based on these advantages, bacterial cellulose is widely used in various medical applications [59], membrane [60], film-coating agent [69], biofilm, composite material [22,70–73], and paper [61]. Bacterial cellulose can be used as a substitute for microplastic in scrubbers in the form of microparticles. Bacterial cellulose microparticles have been successfully produced through various methods, as shown in Table 3.
The synthesis of bi-scrubber from bacterial cellulose ends with the drying process of bacterial cellulose microparticles. The drying process can be divided into several types, namely the supercritical drying method [78] (high-temperature supercritical drying [79] and low-temperature supercritical drying [80]), spray-dried method [31], freezing drying method [81], and ambient pressure drying (APD) method [82].
Table 4 presents various drying methods that can be used to manufacture bio-scrubber from bacterial cellulose. Based on Table 4, the ambient pressure drying method has more advantages than other methods, so this paper recommends ambient pressure drying method to make bio-scrubber from bacterial cellulose.

Table 3. Bacterial cellulose microparticle manufacturing method.

| Method                              | Advantages                                                                                           | Drawbacks                                                                                     | Reference |
|-------------------------------------|------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------|-----------|
| Mechanical method with high-pressure homogenizer | High-pressure homogenizer (HPH) has high efficiency, simple, and free from organic solvents.        | The process runs very difficult and unstable because the narrow space in the homogenizer. The homogenizer tends to be easily blocked. The resulting material is not homogeneous and contains most of the fibers. | [74][75][24] |
| Hydrolysis with acid                | Produces nanoscale characteristics and exhibits good quality hydrophilic and lipophilic properties, and shows a significant effect on reduce surface tension. | Produce microparticles that have lower thermal stability than the initial cellulose, the use of acids can pollute the environment, cause corrosiveness, and can modify the surface of cellulose | [37][38][67] |
| Microbial hydrolysis                | No modification of the cellulose surface                                                              | The yield obtained is still very low at 22%.                                                   | [25]      |
| Cultivation of hydrogel fibers      | Produces bacterial cellulose microparticles that can reach 10 micrometers.                           | Producing bacterial cellulose microparticles is amorphous in shape and requires a fairly complex process. | [16]      |
| Microfluidic process                | Microparticle dimensions can be precisely controlled                                                | Produces bacterial cellulose microparticles with a hollow morphology                           | [29]      |
| Ultrasonication                     | • Do not use solvents that pollute the environment                                                   | There was a significant decrease in the thermal resistance of bacterial cellulose after the first 30 minutes of ultrasonication operation | [30]      |
## Table 4. Bacterial cellulose microparticle drying method.

| Method               | Advantages                                                                 | Drawbacks                                                                                   | Reference |
|----------------------|----------------------------------------------------------------------------|---------------------------------------------------------------------------------------------|-----------|
| Supercritical drying | Supercritical carbon dioxide (scCO₂) drying can avoid changes in the structure of bacterial cellulose | This method has constraints on the use of operating conditions that are quite extreme compared to the ambient pressure drying (APD) method | [83][84] |
| Freezing drying      | Compared to supercritical drying, freeze-drying is safer for the environment and less expensive | • Can reduce swelling by an average factor of 5, while evaporation drying can minimize swelling by an average factor of 50<br>• The aging process must be extended for stabilization and the existing bonds can be destroyed by solvent crystallization in the pores | [85–87] |
| Spray-dried          | This method produces microparticles with low rehydration capacity          | • This method produces bacterial cellulose microparticles with an amorphous shape<br>• Crystallinity index decreased | [31,88] |
| Ambient pressure drying | • Bacterial cellulose microparticles produced by ambient pressure drying have high crystallinity, high mechanical properties, and transparent<br>• The cheapest and easiest method | This method produces microparticles with low specific surface area (SSA) | [33,89] |

### 3.4. Flavonoid

Flavonoids are hydroxylated phenolic substances and are synthesized by plants in response to microbial infection. Flavonoids are a subdivision of polyphenolic compounds having a benzo-γ-pyrene structure, a versatile class of natural compounds that represent secondary metabolites [90]. Flavonoids are widespread in plants and classified into six scaffolds: flavone, flavonol, flavanone, flavanol, isoflavone, and anthocyanidin [91].

Flavonoids belong to the promising groups of bioactive compounds with strong antioxidant and anti-inflammatory properties [36]. In terms of benefits, flavonoids have antibacterial properties because they can release transduction energy to the bacterial cytoplasmic membranes and inhibit the mobility of bacteria [92]. They work by denaturing proteins so that the metabolic activity of bacterial cells stops [93]. Flavanoids have several biochemical and antioxidant effects associated with various diseases such as cancer, Alzheimer's disease (AD), atherosclerosis, etc. [94-96]. Flavonoids are also associated with a broad spectrum of health-promoting effects and are indispensable in various nutraceutical, pharmaceutical, medicinal, and cosmetic applications. This is caused by their properties such as anti-
oxidative, anti-inflammatory, anti-mutagenic, antimicrobial, anti-carcinogenic vascular activities, free radical scavenging abilities, and other medicinal properties coupled with their capacity to modulate essential cellular enzyme functions [97-99].

**Table 5.** Sources of flavonoids and their percentage content.

| Sources of flavonoids          | Flavonoid Content, % | Reference |
|-------------------------------|----------------------|-----------|
| Leaves of *Moringa oleifera*  | 5.530                | [37]      |
| *Hibiscus sabdariffa* from North Luwu | 0.207                | [100]     |
| *Hibiscus sabdariffa* from Kediri | 0.028                | [100]     |
| Leaves of *Terminalia catappa* | 0.293                | [101]     |
| Leaves of *Enhalus acoroides*  | 0.162                | [102]     |
| Leaves of *Camellia sinensis* (green) | 0.056                | [103]     |
| Leaves of *Camellia sinensis* (black) | 0.049                | [103]     |
| Skin of *Persea americana*    | 4.012                | [104]     |
| Leaves of *Ziziphus mauritiana* | 1.531                | [105]     |
| Root of *Eleutherine palmifolia* mesh 40 | 2.410                | [106]     |
| Leaves of *Dendrophthoe pentandra* | 2.480                | [107]     |
| Leaves of *Ruta graveolens*   | 1.670                | [108]     |
| Leaves of *Muntingia calabura* | 0.216                | [109]     |
| Leaves of *Etingerela elatior* | 5.450                | [110]     |
| Leaves of *Momordica charantia* | 2.887                | [111]     |

Based on table 5 above, moringa leaves are a potential source of flavonoids because the flavonoid content is more significant than other sources.

Bio-scrubber products that are applied to human skin also need to be added with antibacterial agents. Moringa leaves have a fairly high flavonoid content of 5.53% [37]. Flavonoids from Moringa leaves have been known to have good antibacterial properties [37]. The bio-scrubber made from bacterial cellulose impregnated with Moringa leaf flavonoids is expected to have superior properties, such as biodegradability, antibacterial ability, and environmental friendliness. Therefore, it can become an innovation as a green product.

4. **Conclusion**

Efforts to substitute plastic-based scrubbers with materials that are more environmentally friendly need to be made. Bacterial cellulose is a material that can be used as a bio-scrubber because of its superior properties. Various methods have been developed to produce bacterial cellulose microparticles. This paper recommends using the ultrasonication method followed by ambient pressure drying to produce bio-scrubber from bacterial cellulose. Based on the literature study, combining these two methods is expected to produce bio-scrubber products with high crystallinity, high mechanical properties, and transparency.

5. **Reference**

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