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Life-Cycle Assessment of Gingko-Wood Three-Dimensional Membrane for Wastewater Treatment

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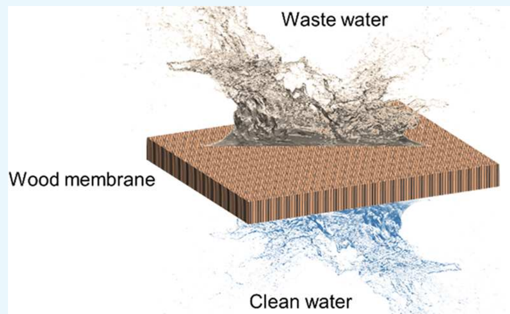
ABSTRACT: Forest is one of nature's most generous gifts to human beings, providing materials and shelters for all living beings with over 30% global land coverage. Apart from being sustainable, biodegradable, and renewable, wood is also extremely fascinating from the application aspect, with numerous advantages including hierarchical and macroporous structure, excellent mechanical performance, and versatile chemistry. The macroporous structure of wood is comprised of numerous long, partially aligned channels along the growth direction. This structure is suitable for a range of emerging applications, especially as a separation/membrane material. In this research, the potentiality of Gingko biloba (Gb) wood in the remediation of wastewater, contaminated with methylene blue (MB), a dye found in the industrial waters, was investigated. We report a macroporous, three-dimensional (3D) Gb-wood membrane decorated with palladium nanoparticles (Pd NPs) for efficient wastewater treatment. The efficiency of the Pd NPs/Gb-wood membrane to remove MB from a flowing aqueous solution was demonstrated. The wastewater treatment rate of the 3D Pd NPs/Gb-wood membrane can reach 0.5 L/min, with a high MB removal efficiency (>99.9%). The 3D Gb-wood macroporous membrane with partially aligned channels exhibits promising results for water treatment and is applicable for an even wider range of separation applications. In addition, the benefit of this 3D-wood membrane system for wastewater treatment was evaluated against the potential impacts on the environment and human health by employing the life-cycle assessment (LCA) approach. The LCA was carried out using the Gabi-education version with the gate-to-grave approach, including industrial wastewater, 3D-wood membrane, and electricity consumption using CML (Centrum VoorMilieukunde Leiden). From the LCA, it can be observed that wastewater filtration using this membrane exhibited a better environmental footprint due to the improved performance of the membrane in treating a higher volume of the permeate. Therefore, this filtration system had outweighed the additional environmental impact of the wastewater treatment process. The energy demand was identified as the main environmental hotspot in the LCA analysis. The analysis revealed that the energy source for electricity generation had a significant influence on the overall sustainability of this system. Additionally, the wood itself, a naturally abundant and eco-friendly material, presented zero environmental hazard to the environment during the filtration process. The experimental and environmental impact results indicate that Gb-wood can be employed as a natural and eco-friendly adsorbent material for the removal of waste from aqueous solutions.



INTRODUCTION

Population increase initiates rapid industrialization which was found to consequently increase the domestic wastewater and effluents into the aquatic ecosystem. Dyes are used today more than ever in industries such as textiles, printing, paper, rubber, pulp mills, cosmetics, plastics, leather tanning, food processing, etc.[1−3] In recent years, the worldwide consumption of dyes has surpassed 700 000 tons annually, and approximately 10−20% is discharged into water.[4] Most of these dyes are released into the aquatic environment, which are toxic and even carcinogenic to humans, causing serious threats to the environment in the long run.[4−6] Therefore, it is necessary to urgently treat the dye-contaminated wastewaters before they are released into the environment. A variety of technologies, including chemical oxidation, physical adsorption, photocatalytic degradation, and membrane filtration, have been implemented for wastewater treatment.[7−10] However, these water treatment methods result in additional challenges such as excessive production of solid waste and low efficiencies.[11] Therefore, there is an urgent demand for methods that are efficient, rapid, recyclable, cost-effective, and eco-friendly for removing dyes from industrial wastewater.

Wood-derived nanomaterials have been successfully prepared and are well suited to expand its application to various fields such as energy, biomedicine, and environment. Beyond biodegradability and abundance, wood does not contribute to

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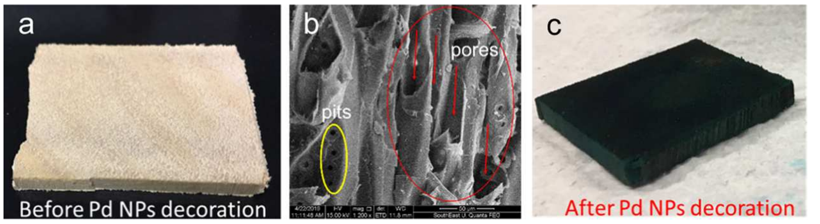

Figure 1. (a) Digital image of the Gb-wood block (3D membrane) before decorating with Pd NPs, and the piece was cut perpendicular to the growth direction, (b) scanning electron microscopy (SEM) image of the mesostructure of the wood, showing many channels aligned through the wood block's thickness, and (c) digital image of the Gb wood after the in situ decoration of the Pd NP block appeared black.

net CO_2 increase due to cyclic carbon fixation during photosynthesis and release during degradation making it a carbon-neutral material. Therefore, wood has been considered as a potential replacement for carbon-based materials to support environmental sustainability. For example, nanomaterials extracted from wood such as cellulose nanofibers and cellulose nanocrystals have attracted much attention due to their mechanical and optical properties for their potential applications. However, directly engineering natural wood can potentially open up a range of opportunities.[12] In this way, the hierarchical wood structure, the internal nanoscale nature, is preserved and utilized, yielding high efficiency without any intensive extraction process.

In this study, we use Ginkgo biloba (Gb) which has served mankind for more than 2000 years and is considered a "living fossil". Gingko wood has a macroporous structure with long and partially aligned channels, inspiring engineers and scientists to design porous materials utilizing its internal structure and mechanical properties.[13−15] It is a natural material with honeycomb-like cells, high strength/stiffness to weight ratio, and high value for hierarchical order. It is estimated that Gb wood is comprised of about 80% (pore volume) of fiber channels, making it a suitable candidate for wastewater treatment and transportation through complex pipelike channels. In addition, Gb wood is of low cost, is environmentally friendly, readily processed, and can be conveniently engineered.

Herein, we report the preparation of robust, macroporous three-dimensional (3D) wood structures for wastewater treatment. The native Gb wood is used directly as a template, decorated with palladium nanoparticles (Pd NPs) by embedding the NPs or inserting into the lumen space or into wood cell walls.[14,16,17] The Pd NPs have shown excellent catalytic efficiency, where materials are often synthesized on nanoporous supports such as silica, zeolite cages, metal oxides, and carbon to avoid agglomeration.[18] The wood 3D substrate supports the in situ formation of Pd NPs, which is composed of long, irregularly shaped wood channels with varying diameter along the growth direction. Hence, the naturally abundant and naturally developed Gb-wood membrane is more advantageous owing to its biodegradability as well as high flux rate, making it a better candidate than the fabricated ceramic and polymeric membranes for sustainable development.

Furthermore, the environmental impact of this 3D Gb-wood-based filtration process was evaluated by employing the life-cycle assessment (LCA) approach.[19] In LCA, by accumulating a set of inventories that is relevant to the inputs and outputs of the objects studied, the process evaluates the impact of a system/product starting from its raw material to production until its end of life toward the sustainable environment and human health.[20] The study takes place in four necessary steps: goal and scope definition, LC inventory analysis (LCI), LC impact assessment (LCIA) followed by interpretation.[21] The environmental impacts were evaluated using the gate-to-grave approach by CML with an endpoint indicator.

LCA has attracted tremendous attention among environmental and water-treatment-related fields and researchers because of its holistic approach. Studies have been conducted to compare different water treatment processes such as conventional treatment versus reverse osmosis, nanofiltration process, etc.[22,23] However, these comparative LCA studies between different treatment processes are still insufficient for public administrations, individuals, business, and policymakers.

Previously, most of the LCA studies were on nano-based products, while few of them were on nanotechnological production methods. To the best of our knowledge, no LCA study has been conducted to investigate the impacts of Gb-wood-based composite membranes. Therefore, this study aims at investigating the benefits of using a Gb-wood membrane to identify the most environment-friendly setup for wastewater treatment. Also, the Pd NPs/wood 3D membrane demonstrated in this study can be readily scaled up for industrial-scale applications. The effects on all of the impact categories in the filtration process stage were evaluated in detail, and further recommendations were made. To determine the most environment-friendly treatment systems based on nanotechnology, these results may be used as a reference for future wastewater treatment projects.

RESULTS AND DISCUSSION

The macroporous structure of Gb wood is unbelievably suitable for water treatment shown in Figure 1. The internal structure of Gb wood is comprised of numerous pits and well-distributed channels (Figure 1b). The pits and spiral thickenings are decorated on the inner surface of the long channels.[13,16] These characteristics are crucial in wastewater treatment for determining the kinetics when water passes through these complex channels. All of these structural characteristics of wood are well preserved in Pd NPs/wood after uniform and conformal in situ Pd NP decoration. The color change of the wood block from yellow to black/light green is due to strong light absorption by the plasmonic effect of the Pd metal NPs anchored at the surfaces of the wood channels (Figure 1c; also see the process in the Supporting Information Figure S1).[24] The Pd decoration of the 3D wood membrane was also confirmed by transmission electron microscopy (TEM), shown in the Supporting Information Figure S3. The resulting Pd NPs/wood 3D membrane exhibits a powerful ability for wastewater treatment, which is to be

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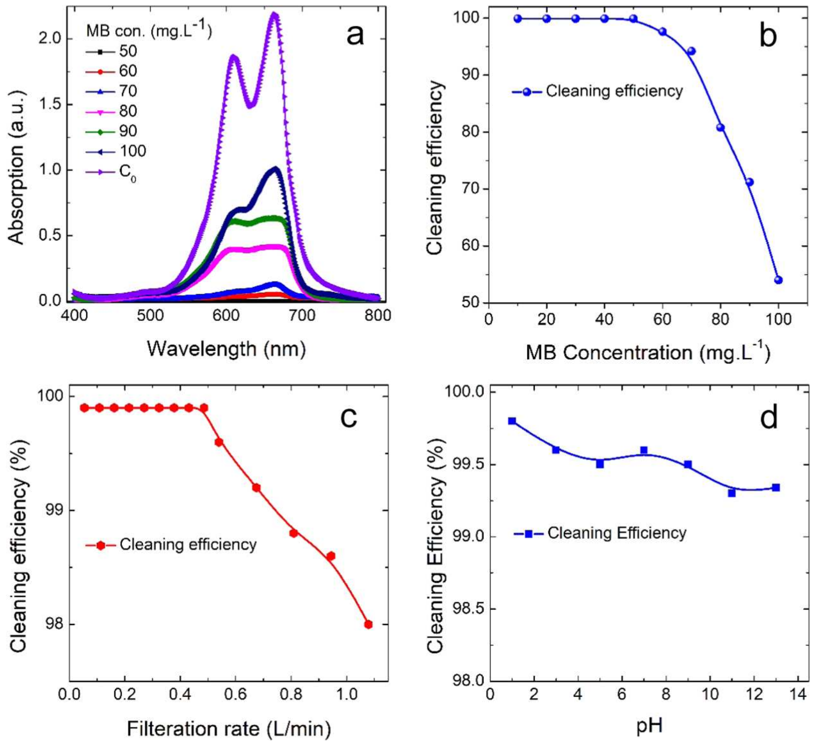

Figure 2. (a) Ultraviolet-visible absorption spectroscopy with MB concentration, (b) calculated cleaning efficiencies of the filtration system with varying MB concentration, (c) cleaning efficiency with respect to the wastewater flow rate, and (d) cleaning efficiency of the wood membrane with respect to the pH change.

demonstrated by methylene blue (MB) degradation characteristics.

In this experiment, MB containing water was modeled as wastewater. MB concentration was varied to measure the waste removal efficiency of the 3D NPs/wood membrane. As MB-contaminated water (wastewater) passes through the Pd/wood membrane, degradation of MB takes place resulting in MB-free water. The efficient MB degradation may be due to two phenomena; (1) the presence of Pd NPs throughout the wood channels serves as an efficient electron relay between the electrophilic MB and nucleophilic $NaBH_4$ for catalytic reduction following the degradation process, (2) the irregular long channels and the perforation plates within the wood membrane result in efficient contact between the MB solution and the Pd NPs/wood membrane. This enables a highly efficient reaction toward the degradation of MB. The degradation ability of the 3D membrane toward MB is quantitatively confirmed by UV−vis measurements as shown in Figure 2a. After the MB solution was treated with the wood membrane, the characteristic absorbance band for MB (664 nm) completely disappeared. It can be seen that in aqueous solution with MB content less than 50 mg L^{-1}, the removal efficiency was nearly 100% (Figure 2b). As the concentration increases, the removal efficiency gradually decreases reaching 54% for a very high concentration of 100 mg L^{-1}. From these results, significantly high efficiency is demonstrated, which is prominently higher than the previously reported works with similar filtration systems.[16] The wood membrane showed great performance for MB cleaning with an efficiency of around 99.9% as shown quantitatively in Table 1.

The Gb-wood membrane also shows a high water treatment rate while maintaining high efficiency. The efficiency rate is evaluated by passing the aqueous solution through this 3D wood membrane at various rates, shown in Figure 2c. There is a suction pump attached to the equipment at the filtrate side, and by controlling the sucking via a manually controlled pressure gauge, we can tailor the flow rate of the filtrate. The suction pump increases the vacuum/air pressure in the chamber where the filtrate is accumulated, so when the suck is low, the flow is low and vice versa. It can be observed in the video shown in the Supporting information Video 1 (a filtration rate of 0.2×10^5 Lm^{-2} h^{-1} with an MB concentration of 50 mg L^{-1}) and Video 2 (a filtration rate of 1.5×10^5 Lm^{-2} h^{-1} with an MB concentration of 50 mg L^{-1}) that the initial blue color solution became colorless after flowing through the 3D wood membrane. A high degradation efficiency of 99.9% was maintained for a flow rate of 0.5 L min^{-1} and slowly decreased to more than 98% for a very high flow rate of 1 L min^{-1}. In addition, the NPs/Gb-wood membrane maintains the high treatment efficiencies even for a wide range of pH variations (Figure 2d). The cleaning efficiency remains high (>99%) across all pH values ranging from 1 to 13, which is

Table 1. Cleaning Efficiency Calculation

MB water concentration (mg L^{-1})	C_0 at 664 nm	C at 664 nm	cleaning efficiency (%)
10		0.00014	99.9
20		0.00022	99.9
30		0.00028	99.9
40		0.00069	99.9
50	2.188	0.00138	99.9
60		0.05333	97.6
70		0.12612	94.2
80		0.42032	80.8
90		0.63055	71.2
100		1.00654	54

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essential for industrial applications. The efficient degradation of MB by the proposed 3D Gb-wood membrane may be ascribed by two factors. First, the irregular and long channels within the wood membrane result in intimate and efficient contact between the MB solution and the Pd NPs/wood membrane. Second, the presence of Pd NPs throughout the wood channels serves as an efficient electron relay between electrophilic MB and nucleophilic $NaBH_4$ for the catalytic reduction−degradation process to occur, thus enabling a highly efficient reaction toward the degradation of MB.

The promising water treatment performance of the Pd NPs/Gb-wood membrane originates from the fluid dynamics induced by the macroporous structure of the 3D wood as water passes through it. Figure 3 shows the scanning electron

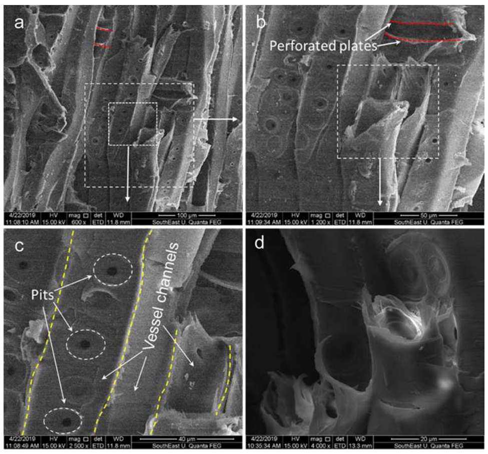

Figure 3. SEM image showing irregular and long channels in the radial section of the wood. (a−c) Some of the perforation plates and vessel channels are highlighted with red and yellow dashed lines. Vessel pits within the Gb-wood are highlighted with white circular shapes. (d) Zoom-in SEM image of the Gb-wood, showing weaved vessel channels.

microscopy (SEM) images illustrating the 3D mesostructure of Gb-wood, where long, irregular vessel channels with nonuniform diameters travel throughout the basswood. The vessel channels are connected by perforation plates, highlighted by red dashed lines in Figure 3a,b. Furthermore, water flows through the vessel pits in a more complex manner and exists inside the wood channels, highlighted by yellow dashed lines in Figure 3c. Hence, the water flow can be changed by tailoring the wood channels, e.g., the number of vessel pits and irregular channel shapes with varying diameters. The water transport through the wood channels is dictated by the vessel network in Gb-wood which is a type of hardwood.[25] In contrast, the pits connecting the tapered ends of longitudinal cells are important for water transport.[26] The vessel network in the hardwood consists of bundles of vessels that are weaved to form a 3D network that dictates transport in longitudinal and tangential directions. These perforated plates as well as porous vessel walls interconnect the individual vessels to form this vessel network. In-depth mechanism exploration involving water diffusive transport in such a complicated vessel network is beyond the scope of this study.

Environmental Analysis. As the designed membranes used in the system were not available in the Gabi-education open database, the information of the system was created and the results were compared. The functional unit for the 3D filtration membrane used was 30 × 30 × 5 mm^3, and the operational volume of the wastewater was 1 L, with an MB concentration of 50 mg L^{-1}. Therefore, all of the raw material inputs were energy consumption and wastewater. The system boundaries of the 3D wood-membrane unit are illustrated in the schematics shown in Figure 4. The activities and components outside the dotted line were not considered in the LCA. The influent water from the environment and disposal to the environment were excluded.

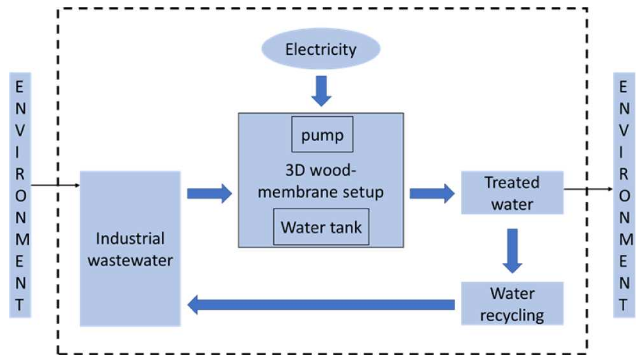

Figure 4. System boundaries of the 3D wood-membrane wastewater treatment unit for LCA analysis.

Wastewater. The wastewater sources are assumed to be the industrial wastewater in China, which is the byproduct of paper, leather, plastic, textile, and other dye industries. All of these industries release contaminated water (organic waste) with various types of contaminants including dyes like methylene orange (MO) or Rhodamine 6 G (Rh6G) and methylene blue (focused in this study). This synthetic wastewater was prepared at different concentrations (10−50 mg L^{-1}) for treatment (Table 2).

Table 2. Simplified Inventories of the System

resources/emissions	unit	amount
	Inputs	
wastewater flow	m^3	1×10^{-3}
methylene blue	mg L^{-1}	50
electricity	kW h	1
	Outputs	
treated water flow	m^3	1×10^{-3}

Operation of the Wastewater Treatment Unit. Emissions of direct substances like methane, sulfur dioxide, and carbon dioxide were calculated using the emission factors defined by Chong et al. for wastewater treatment using poly(vinylidene fluoride) (PVDF)-based membranes.[27] Our study is based on a similar approach to the neat 3D Gb-wood membrane and its comparison with the composite 3D wood-nanomaterial (palladium-incorporated Gb-wood).

Comparison of 3D Gb-Wood Membranes with and without Pd NPs. Figure 5 presents the results of CML three endpoint indicators that consist of human toxicity, acidification, and eutrophication. The 3D Gb-wood membrane decorated with Pd nanoparticles showed significantly large effects on all of the three categories compared to the neat wood membrane. The total environmental footprint of the 3D wood membrane was 740 mPt L^{-1} of contaminated water. On the other hand, the environmental footprint of the 3D wood

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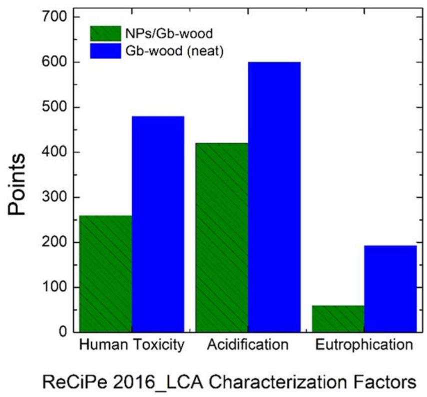

Figure 5. Endpoint impact categories of 3D Gb-wood and Pd/Gb-wood membranes using CML. *Analysis based on China mixed grid in Gabi.

membrane treated with nanoparticles has reduced by 72%, to 1273 mPt L^{-1} of contaminated water. This demonstrated that the nanoparticle incorporation into the macroporous wood membrane was more environment friendly and showed significantly encouraging results in reducing the environmental impacts. As shown in Figure 5, acidification exhibited the highest impact, followed by human toxicity and eutrophication for both of the membranes having similar trends to the other studies (Table 3).[28,29] The results could be interpreted as that the system prevented environmental pollution due to the use of fossil fuel which is the main source of energy generation.

It is assumed, in China, that electricity is produced from hard coal (bituminous and sub-bituminous).[3,30−34] Also, it is assumed that a net calorific value of 11.3 MJ kg^{-1} coal, i.e., 319 g of hard coal, was needed to produce 1 kW h of electricity.[35] These electricity-generation units contain an average sulfur content of 1.01%, resulting in approximately 15.15 kg SO_2/t coal.[32] The coal extraction, processing, burning, and waste disposal of this fossil fuel deteriorated the environment in various forms (i.e., release of toxic substances, particulate matter, and greenhouse gases). These pollutants in turn come into contact with humans directly via food intake or skin contact or indirectly via inhalation. Also, the continuous extraction of fossil fuel would definitely lead to resource depletion as coal, petroleum, and natural gas are nonrenewable energy sources on earth. The use of 100% renewable energy grids such as geothermal, hydropower, and solar photovoltaic will be able to mitigate the CO_2 emission by more than 90% (Table 4).

Table 4. Mixed Energy Grid Composition

		emission factors (kg kW h^{-1})					
energy Mix	percentage	CO_2	CO	NO_x	SO_2	N_2O	CH_4
coal	64.70	10.00	0.50	0.30	0.40	0.20	0.10
natural Gas	3.20	0.10	0.05	0.03	0.04	0.02	0.01
other-thermal	1.90	3.00	0.20	0.20	0.30	0.10	0.10
hydro	18.10						
wind	4.70						
solar	1.80						
nuclear	3.90						
pump-hydro	0.50						
biomass	1.20						

CONCLUSIONS

We demonstrated a macroporous wood membrane decorated with palladium (Pd) nanoparticle (NP)-based wastewater treatment unit, which imposed a significantly positive effect on environmental sustainability. The 3D Gingko biloba (Gb) wood membrane can function as an excellent wastewater treatment material, which is attributed to the channel structure of the wood and the synergistic effect of the evenly distributed nanosized catalyst particles. The wastewater used was an aqueous mixture of $NaBH_4$ and MB. The MB degradation efficiency reached 99.9% at a treatment flow rate up to 0.5 L min^{-1}. Additionally, we also investigated the environmental effects of this filtration process via life-cycle impact assessment (LCA) analysis. As a crucial parameter in the LCA analysis, energy consumption was found to be the main cause of environmental effects in this wood-membrane treatment unit. The analysis shows that the used Gb-wood filter for wastewater treatment presents no environmental hazardous effects. The estimated effects on human health, acidification, and eutrophication arose from the use of a mixed electricity grid, which is the main source of energy. Therefore, it is suggested that efforts should be made to eradicate the bottom cause of these effects by shifting the current electricity mix toward renewable energy such as hydropower, fuel cell, solar photovoltaic, and wind power in preventing environmental pollution while enjoying the economic benefit.

METHODOLOGY

Experimental Section. *Materials and Chemicals.* The Gb wood used in this study, having an age of 30 years, was received from Taixing Company, Jiangsu, China. Methylene blue (MB, $C_{16}H_{18}N_3ClS$), deionized (DI) water, palladium (II) chloride ($PdCl_2$), hydrochloric acid (HCl, 37%), sodium borohydride ($NaBH_4$), and sodium hydroxide (NaOH) were purchased from InnoChem (Beijing InnoChem Science & Technology Co., Ltd., China) and used as received.

Preparation of 3D Pd NPs/Wood Membrane. The Pd nanoparticle decoration of the wood block was adopted from the study by Chen et al. with some modifications.[16] An aqueous solution (1.5 mg L^{-1}) of $PdCl_2$ was prepared by adding $PdCl_2$ powder (150 mg) into 100 mL of 20 mM HCl solution, followed by 1 h heat treatment under continuous stirring until the powder was dissolved completely. After obtaining a completely uniform solution, the wood slices were immersed in it and heated overnight at 80 °C to obtain a Pd

Table 3. LCA Characterization Factors Using CML

LCA characterization factors (CML 2000)	unit	CO_2	CO	NO_x	SO_2	PM 10	N_2O	CH_4
human toxicity	g 1,4 dichlorobenzene eq.			1.2	0.096			
acidification	g SO_2 equivalent			0.5	1.2			
eutrophication	g PO_4 equivalent			0.13			0.27	

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NPs/wood 3D membrane. It is estimated that the Pd NP content was 0.19 wt %.

Water Treatment Measurements. The waste removal performance of the 3D Pd NPs/wood membrane was evaluated using the degradation of MB in an aqueous solution in the presence of $NaBH_4$ as the model reaction. Nominally, 300 mL of MB (~40 mg L^{-1}) and 30 mL of $NaBH_4$ (100 mg L^{-1}) were mixed with DI water to form an aqueous solution. The 3D Pd NPs/wood membrane was used as a filter for this MB-contaminated aqueous solution. Ultraviolet-visible (UV−vis) spectroscopy measurements were conducted before and after filtration by employing a LAMBDA 650 UV−vis spectrophotometer.[4,36] The MB concentration was monitored with an absorption peak at 664 nm, and the MB degradation efficiency was calculated as follows

$$\text{removal efficiency (\%)} = 100 \times (C_0 - C)/C_0 \quad (1)$$

where C_0 is the initial MB concentration, and C is the MB concentration after filtration. Scanning electron microscopy (SEM-Quanta FEG-250) was used to study the internal structure of the 3D wood.

Environmental Analysis. *Goal and Scope Definition.* The prime objective of this study was to compare the environmental impact of a wood-based filtration system operated with different sets of membranes: neat 3D Gb-wood membrane and Pd/Gb-wood membrane. Life-cycle impact assessment (LCA) of the designed wood membrane was used for the environmental impact analysis. Gabi-education 2018 version was utilized to classify these impacts based on the CML methodology with an endpoint gate-to-grave approach.

Inventory Analysis. Life-cycle inventory analysis (LCI) was conducted regarding mainly wastewater flow and energy consumption in the input items and treated water as the output item. The wastewater input, energy consumption, recycling involved in the construction and operation of the wastewater treatment unit were collected and categorized. Inventories of the system are shown in Table 2. Most LCA data related to individual units were taken from the Gabi database.

Impact Assessment. Impact assessment is one of the important steps in measuring the environmental impacts in LCA. CML was employed as the life-cycle impact assessment method in this study. The CML approach was used to indicate the impacts of the activities in a broad set of environmental issues such as human health, ecosystem, and resources.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b03722.

Membrane preparation process; solutions prepared with various concentrations of methylene blue; transmission electron microscopy (TEM) analysis (PDF)

Video 1. Filtration rate of 0.2 × 105 Lm^{-2} h^{-1} with an MB concentration (MP4)

Video 2. Filtration rate of 1.5 × 105 Lm^{-2} h^{-1} with an MB concentration (MP4)

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Notes

The authors declare no competing financial interest.

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REFERENCES

(1) Sokolowska-Gajda, J.; Freeman, H. S.; Reife, A. Synthetic dyes based on environmental considerations. Part 2: Iron complexes formazan dyes. *Dyes Pigm.* **1996**, *30*, 1−20.

(2) Kabdaşli, I.; Tünay, O.; Orhon, D. Wastewater control and management in a leather tanning district. *Water Sci. Technol.* **1999**, *40*, 261−267.

(3) He, Z.; Xu, S.; Shen, W.; Wang, M.; Li, C. Exploring external and internal pressures on the environmental behavior of paper enterprises in China: A qualitative study. *Bus. Strategy Environ.* **2019**, *28*, 951−969.

(4) Jin, Q.; Li, Y.; Yang, D.; Cui, J. Chitosan-derived three-dimensional porous carbon for fast removal of methylene blue from wastewater. *RSC Adv.* **2018**, *8*, 1255−1264.

(5) Cheremisinoff, P. N.; Ellerbusch, F. *Carbon Adsorption Handbook*; Ann Arbor Science Publishers, Second Printing, Michigan, Collingwood, 1980; Vol. *1054* S., 393 Abb.ISBN 0-250-40236.

(6) Pollard, S.; Fowler, G.; Sollars, C.; Perry, R. Low-cost adsorbents for waste and wastewater treatment: a review. *Sci. Total Environ.* **1992**, *116*, 31−52.

(7) Ali, I. New generation adsorbents for water treatment. *Chem. Rev.* **2012**, *112*, 5073−5091.

(8) Panizza, M.; Cerisola, G. Direct and mediated anodic oxidation of organic pollutants. *Chem. Rev.* **2009**, *109*, 6541−6569.

(9) Huang, L.; Chen, J.; Gao, T.; Zhang, M.; Li, Y.; Dai, L.; Qu, L.; Shi, G. Reduced graphene oxide membranes for ultrafast organic solvent nanofiltration. *Adv. Mater.* **2016**, *28*, 8669−8674.

(10) Thebo, K. H.; Qian, X.; Zhang, Q.; Chen, L.; Cheng, H.-M.; Ren, W. Highly stable graphene-oxide-based membranes with superior permeability. *Nat. Commun.* **2018**, *9*, No. 1486.

(11) Li, S.; Li, H.; Liu, J.; Zhang, H.; Yang, Y.; Yang, Z.; Wang, L.; Wang, B. Highly efficient degradation of organic dyes by palladium nanoparticles decorated on 2D magnetic reduced graphene oxide nanosheets. *Dalton Trans.* **2015**, *44*, 9193−9199.

(12) Zeb, M. H.; Shabbir, B.; Sagar, R. U. R.; Mahmood, N.; Chen, K.; Qasim, I.; Malik, M. I.; Yu, W.; Hossain, M. M.; Dai, Z.; et al. Superior Magnetoresistance Performance of Hybrid Graphene Foam/Metal Sulfide Nanocrystal Devices. *ACS Appl. Mater. Interfaces* **2019**, *11*, 19397−19403.

(13) Fu, Q.; Ansari, F.; Zhou, Q.; Berglund, L. A. Wood nanotechnology for strong, mesoporous, and hydrophobic biocomposites for selective separation of oil/water mixtures. *ACS Nano* **2018**, *12*, 2222−2230.

(14) Gu, Z.; Shi, X.; Omari-Siaw, E.; Zhu, Y.; Li, H.; Guo, M.; Yang, X.; Yu, J.; Xu, X. Self-microemulsifying sustained-release pellet of Ginkgo biloba extract: Preparation, in vitro drug release and pharmacokinetics study in beagle dogs. *J. Drug Delivery Sci. Technol.* **2017**, *37*, 184−193.

(15) Sagar, R. U. R.; Galluzzi, M.; Wan, C.; Shehzad, K.; Navale, S. T.; Anwar, T.; Mane, R. S.; Piao, H.-G.; Ali, A.; Stadler, F. J. Large, linear, and tunable positive magnetoresistance of mechanically stable graphene foam−toward high-performance magnetic field sensors. *ACS Appl. Mater. Interfaces* **2017**, *9*, 1891−1898.

(16) Chen, F.; Gong, A. S.; Zhu, M.; Chen, G.; Lacey, S. D.; Jiang, F.; Li, Y.; Wang, Y.; Dai, J.; Yao, Y.; et al. Mesoporous, three-dimensional wood membrane decorated with nanoparticles for highly efficient water treatment. *ACS Nano* **2017**, *11*, 4275−4282.

(17) Zhou, L.; Zhuang, S.; He, C.; Tan, Y.; Wang, Z.; Zhu, J. Self-assembled spectrum selective plasmonic absorbers with tunable bandwidth for solar energy conversion. *Nano Energy* **2017**, *32*, 195−200.

(18) Rehman Sagar, R. U.; Shehzad, K.; Ali, A.; Stadler, F. J.; Khan, Q.; Zhao, J.; Wang, X.; Zhang, M. Defect-induced, temperature-independent, tunable magnetoresistance of partially fluorinated graphene foam. *Carbon* **2019**, *143*, 179−188.

(19) Lemos, D.; Dias, A. C.; Gabarrell, X.; Arroja, L. Environmental assessment of an urban water system. *J. Cleaner Prod.* **2013**, *54*, 157−165.

(20) Rodríguez, R.; Espada, J.; Pariente, M.; Melero, J.; Martínez, F.; Molina, R. Comparative life cycle assessment (LCA) study of heterogeneous and homogenous Fenton processes for the treatment of pharmaceutical wastewater. *J. Cleaner Prod.* **2016**, *124*, 21−29.

(21) Jacquemin, L.; Pontalier, P.-Y.; Sablayrolles, C. Life cycle assessment (LCA) applied to the process industry: a review. *Int. J. Life Cycle Assess.* **2012**, *17*, 1028−1041.

(22) Beavis, P.; Lundie, S. Integrated environmental assessment of tertiary and residuals treatment-LCA in the wastewater industry. *Water Sci. Technol.* **2003**, *47*, 109−116.

(23) Van der Bruggen, B.; Vandecasteele, C. Removal of pollutants from surface water and groundwater by nanofiltration: overview of possible applications in the drinking water industry. *Environ. Pollut.* **2003**, *122*, 435−445.

(24) Fu, G.; Tao, L.; Zhang, M.; Chen, Y.; Tang, Y.; Lin, J.; Lu, T. One-pot, water-based and high-yield synthesis of tetrahedral palladium nanocrystal decorated graphene. *Nanoscale* **2013**, *5*, 8007−8014.

(25) Hass, P.; Wittel, F. K.; Mendoza, M.; Herrmann, H. J.; Niemz, P. Adhesive penetration in beech wood: experiments. *Wood Sci. Technol.* **2012**, *46*, 243−256.

(26) Plumb, O.; Spolek, G.; Olmstead, B. Heat and mass transfer in wood during drying. *Int. J. Heat Mass Transfer* **1985**, *28*, 1669−1678.

(27) Chong, W. C.; Chung, Y. T.; Teow, Y. H.; Zain, M. M.; Mahmoudi, E.; Mohammad, A. W. Environmental impact of nanomaterials in composite membranes: Life cycle assessment of algal membrane photoreactor using polyvinylidene fluoride−composite membrane. *J. Cleaner Prod.* **2018**, *202*, 591−600.

(28) Pretel, R.; Robles, A.; Ruano, M.; Seco, A.; Ferrer, J. Environmental impact of submerged anaerobic MBR (SAnMBR) technology used to treat urban wastewater at different temperatures. *Bioresour. Technol.* **2013**, *149*, 532−540.

(29) Ioannou-Ttofa, L.; Foteinis, S.; Chatzisymeon, E.; Fatta-Kassinos, D. The environmental footprint of a membrane bioreactor treatment process through life cycle analysis. *Sci. Total Environ.* **2016**, *568*, 306−318.

(30) Foley, S.; Crowley, C.; Smaihi, M.; Bonfils, C.; Erlanger, B. F.; Seta, P.; Larroque, C. Cellular localisation of a water-soluble fullerene derivative. *Biochem. Biophys. Res. Commun.* **2002**, *294*, 116−119.

(31) Wang, H.; Wick, R. L.; Xing, B. Toxicity of nanoparticulate and bulk ZnO, Al2O3 and TiO2 to the nematode Caenorhabditis elegans. *Environ. Pollut.* **2009**, *157*, 1171−1177.

(32) Bai, W.; Zhang, Z.; Tian, W.; He, X.; Ma, Y.; Zhao, Y.; Chai, Z. Toxicity of zinc oxide nanoparticles to zebrafish embryo: a physicochemical study of toxicity mechanism. *J. Nanopart. Res.* **2010**, *12*, 1645−1654.

(33) Li, Y.; Luo, X.; Huang, X.; Wang, D.; Zhang, W. Life Cycle Assessment of a municipal wastewater treatment plant: a case study in Suzhou, China. *J. Cleaner Prod.* **2013**, *57*, 221−227.

(34) Xu, S.-C.; Gao, C.; Miao, Y.-M.; Shen, W.-X.; Long, R.-Y.; Chen, H.; Zhao, B.; Wang, S.-X. Calculation and decomposition of China's embodied air pollutants in Sino-US trade. *J. Cleaner Prod.* **2019**, *209*, 978−994.

(35) Tian, F.; Cui, D.; Schwarz, H.; Estrada, G. G.; Kobayashi, H. Cytotoxicity of single-wall carbon nanotubes on human fibroblasts. *Toxicol. In Vitro* **2006**, *20*, 1202−1212.

(36) Almaamary, E. A. S.; Abdullah, S. R. S.; Hasan, H.; Rahim, R. A. A.; Idris, M. Treatment of methylene blue in wastewater using Scirpus grossus. *Malays. J. Anal. Sci.* **2017**, *21*, 182−187.

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