Water Research 222 (2022) 118952

Contents lists available at ScienceDirect

Water Research

journal homepage: www.elsevier.com/locate/watres

Bacterial cellulose biopolymers: The sustainable solution to water-polluting microplastics

Marisa Faria [a,b,1], César Cunha [a,1], Madalena Gomes [a], Ivana Mendonça [a], Manfred Kaufmann [b,c], Artur Ferreira [d], Nereida Cordeiro [a,b,*]

[a] LB3-Faculty of Science and Engineering, University of Madeira, Portugal
[b] CIIMAR-Interdisciplinary Centre of Marine and Environmental Research, University of Porto, Portugal
[c] Marine Biology Station of Funchal, Faculty of Life Sciences, University of Madeira, Portugal
[d] CICECO-Aveiro Institute of Materials and Águeda School of Technology and Management, University of Aveiro, Portugal

ARTICLE INFO

Keywords:
Bacterial cellulose
Biopolymers
Microplastics
Environmental biotechnology
Sustainability

ABSTRACT

Microplastics (MPs) pollution has become one of our time's most consequential issue. These micropolymeric particles are ubiquitously distributed across all natural and urban ecosystems. Current filtration systems in wastewater treatment plants (WWTPs) rely on non-biodegradable fossil-based polymeric filters whose maintenance procedures are environmentally damaging and unsustainable. Following the need to develop sustainable filtration frameworks for MPs water removal, years of R&D lead to the conception of bacterial cellulose (BC) biopolymers. These bacterial-based naturally secreted polymers display unique features for biotechnological applications, such as straightforward production, large surface areas, nanoporous structures, biodegradability, and utilitarian circularity. Diligently, techniques such as flow cytometry, scanning electron microscopy and fluorescence microscopy were used to evaluate the feasibility and characterise the removal dynamics of highly concentrated MPs-polluted water by BC biopolymers. Results show that BC biopolymers display removal efficiencies of MPs of up to 99%, maintaining high performance for several continuous cycles. The polymer's characterisation showed that MPs were both adsorbed and incorporated in the 3D nanofibrillar network. The use of more economically- and logistics-favourable dried BC biopolymers preserves their physicochemical properties while maintaining high efficiency (93–96%). These polymers exhibited exceptional structural preservation, conserving a high water uptake capacity which drives microparticle retention. In sum, this study provides clear evidence that BC biopolymers are high performing, multifaceted and genuinely sustainable/circular alternatives to synthetic water treatment MPs-removal technologies.

1. Introduction

Plastic is everywhere, given its high durability, stability, economic viability and applicability in several economic sectors, predominantly in industrial, constructional, electronic, medical, and retail (PlasticsEurope, 2018). Despite concerns, plastic production has been exponentially increasing to meet the escalating demand of societal consumerism. It has been estimated that 10% of all human-generated waste is plastic (UN environment, 2018). Despite estimates that the total amount of plastic ever produced exceeds 8000 million metric tons (Mt) (Geyer et al., 2017), global plastic production is projected to double in the next 10–15 years (UN environment, 2018). As plastic production incessantly increases, plastic pollution has become a major concern since these polymers tend to accumulate in the environment. In fact, from the 6300 Mt of plastic waste generated as of 2015, roughly 79% (4977 Mt) was accumulated in the natural environment (Geyer et al., 2017), with approximately 13 Mt of plastic being leaked to aquatic environments in 2018 alone, through indiscriminate disposal (UN environment, 2018). At the current rate, the environment is estimated to become home to roughly 12,000 Mt of plastic waste by 2050 (Geyer et al., 2017), with plastic debris already comprising 80% of the total waste found in aquatic ecosystems (Venghaus and Barjenbruch, 2017). Alongside several other critical environmental concerns, plastic waste is profiling itself as one of the biggest generational challenges of our time. Accordingly, recent

* Corresponding author at: LB3-Faculty of Science and Engineering, University of Madeira, Portugal.
E-mail address: ncordeiro@staff.uma.pt (N. Cordeiro).
[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.watres.2022.118952
Received 16 May 2022; Received in revised form 2 August 2022; Accepted 4 August 2022
Available online 5 August 2022

years have sparked another concern within the scientific community: microplastics (MPs) pollution. MPs are defined by the National Oceanic and Atmospheric Administration (NOAA) as small-scale plastic debris, of all shapes and sizes, with a diameter inferior to 5 mm (NOAA, 2020). MPs are divided by their origin into two major categories: primary and secondary. Primary MPs are intentionally manufactured as small microbeads, microspheres, and synthetic microparticles aimed at cosmetic, medical, and general applications (Galloway et al., 2017). On the other hand, secondary MPs are smaller-sized low molecular-weight polymer fragments derived from macroplastics undergoing biological, chemical, and mechanical degradation substantially driven by UV-induced photooxidation (Gewert et al., 2015).

Consequently, sites like wastewater treatment plants (WWTPs) have converged a lot of notice in recent times. Central to urban water distribution, these infrastructures have been documented to be massive focal points of MP release (Ben-David et al., 2021; Edo et al., 2020; Gatidou et al., 2019; Horton et al., 2021; Kay et al., 2018; Li et al., 2018; Mintenig et al., 2017; Ngo et al., 2019; Ou and Zeng, 2018; Rolsky et al., 2020; Sun et al., 2019; Turan et al., 2021; Xu et al., 2019; Ziajahromi et al., 2017). Previous studies have reported that, despite the use of both filtration and mechano-biochemical removal processes, an average of 2 million MP particles with less of 300 μm are discharged to urban networks *per* day (Verschoor et al., 2014; Mintenig et al., 2017; Sun et al., 2019). Consequently, MPs have been documented in both raw and treated drinking water (Koelmans et al., 2019; Na et al., 2021; Pivokonsky et al., 2018), with polystyrene being one of the most commonly used and hazardous plastic polymers detected in WWTPs (Sun et al., 2019) and in the environment (Alimi et al., 2018; Sharma et al., 2021).

Aside from the widely documented deleterious effects of MPs in aquatic and terrestrial biota (Hale et al., 2020; Rezania et al., 2018; Sun et al., 2021), a growing body of evidence has strongly hinted at the direct implication of MPs in human health (Campanale et al., 2020; Carbery et al., 2018; Chen et al., 2020; Cox et al., 2019; Gupta et al., 2019; Jin et al., 2019; Lehner et al., 2019; Li et al., 2020; Lu et al., 2019, 2018; Prata, 2018; Prata et al., 2020; Schwabl, 2020; Smith et al., 2018). Accordingly, the first direct evidence of MPs in the human placenta of unborn babies has surfaced (Ragusa et al., 2021). These findings were followed by the discovery of MPs in the human blood, sending shockwaves throughout the world. The fact that MPs are also vectors for the transport of hazardous chemicals and organic pollutants (Wang et al., 2020) but still represent emerging contaminants with unknown health significance raises extreme concern regarding their potential health risks to permanently exposed humans (Dick Vethaak and Legler, 2021; Gruber et al., 2022). It is therefore imperative to find sustainable and environmental-fit solutions to tackle this issue.

Most studies on the removal of MPs from waters focus on technologies such as coagulation, ultrafiltration membranes, dynamic membranes, or more advanced technological approaches such as membrane bioreactors (MBRs) (Poerio et al., 2019). Unfortunately, techniques such as coagulation and ultrafiltration have demonstrated poor removal efficiencies (Ma et al., 2019). More recently, approaches such as surface-functionalised microbubbles have shown promise in removing MPs but present considerable applicability and sustainability hurdles (Zhang et al., 2021). Poerio et al. have argued that the water and wastewater treatment industries still lack experience and technological expertise to efficiently separate MPs from effluents (Poerio et al., 2019). Despite MBRs representing the current most promising technology, far outperforming conventional hazardous WWTP-applied MP removal methods, its membranes still represent a focal point of concern (Poerio et al., 2019). These synthetic and organic membranes are non-biodegradable fossil-based polymers whose fouling and high costs represent major obstacles for the broader adoption of MBRs (Meng et al., 2009). Aside, both *in-situ* and *ex-situ* fouling removal mechanisms are economically, energetically, and environmentally unsustainable (Wang et al., 2014). Recent data suggest that membrane filtration systems might also be acting as sources of nano- and microplastics, given their polymeric composition. Alongside problems raised by the wear stress, aging and harsh cleaning cycles throughout the membrane's life cycle, the authors emphasise the need to accelerate the development of biomembranes as solutions to this problem (Ding et al., 2021).

Appropriately, bacterial cellulose (BC) is a polysaccharide produced by specific bacteria genera such as *Acetobacter* spp., *Agrobacterium* spp., *Azotobacter, Rhizobium* spp., *Sarcina, Alcaligenes*, and *Pseudomonas* (Lahiri et al., 2021). More recently, the genus *Komagataeibacter* has garnered considerable interest regarding BC production (Chen et al., 2018; He et al., 2020; Raiszadeh-Jahromi et al., 2020; Singhsa et al., 2018). These BC-producing bacteria have been shown to produce characteristically similar BC using alternative and sustainable carbon sources (Vazquez et al., 2013), highlighting the fact that production costs can be immensely reduced in industrial setups. This exopolymer has been attracting a lot of scientific and industrial interest given its wide range of morphologies, physicochemical properties, and applications (Wang et al., 2019). Structurally, BC consists of hydrogen-bond $(C_6H_{10}O_5)_n$ β-1,4-glucan chains (Ul-Islam et al., 2012). Further, the process of BC synthesis is centred around the exocytosis of glucose chains through small pores in the bacterial cell wall. These glucose chains form nano- and microfibrils that aggregate to form a porous three-dimensional network (Esa et al., 2014; Shah et al., 2013), resulting in the formation of hydrogel matrixes with large surface area and high porosity (Esa et al., 2014). When compared to the cellulose produced by other organisms, its high purity, peculiar physical properties, biocompatibility, biodegradability, and renewable character makes BC extremely attractive for industrial use (Wang et al., 2019), particularly in the food, biomedical, pharmaceutical, cosmetic, and electronic industries (Portela et al., 2019; Shah et al., 2013). Nevertheless, it was the absorbent nature of BC that has recently attracted the interest of specialists in the wastewater treatment field regarding its potential use in the removal of contaminants such as dyes and heavy metals (Isik et al., 2018; Kurniawan and Yamamoto, 2013; Mohite and Patil, 2014; Wanichapichart et al., 2002).

Given the theoretical physicochemical properties of BC regarding organic micropolymeric particles such as MPs, these biopolymers were investigated as potential solutions to MPs pollution. Accordingly, this laboratory-scale work aims to assess the viability of previously unexplored BC biopolymers as removal matrixes of MPs from contaminated waters. To the best of our knowledge, this is the first-time bacterial cellulose biopolymers have been explored concerning its remediation potential of microplastics. The quantification of several differently-treated BC biopolymers regarding removal efficiency and MPs retention was determined using flow cytometry. To characterise the physicochemical properties of the biopolymer and understand the mechanisms driving the retention of MPs, techniques such as infrared spectroscopy (ATR-FTIR), x-ray diffraction (XRD), inverse gas chromatography (IGC), atomic force microscopy (AFM), fluorescence microscopy and scanning electron microscopy (SEM) were used.

2. Materials and methods

2.1. BC production

BC biopolymers were produced by growing *Komagataeibacter saccharivorans* in static conditions, in Hestrin and Schramm (HS) liquid medium [2% (w/v) glucose (G7021/Sigma-Aldrich), 0.5% (w/v) peptone (84616.0500/VWR Chemicals), 0.5% (w/v) yeast extract (84601.0500/VWR Chemicals), 0.27% (w/v) disodium hydrogen phosphate (02494C/VWR Chemicals), 0.115% (w/v) citric acid (0529–500 G/VWR Chemicals), 0.4% (w/v) ethanol, pH 3.25] at 30 °C and with an initial OD_{600} of 0.4 (UV-6300 PC Double Beam). HS medium was sterilized at 121 °C for 15 min. After incubation, to remove adsorbed cells and culture medium components retained, the BC biopolymers were treated with 0.5 M NaOH (80 °C for 45 min.) and washed with distilled water until neutral pH. BC biopolymers exhibited a wet weight of 22.715

± 1.519 g and a dry weight of 0.055 ± 0.003 g (99.76% of water). BC membrane thickness was measured with a calliper rule (Electronic Digital Calliper), and the dry weight was determined through a moisture balance (Gibertini, Eurotherm) at 105 °C for 45 min. The BC biopolymers were characterized by scanning electron microscopy (SEM), fluorescence microscopy, atomic force microscopy (AFM), inverse gas chromatography (IGC), x-ray diffraction (XRD) and infrared spectroscopy (ATR-FTIR). The used experimental methodology was described in Supplementary Material S1.

2.2. BC biopolymer treatment

BC biopolymers were treated in this study in five different forms, to evaluate its effect on filtration efficiency: *(i)* wet biopolymer (W-BC); *(ii)* oven-dried (40 ± 2 °C) biopolymer (OD-BC); *(iii)* freeze-dried (Savant RT 400 Refrigerated Condensation Trap) biopolymer (FD-BC); *(iv)* filtered (80% of water content removed under vacuum filtration) biopolymer (F-BC); and *(v)* drained (80% of water content removed under compression) biopolymer (D-BC).

A porous plate glass crucible was used to support BC biopolymers in the filtration system. During the filtration procedure, the MPs-contaminated water was inserted into the filtration system and passed to a pressure of 7 mbar (Vacuubrand MZ 2C). An aliquot of the filtered solution was collected for flow cytometry analysis.

2.3. Microplastics specimens

Spherical polystyrene microplastics (PS-MPs) with a diameter of 10 μm were purchased from Thermo Scientific™ (G1000) as a 1% (w/v) suspension with excitation and emission wavelengths of 468 and 508 nm, respectively. A high concentration MPs stock solution was prepared at 10 mg L^{-1} and stored at 4 °C until further use. All solutions were prepared in glass flasks to minimize the establishment of electrostatic interactions with their walls. Also, solutions were vortexed before use to guarantee homogeneity.

2.4. Microplastics removal

The MPs in the MPs-contaminated water and the not retained MPs in the BC biopolymer were quantified using flow cytometry (CytoFLEX, Beckman Coulter) with a blue laser (excitation radiation of 488 nm), based on recent methodologies (Kaile et al., 2020; Tse et al., 2022). The PS-MPs were characterized according to "forward scatter" (FSC), "side scatter" (SSC) and "fluorescein isothiocyanate" (FTIC; 525/550 nm emission detection filter). The CytExpert software (Beckman Coulter) was used to analyze the output data. The calibration line was obtained using 0, 2.5, 5, 10, 50 and 100 mg L^{-1} of MPs and applied to calculate the concentration of MPs in pre- and post-filtrated water samples.

2.5. Biopolymer efficiency and flux analysis

Biopolymer efficiency and continued water absolute and relative fluxes of BC biopolymer were evaluated. The biopolymer efficiency assessment was carried out as follows: 20 cycles (filtration number) of 25 mL of MPs-contaminated water were passed through the BC biopolymers. Filtered solution aliquots were collected for flow cytometry analysis every two filtration cycles. The efficiency was calculated following the Eq. (1), were n_i is the initial number of MPs and n_f is the number of MPs after filtration:

$$Efficiency = \frac{n_i - n_f}{n_i} \times 100 \quad (1)$$

The absolute and relative water fluxes were calculated according to Kim et al., with some adjustments, and following Eqs. (2) and (3), where t_f is the filtration time of each cycle, V_f is the throughput volume of the MPs-contaminated water solution, J is the flux of each cycle and J_0 is the initial flux (cycle 1) of the solution (Kim et al., 2017).

$$Flux\ (ml/\min) = \frac{t_f}{V_f} \quad (2)$$

$$Relative\ flux = \frac{J}{J_0} \quad (3)$$

2.6. Biopolymer resistance

The resistance of the polymer to successive filtration cycles (50) was calculated (Eq. (4)) based on the filtration time of MPs-contaminated water by the BC biopolymer, where t_f is the initial filtration time and t_i is the filtration time of each cycle.

$$Resistance = \frac{(t_f * 100)}{t_i} - 100 \quad (4)$$

2.7. Microscopy characterisation

2.7.1. Scanning electron microscopy

Dry BC biopolymers (OD-BC and FD-BC) were coated with a thin layer of carbon using EMITECH K950X Turbo Evaporator and deposited on a steel plate. SEM micrographs were obtained using the HR-FESEM SU-70 Hitachi Scanning Electron Microscopy equipment, operating with a 5 kV beam, at a 15.6 mm working distance, in the field emission mode. Images were collected at several magnifications, from x500 to x30 000.

2.7.2. Fluorescence microscopy

The retention/incorporation of MPs in the BC network was observed using epifluorescence microscopy on a Leica DM2700 P device coupled with a Leica DFC450 C digital camera and a CoolLED pE-300 lite lighting system. The fluorescent signal was captured using the I3 450–490 nm/515–565 nm excitation/emission filter. BC biopolymers were stained with Calcofluor White (18909 - Sigma-Aldrich) for enhanced observation.

2.8. Data and statistical analysis

Data representation and statistics were carried out using GraphPad Prism 8. The D'Agostino-Pearson omnibus and Kolmogorov-Smirnov normality tests were used to assess the gaussian distribution of data. Parametric unpaired t-tests (or one-way ANOVA) were applied for normally distributed data, while non-parametric unpaired Mann-Whitney (or Kruskal-Wallis) tests were applied for non-gaussian distributed data. Statistical analysis was performed in at least three independent experiments.

3. Results and discussion

In recent years, severe environmental and health-related concerns have been raised regarding an "invisible" but highly abundant, ubiquitous, and still poorly characterized pollutant: microplastics (MPs). Despite technological advances in MPs removal in locations such as wastewater treatment plants (WWTPs), synthetic membrane filtration systems still represent the major point of contention and inviability. Therefore, it is critical to develop solutions that align with biosustainability and circularity expectations. Here, several types of semi- or fully-dried bacterial cellulose (BC) biopolymers produced by *Komagataeibacter saccharivorans* were studied to not only to understand its potential as a raw MPs-removing biopolymer but also in forms that would enable and facilitate industrial adoption and scaling. These include wet BC (W-BC) biopolymers, oven-dried BC (OD-BC) biopolymers, freeze-dried BC (FD-BC) biopolymers, filtered BC (F-BC)

biopolymers and drained BC (D-BC) biopolymers (Section 2.2. for details). It is important to note that technique applicability is always selectively dependent on the wet or dried nature of the BC biopolymer, highlighting that some techniques cannot be applied to wet forms of the biopolymer.

3.1. BC biopolymer production and characterization

3.1.1. BC biopolymer production

Important for understanding its interactions with microplastics (MPs) and industrial applicability/scalability, assessing a wide range of physicochemical properties regarding bacterial cellulose (BC) biopolymers is crucial. Accordingly, the optimal culturing conditions of BC biopolymers produced by *Komagataeibacter saccharivorans* were evaluated. It was concluded that 2% glucose, pH of 3.25 and 7 days of cultivation yielded a maximised time-efficient concentration of roughly 2.95 g L^{-1} (Fig. S1), in line with the literature (Revin et al., 2018; Volova et al., 2018). The raw wet BC (W-BC) biopolymers exhibited an averaged thickness of 11.583 ± 0.685 mm, where 99.76% of the biopolymer's net weight was water. Given these values, and considering the potential industrial applicability of these biopolymers, logistics-facilitating drying processes were extensively characterised.

3.1.2. BC characterization

Attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR) analysis (Fig. S2) showed that structurally, both oven-drying (OD-) and freeze-dried (FD-) bacterial cellulose (BC) biopolymers displayed the expected absorption bands of cellulose around 3347 cm^{-1} (O—H stretching vibrations); 2899, 1434, 1373 and 1312 cm^{-1} (C—H and C—H_2 stretching vibrations); 1632 and 666 cm^{-1} (C—H or O—H bending vibration), and 1064 cm^{-1} (C—O stretching vibration) of ether groups, which is often found within the glucopyranose ring and between the glucose monomers (Alonso et al., 2018; Fan et al., 2012; Faria et al., 2019). The sharper peaks observed on the ATR-FTIR spectra, corresponding to FD-BC biopolymers, suggest a more regular structure preserved during the freeze-drying process.

Both dried BC biopolymers (OD-BC and FD-BC) showed similar x-ray diffraction profiles (Fig. S3) of cellulose type I, with characteristic peaks found at 14.5, 16.73 and 22.7°, corresponding to the (100), (010) and (110) planes, respectively. Despite both exhibiting similar crystallinity levels, FD-BC biopolymers exhibited considerably larger surface areas (Table 1) determined by Inverse Gas Chromatography (IGC). IGC showed linear adsorption isotherms for both the BC biopolymers, corresponding to a type I isotherm typically associated with microporous materials. This is strengthened by a high degree of permeability values (76.78 *vs* 58.10 cm^2 min^{-1}), which indicates that the FD-BC biopolymers exhibit a higher number of smaller, structurally conserved pores. Consistently, the 78–79% crystallinity values registered here are roughly in line with the ones reported in the literature (lowest to highest): 63.2% (Ul-Islam et al., 2012), 75.2% (Phisalaphong and Jatupaiboon, 2008), 86.9% (Andritsou et al., 2018), 88.0% (Tsouko et al., 2015), and 88.4% (Jia et al., 2017), while OD-BC biopolymers' S_{BET} was considerably higher than the values reported in the literature (Castro et al., 2015; Faria et al., 2019; Mohammadkazemi et al., 2017).

Moreover, atomic force microscopy (AFM) was used to characterize the surface topography of the BC biopolymers. This ultra-high-resolution technique highlighted that OD-BC biopolymers exhibit a rougher surface than FD-BC biopolymers (Fig. S4; Table 1). Also, the hypothesis that FD-BC biopolymers would present greater preservation of their structural network is confirmed. A more substantial pore collapse during oven-drying, compared to freeze-drying, is observed. These morphological characteristics underscore that the drying process plays a critical role in modulating structural integrity.

Furthermore, considering that the BC biopolymers are immersed in water during the filtration process, studying the water uptake capacity (swelling) becomes critical to understanding their behavior during remediation cycles. OD-BC and FD-BC biopolymers manifested considerably different swelling behaviors: FD-BC biopolymers displayed an inflated swelling performance from 659 to 1011% compared to OD-BC biopolymers, taking 3 h to saturate, while their oven-dried counterparts took 8 h to achieve statistically constant saturation (Fig. S5). This greatly enhanced water uptake ability is likely observed due to the structural conservation of pores during freeze-drying, unlike their collapse during oven-drying, effectively corroborating previous data.

Table 1
Bacterial cellulose (BC) properties obtained from oven-dried (OD–BC) and freeze-dried (FD–BC) BC biopolymers.

Properties	OD-BC	FD-BC
Degree of crystallinity (%)	79.15	78.45
Surface area (m^2 g^{-1})	4.59	7.05
Degree of permeability (cm^2 min^{-1})	58.10	76.78
Surface roughness (nm)	25.20	16.20
Water uptake capacity (%; 10 h)	580	4450

3.2. Removal efficiency of microplastics

First, the ability of bacterial cellulose (BC) biopolymers to remove polystyrene microplastics (PS-MPs) from contaminated water was evaluated. Given the venture-style nature of the research, well characterized and homogeneous spherical PS-MPs were used in order to focus and streamline the characterization of the biopolymer's remediation potential.

Even macroscopically, it was easily discernible to the naked eye that BC biopolymers removed virtually all MPs post-filtration (as schematised in Fig. S6). Flow cytometry was used to quantify the removal efficiency and the concentration of MPs retained in the BC biopolymer. Fig. 1 illustrates the removal efficiency of the wet BC (W-BC) biopolymer and each of its treatment variations (oven-dried: OD-BC, freeze-dried: FD-BC, filtered: F-BC, and drained: D-BC) after a single filtration cycle. The W-BC biopolymer displayed the highest removal efficiency, at 98.63 ± 0.39%, being able to retain 9.47 ± 0.03 mg L^{-1} of 10 μm PS MPs (Fig. 1b), respectively. Moreover, the dried OD-BC and FD-BC biopolymers exhibited removal efficiencies of 93.04 ± 2.07 and 96.45 ± 2.22%, equating to a retained concentration of 9.05 ± 0.16 and 9.31 ± 0.17 mg L^{-1} (Fig. 1b), correspondingly. The semi-dried F-BC and D-BC biopolymers revealed lower removal efficiencies of 92.49 ± 1.69 and 84.15 ± 1.91%, retaining 9.01 ± 0.13 and 8.38 ± 0.14 mg L^{-1} (Fig. 1b), respectively. Despite field studies generally presenting MP concentrations in particles/L, making the transition to laboratory-scale studies difficult, concentrations around 9–10 mg L^{-1} are far higher than those expected at any treatment stage in WWTPs (Blair et al., 2019; Sun et al., 2019). This highlights that BC biopolymers have gigantic MPs-aggregating capacity and would not act as the limiting factor in the removal of MPs. Still, it is important to acknowledge that more complex water matrixes present different challenges regarding the multiplicity of particles and pollutants that might interplay to change the remediation potential of any filter or biopolymer. Unfortunately, this is an issue that is yet to be addressed in different experimental designs to provide critical knowledge before the translation of any environmental-friendly biotechnologically enabled solution to real wastewater settings. Be that as it may, representative scattering profiles of PS-MPs in pre-and post-filtrated water by OD-BC biopolymers are shown in Fig. 1c,d, exhibiting the exceptionally high retention of MPs by and in the biopolymers.

W-BC biopolymers are highly hydrated as the abundance of hydroxyl groups available in cellulose interact with water molecules, occupying the entirety of the pores within the BC matrix (Portela et al., 2019). Accordingly, these characteristics are highly favourable for the interaction and aggregation of MPs on and within the hydrogel matrix.

In view of the industrial applicability and economic scalability that

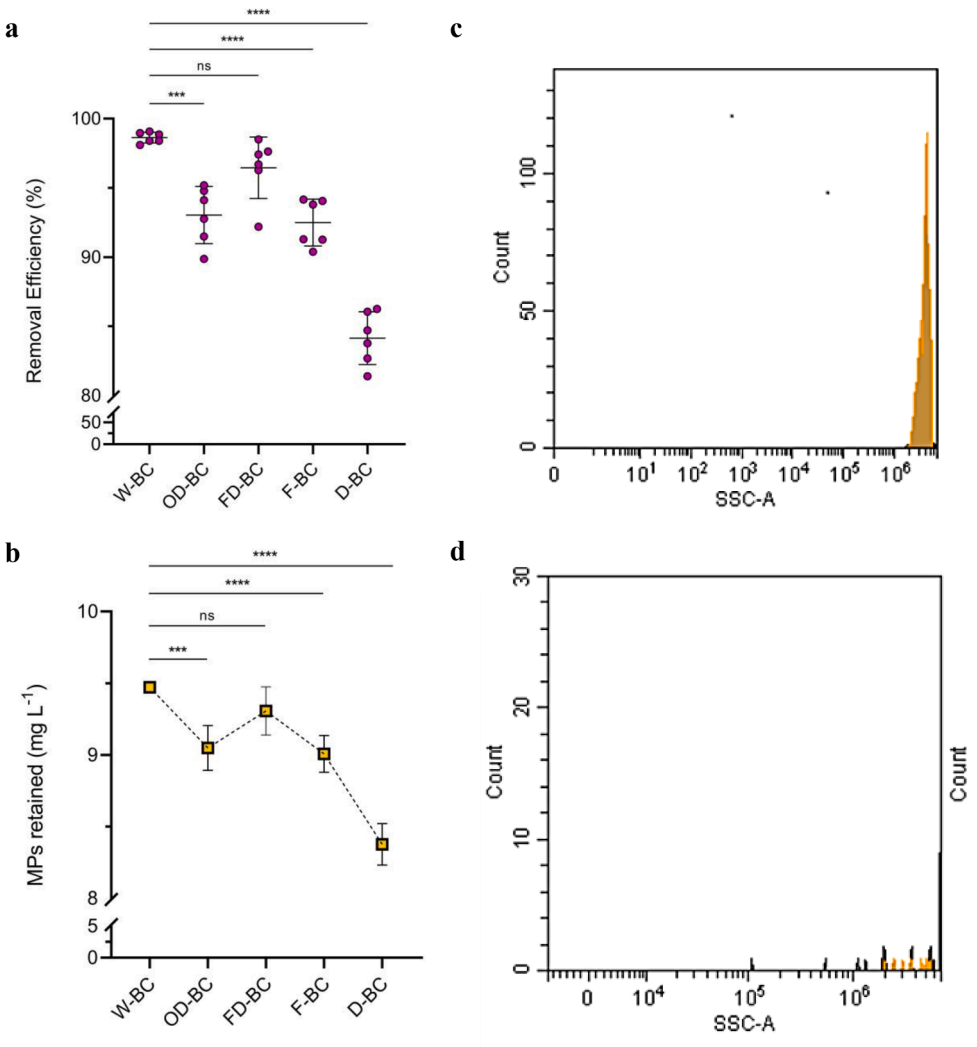

Fig. 1. (**a**) Removal efficiency (%) of microplastics (MPs) and (**b**) MPs retained (mg L^{-1}) in each treated bacterial cellulose (BC) biopolymer: wet (W-BC); oven-dried (OD-BC); freeze-dried (FD-BC); filtered (F-BC) and drained (D-BC). Representative side scattering histograms of the polystyrene MPs in pre- (**c**) and post- (**d**) filtrated water with OD-BC biopolymers. (**a**/**b**) One-way ANOVA/Kruskal-Wallis test.

would most likely benefit from dry rather than wet biopolymers, the focus was put on the potential use of dry biopolymeric biopolymers. Hence, it was noted that OD-BC biopolymers exhibited a significantly decreased removal efficiency ($p = 0.0001$) compared to W-BC biopolymers. However, no statistical differences were found when comparing W-BC to FD-BC biopolymers ($p = 0.2439$). Statistical differences between both dried BC biopolymers (OD-BC and FD-BC) were reported ($p = 0.0211$). These are still highly robust numbers despite statistically lower removal efficiencies around the 95% mark. From a microscopical viewpoint, FD-BC biopolymers are expected to present a more porous structure than an OD-BC biopolymer, given the collapse of the cellulosic fibers due to the oven-drying process. However, as discussed previously, these dried biopolymers can swell and re-hydrate rapidly to recuperate their complete structural integrity during the filtration process. Further, particle retention is known to result from swelling behaviors typical of hydrogel biopolymers such as BC (Hamidi et al., 2008).

These results highlight that water content indeed influences the retention capability of MPs in the BC network. The superior water uptake capacity exhibited by both OD-BC and FD-BC biopolymers enhances membrane hydration due to high affinity with water molecules, effectively potentiating the retention of particles, such as MPs, within the BC matrix in an aqueous solution. The preservation of the dried BC microporous structure likely acts as the trap allowing for the retention of water-polluting MPs. Therefore, and even though drying the BC biopolymers decreased removal efficiencies by 2.18–5.59%, this would most likely be outweighed by logistics and economic advantages, as they can be more easily transported and stored.

Semi-dried BC biopolymers were tested, where 80% of their water content was removed by vacuum filtration (F-BC) or mechanical draining (D-BC). The F-BC biopolymers presented significantly lower removal efficiencies compared to fully dried W-BC ($p < 0.0001$) and FD-BC biopolymers ($p = 0.0060$), but not to OD-BC biopolymers ($p = 0.9835$). On the other hand, D-BC biopolymers display considerably lower removal efficiencies (less 8.34–14.48%) compared to every other biopolymer ($p < 0.0001$). The vacuum filtration process is only likely to remove the unbound water molecules, which play a critical role in maintaining proper hydration levels in the biopolymer. These unbound molecules also act as stabilizing intermediates of the pore structure within the network, given the establishment of hydrogen bonds with cellulose (Portela et al., 2019; Stanisławska et al., 2020). Removing these molecules results in a spatial narrowing between microfibrils, suggesting a decrease in membrane porosity and a lower retention capacity of MPs, similar to the structural changes observed in the OD-BC biopolymers. It is noteworthy that water affects the polymer's charge, which can also affect the adsorption/absorption of particles due to

electrostatic interactions, that is likely to result in the repulsion of particles that settle into the first layers of the polymer. Despite presenting a more favourable economic and least resource-intensive outlook, the compression process is the most likely to physically damage the tightly structured porous network of the BC, effectively compromising its utility as a filter. Thus, the D-BC biopolymers were no longer taken into consideration moving forward.

Overall, and to the best of our knowledge, this is the first study evaluating the potential of BC biopolymers as removal filters for ubiquitous water-polluting MPs. Still, BC biopolymers have been successfully applied to other remediation ends. Particularly, BC has shown promise as a bioadsorbent matrix for removing dyes, proteins, and heavy metals (Kurniawan and Yamamoto, 2013; Mohite and Patil, 2014; Wanichapichart et al., 2002). This biopolymer has also been shown to be a suitable candidate as an environmental-friendly ultrafiltration membrane (Hassan et al., 2017; Isik et al., 2018). Hassan et al. documented that BC acts as an exquisite ultrafiltration membrane to remove oil emulsions from water, yielding a removal efficiency of 98.3 and 99.3% for stabilized and non-stabilized oil emulsions, respectively (Hassan et al., 2017). Moreover, Isik et al. successfully implemented BC membranes to filter water dyes from the textile industry, achieving a 90.9% removal rate (Isik et al., 2018). All these data highlight that BC biopolymers exhibit several other remediation purposes aside from the novel removal of MPs shown here. These biopolymeric membranes demonstrate a considerable promise as multifaceted, genuinely sustainable and circular alternatives to synthetic membrane separation technologies, in which membrane bioreactors (MBRs) stand out as the most prolific scientific solution to date (Poerio et al., 2019). The extremely high removal efficiency numbers coupled with the biosustainable nature and easy economic scalability places BC biopolymers as the best solution to the numerous central issues with fossil-based polymeric membranes (Meng et al., 2009; Poerio et al., 2019; Wang et al., 2014) that have now called for the urgent development of biomembranes (Ding et al., 2021). This follows reports from our laboratory of similarly effective microalgal-based extracellular polymeric substances in aggregating and removing MPs (Cunha et al., 2019, 2020), highlighting that a multitude of biocompatible alternatives are still in the infancy stages of research and development despite tremendous promise.

3.3. BC biopolymers' efficiency

The implementation viability of new biopolymers is tightly correlated with their resistance and consequent longevity. The water uptake ability has been shown to be an important material property to determine hydraulic resistance, which is a measure of filtration performance of the bacterial cellulose (BC) biopolymer (Kim et al., 2017). So, to evaluate the hydraulic resistance and removal efficiency of the wet: W-BC, oven-dried: OD-BC, freeze-dried: FD-BC, and filtered: F-BC biopolymers throughout several (20) filtration cycles, a microplastics (MPs)-contaminated water solution was continuously passed through the biopolymers, with the results summarized in Fig. 2.

Given that both OD-BC and FD-BC biopolymers showed remarkably similar behaviors, the OD-BC biopolymer was chosen as the benchmark going forward due to its lower input costs. The absolute flux and relative flux were also assessed throughout the 20 filtration cycles (Fig. 3). Both the absolute (Fig. 3a) and relative (Fig. 3b) fluxes were shown to decrease across all biopolymers. Interestingly, the W-BC and F-BC biopolymers exhibit a flux decrease of 15.22 and 14.57%, respectively, from the first (1) to the last cycle (20), unlike the OD-BC biopolymers that exhibit a decrease of 88.30%. However, this massive decrease is due to comparatively high initial flux values (hydrated biopolymer). At the end of the 20 cycles, the absolute flux value sits at 2.34 mL min^{-1}, between the 1.50 and 3.99 mL min^{-1} exhibited by the W-BC and F-BC biopolymers. Accordingly, these results are reflected in Fig. 3b, as the relative flux decreases as described. Therefore, it is logical that initial hydration levels play an essential role in the flux registered here, with the effect normalizing as the dried (OD-BC) or partially dried filtered (F-BC) biopolymers re-hydrate. One of the likely reasons is that lower negative zeta potential on the membrane surface acts as an electrostatic barrier against the negatively charged MPs particles, which results in the repulsion of particles settling in the first layers of the biopolymer (Kim et al., 2017). It is also noteworthy that biopolymers did not present any signs of degradation throughout the 20 filtration cycles. The W-BC biopolymer maintained the flux constant for 50 cycles.

3.4. BC biopolymer resistance

The bacterial cellulose (BC) biopolymer's resistance was assessed throughout 50 filtration cycles to evaluate its permeability to polystyrene-microplastics (PS-MPs) in the long term (Fig. 4a). Since all

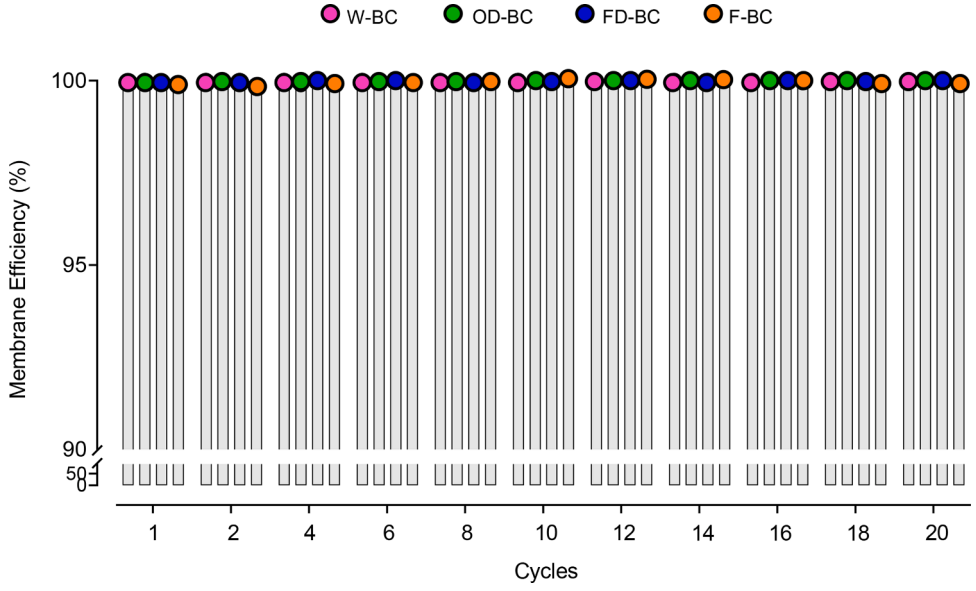

Fig. 2. Biopolymer efficiency in the removal of microplastics (MPs) through 20 filtration cycles, for each treated BC biopolymer: wet (W-BC); oven-dried (OD-BC); freeze-dried (FD-BC) and filtered (F-BC).

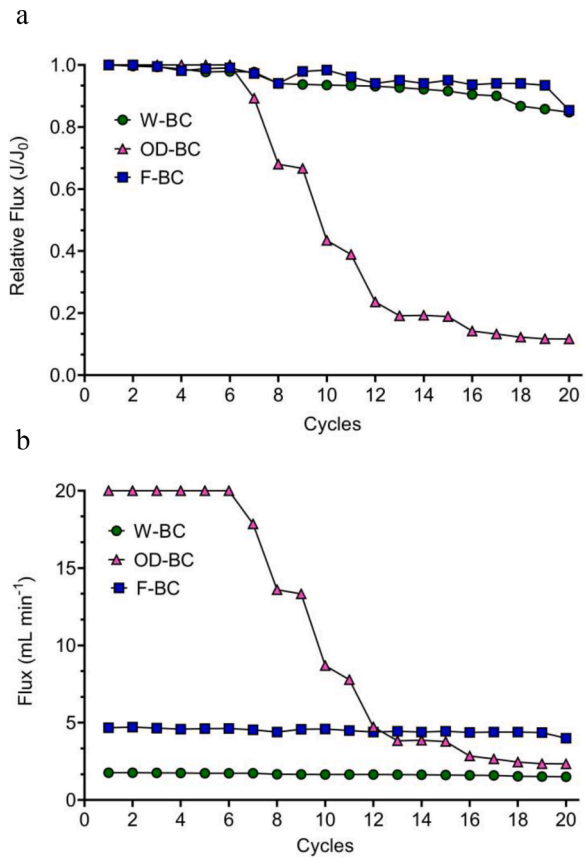

Fig. 3. (**a**) Water flux (mL min^{-1}) and (**b**) Relative water flux (J/J_0) through 20 filtration cycles for each treated BC biopolymer: wet (W-BC), oven-dried (OD-BC), and filtered (F-BC).

BC biopolymers behaved very similarly, only the W-BC biopolymers' data is shown here. It turns out that the pore clogging is shown to increase more rapidly until the 25th cycle, after which the increase is less marked. After 50 filtration cycles, the membrane showed no deformation or signs of destruction, effectively preserving its structural integrity.

In correlation with previously presented data, it is shown that, despite membrane resistance/pore clogging hitting 43.5% after 50 filtration cycles (Fig. 4a), the flux remains stable during 16 cycles. It indicates that these BC biopolymers enable an efficient re-utilization for several filtration cycles, maintaining an extremely high removal efficiency of MPs (Fig. 2) despite flux reduction (Fig. 3) and pore-clogging (Fig. 4a). This highlights that despite clogging roughly half of the BC biopolymer's pores after 50 cycles, a large network area within the membrane is still available to capture and remove MPs. These results underline that membrane fouling is somewhat negligible to the biopolymer's MPs-removal performance. Still, it is important to acknowledge that these results still represent an experimental laboratory-scale studies that will require further testing with more complex water matrixes.

Relative to the release of MPs, it was noted that after 20 filtration cycles, only a small portion of MPs were organically released (<7.30%), with OD-BC biopolymers displaying the best retention (99.32%) (Fig. 4b). This means that BC biopolymers are able not only to capture MPs, but to firmly imprison these micropolymeric particles on and within the network. This raises a critical question regarding the application scalability of these biopolymeric membranes. After reaching its end of life, a close to harmless and environmental undamaging solution for MP-saturated membranes must be delineated. Several solutions would be suggested: *(1)* the compression moulding into high-density pellets which could be melt-processed or used in composites; *(2)* the carbonization in inert atmosphere to obtain active carbon absorbers since cellulose can be easily carbonized; *(3)* the melt- or electrospinning into a new type of secondary filters.

3.5. MPs retention in BC biopolymers

To characterize the retention of microplastics (MPs) in the nanoporous bacterial cellulose (BC) biopolymeric network, scanning electron (SEM) and fluorescence microscopy were employed. W-BC biopolymers were analyzed using SEM, from both the cross-section (Fig. 5a) and top-view (Fig. 5b) of the polymers. These biopolymeric membranes presented a characteristic three-dimensional nanofibrillar network and laminar microstructure, with a dense and fibrillar microporous structure in ultra-fine detail (1–5 μm) (Fig. S7a–d). As expected, and predicted during the BC characterization, severe structural changes in the oven-dried (OD-BC) and freeze-dried (FD-BC) biopolymers are observed. The OD-BC biopolymers exhibited a compact structure constituted by ribbon-shaped fibrils, providing a larger surface area and high-water absorption ability (Fig. 5c). On the other hand, FD-BC biopolymers displayed an increase in the number of fibril networks, resulting in an increasingly porous matrix and greater structural preservation (Fig. 5d, e). This structural conservation is likely the reason the removal efficiency is closer to the one observed in wet (W-BC) biopolymers (Fig. 1a,

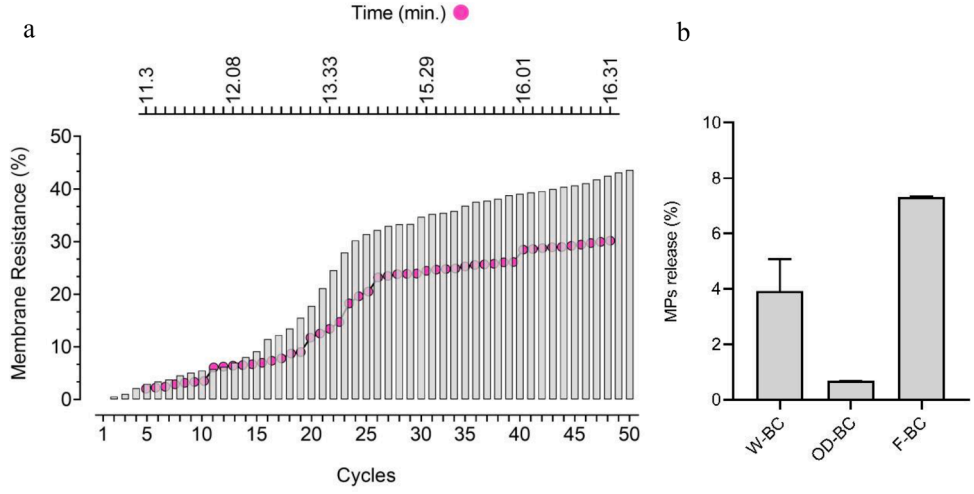

Fig. 4. (**a**) Membrane resistance/pore clogging of a W-BC biopolymer throughout 50 filtration cycles, in a time-dependant manner. (**b**) Release of microplastics (MPs) after 20 filtration cycles assessed via flow cytometry for each treated BC biopolymer: wet (W-BC); oven-dried (OD-BC) and filtered (F-BC).

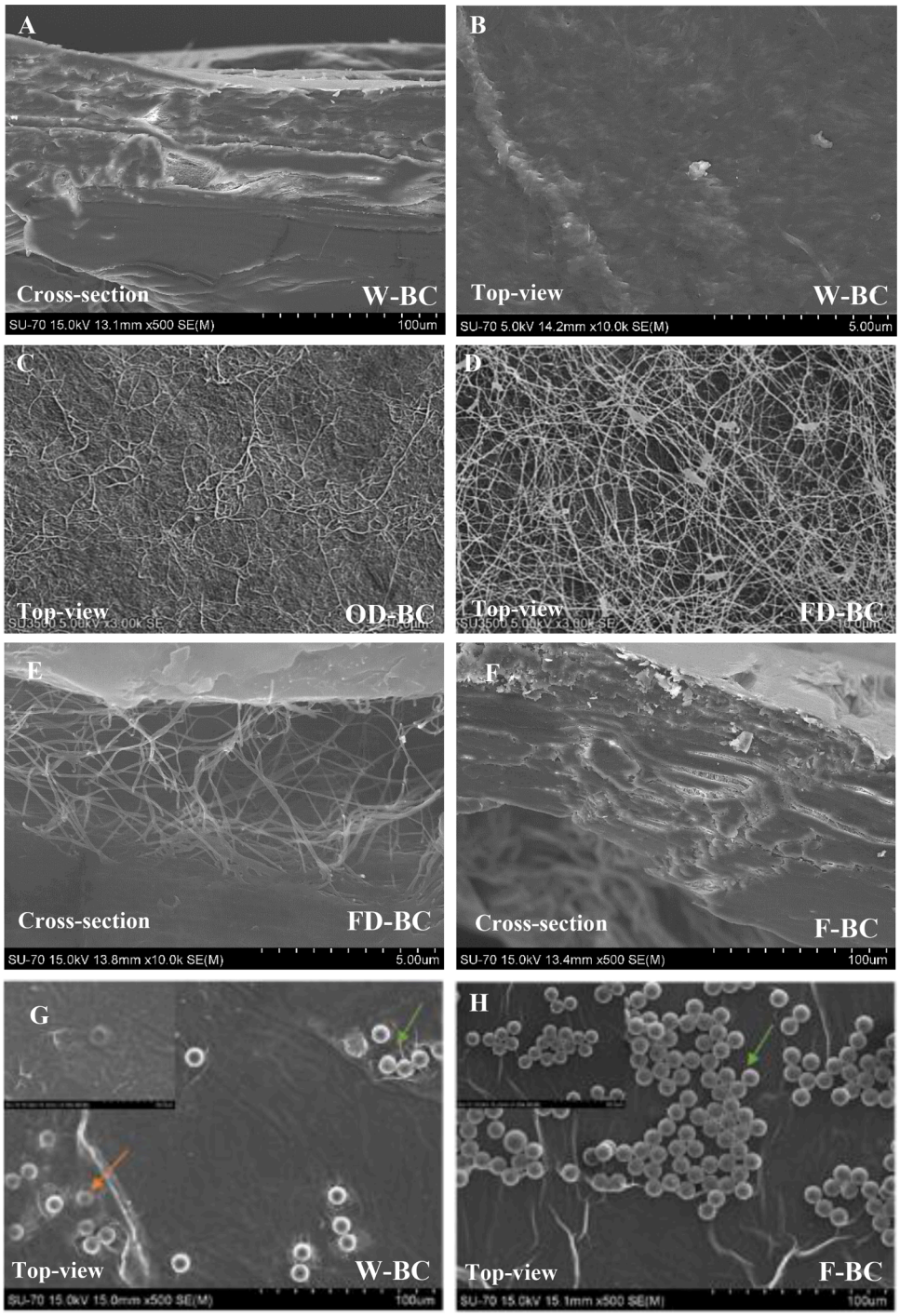

Fig. 5. (**a**) Representative cross-section scanning electron microscopy (SEM) image of a wet bacterial cellulose (W-BC) biopolymer (x500; 100 μm). (**b**) Representative top-view SEM image of a wet bacterial cellulose (W-BC) biopolymer (x10.000; 5 μm). (**c**) Representative top-view SEM image of an oven-dried bacterial cellulose (OD-BC) biopolymer (x3000; 10 μm). (**d**) Representative top-view SEM image of a freeze-dried bacterial cellulose (FD-BC) biopolymer (x3000; 10 μm). (**e**) Representative cross-section SEM image of a FD-BC biopolymer (x10.000; 5 μm). (**f**) Representative cross-section SEM image of a filtered bacterial cellulose (F-BC) biopolymer (x500; 100 μm). (**g**) Representative top-view SEM image of a W-BC biopolymer with MPs (x500; 100 μm). (**h**) Representative top-view SEM image of a F-BC biopolymer with MPs (x500; 100 μm).

b), with equally preserved membrane efficiency across a great number of cycles (Fig. 2). Nevertheless, if the input and economic scalability costs of FD-BC biopolymers would be massively different from OD-BC biopolymers, the latter still presents an excellent and far superior alternative to current solutions.

The adsorption of PS microplastic particles across the polymer's surface was confirmed as observable in Fig. 5g,h. In finer detail (1–10 μm), the interaction between the PS-MPs and the dried hydrogel matrix of the BC hydrogel (Fig. S7e–h) was clear. Additionally, fluorescence microscopy revealed that, aside from adsorbing on the surface of the BC biopolymers, MPs were also thoroughly incorporated (Fig. S7e1,g1).

3.6. Call for research

The next years are expected to surface even more data reinforcing the concerns surrounding the health hazards of microplastics (MPs) following the reports of their presence in human placenta and blood (Ragusa et al., 2021; Leslie et al., 2022). Despite environmental pollution being a more considerable ramification of the whole plastic issue, the urban and more human-correlated contamination outgrowth are still tremendously troubling. As the demand for biosustainable and circular alternatives grows across all industrial and technological sectors of the economy, a novel bacterial-based alternative for the remediation of MP-polluted waters is shown here. The discovery of this biopolymer follows other microorganismal-based biopolymers pioneered by our

laboratory such as microalgal-based exopolymers/extracellular polymeric substances in aggregating and removing MPs from contaminated waters (Cunha et al., 2020, 2019). This highlights that a wide array of environmentally-compatible biotechnologically-advanced alternatives are still in early stages of R&D despite enormous potential. This experimental design focuses on characterizing and analysing the effectiveness of the biopolymers in an accurate and controlled manner by focusing on a simpler water matrix composed of clean water spiked with microplastics. More complex water matrixes, including real wastewater samples, present severe analytical challenges that would inherently defeat the purpose of trying to accurately measure the removal efficiency of MPs. A wide range of pollutants might synergistically influence the effectiveness of the biopolymer in removing other types of pollutants. Artificially recreated wastewater matrixes should be developed before transitioning any type of biotechnological development to real wastewaters or any other industrial setting is that it is completely unknown how different compounds would interact within real wastewaters to promote compound degradation and stability. The experimental setup required to understand these synergistic effects serves a different purpose from the one employed in the present work and requires intricate scientific thinking going forward. We are working towards identifying the knowledge gaps regarding particle and compound interactions and urge others to do the same. The fact that no one has been able to recreate and understand these dynamics are reflective of the analytical intricacy of the experimental design. However, that should act as a catalyst for decoding the last step of the chain from the bench to industrial application. Noticeably, the results presented here are extremely encouraging, but several whole new research avenues still need to be explored. On the biotechnological front, it is now important to understand the cake layer, fouling and cleaning behaviors of the bacterial cellulose (BC) biopolymers under the filtration of more complex water matrixes. On the financial front, it is now necessary to calculate the CAPEX (capital expenditure) to achieve economies of scale. It would be inherently inaccurate to calculate and compare the costs of current inorganic and synthetic polymeric membranes against BC biopolymers given the lack of data and the multitude of factors involved in production and scaling costs that are ultimately very specific to each operation. It is however important to understand how to optimize culture conditions at an industrial scale to produce bacterial cellulose (BC) biopolymers at cost-efficiency. Still, it is vital to understand that the economic framework is only one of the many constants of the equation. It cannot outweigh or outshine the principal issue: the development of biodegradable, biosustainable and truly circular biomembrane alternatives to current water treatment applications. The fact that BC biopolymers can be easily morphed to create all types of custom-size membranes further strengthens its application narrative. All and all, we have to realize that this is not just another human-prompted issue. This is one of our biggest generational issues and should be addressed as such. And turning a blind eye is not an option anymore.

4. Conclusion

The focus of this experimental laboratory-scale study was to evaluate the potential of bacterial cellulose (BC) biopolymers as biosustainable and ecologically inert alternatives to conventional fossil-based polymeric membranes used in water-treatment sites such as wastewater treatment plants (WWTPs) for microplastics (MPs) removal. Results show that wet BC (W-BC), oven-dried BC (OD-BC) and freeze-dried BC (FD-BC) biopolymers display the best compromise between efficiency and applicability. Both wet and dried BC biopolymers present removal efficiencies ranging from 93 to 99%. However, drying the biopolymers greatly increases logistics and implementation practicality, with removal efficiencies reduced by only 2–6%. These membranes rehydrate rapidly and show no signs of degradation throughout filtration cycles, preserving exceptionally high removal efficiencies despite flux reduction and pore-clogging. The extensive surface area available for MPs particle interaction in these biopolymeric networks enables a sustainable re-utilization throughout several filtration cycles. BC biopolymers exhibit exceedingly high levels of MPs retention, not only by capturing micropolymeric particles but by also firmly imprisoning them within the laminar 3D microstructure nanofibrillar matrix. Altogether, this study shows that BC biopolymers have a massive potential as true and feasible biosustainable solutions to membrane-based water treatment processes.

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.

Data availability

Data will be made available on request.

Acknowledgments

This research was supported by Foundation for Science and Technology (FCT), through CIIMAR - UIDB/04423/2020 and UIDP/04423/2020, and by the European Territorial Cooperation Programme PCT-MAC 2014–2020 through REBECA-CCT (MAC/1.1.B/269) project. Marisa Faria was financially supported by a doctoral grant (BD/6615/2020) from FCT. Ivana Mendonça acknowledges a research grant financed by Fundação Amadeu Dias under the scope of the programme BYT+, promoted by CIIMAR.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.118952.

References

Alimi, O.S., Farner Budarz, J., Hernandez, L.M., Tufenkji, N., 2018. Microplastics and nanoplastics in aquatic environments: aggregation, deposition, and enhanced contaminant transport. Environ. Sci. Technol. 52, 1704–1724. https://doi.org/10.1021/acs.est.7b05559.

Alonso, E., Faria, M., Ferreira, A., Cordeiro, N., 2018. Influence of the matrix and polymerization methods on the synthesis of BC/PANi nanocomposites: an IGC study. Cellulose 25, 2343–2354. https://doi.org/10.1007/s10570-018-1736-0.

Andritsou, V., de Melo, E.M., Tsouko, E., Ladakis, D., Maragkoudaki, S., Koutinas, A.A., Matharu, A.S., 2018. Synthesis and characterization of bacterial cellulose from citrus-based sustainable resources. ACS Omega 3, 10365–10373. https://doi.org/10.1021/acsomega.8b01315.

Ben-David, E.A., Habibi, M., Haddad, E., Hasanin, M., Angel, D.L., Booth, A.M., Sabbah, I., 2021. Microplastic distributions in a domestic wastewater treatment plant: removal efficiency, seasonal variation and influence of sampling technique. Sci. Total Environ. 752, 141880 https://doi.org/10.1016/j.scitotenv.2020.141880.

Blair, R.M., Waldron, S., Gauchotte-Lindsay, C., 2019. Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period. Water Res. 163, 114909 https://doi.org/10.1016/j.watres.2019.114909.

Campanale, C., Massarelli, C., Savino, I., Locaputo, V., Uricchio, V.F., 2020. A detailed review study on potential effects of microplastics and additives of concern on human health. Int. J. Environ. Res. Public Health 17, 1212. https://doi.org/10.3390/ijerph17041212.

Carbery, M., O'Connor, W., Palanisami, T., 2018. Trophic transfer of microplastics and mixed contaminants in the marine food web and implications for human health. Environ. Int. 115, 400–409. https://doi.org/10.1016/j.envint.2018.03.007.

Castro, C., Cordeiro, N., Faria, M., Zuluaga, R., Putaux, J.L., Filpponen, I., Velez, L., Rojas, O.J., Gañán, P., 2015. *In-situ* glyoxalization during biosynthesis of bacterial cellulose. Carbohydr. Polym. 126, 32–39. https://doi.org/10.1016/j.carbpol.2015.03.014.

Chen, G., Feng, Q., Wang, J., 2020. Mini-review of microplastics in the atmosphere and their risks to humans. Sci. Total Environ. 703, 135504 https://doi.org/10.1016/j.scitotenv.2019.135504.

Chen, S.Q., Lopez-Sanchez, P., Wang, D., Mikkelsen, D., Gidley, M.J., 2018. Mechanical properties of bacterial cellulose synthesised by diverse strains of the genus Komagataeibacter. Food Hydrocoll. 81, 87–95. https://doi.org/10.1016/j.foodhyd.2018.02.031.

Cox, K.D., Covernton, G.A., Davies, H.L., Dower, J.F., Juanes, F., Dudas, S.E., 2019. Human consumption of microplastics. Environ. Sci. Technol. 53, 7068–7074. https://doi.org/10.1021/acs.est.9b01517.

Cunha, C., Faria, M., Nogueira, N., Ferreira, A., Cordeiro, N., 2019. Marine vs freshwater microalgae exopolymers as biosolutions to microplastics pollution. Environ. Pollut. 249, 372–380. https://doi.org/10.1016/j.envpol.2019.03.046.

Cunha, C., Silva, L., Paulo, J., Faria, M., Nogueira, N., Cordeiro, N., 2020. Microalgal-based biopolymer for nano- and microplastic removal: a possible biosolution for wastewater treatment. Environ. Pollut. 263, 114385 https://doi.org/10.1016/j.envpol.2020.114385.

Dick Vethaak, A., Legler, J., 2021. Microplastics and human health. Science 371 (80), 672–674. https://doi.org/10.1126/SCIENCE.ABE5041.

Ding, H., Zhang, J., He, H., Zhu, Y., Dionysiou, D.D., Liu, Z., Zhao, C., 2021. Do membrane filtration systems in drinking water treatment plants release nano/microplastics? Sci. Total Environ. 755, 142658 https://doi.org/10.1016/j.scitotenv.2020.142658.

Edo, C., González-Pleiter, M., Leganés, F., Fernández-Piñas, F., Rosal, R., 2020. Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. Environ. Pollut. 259, 113837 https://doi.org/10.1016/j.envpol.2019.113837.

Esa, F., Tasirin, S.M., Rahman, N.A., 2014. Overview of bacterial cellulose production and application. Agric. Agric. Sci. Procedia 2, 113–119. https://doi.org/10.1016/j.aaspro.2014.11.017.

Fan, M., Dai, D., Huang, B., 2012. Fourier transform infrared spectroscopy for natural fibres. Fourier Transform - Materials Analysis. InTech, pp. 45–68.

Faria, M., Vilela, C., Silvestre, A.J.D., Deepa, B., Resnik, M., Freire, C.S.R., Cordeiro, N., 2019. Physicochemical surface properties of bacterial cellulose/polymethacrylate nanocomposites: an approach by inverse gas chromatography. Carbohydr. Polym. 206, 86–93. https://doi.org/10.1016/j.carbpol.2018.10.110.

Galloway, T.S., Cole, M., Lewis, C., 2017. Interactions of microplastic debris throughout the marine ecosystem. Nat. Ecol. Evol. 1, 1–8. https://doi.org/10.1038/s41559-017-0116.

Gatidou, G., Arvaniti, O.S., Stasinakis, A.S., 2019. Review on the occurrence and fate of microplastics in sewage treatment plants. J. Hazard. Mater. 367, 504–512. https://doi.org/10.1016/j.jhazmat.2018.12.081.

Gewert, B., Plassmann, M.M., Macleod, M., 2015. Pathways for degradation of plastic polymers floating in the marine environment. Environ. Sci. Process. Impacts 17, 1513–1521. https://doi.org/10.1039/c5em00207a.

Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use, and fate of all plastics ever made. Sci. Adv. 3, 1700782 https://doi.org/10.1126/sciadv.1700782.

Gruber, E.S., Stadlbauer, V., Pichler, V., Resch-Fauster, K., Todorovic, A., Meisel, T.C., Trawoeger, S., Hollóczki, O., Turner, S.D., Wadsak, W., Vethaak, A.D., Kenner, L., 2022. To waste or not to waste: questioning potential health risks of micro- and nanoplastics with a focus on their ingestion and potential carcinogenicity. Expo Health 15, 1–19. https://doi.org/10.1007/S12403-022-00470-8/FIGURES/1.

Gupta, J., Hurley, F., Grobicki, A., Keating, T., Stoett, P., Baker, E., Guhl, A., Davies, J., Ekins, P., 2019. Communicating the health of the planet and its links to human health. Lancet Planet. Health 3, 204–206. https://doi.org/10.1016/S2542-5196(19)30040-3/ATTACHMENT/7C936248-529B-435B-A684-49E12E3F0ECD/MMC1.PDF.

Hale, R.C., Seeley, M.E., La Guardia, M.J., Mai, L., Zeng, E.Y., 2020. A global perspective on microplastics. J. Geophys. Res. Ocean. 125, e2018JC014719 https://doi.org/10.1029/2018JC014719.

Hamidi, M., Azadi, A., Rafiei, P., 2008. Hydrogel nanoparticles in drug delivery. Adv. Drug Deliv. Rev. 60, 1638–1649. https://doi.org/10.1016/j.addr.2008.08.002.

Hassan, E., Hassan, M., Abou-zeid, R., Berglund, L., Oksman, K., 2017. Use of bacterial cellulose and crosslinked cellulose nanofibers membranes for removal of oil from oil-in-water emulsions. Polymers 9, 388. https://doi.org/10.3390/polym9090388 (Basel).

He, X., Meng, H., Song, H., Deng, S., He, T., Wang, S., Wei, D., Zhang, Z., 2020. Novel bacterial cellulose membrane biosynthesized by a new and highly efficient producer Komagataeibacter rhaeticus TJPU03. Carbohydr. Res. 493, 108030 https://doi.org/10.1016/j.carres.2020.108030.

Horton, A.A., Cross, R.K., Read, D.S., Jürgens, M.D., Ball, H.L., Svendsen, C., Vollertsen, J., Johnson, A.C., 2021. Semi-automated analysis of microplastics in complex wastewater samples. Environ. Pollut. 268, 115841 https://doi.org/10.1016/j.envpol.2020.115841.

Isik, Z., Unyayar, A., Dizge, N., 2018. Filtration and antibacterial properties of bacterial cellulose membranes for textile wastewater treatment. Avicenna J. Environ. Health Eng. 5, 106–114. https://doi.org/10.15171/ajehe.2018.14.

Jia, Y., Wang, X., Huo, M., Zhai, X., Li, F., Zhong, C., 2017. Preparation and characterization of a novel bacterial cellulose/chitosan bio-hydrogel. Nanomater. Nanotechnol. 7, 184798041770717 https://doi.org/10.1177/1847980417707172.

Jin, Y., Lu, L., Tu, W., Luo, T., Fu, Z., 2019. Impacts of polystyrene microplastic on the gut barrier, microbiota and metabolism of mice. Sci. Total Environ. 649, 308–317. https://doi.org/10.1016/j.scitotenv.2018.08.353.

Kaile, N., Lindivat, M., Elio, J., Thuestad, G., Crowley, Q., Hoell, I., 2020. Preliminary results from detection of microplastics in liquid samples using flow cytometry. Front. Mar. Sci. 7, 552688 https://doi.org/10.3389/fmars.2020.552688.

Kay, P., Hiscoe, R., Moberley, I., Bajic, L., McKenna, N., 2018. Wastewater treatment plants as a source of microplastics in river catchments. Environ. Sci. Pollut. Res. 25, 20264–20267. https://doi.org/10.1007/s11356-018-2070-7.

Kim, H., Kim, K., Lee, S.J., 2017. Nature-inspired thermo-responsive multifunctional membrane adaptively hybridized with pnipam and ppy. NPG Asia Mater. 9, 445. https://doi.org/10.1038/am.2017.168.

Koelmans, A.A., Mohamed Nor, N.H., Hermsen, E., Kooi, M., Mintenig, S.M., De France, J., 2019. Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res. 155, 410–422. https://doi.org/10.1016/j.watres.2019.02.054.

Kurniawan, A., Yamamoto, T., 2013. Biofilm polymer for biosorption of pollutant ions. Procedia Environ. Sci. 17, 179–187. https://doi.org/10.1016/j.proenv.2013.02.027.

Lahiri, D., Nag, M., Dutta, B., Dey, A., Sarkar, T., Pati, S., Edinur, H.A., Kari, Z.A., Noor, N.H.M., Ray, R.R., 2021. Bacterial cellulose: production, characterization and application as antimicrobial agent. Int. J. Mol. Sci. 22, 12984. https://doi.org/10.3390/ijms222312984.

Lehner, R., Weder, C., Petri-Fink, A., Rothen-Rutishauser, B., 2019. Emergence of nanoplastic in the environment and possible impact on human health. Environ. Sci. Technol. 53, 1748–1765. https://doi.org/10.1021/acs.est.8b05512.

Li, D., Shi, Y., Yang, L., Xiao, L., Kehoe, D.K., Gun'ko, Y.K., Boland, J.J., Wang, J.J., 2020. Microplastic release from the degradation of polypropylene feeding bottles during infant formula preparation. Nat. Food 1, 746–754. https://doi.org/10.1038/s43016-020-00171-y.

Leslie, H.A., van Velzen, M.J.M., Brandsma, S.H., Vethaak, A.D., Garcia-Vallejo, J.J., Lamoree, M.H., 2022. Discovery and quantification of plastic particle pollution in human blood. Environ. Int. 163, 107199. https://doi.org/10.1016/j.envint.2022.107199.

Li, X., Chen, L., Mei, Q., Dong, B., Dai, X., Ding, G., Zeng, E.Y., 2018. Microplastics in sewage sludge from the wastewater treatment plants in China. Water Res. 142, 75–85. https://doi.org/10.1016/j.watres.2018.05.034.

Lu, L., Luo, T., Zhao, Y., Cai, C., Fu, Z., Jin, Y., 2019. Interaction between microplastics and microorganism as well as gut microbiota: a consideration on environmental animal and human health. Sci. Total Environ. 667, 94–100. https://doi.org/10.1016/j.scitotenv.2019.02.380.

Lu, L., Wan, Z., Luo, T., Fu, Z., Jin, Y., 2018. Polystyrene microplastics induce gut microbiota dysbiosis and hepatic lipid metabolism disorder in mice. Sci. Total Environ. 631–632, 449–458. https://doi.org/10.1016/j.scitotenv.2018.03.051.

Ma, B., Xue, W., Hu, C., Liu, H., Qu, J., Li, L., 2019. Characteristics of microplastic removal via coagulation and ultrafiltration during drinking water treatment. Chem. Eng. J. 359, 159–167. https://doi.org/10.1016/j.cej.2018.11.155.

Meng, F., Chae, S.R., Drews, A., Kraume, M., Shin, H.S., Yang, F., 2009. Recent advances in membrane bioreactors (MBRs): membrane fouling and membrane material. Water Res. 43, 1489–1512. https://doi.org/10.1016/j.watres.2008.12.044.

Mintenig, S.M., Int-Veen, I., Löder, M.G.J., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res. 108, 365–372. https://doi.org/10.1016/j.watres.2016.11.015.

Mohammadkazemi, F., Aguiar, R., Cordeiro, N., 2017. Improvement of bagasse fiber–cement composites by addition of bacterial nanocellulose: an inverse gas chromatography study. Cellulose 24, 1803–1814. https://doi.org/10.1007/s10570-017-1210-4.

Mohite, B.V., Patil, S.V., 2014. Bacterial cellulose of Gluconoacetobacter hansenii as a potential bioadsorption agent for its green environment applications. J. Biomater. Sci. Polym. Ed. 25, 2053–2065. https://doi.org/10.1080/09205063.2014.970063.

Na, S.H., Kim, M.J., Kim, J.T., Jeong, S., Lee, S., Chung, J., Kim, E.J., 2021. Microplastic removal in conventional drinking water treatment processes: performance, mechanism, and potential risk. Water Res. 202, 117417 https://doi.org/10.1016/j.watres.2021.117417.

Ngo, P.L., Pramanik, B.K., Shah, K., Roychand, R., 2019. Pathway, classification and removal efficiency of microplastics in wastewater treatment plants. Environ. Pollut. 255, 113326 https://doi.org/10.1016/j.envpol.2019.113326.

NOAA, 2020. Ocean Facts: what are microplastics? Available at: https://oceanservice.noaa.gov/facts/microplastics.html.

Ou, H., Zeng, E.Y., 2018. Occurrence and fate of microplastics in wastewater treatment plants,. Microplastic Contamination in Aquatic Environments: An Emerging Matter of Environmental Urgency. Elsevier, pp. 317–338. https://doi.org/10.1016/B978-0-12-813747-5.00010-2.

Phisalaphong, M., Jatupaiboon, N., 2008. Biosynthesis and characterization of bacteria cellulose-chitosan film. Carbohydr. Polym. 74, 482–488. https://doi.org/10.1016/j.carbpol.2008.04.004.

Pivokonsky, M., Cermakova, L., Novotna, K., Peer, P., Cajthaml, T., Janda, V., 2018. Occurrence of microplastics in raw and treated drinking water. Sci. Total Environ. 643, 1644–1651. https://doi.org/10.1016/J.SCITOTENV.2018.08.102.

PlasticsEurope, 2018. Plastics -The facts 2018. An analysis of European plastics production, demand and waste data. PlasticsEurope 1–30.

Poerio, T., Piacentini, E., Mazzei, R., 2019. Membrane processes for microplastic removal. Molecules 24, 4148. https://doi.org/10.3390/molecules24224148.

Portela, R., Leal, C.R., Almeida, P.L., Sobral, R.G., 2019. Bacterial cellulose: a versatile biopolymer for wound dressing applications. Microb. Biotechnol. 12, 586–610. https://doi.org/10.1111/1751-7915.13392.

Prata, J.C., 2018. Airborne microplastics: consequences to human health? Environ. Pollut. 234, 115–126. https://doi.org/10.1016/J.ENVPOL.2017.11.043.

Prata, J.C., da Costa, J.P., Lopes, I., Duarte, A.C., Rocha-Santos, T., 2020. Environmental exposure to microplastics: an overview on possible human health effects. Sci. Total Environ. 702, 134455 https://doi.org/10.1016/j.scitotenv.2019.134455.

Ragusa, A., Svelato, A., Santacroce, C., Catalano, P., Notarstefano, V., Carnevali, O., Papa, F., Rongioletti, M.C.A., Baiocco, F., Draghi, S., D'Amore, E., Rinaldo, D., Matta, M., Giorgini, E., 2021. Plasticenta: first evidence of microplastics in human placenta. Environ. Int. 146, 106274 https://doi.org/10.1016/J.ENVINT.2020.106274.

Raiszadeh-Jahromi, Y., Rezazadeh-Bari, M., Almasi, H., Amiri, S., 2020. Optimization of bacterial cellulose production by Komagataeibacter xylinus PTCC 1734 in a low-cost medium using optimal combined design. J. Food Sci. Technol. 57, 2524–2533. https://doi.org/10.1007/s13197-020-04289-6.

Revin, V., Liyaskina, E., Nazarkina, M., Bogatyreva, A., Shchankin, M., 2018. Cost-effective production of bacterial cellulose using acidic food industry by-products. Braz. J. Microbiol. 49, 151–159. https://doi.org/10.1016/j.bjm.2017.12.012.

Rezania, S., Park, J., Md Din, M.F., Mat Taib, S., Talaiekhozani, A., Kumar Yadav, K., Kamyab, H., 2018. Microplastics pollution in different aquatic environments and biota: a review of recent studies. Mar. Pollut. Bull. 133, 191–208. https://doi.org/10.1016/J.MARPOLBUL.2018.05.022.

Rolsky, C., Kelkar, V., Driver, E., Halden, R.U., 2020. Municipal sewage sludge as a source of microplastics in the environment. Curr. Opin. Environ. Sci. Health 14, 16–22. https://doi.org/10.1016/j.coesh.2019.12.001.

Schwabl, P., 2020. Microplastics in hot water. Nat. Food 1, 671–672. https://doi.org/10.1038/s43016-020-00174-9.

Shah, N., Ul-Islam, M., Khattak, W.A., Park, J.K., 2013. Overview of bacterial cellulose composites: a multipurpose advanced material. Carbohydr. Polym. 98, 1585–1598. https://doi.org/10.1016/j.carbpol.2013.08.018.

Sharma, S., Basu, S., Shetti, N.P., Nadagouda, M.N., Aminabhavi, T.M., 2021. Microplastics in the environment: occurrence, perils, and eradication. Chem. Eng. J. 408, 127317 https://doi.org/10.1016/j.cej.2020.127317.

Singhsa, P., Narain, R., Manuspiya, H., 2018. Physical structure variations of bacterial cellulose produced by different Komagataeibacter xylinus strains and carbon sources in static and agitated conditions. Cellulose 25, 1571–1581. https://doi.org/10.1007/s10570-018-1699-1.

Smith, M., Love, D.C., Rochman, C.M., Neff, R.A., 2018. Microplastics in seafood and the implications for human health. Curr. Environ. Health Rep. 5, 375–386. https://doi.org/10.1007/s40572-018-0206-z.

Stanisławska, A., Staroszczyk, H., Szkodo, M., 2020. The effect of dehydration/rehydration of bacterial nanocellulose on its tensile strength and physicochemical properties. Carbohydr. Polym. 236, 116023 https://doi.org/10.1016/j.carbpol.2020.116023.

Sun, J., Dai, X., Wang, Q., van Loosdrecht, M.C.M., Ni, B.J., 2019. Microplastics in wastewater treatment plants: detection, occurrence and removal. Water Res. 152, 21–37. https://doi.org/10.1016/j.watres.2018.12.050.

Sun, T., Zhan, J., Li, F., Ji, C., Wu, H., 2021. Effect of microplastics on aquatic biota: a hormetic perspective. Environ. Pollut. 285, 117206 https://doi.org/10.1016/J.ENVPOL.2021.117206.

Tse, Y., Lo, H., Chan, S., Sze, E., 2022. Flow cytometry as a rapid alternative to quantify small microplastics in environmental water samples. Water 14, 1436. https://doi.org/10.3390/w14091436.

Tsouko, E., Kourmentza, C., Ladakis, D., Kopsahelis, N., Mandala, I., Papanikolaou, S., Paloukis, F., Alves, V., Koutinas, A., 2015. Bacterial cellulose production from industrial waste and by-product streams. Int. J. Mol. Sci. 16, 14832–14849. https://doi.org/10.3390/ijms160714832.

Turan, N.B., Erkan, H.S., Engin, G.O., 2021. Microplastics in wastewater treatment plants: occurrence, fate and identification. Process Saf. Environ. Prot. 146, 77–84. https://doi.org/10.1016/J.PSEP.2020.08.039.

Ul-Islam, M., Khan, T., Park, J.K., 2012. Nanoreinforced bacterial cellulose-montmorillonite composites for biomedical applications. Carbohydr. Polym. 89, 1189–1197. https://doi.org/10.1016/j.carbpol.2012.03.093.

UN environment, 2018. World environment day 2018: overview. United Nations. Available at: https://wedocs.unep.org/bitstream/handle/20.500.11822/25398/WEDMessagingTwo-Page27April.pdf?sequence=12&isAllowed=y.

Vazquez, A., Foresti, M.L., Cerrutti, P., Galvagno, M., 2013. Bacterial cellulose from simple and low cost production media by gluconacetobacter xylinus. J. Polym. Environ. 21, 545–554. https://doi.org/10.1007/s10924-012-0541-3.

Venghaus, D.V., Barjenbruch, M., 2017. Microplastics in urban water management. Czas. Tech. 1, 137–146. https://doi.org/10.4467/2353737xct.17.011.6108.

Verschoor, A, de Poorter, L, Roex, E, Bellert, B., 2014. Quick scan and prioritization of microplastic sources and emissions. RIVM Letter report 5, 250012001.

Volova, T.G., Prudnikova, S.V., Sukovatyi, A.G., Shishatskaya, E.I., 2018. Production and properties of bacterial cellulose by the strain Komagataeibacter xylinus B-12068. Appl. Microbiol. Biotechnol. 102, 7417–7428. https://doi.org/10.1007/s00253-018-9198-8.

Wang, J., Tavakoli, J., Tang, Y., 2019. Bacterial cellulose production, properties and applications with different culture methods – a review. Carbohydr. Polym. 219, 63–76. https://doi.org/10.1016/j.carbpol.2019.05.008.

Wang, T., Wang, L., Chen, Q., Kalogerakis, N., Ji, R., Ma, Y., 2020. Interactions between microplastics and organic pollutants: effects on toxicity, bioaccumulation, degradation, and transport. Sci. Total Environ. 748, 142427 https://doi.org/10.1016/j.scitotenv.2020.142427.

Wang, Z., Ma, J., Tang, C.Y., Kimura, K., Wang, Q., Han, X., 2014. Membrane cleaning in membrane bioreactors: a review. J. Membr. Sci. 468, 276–307. https://doi.org/10.1016/j.memsci.2014.05.060.

Wanichapichart, P., Kaewnopparat, S., Buaking, K., Puthai, W., 2002. Characterization of cellulose membranes produced by acetobacter xyllinum. Songklanakarin J. Sci. Technol. 24, 855–862.

Xu, X., Jian, Y., Xue, Y., Hou, Q., Wang, L.P., 2019. Microplastics in the wastewater treatment plants (WWTPs): occurrence and removal. Chemosphere 235, 1089–1096. https://doi.org/10.1016/j.chemosphere.2019.06.197.

Zhang, M., Yang, J., Kang, Z., Wu, X., Tang, L., Qiang, Z., Zhang, D., Pan, X., 2021. Removal of micron-scale microplastic particles from different waters with efficient tool of surface-functionalized microbubbles. J. Hazard. Mater. 404, 124095 https://doi.org/10.1016/j.jhazmat.2020.124095.

Ziajahromi, S., Neale, P.A., Rintoul, L., Leusch, F.D.L., 2017. Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics. Water Res. 112, 93–99. https://doi.org/10.1016/j.watres.2017.01.042.