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Promising sheets of Bacterial cellulose/ Graphene Oxide for oil-water separation

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Abstract

One of the major issues with a sustainable environment is oil spills. Membrane technologies are increasingly being used to purify water and treat wastewater. A hydrophilic and extremely porous substance, bacterial cellulose (BC) has promise for oil-water separation from contaminants. In our investigation, *Komagataeibacter intermedius* MO generated BC. Based on graphene oxide (GO) and bacterial cellulose (BC), an oil-water separation sheet has been created. The modified Hummer method was used to prepare GO, which was then added to BC sheets. The prepared sheets have been investigated by FTIR, SEM, and XRD. Scanning electron microscope analysis showed that the BC surface appears as a homogenous surface with small and middle fibers, while these fibers disappeared due to the presence of oil on the surface of BC/GO sheet after oil separation. Results revealed that in just five seconds, the generated sheets demonstrated remarkable efficiency in separating water from oil. Therefore, this study offers a promising technique for the application of BC-GO membranes for water and oil separation.

Keywords: Bacterial cellulose; Graphene oxide; Characterization; Oil-water Separation.

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1. Introduction

Oil-water purification is one of the most significant challenges in water treatment. Oil industry pollution has long been a major environmental problem that has harmed soil, water, and air over the short, medium, and long terms. In addition, It causes economic difficulties, especially in areas like tourism, fishing, and coastal communities [1]. Oil in wastewater poses special difficulties for elimination and suitable care. Various kinds of oil might be found, and the specifics of the oil types involved have a significant impact on how well separation techniques work [2]. Because emulsions are stable water-in-oil or oil-in-water mixes that resist dissolution, they are particularly challenging to work with. Effective treatment is made much more difficult by the presence of emulsifying and antifoaming chemicals, which complicate the separation process [3].

A global commitment to switching to renewable energy sources, stringent laws, and developments in clean technologies can all help to lessen the negative effects of the oil business [4]. Indeed, cellulose is a biopolymer that is both abundant and adaptable, making it a perfect raw material for a variety of applications. It comes mostly from plant biomass, particularly wood, cotton, and other lignocellulosic materials, and is the most abundant carbohydrate polymer on Earth. For the development of sustainable alternatives in a variety of industries, its availability and renewability make it a desirable option [4].

Membrane technologies are increasingly being used to purify water and treat wastewater [5]. Bacterial cellulose (BC) membranes are becoming more and more popular as an environmentally friendly substitute for membranes made of synthetic polymers, especially in water treatment applications. These membranes are created by the metabolic processes of some bacteria, including *Komagataeibacter* or *Acetobacter*, which directly make cellulose from glucose or other carbohydrates **[6]**. Other microbes, particularly bacteria belonging to the genera *Sarcina, Komagataeibacter,* and *Agrobacterium*, ferment to generate BC. [7].

There are a number of benefits to using bacterial cellulose instead of conventional synthetic membranes. One important advantage is that BC membranes are environmentally benign and bio-based, which fits well with the expanding need for sustainable materials across a range of industries. In order to produce BC membranes, carbohydrates must be fermented, which is consistent with a bio-based economy. As a result, there is less dependence on fossil fuels because chemicals and materials are made from renewable carbon sources [8]. Additionally, the production of BC membranes can be achieved by employing agro-industrial wastes as feedstocks, which makes the process more sustainable and profitable. In recent reviews, this scalability has been highlighted [9], demonstrating the possibility of using cheap, plentiful organic materials as raw materials for the synthesis of BC. In addition to cutting waste, this process offers a more affordable option for producing membranes than more traditional techniques.

While plant cellulose is the primary source, BC is a novel form of natural polymer [10]. It is superior to plant cellulose in terms of its mechanical qualities and microstructure, including mechanical strength, degree of polymerization, water-absorbing capacity, crystallinity, and finer web-like network [11]. Moreover, BC membranes are good in treating water because of their high porosity, mechanical strength, and surface area. Because of these qualities, they are efficient at removing impurities, which may make wastewater treatment and other filtration procedures more environmentally friendly. BC membranes may play a crucial role in the creation of bio-based, more sustainable filtering systems because of these characteristics.

Effluent treatment research has concentrated on resolving the shortcomings of the industry's present approaches to eliminating pollutants, to streamline procedures, and/or utilizing cutting-edge, environmentally friendly materials. These developments have made it possible to employ biotechnological materials as filtering membranes or to agglutinate oily molecules A flexible biomaterial that can be used in these treatments is bacterial cellulose (BC).

According to [14, 15], numerous industries have used BC bio membranes and biofilms, which offer improved performance and replace conventional synthetic materials. For high-quality filtration, BC membranes are perfect because of their high porosity and network structure, which includes tiny pores (less than 100 nm) [12, 13].

Additionally, compared to conventional techniques, bacterial cellulose membranes offer a number of benefits for filtering oily wastewater. They efficiently capture oil while permitting water to flow through due to their high hydrophilia and oleophobia, which produces a high degree of separation efficiency.

Because BC membranes may be made in a variety of shapes, including fibers, tubes, and flat sheets, they provide more design freedom for filtration system configurations. Furthermore, BC membranes can have their characteristics customized to meet certain filtration requirements. These membranes, which offer greater efficiency, selectivity, and sustainability than traditional techniques, have demonstrated encouraging results in the filtering of oily wastewater [14]**.** Our goal in this study is to prepare and purify BC membranes. Then, graphene oxide is added to BC sheets. Following their characterization, the generated BC-GO sheets were employed as a water decontamination filter.

2. Experimental

2.1. Materials

Graphite (G) powder (99.9%) was provided by Fisher Scientific UK. Potassium permanganate (> 99%) and hydrogen peroxide (30%) were bought from Bio Basic Canada Inc. and Carl Roth GmbH.Analytical-grade chemicals and reagents were utilized without additional purification.

2.2. Methods

Graphical abstract of BC-GO sheet under study was showed in Fig. 1. As, graphene oxide is added to BC sheets. Following their characterization, the generated BC-GO sheets were employed as a water decontamination filter.

2.2.1. Collection of bacterial sample

Komagataeibacter intermedius MO was obtained kindly from the Faculty of Science, Benha University, Egypt.

2.2.2. Preparation of bacterial cellulose

Distinct purified colonies of *Komagataeibacter intermedius* MO were inoculated individually in sterilized modified seed medium: Peptone 15 g/L, cheese whey 15 ml, Yeast extract 15 g/L, ethanol 40 ml/L, glycerol 15 ml/L, & pH 7 [15] subsequently incubation for 7 days at 30˚C in static conditions.

2.2.3. Purification of BC sheets

The pellicles that developed at the production medium's air-liquid contact were gathered and cleaned three times using water. After that pellicles were treated with 1 N NaOH for 20 minutes at 80°C. To neutralize the NaOH, the sheets were treated with a 5% acetic acid solution. Three more water washes were performed on them. After being cleaned, the produced pellicles were dried at 60°C until their weight stayed constant, which was expressed as g/L dry BC weight **[16].**

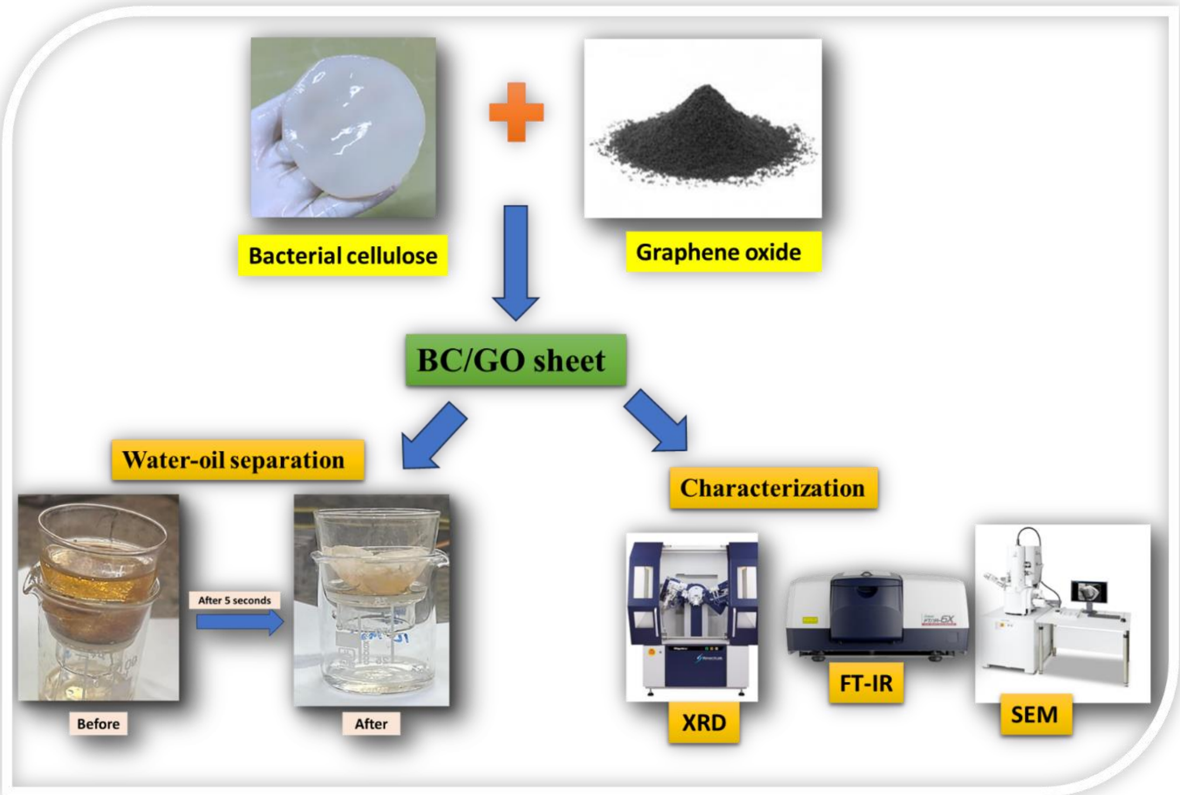

Fig. 1. Graphical abstract of BC-GO sheet under study.

2.2.4. Graphene oxide (GO) preparation

GO was created by applying the modified Hammar technique. In this, three milliliters of phosphoric acid (H_3PO_4) and 27 milliliters of sulfuric acid (H_2SO_4) were mixed and stirred (9:1). Then, 0.225 g of graphite powder was added. A 1.32 g addition of potassium permanganate ($KMnO_4$) was made gradually to the mixture. The mixture was allowed to agitate until the solution turned dark green within six hours. The excess $KMnO_4$ was eliminated by slowly adding 0.675 mL of hydrogen peroxide and swirling for ten minutes. Next, 10 mL of hydrochloric acid and 30 mL of deionized water were added. The supernatant was washed three times with water after it was decanted. The black GO product was dried in an oven set to 90°C for 24 hours [17].

2.2.5. Preparation of BC/GO sheet

0.1 gram of GO was dispersed in 10 mL of distilled water to create a homogeneous solution, which was then sonicated for five minutes. After that, GO was precipitated on the BC sheet by adding it while stirring constantly for 24 hours. At room temperature, the BC/GO sheets had dried.

2.2.6. Characterizations

- FT-IR spectra were obtained within the range of 400–4000 cm^{-1} using a Shimadzu 8400S FT-IR Spectrophotometer.
- The surface morphology was examined utilizing an FEI IN SPECTS Company SEM electron microscope from Philips, Poland, employing environmental scanning without coating, alongside a JEOL JEM-2100 electron microscope at a magnification of 100,000x and an acceleration voltage of 120 kV.
- The XRD patterns were analyzed using a Diano X-ray diffractometer with a CuKα radiation source operating at 45 kV and a Philips X-ray diffractometer (PW 1930 generator, PW 1820 goniometer) with a CuK radiation source (λ=0.15418 nm), over a diffraction angle range of 2θ from 10 to 80° in reflection mode.

Crystalinity has calculated from the equation

$$Cr_I = \frac{(I_{002} - I_{am})}{I_{002}} X\ 100$$

Where I_{002} and I_{am} are the maximum intensity of the 002 lattice diffraction and lam is the intensity of diffraction in the same units at 28 = 18°.

2.2.7. BC/GO sheet for oil water separation

Oil-water separation was achieved using a BC/GO sheet as a filter. The 50:50 oil (food oil) and water mixture was prepared and then poured into a glass funnel 5 cm dimension with a BC/GO sheet to separate the mixture.

3. Results and discussion

3.1. preparation and purification of bacterial cellulose

The modified seed medium, which produced 17.8 g/L of dry weight using isolated *Komagataeibacter intermedius* MO, was an appropriate growth medium for the production yield of BC. The value of cheese whey in BC production will have an impact on both environmental regulations and production costs. Environmental contamination resulted from the dairy industry's by-products and their disposal [18].

3.2. Preparation of GO

Many graphite layers were transformed into a single layer throughout the GO preparation process, adding new groups like C=O, COOH, and OH to the GO matrix. According to **Figure 2**, XRD has been employed to study how graphite forms GO, which has a distinctive peak at 2θ = 26, while GO has a characteristic peak at 2θ = 10 due to new group formation which confirms the successful oxidation of graphite [19].

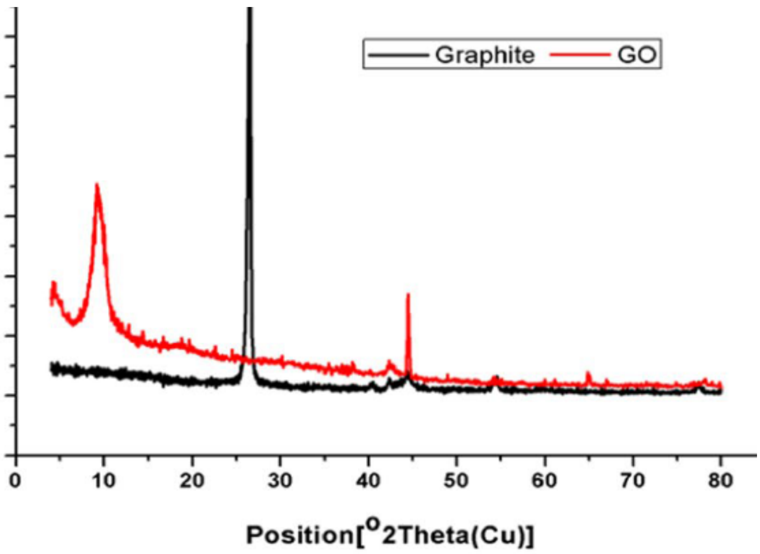

Fig. 2. XRD of GO and Graphite.

3.3. Characterization

3.3.1. FTIR – analysis

One such method for determining the functional groups in the materials being studied is FTIR. **Figure 3** shows the FTIR analysis of BC. The broad peak between 3000 and 3500 cm^{-1} frequencies is indicative of OH stretching, whereas CH stretching is represented by the peak at 2900 cm^{-1} The peaks at 1650 cm^{-1} and 1500cm^{-1} due to absorbed water and C=O, and ether linkage C-O-C at 1100 cm^{-1}. After GO incorporation, many peaks disappeared and a new peak was caused by the C=O of cellulose, and GO emerged at 1700 cm^{-1} **[20, 21].** Due to presence of GO on the surface of the sheet and GO/BC interaction the others peaks slightly appear

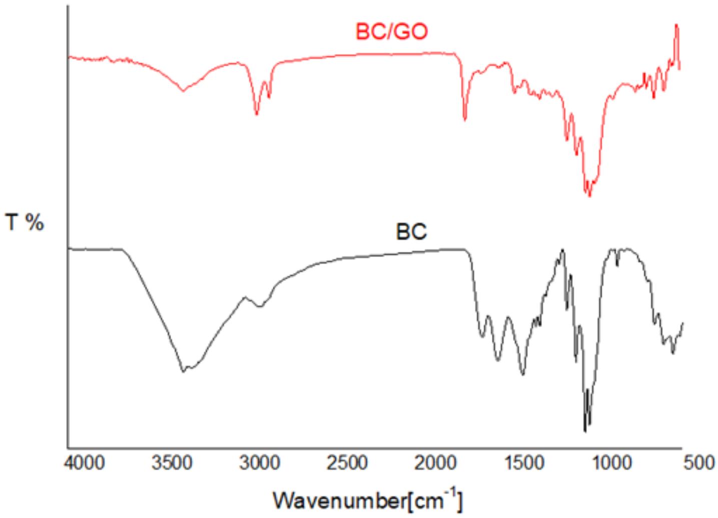

Fig. 3. FTIR of BC and BC/GO sheet.

3.3.2. Scanning electron microscope (SEM)

Figure 4 shows the surface morphology of BC sheet before and after oil separation. The BC surface appears as a homogenous surface with small and middle fibers, while these fibers disappeared due to the presence of oil on the surface of BC/GO sheet after oil separation. The oil appears as a lighting point on the surface.

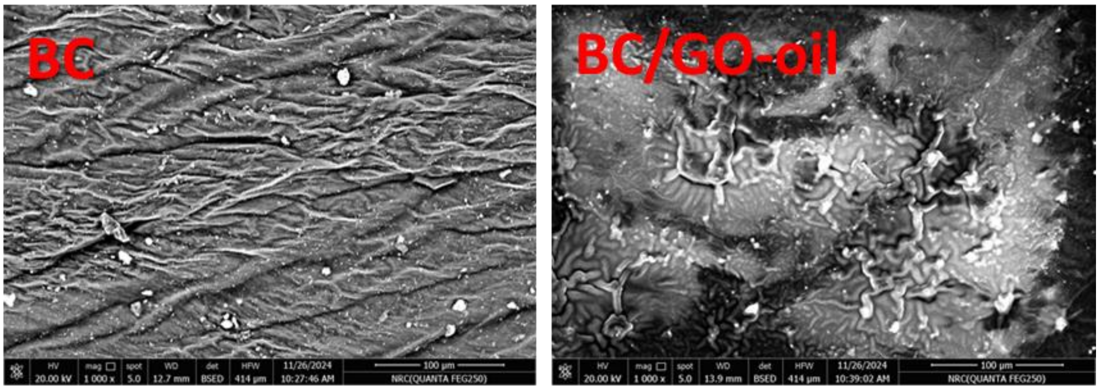

Fig. 4. SEM of BC and BC/GO-oil sheet

3.3.3. XRD- analysis

Figure 5 depicts the BC/GO XRD pattern. The BC pattern displays two cellulose peaks at $2\theta = 15$ and a peak at $2\theta = 22$ °. These peaks appear slightly in BC/GO sheet due to GO incorporation, as well as the characteristic peak of GO at $2\theta = 10°$ appears clearly [22]. The crystalinity has increased after GO added to became 80% after 35% due to GO coating of BC surface.

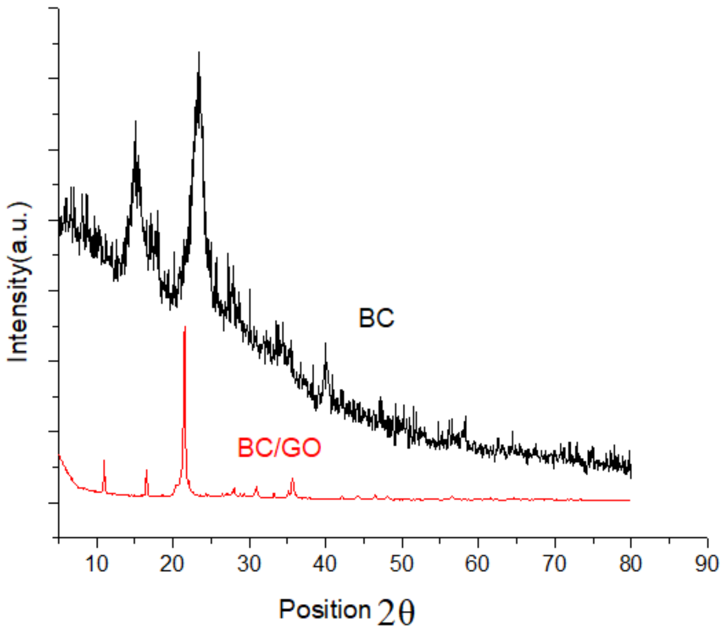

Fig. 5. XRD of BC and BC/GO sheet

3.4. BC/GO sheet for oil water separation

The prepared sheet represented good ability to remove oil from water. The food oil has been used in the separation process. Although this oil is very light, it was separated easily from water. As in **Figure 6** BC/GO sheet separated oil from water throughout five seconds with 90% of oil has removed and the sheet keeps its durability and reusability during the process. The comparison of oil absorption by different materials was shown in Table 1.

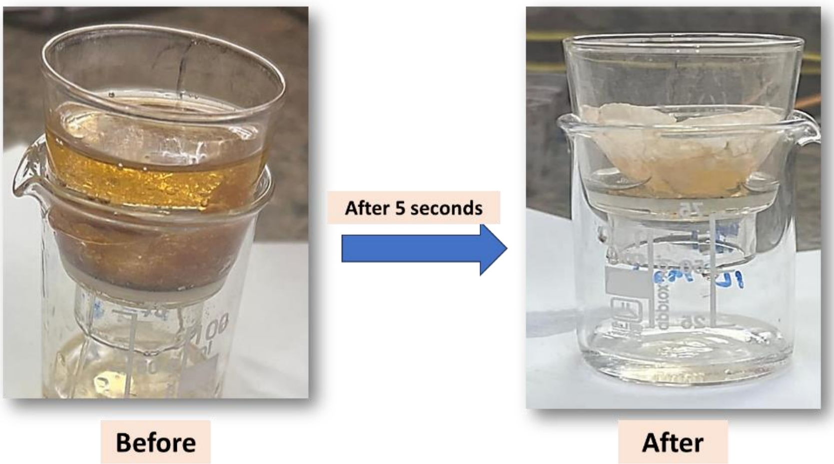

Fig. 6. Water-oil separation before and after addition of BC/GO sheet.

Table 1. Comparison of oil absorption by different materials.

Sorbents	Absorption capacity (g g−1)	References
Cotton grass	(19 g/g)	Suni et al. [23]
Carbonized bagasse	(23.86 g/g)	Hussein et al. [24]
DCC/GO foam	26 g/g	Dacrory [25]
BC/GO	90%	Present study

4. Conclusion

The exceptional efficiency, variable pore structure, chemical stability, and environmental friendliness of BC-GO sheets in oil-water separation make them a practical and sustainable substitute. GO has been successfully prepared by modified Hummer method and then incorporated into the BC sheet. Oil-water separation sheet has been prepared based on bacterial cellulose (BC) and graphene oxide GO. The prepared sheet has been examined by FTIR, SEM, and XRD, and the ability of the prepared sheet to separate oil from water has been recorded by applying the sheet in the separation process. Thus, environmentally friendly filtering membranes can benefit from BC's high wet strength, nanoporous structure, and capacity to eliminate micro-sized contaminants. Future studies should focus on the difference between the filtration of a stabilized oily emulsion and a non-emulsified solution. The additional characterization of BC membranes will support the use of this biomaterial as an industrial biotechnological filter.

Declaration of competing interests

The authors state that none of the work described in this article could have been influenced by any known competing financial interests or personal relationships.

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