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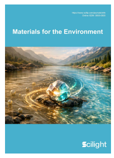

Review

Green Materials for Biological Wastewater Treatment: Roles, Applications, and Sustainability Challenges

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Abstract: Wastewater is produced around the world in amounts that need treatment methods which are both environmentally friendly and affordable. Despite the progress made in existing reviews, comprehensive assessments of biodegradable, non-toxic, widely available, and biocompatible materials for biological wastewater treatment are still lacking. This review focuses on the roles of green materials, such as bio-based, natural, and waste-derived materials, as support media, stimulants, and adsorbents in biological wastewater treatment. It also examines their contributions to pollutant removal efficiency and overall treatment performance. These materials have high porosity, many functional groups, and good surface properties (e.g., specific surface area of biochar, up to 350 m^2/g). They can improve microbial or enzymatic immobilization, boost biological activity, and enhance pollutant adsorption and biodegradation. The review also points out the combined benefits of microalgae-bacteria groups for better pollutant removal, reaching approximately 100% removal of sulfamethoxazole. A significant part of this review is its overall assessment of the performance, reusability, and sustainability of various green materials across different biological treatment systems. While some materials, like alginate-based carriers and natural carriers, show good reusability (e.g., reusing light expanded clay aggregate for 19 cycles), others have issues related to stability, saturation, biomass detachment, or possible toxic risks. Overall, green materials have shown great promise for improving the removal of nutrients and micropollutants in biological treatment processes. However, more research is needed to improve the durability, scalability, cost effectiveness, and long-term sustainability of green materials for large-scale applications in pollutant removal and resource recovery. Further studies are also needed to evaluate their economic and environmental sustainability.

Keywords: green materials; biological wastewater treatment; immobilization; stimulation; biodegradation; reusability

1. Introduction

The production of wastewater is expected to rise by at least 50%, reaching about 497 billion m^3 per year by 2030 compared to 2013 levels [1]. However, only around 52% of wastewater is treated globally. The amount of safely treated wastewater varies widely among countries with different income levels: 74% in high-income countries, 26% in lower-middle-income countries, and just 4.3% in low-income countries [2]. As a result, a large volume of wastewater continues to be released into the environment without adequate treatment, which poses significant risks to ecosystems and public health. This has led to stricter regulations and standards for wastewater management. These challenges also highlight the need for wastewater treatment technologies that are efficient, affordable, and environmentally friendly. The wastewater reuse market is projected to grow from $18.4 billion in 2025 to $44.1 billion by 2034, with an annual growth rate of 10.2% [3]. Among the available technologies, biological wastewater treatment is gaining more attention due to its cost-effectiveness, ease of operation, and lower environmental impact [4]. Biological methods are estimated to account for 18% of the water reuse and recycling market in 2025. This share is close to that of physical methods at 20%, advanced hybrid systems combining biological and membrane technologies at 15%, and advanced treatment for final treatment and pathogen removal at 15%. Membrane filtration is expected to account for the largest market share at 32% [3].

Existing reviews have mainly looked at aerobic and anaerobic treatment processes [5,6], biological treatment technologies for specific types of wastewaters such as saline organic wastewater [7] and textile wastewater [8], and the application of artificial intelligence and machine learning in system modeling and optimization [9,10]. Some latest studies also explore the applications of biofilm in wastewater treatment. More specifically, Liu et al. [11] reviewed the formation of microalgal biofilms and examined the factors affecting microalgal adhesion and pollutant removal during wastewater treatment. The biofilm-mediated remediation method was also discussed by Saini et al. [12], with the focus on various types of biofilms (e.g., bacterial biofilms, algal biofilms, fungi biofilms, symbiotic biofilms), their applications in bioreactors for wastewater treatment and in the removal of pollutants, and factors affecting biofilm formation. However, biofilm carriers are out of scope of these articles. A recent review article conducted by Lago et al. [13] presented general mechanisms about cell adhesion and biofilm formation, and some aspects of various biofilm carriers, including commercial supports, synthetic supports, and sustainable carriers (i.e., natural resources, waste-derived materials, surface modification), mainly highlighting their physicochemical properties responsible for microbial adhesion, characteristics of biofilm, and pollutant removal abilities. Table 1 shows that these review papers have covered a wide range of biofilm carriers. However, they mainly focused on the properties of carriers related to microbial adhesion and biofilm development. Less attention has been paid to the sustainability and functional integration of the supporting materials. In addition, these reviews did not provide detailed and systematic explanations of the removal mechanisms of biofilm carriers in wastewater treatment. A comprehensive evaluation of green materials as bio-support media in biological wastewater treatment systems is also lacking. In particular, their key functional roles in wastewater treatment and their implications for the coupled adsorption and biodegradation process have not been thoroughly evaluated.

Studies are scattered across different material types, immobilization methods, and treatment applications. This makes it difficult to develop a clear understanding of how material properties influence microbial activity, pollutant removal mechanisms, environmental compatibility, and long-term sustainability. Integrating material functionality with sustainability is important for modern wastewater treatment. The environmental benefits of biological processes may be limited by the use of non-renewable, expensive, or environmentally harmful support materials. Green materials from renewable or waste-based sources offer several advantages, including biodegradability, low toxicity, wide availability, and compatibility with microbial and enzymatic immobilization. Besides improving pollutant removal, these materials also support the principles of the circular economy, resource recovery, and reduced environmental impacts. These advantages are becoming increasingly important for large scale wastewater treatment, especially in resource limited areas. Therefore, a focused review of green materials for biological wastewater treatment is timely and necessary.

This review provides an overview of recent developments in green materials used for microorganism or enzyme immobilization, pollutant adsorption, and the provision of stimulants for microorganisms. It also helps researchers better understand the effectiveness of these materials in biological wastewater treatment. Specifically, this review classifies green materials into three categories according to their feedstocks, including biological sources, natural sources, and biowastes. It explains how these materials contribute to pollutant removal when combined with immobilized microorganisms or enzymes, with a focus on their roles as support media, adsorbents, and stimulants, as well as the biodegradation process. This review also summarizes their typical applications and discusses their sustainability, current challenges, and future research directions. Figure 1 shows the scope and framework of this review.

Table 1. Comparison between previous reviews on biofilm-based wastewater treatment and this review.

Review Topics	Materials Covered [a]	Immobilization Focus	References
• General formation process and technical advantages of microalgal biofilms • Mechanism and kinetic prediction model of microalgal adhesion • Factors affecting microalgal adhesion • Advanced technologies for microalgal adhesion	• Poplar sawdust • Glass • PVC plastic sheet • Polyethylene • Polyurethane foam • Polyethylene filled wall • Polyester fibre flannel • Geotextile	Physio-chemical properties of biofilm carriers, especially the water contact angle and zeta potential, influencing the adhesion of microalgal cells	[11]

Table 1. *Cont.*

Review Topics	Materials Covered [a]	Immobilization Focus	References
• Different types of biofilms • Applications of biofilms in bioreactors for wastewater treatment and in the removal of pollutants • Factors affecting biofilm formation	• GAC • Sponge • Cord media • Plastic media • Polyethylene • RBC media • Silica-based compounds • Zeolite • Ceramsite	Physio-chemical properties of biofilm carriers, e.g., surface area, construction materials, etc., influencing the growth and adhesion of microbial cells	[12]
• Cell adhesion mechanisms and biofilm development • Commercial and synthetic supports, and sustainable carriers, and their properties responsible for biofilm formation and development • Applications of different carriers in biofilm reactors for wastewater treatment	• Polyvinyl alcohol carriers • Polyurethane carriers • Polyethylene carriers • Polypropylene carriers • Biofilm carriers derived from inorganic materials, organic materials, forest wastes, agro-food wastes, industrial/urban wastes, and mixed wastes	Physico-chemical properties of biofilm carriers, e.g., porosity, surface area, roughness, hydrophilicity, functional groups, etc., influencing for biofilm formation and development	[13]
Green materials • Types, properties and functional roles of green materials for biological treatment • Mechanisms about removal of varied pollutants when integrating green materials into biological wastewater treatment • Discussions on reusability and stability of green materials • Representative applications of green materials in different systems	• Bio-based materials, including SA or CA carriers, GG and CG carriers, and chitosan-based aerogel materials • Natural materials, such as lignocellulosic materials (e.g., wood) and LECA • Biochar that is made from biowastes (e.g., bamboo, etc.) and agricultural wastes (e.g., pig manure, peanut shells, spent mushroom substrates (SMSs), etc.) • Agricultural wastes, such as cashew apple bagasse, sugarcane bagasse, dried *Luffa* fruits (*Luffa cylindrica*), corncobs, and coir, etc.	Contributions of physicochemical properties of green materials to their functional roles as the support media, stimulants and adsorbents during biological wastewater treatment	This review

[a] CG, carrageenan; GAC, granular activated carbon; GG, guar gum; LECA, light expanded clay aggregate; RBC, rotating biological contactor; SMSs, spent mushroom substrates.

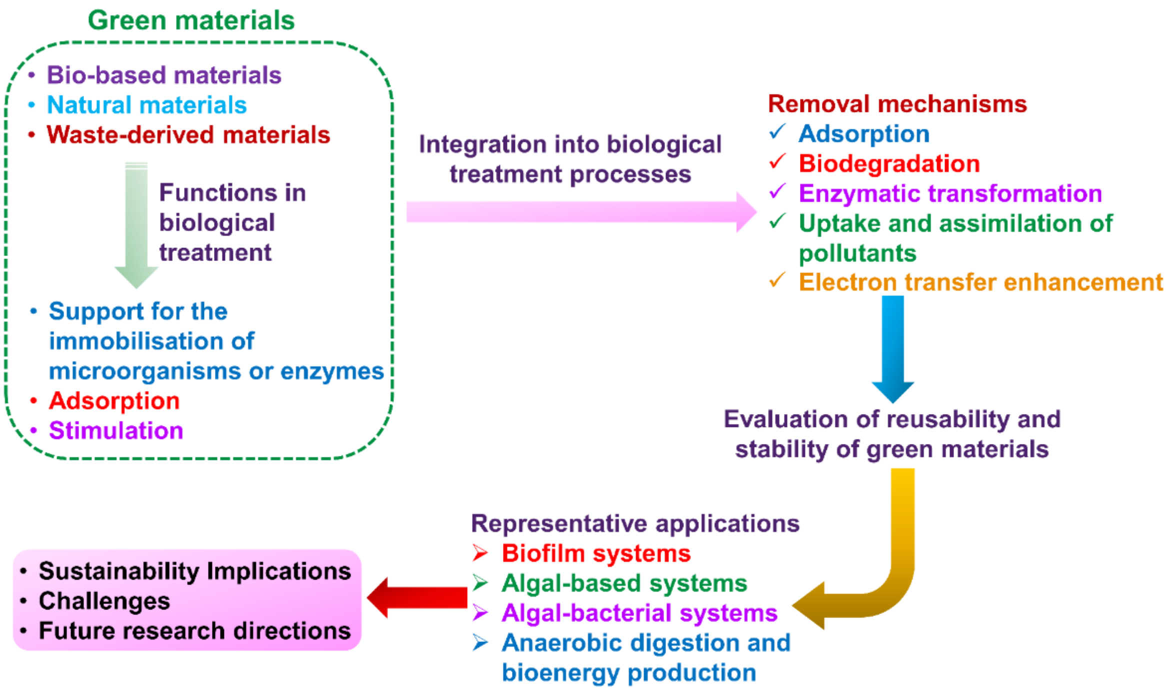

Figure 1. The scope and framework of this review.

2. Green Materials for Biological Treatment: Types and Functions

Green materials for biological treatment fall into three types: bio-based, natural, and waste-derived. Bio-based materials include sodium alginate (SA) or calcium alginate (CA) carriers, guar gum (GG) and carrageenan

(CG) carriers, and chitosan-based aerogel materials. These are commonly used to immobilize microorganisms and enzymes. SA and CA carriers are the most widely used supports. They have high porosity, a rough surface, a porous network structure, high mechanical strength, good elasticity, and many functional groups, such as -OH, -COOH, and -NH_2. These features help immobilize microorganisms and enzymes, enhance interactions among microbial cells, produce more enzymes, and create a stable microbial community. They also protect microorganisms and enzymes while increasing their resistance to pollutants. Additionally, these materials support the adsorption of targeted pollutants, allowing them to diffuse into the active sites of the carriers. This process promotes direct contact between microorganisms or enzymes and the pollutants [14–25] (Figure 2A). The addition of polymers (e.g., polyvinyl alcohol (PVA), N, N-methylene bisacrylamide, polyethylene glycol, etc.) during the synthesis of SA- or CA-based carriers can further improve the characteristics of purely SA or CA beads by (1) providing lots of polymer chains on the outer and inner layers of the carriers to form a three-dimensional space network and porous structure, which improves mass transfer efficiency and mechanical strength, as well as extending their life span; (2) increasing the hydrophilicity and the number of hydroxyl groups of the carriers, which facilitates the entrapment of microbes into the molecular chain provided by polymers; (3) helping the carriers reduce their hydraulic resistance due to their similar density with that of water, which enables microbes to firmly adhere and grow on the surface of the carriers and inside the carriers under the aeration condition, and to maintain a stable and high microbial activity [16,18,22]. The complex matrix structure of SA- and CA-based carriers, as discussed above, may be the main reason why these carriers can immobilize microorganisms more stably and provide better protection than other materials. Fungal pellets, a type of bio-based material, contain a great amount of loosely structured mycelium. They have highly porous structure and network voids that aid in the growth of fungi or bacteria, as well as adsorb targeted pollutants, support mass and oxygen transfer within the materials, and secrete enzymes [26,27] (Figure 2A).

Natural materials, such as lignocellulosic materials (e.g., wood) and light expanded clay aggregate (LECA), are chosen as supports for microorganisms, especially fungi (Figure 2B). Lignocellulosic materials can provide a carbon source for the immobilized fungi and further prompt their growth, while LECA shows high porosity for immobilizing fungi [28–31]. There are two types of waste derived materials used as support media: biochar and waste materials. Biochar is commonly produced from biowastes, such as bamboo wastes, and agricultural wastes, such as pig manure, corn straw, and spent mushroom substrates (SMSs) [32–37]. Biochar has three different functions, including (1) the support for immobilizing microorganisms, especially bacteria; (2) the adsorbent since biochar possesses highly porous structure, a heterogeneous pore network, abundant functional groups (e.g., -OH, -NH, -C=O, O-C=O, etc.), folds, and a rough surface. These characteristics provide favourable microenvironments for the growth of microorganisms, protect the immobilized microorganisms, facilitate the adsorption of pollutants, increase the resistance of microorganisms to the toxicity of pollutants, enhance electron transfer between immobilized microorganisms and pollutants, and improve mass transfer. They also enhance the capabilities of microorganisms in removing pollutants, and promote the growth of selective microorganisms and enzymes, which contribute to excellent treatment performance and bioenergy production. For example, the addition of biochar increased the adsorption and dechlorination abilities of microorganisms under anaerobic conditions, enabling the complete removal of trichloroethylene (TCE). Biochar also enriched microorganisms and enzymes associated with methane production, such as hydrogenotrophic methanogens (e.g., *Methanobacterium*), syntrophic bacteria (e.g., *Syntrophobacter*), electron active genes (e.g., *PilB* and *PilC*), and methanogenic enzymes (e.g., *frhB*) [32–34,38].

It should be noted that the surface area, pore volume, and pore diameter of biochar varied greatly with the feedstock. For example, biochar derived from the herbaceous plant *Solidago canadensis* L. had a large specific surface area of about 348 m^2/g and a micropore volume of 0.17 cm^3/g, but a small pore diameter of 1.73 nm [36]. In comparison, biochar produced from corn straw had a specific surface area of no more than 1 m^2/g [34]. Sludge derived biochar also showed a relatively small specific surface area of 10 to 16 m^2/g but a large pore diameter of 13 to 17 nm [38]. Nevertheless, all of the biochars described above showed excellent pollutant removal and bioenergy production, including almost complete removal of chemical oxygen demand (COD), TCE, and pyridine, as well as methane production of 1230 mL/d. These results are discussed in the following sections. This suggests that a large specific surface area or a large pore diameter may partly contribute to the good performance of biochar; (3) the biological stimulatory substances that provide minerals and small-molecular organic matters as nutrients and carbon sources for the immobilized microorganisms, increase the generation of antioxidases, and reduce the oxidative stress inside the microorganisms [32,34,36,38] (Figure 2C).

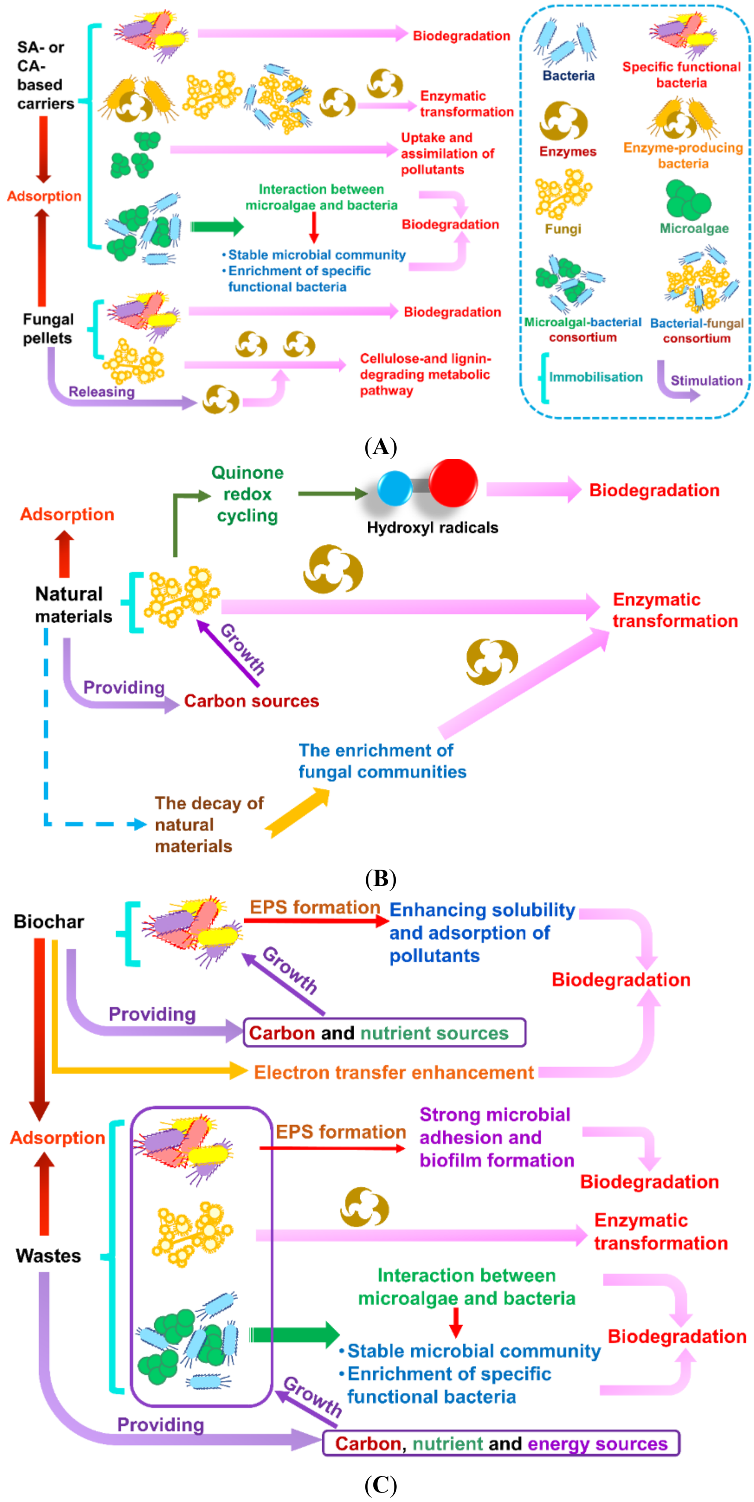

Figure 2. The functional roles of (**A**) bio-based materials (including SA- or CA-based carriers, and fungal pellets), (**B**) natural materials, and (**C**) waste-derived materials (including biochar and wastes) in biological wastewater treatment, and their corresponding pollutant removal pathways.

The most commonly used wastes for immobilizing microorganisms are agricultural wastes, such as cashew apple bagasse, sugarcane bagasse, dried *Luffa* fruits (*Luffa cylindrica*), corncobs, and coir, etc. Agricultural wastes exhibit high porosity, a fibrous network, and vascular strands, allowing them to be used as supports for immobilizing specific functional microorganisms, to adsorb targeted pollutants, and to act as the biological stimulatory substances that are consumed by immobilized microorganisms such as carbon, nutrient, and energy sources (Figure 2C). Moreover, agricultural wastes can protect microorganisms, reduce pollutant toxicity to immobilized microorganisms, and enhance their adaptability to adverse conditions (e.g., high pollutant levels) [39–42]. Table 2 compares the key differences among green materials.

Table 2. Types of green materials, and their key functional roles, major advantages and key limitations.

Types of Green Materials and Typical Examples [a]		Key Functional Roles	Representative Microorganisms	Major Advantages	Key Limitations	References
Bio-based materials	SA- or CA-based carriers	• Support for immobilizing microorganisms (i.e., bacteria, fungi, microalgae, bacterial-fungal consortium, microalgal-bacterial consortium) and/or enzymes • Adsorption	• Bacteria: *Pseudomonas* • Fungi: *Phanerochaete chrysosporium* • Microalgae: *Chlorella vulgaris* and *Chlorella* sp.	✓ Porous and three-dimensional spatial network structure, and complex matrix structure that favour the immobilization of microorganisms and their growth ✓ High strength of carriers obtained at relatively higher alginate concentration (e.g., 3.0%) ✓ Abundant functional groups ✓ Good hydrophilicity ✓ Can be reused for several times (e.g., up to 12 cycles)	× Shading effect caused by the increased microalgal cell density inside carriers, leading to a decline in nutrient removal × Not for treatment of diversified pollutants due to the breakage and dissolution of the carriers over time	[14–25]
	Fungal pellets	• Support for immobilizing fungi or bacteria • Adsorption • Stimulation	• Bacteria: *Pseudomonas* • Fungi: *Pestalotiopsis* sp.	✓ High porosity and reticular voids favouring mass and oxygen transfer, and the immobilization and growth of microorganisms ✓ Release of enzymes	× Cannot be reused × Should be frequently replaced due to their release of unexpected secondary metabolites	[26,27]
Natural materials	LECA	• Support for immobilizing fungi • Adsorption	• Fungi: *Trametes hirsuta*	✓ Low density, no toxicity, and low cost ✓ Resistance to chemical compounds ✓ Meso and macro-porosity favouring fungal immobilization ✓ Can be recycled for many times (e.g., up to 19 cycles)	× The erosion of LECA due to collision among LECA particles, especially during shaking experiments	[28,31]
	Lignocellulosic materials (i.e., wood)	• Support for immobilizing fungi • Adsorption • Stimulation	• Fungi: *Trametes versicolor*	✓ Low cost ✓ Resistance to the environmental disturbance and toxic pollutants ✓ Providing carbon source for the immobilized fungi	× The decay of wood during the long-term operation	[29,30]
Waste-derived materials	Biochar	• Support for immobilizing bacteria • Adsorption • Stimulation	• Bacteria: *Bacillus halotolerans*, and specific functional bacteria for biodegradation of targeted pollutants	✓ Large and small pores favouring the immobilization of bacteria ✓ Porous structure, large surface area, abundant functional groups, and aliphatic or aromatic carbon structure for pollutant adsorption ✓ Can be reused for several times (e.g., 3 cycles)	× Limited cycle times due to saturation of the biochar surface, and/or death or detachment of immobilized cells × Residual pollutants left on the biochar surface	[32–38]
	Wastes	• Support for immobilizing bacteria, fungi, and microalgal-bacterial consortium • Adsorption • Stimulation	• Bacteria: Specific functional bacteria for biodegradation of targeted pollutants • Fungi: *Candida tropicalis*, *Trametes versicolor, Aspergillus niger* and *Aspergillus tubingenesis* • Microalgae: *Chlorella* sp. and *Scenedesmus* sp.	✓ Low cost, abundant resources, and no toxicity ✓ High porosity and fibrous network for immobilization and growth of microorganisms, and for mass transfer ✓ Helping microorganisms resist the hydrodynamic shearing forces ✓ Can be reused for several times	× Limited cycle times when treating pollutants with high concentrations due to high toxicity	[39–44]

[a] CA, calcium alginate; LECA, light expanded clay aggregate; SA, sodium alginate.

3. Integration of Green Materials into Biological Treatment Processes

The integration of green materials into biological treatment processes includes a combined adsorption and biodegradation process. Pollutants are adsorbed onto green materials mainly through electrostatic interactions, hydrogen bonding, pore filling, extracellular complexation, and π-π interactions [14,17,24,40]. Biodegradation serves as the primary removal method, especially during long-term experiments. For bio-based carriers, such as SA- and CA-based carriers, biodegradation occurs after pollutant adsorption reaches equilibrium. In addition, adsorption occurs rapidly and at a high rate when the initial pollutant concentration is relatively high, such as trinitrotoluene (TNT) at 60 mg/L, Reactive Blue 19 at 100 mg/L, and phenol at 50 mg/L [20,22,45]. In contrast, biodegradation is the main mechanism for pollutant removal when natural materials, such as LECA, or agricultural wastes with limited adsorption capacity are used. Examples include decolourisation of 44 to 55% for LECA alone compared with 90 to 95% for LECA with immobilized fungi, and an adsorption capacity of no more than 6.41 mg/g for sugarcane bagasse [31,39]. For biochar, pollutant removal mainly relies on the continuous cycle of coupled adsorption and biodegradation. In this process, biodegradation is followed by the re-establishment of adsorption equilibrium [35,36].

Specific immobilized bacteria on the surface of and inside green materials (e.g., SA-based carriers, fungal pellets, *Luffa* cubes, and biochar) are key for breaking down a variety of pollutants [16,18,27,42]. More specifically, the microbes immobilized onto the SA-based carriers promoted the removal of COD and NH_4^+-N. This was attributed to the distribution of aerobic microbes on the surface of the carriers (aerobic microenvironment), and the anaerobes and hypoxic bacteria inside the carriers (anoxic or anaerobic microenvironment) due to the diffusion of dissolved oxygen into the carriers [18]. Other studies also employed SA-based carriers for immobilizing a TNT-degrading bacterium for TNT breakdown [20], and a laccase enzyme-producing bacterial strain for the elimination of biochemical oxygen demand, COD, total Kjeldahl nitrogen (TKN), total organic carbon (TOC), phenol, lignin, and color [16]. A great number of denitrifying bacteria could be successfully attached onto the fungal pellets, thereby ensuring highly effective removal of NO_3^--N [27]. When employing *Luffa* cubes for the immobilization of microorganisms, the bacterial consortium collected near the pharmaceutical industry gradually got used to the *Luffa* cubes over time, and secreted extracellular polymeric substances (EPS) that favoured the strong attachment of microbes and formation of biofilm. Additionally, the immobilized bacterial consortium released a series of enzymes that helped biodegrade phenol compounds [42]. Biochar encouraged the immobilization and the growth of the dense, diversely distributed and rod-shaped microbes inside the pores. Moreover, the biochar with a larger mesopore volume (e.g., bamboo-derived biochar (BC) with 0.13 m^3/g) facilitated the immobilization of a larger number of microorganisms than the biochar with a smaller mesopore volume (e.g., the wood-derived biochar (WC) with 0.002 m^3/g). The growth of immobilized microorganisms promoted the release of EPS, which increased the solubility of the target pollutants. This enhanced pollutant adsorption onto the biochar and biodegradation by the immobilized microorganisms. Therefore, the immobilized bacteria on biochar can improve the removal of targeted pollutants and shorten the operational duration [35]. For example, the bamboo-derived biochar with immobilized nonylphenol (NP)-degrading bacteria ensured the excellent cumulative removal rate of NP (93.95%) after 24 days of operation [35]. It was also found that biochar derived from corn straw reduced the stagnation phase of microbes for biodegradation of TCE by approximately 10 days [34].

The most commonly used materials for immobilizing fungi are SA- and CA-based carriers, and the immobilized fungi can effectively remove various emerging pollutants, such as dyes, bisphenol A (BPA), phenol compounds (i.e., diclofenac, and pentachlorophenol), 2,4-dichlorophenol (2,4-DCP), and pesticide mixtures (i.e., atrazine, iprodione, and chlorpyrifos). This can be attributed to the fact that the immobilized fungi secrete enzymes involved in the biodegradation process, mainly lignin-degrading enzymes, such as laccase, lignin peroxidase, manganese peroxidase, and manganese-independent peroxidase, etc. [17,23,45–47]. It should be noted that the concentrations of heavy metals have a considerable impact on the removal of targeted pollutants by the immobilized fungi. For example, when treating wastewater including Cd(II) and 2,4-DCP, Cd(II) at high concentration (>20 mg/L) limited the growth and biological activity of the fungi, thereby reducing the activities of extracellular enzymes released from fungi, which in turn impaired the biodegradation of 2,4-DCP. When Cd(II) level decreased to less than 20 mg/L, 2,4-DCP concentration at low concentration (≤40 mg/L) could be used as carbon and energy source for the immobilized fungi, enhancing biological activity and improving secretion of extracellular proteins. These proteins were employed not only for the immobilized fungi as nitrogen source, but also for the adsorption of Cd(II) via extracellular complexation [17]. The high decolorization of textile industry effluent by the bacterial-fungal consortium on CA-based carriers has also been found (>95%) due to their release of diversified enzymes, e.g., laccase, riboflavin reductase, tyrosinase, etc. [48]. It was also found that when treating

Reactive Blue 19, the enzyme (e.g., laccase) immobilized onto SA-based carriers showed higher enzymatic activity (6.88 times) than the free laccase [22].

What is noticeable is that when using the fungal pellets (i.e., *Penicillium janthinellum* P1) for immobilizing fungi (i.e., *Pestalotiopsis* sp. J63), biodegradation of complex emerging pollutants (e.g., insoluble fine fibres in paper mill effluent) was realised by cellulose- and lignin-degrading metabolic pathways [26]. The immobilized fungi on natural materials (e.g., wood, LECA) stimulate quinone redox cycling that prompts the production of hydroxyl radicals which can attack targeted pollutants (e.g., dyes, pharmaceutical active compounds) for biodegradation. Additionally, the decay of natural materials over time also promotes the enrichment of fungal communities, which contributes to the biodegradation process. One example is the presence of the wood decaying fungal genus *Coniochaeta* as the wood-decaying fungi [29]. Besides, the immobilized fungi can also produce biosurfactants that simultaneously enhance the solubility of oil during the emulsification process and decrease the hydrophobicity of the fungal cell surface, thereby increasing the biodegradation rate of oil (up to 64%) [49].

The immobilized microalgae on green materials, mainly SA- and CA-based carriers, remove nitrogen (i.e., NH_4^+-N, NO_3^--N, NO_2^--N) and phosphorus (mainly PO_4^{3-}-P) via uptake and assimilation processes [50,51]. Additionally, the synergistic interaction between microalgae and bacteria also assists nutrient removal, e.g., oxygen synthesised by microalgae for the nitrification process with bacteria, anoxic or anaerobic conditions created by microalgae during the light/dark period for the denitrification process with bacteria, uptake and release of PO_4^{3-}-P by bacteria for subsequent uptake by microalgae, and assimilation of phosphorus by denitrifying phosphate accumulating organisms under anoxic conditions, etc. [52,53]. It should be noted that during long-term experiments (e.g., 30 days of operation), phosphorus removal declines owing to the saturation of the uptake capacity of the microalgae [51]. Although microalgae and bacteria use organic matter for their growth [54], the TOC level in the treated water is still higher than that in the raw wastewater. This can be attributed to the fact that (1) inorganic carbon (IC, e.g., CO_2) is used by microalgae for the synthesis of organic matter [14,51]; (2) the death of microalgal cells and/or dissolution of carriers over time release organic matter [50]. When treating emerging pollutants, the green materials (e.g., SA-based carriers, CA-based carriers) reduce the direct contact of the immobilized microalgae with the pollutants (e.g., carbamazepine (CBZ), 17β-estradiol), and prolong the penetration of pollutants into the carriers, thereby creating a microenvironment containing lower concentrations of pollutants around the microalgal cells in the materials than that in the suspended solution. This protects the antioxidant enzyme system, limits the cell damage, reduces excessive generation of reactive oxygen species, and maintains the photosynthetic capacity and cell activity of microalgae, thereby enhancing the removal of pollutants, e.g., CBZ removal of 84% after immobilization vs. 67% before immobilization [55,56]. The interaction between microalgae and bacteria helps the formation of a stable microbial community and the enrichment of specific functional bacteria on green materials for the biodegradation of pollutants (e.g., sulfamethoxazole (SMX)) [25]. Under harsh conditions (e.g., acidic pH), the photosynthetic process induced by the immobilized microalgae increases pH inside the carriers for the effective removal of emerging pollutants. Moreover, the metabolites released from bacteria are used as nutrients for the growth of microalgae. This leads to a considerable increase in the removal performance, e.g., 40.57% removal of atrazine using an immobilized microalgal-bacterial consortium vs. 4.35% removal with the free consortium [19]. Agricultural wastes are also used as the support for microalgal-bacterial consortium. Compared to the free microalgal-bacterial consortium, the immobilized microalgal-bacterial consortium on agricultural wastes (i.e., the matured dried fruit of *Luffa cylindrica*) demonstrated the enhanced removal performance of pesticides due to the symbiotic interaction of microalgae and bacteria, e.g., bromacil, atrazine, diuron, and bentazon removals of 89%, 76%, 77%, and 63% (average) for the immobilized consortium vs. 79%, 51%, 58%, and 42% (average) for the free consortium. However, the wastes limited the removal of antibiotics as they prohibited the light penetration [41].

4. Reusability and Stability

The reusability and long-term stability of immobilized systems depend largely on material properties, microbial and enzyme activity, and pollutant concentration and toxicity. Table 3 summarizes the key characteristics of the green materials discussed in this review, together with their reusability and Technology Readiness Levels (TRLs). TRLs were first introduced by NASA in the 1970s and are now widely used to assess the maturity of a technology. They support risk management, improve communication among innovators, stakeholders, and investors, and help align funding with technology development. Therefore, TRLs provide a useful framework for narrowing the gap between scientific concepts and commercial applications [57].

Table 3. The reusability of green materials and their Technology Readiness Levels (TRLs).

Green Materials [a]	Surface Area *	Pore Volume *	Regeneration Method *	Reusability Cycles [b]	Pollutant Removal at the Last Cycle [c]	Cost *	TRL [d]	References
SA-based carriers	-	-	• Decanting the medium containing 100 mg/L TNT in the previous cycle; • Washing carriers with sterile water; • Transferring carriers into a fresh medium	12 (8 h/cycle)	99.5% of TNT removal	-	TRL 3	[20]
	-	-	• Withdrawing the carriers from the medium in the previous cycle; • Washing the carriers with de-ionised water; • Reusing the carriers in a fresh medium	6	Maintaining 90% of initial activity	-	TRL 3	[22]
CA-based carriers	-	-	• Separating carriers with immobilized microalgae from effluent containing activated sludge; • Harvesting activated sludge by centrifugation; • Washing carriers with the sterile sodium chloride solution; • Adding carriers and activated sludge into a fresh medium	3 (48 h/cycle)	~100% of NH_4^+-N and PO_4^{3-}-P removal	-	TRL 4	[54]
	-	-	-	9 (12 h/cycle)	100% of BPA removal	-	TRL 3	[23]
	-	-	-	5 (18 h/cycle)	89% of phenol removal	-	TRL 3	[45]
	-	-	**During the first two cycles** • Washing carriers two times with sterile distilled water; • Transferring the carriers into the fresh medium; **After the second cycle** • Washing the carriers with sterile distilled water; • Storing the carriers in sterile calcium chloride at 4 °C for three weeks; • Recovering the carriers by filtration and washing with sterile distilled water; • Adding the carriers into the fresh medium	3 (96 h/cycle)	68–75% of decolourisation	-	TRL 3	[58]
	-		• Feeding the reactor containing carriers with nutrient medium after each cycle for regenerating the cellular activity	5 (168 h/cycle)	>76% of Remazol red removal	-	TRL 4	[48]
LECA	-	-	• Filtering the mixture from the previous cycle; • Reusing the solid phase after filtration in the next cycle	19 (72 h/cycle)	96% of decolourisation	-	TRL 3	[31]
Bamboo-derived biochar	247.33 m^2/g	• Average, 0.206 m^3/g; • Mesopore, 0.131 m^3/g; • Micropore, 0.055 m^3/g	• Centrifuging the tubes containing the mixture in the previous cycle to separate the aqueous and solid phases; • Washing the biochar three times with deionised water; • Reusing the biochar into a fresh medium	8 (72 h/cycle)	93.95% of NP removal	-	TRL 3	[35]
Sugarcane bagasse	1.75 m^2/g	-	-	6 (54 h/cycle)	>94% removal of phenol	-	TRL 4	[39]

* The related data were not given in the manuscript; [a] CA, calcium alginate; SA, sodium alginate; LECA, light expanded clay aggregate; [b] h, hours; [c] BPA, bisphenol A; NP, nonylphenol; TNT, trinitrotoluene; [d] TRL 3, simple tests under controlled laboratory conditions to provide experimental proof; TRL 4, performance testing of a small-scale prototype under controlled laboratory conditions.

TRLs consist of nine levels. TRL 1 represents basic research. TRL 2 refers to technology formulation. TRL 3 involves simple tests under controlled laboratory conditions to provide experimental proof. TRL 4 refers to performance testing of a small-scale prototype under controlled laboratory conditions. TRL 5 involves testing and validation of a large-scale prototype in environments that simulate real operating conditions. TRL 6 demonstrates a full-scale prototype system under near real operating conditions. TRL 7 represents a pre-commercial demonstration system. TRL 8 refers to the first commercial system, where the technology is completed, its performance is verified, and manufacturing issues are addressed. TRL 9 represents full commercial application [57].

Most of the green materials can be reused due to the fact that the pollutants adsorbed onto the green materials are effectively biodegraded by the immobilized microorganisms, which facilitates the release of some adsorption sites on the materials for further coupled adsorption-biodegradation process. The lifespan of bio-based materials (e.g., CA-based carriers) can be prolonged by the immobilized microalgae due to their capability in removing inorganic carbon and phosphorus, which limits the interaction of carbonate and phosphate ions with the matrix of the materials, thereby reducing carrier deterioration [50]. Hence, these carriers can be reutilized after several cycles, e.g., almost complete removal of NH_4^+-N and PO_4^{3-}-P after 3 cycles with the immobilized microalgal-bacterial consortium [54]. Similarly, immobilized bacteria, fungi, and enzymes onto the bio-based carriers, especially SA- or CA-based carriers, usually keep a high treatment efficiency over repeated cycles because the immobilization matrix enhances biomass retention and protects microorganisms from environmental stress, e.g., TNT removal of 99.5% with immobilized bacteria after 12 cycles [20], 90% of the initial activity after 6 cycles with immobilized laccase for the treatment of dyeing wastewater [22], and complete removal of BPA after 9 cycles when using CA-immobilized fungi [23], >76% of decolourisation for 5 cycles with immobilized bacterial-fungal consortium for treatment of textile industry effluent [49], etc. However, when used to treat atrazine containing water, the SA-based carriers broke down and even dissolved after four cycles [19]. This indicates that these carriers can be reused only for a limited range of pollutants. Fungal pellets should be frequently replaced as the treated carriers may release unexpected secondary metabolites during the subsequent treatment process [26]. Natural carriers used for fungal immobilization can achieve high durability because of the strong adaptability of fungi and the continuous growth and regeneration of fungal biomass. For example, LECA with immobilized fungal mycelia was reused for up to 19 cycles while maintaining a decolourisation efficiency of 96% [31].

The immobilization of bacteria onto biochar can improve the biochar's adsorption capacity as the biodegradation process helps release adsorption sites on the biochar for further adsorption of pollutants [35]. However, long term reuse still faces challenges, including carrier saturation, biomass detachment, and microbial inhibition caused by toxic pollutants. Pollutant accumulation in porous materials may also increase the risk of secondary pollution during repeated use [35]. In addition, high pollutant concentrations can inhibit microbial metabolism, damage biofilm integrity, and reduce EPS production. These changes ultimately decrease treatment performance. For example, when the pollutant concentration increases to a high level (e.g., 2600 mg/L phenol), the removal efficiency declines after reutilisation (e.g., only 24% removal after 6 cycles) due to the inhibited growth of the immobilized biomass and their activity, as well as cell washout caused by the toxicity of pollutants [39].

As shown in Table 3, natural materials, such as LECA, and agricultural wastes, such as sugarcane bagasse, have a longer service life than bio-based materials and biochar during reusability assessment. The service life of bio-based materials and biochar is no more than 35 days, whereas that of natural materials and agricultural wastes can reach 13.5 days. This difference may be related to their limited adsorption capacity, as discussed in Section 3. Limited adsorption reduces the need for regeneration by maintaining high biodegradation efficiency, which helps extend the service life.

The regeneration method depends mainly on the type of green material and the immobilized microorganisms. For bio-based carriers, SA based carriers are regenerated by washing with sterile or deionised water. When microalgal bacterial consortia are immobilized on CA based carriers, the carriers are first separated from activated sludge. They are then washed with or stored in sterile salt solution, followed by filtration and washing with sterile water [20,22,58]. Natural materials, such as LECA, are regenerated by simple filtration [31]. For waste derived materials, centrifugation is first used to separate the biochar from the mixture. The biochar is then washed with deionised water [35]. Future studies should concentrate on improving carrier durability, boosting microbial resilience, and fine-tuning microbial ecology to ensure stable long-term operation of immobilized biological treatment systems.

5. Representative Applications

Green materials with immobilized microorganisms or enzymes are commonly used in biofilm systems, algal-based systems, algal bacterial systems, anaerobic digestion, and bioenergy production (Table 4).

Table 4. Representative applications of green materials (lab-scale experiments) and their performance during biological wastewater treatment processes.

Representative Applications of Green Materials (Examples)			Wastewater [c]	HRT (h) [d]	Operational Duration (Days)	Advantages (✓)/Challenges (×)	References
Systems	Green Materials [a]	Configurations [b]					
Biofilm systems	SA-based carriers	SBBR	Synthetic sewage (COD, 260 ± 30 mg/L; NH_4^+-N, 22.5 ± 1.5 mg/L; C/N ratio of 9.2–11.6)	N.G.	~ 25	✓ Excellent removals of COD, NH_4^+-N and TN (> 70%) ✓ Saving the aeration time, footprint, and construction cost ✓ Reducing power consumption by 50%	[18]
	Wood	RBC	Real hospital wastewater (TOC, from 71 ± 17 mg/L to 77 ± 18 mg/L; COD, from 181 ± 40 mg/L to 195 ± 49 mg/L; NH_4^+-N, from 47 ± 6 mg/L to 49 ± 8 mg/L; TKN, from 74 ± 16 mg/L to 77 ± 16 mg/L)	24	75	✓ Excellent removals of TOC and PO_4^{3-}-P (> 73%), and of azithromycin, metronidazole, and sulfamethoxazole (> 76%) × Limited nitrogen removal (TN removal, ≤ 41%) that could be ascribed to the fact that 1) wood decaying fungi employed the wood as substrate for nitrogen conserving and recycling at nitrogen-limited conditions; 2) NH_4-N from the wastewater inhibits the use of other nitrogen sources by enzymes	[29]
		PBBR		72	50	✓ Excellent removals of ibuprofen, ketoprofen and naproxen (60–90%)	[30]
	Fungal pellets	Bioreactor	Actual sewage (C/N ratio of 1–2)	4–8	150	✓ Denitrification efficiency, 96%	[27]
	Biochar	PBBR	Diluted raw piggery wastewater (COD, 378–530 mg/L; NH_4^+-N, 393–535 mg/L; NO_2^--N, 5.85–12.01 mg/L; NO_3^--N, 31.68–54.55 mg/L)	1.67	7.5	✓ Excellent removals of NH_4^+-N and TN (73–95%) × Large variation in COD removal (54–82%)	[37]
	Agricultural wastes	PBBR	Pharmaceutical wastewater (OLR, 0.56–3.35 kg COD/m^3·d)	10–24	61	✓ Excellent removals of phenol (≥ 93%) and COD (> 96%) × Declined removal performance over time due to the high phenol concentrations and cell washout	[42]
			Real phenol-containing wastewater (phenol concentration, 1500–2400 mg/L)	0.05–0.28	18 13.5		[39]
Algal-based systems	CA-based carriers	iMBR, FBBR	Secondary effluent of wastewater treatment plants (TP, 8.9 ± 0.4 mg/L; PO_4^{3-}-P, 8.5 ± 0.4 mg/L; NO_3^--N, 5.3 ± 0.4 mg/L; NH_4^+-N, 10–20 mg/L; TN, 18.3–27.4 mg/L)	N.G.	~ 30	✓ Excellent removals of NH_4^+-N, PO_4^{3-}-P, and TP (up to 100%) × Regular replacement of the carriers due to their deterioration over time	[14,50]
Algal-bacterial systems	CA-based carriers	PBR	Anaerobically digested centrate (COD, 1168.92 ± 3.14 mg/L; TN, 55.04 ± 0.39 mg/L; TP, 27.16 ± 1.24 mg/L)	N.G.	7	✓ Excellent removals of sulfamethoxazole (99.0%), and COD, TN, and TP (> 72%)	[25]
	GG and CG-based carriers	Living biofilter	Synthetic municipal wastewater (NH_4^+-N, 67.1 mg/L; PO_4^{3-}-P, 29.9 mg/L)	N.G.	7	✓ Excellent removals of NH_4^+-N and PO_4^{3-}-P (> 86%)	[59]
Anaerobic digestion and bioenergy processes	CA-based carriers	Two-stage immobilized bioreactor consisting of AC and BC	Synthetic medium (glucose,10 g/L)	N.G.	7	✓ Maximum bioH_2 production, 9.8 mL/L in AC and 222.7 mL/L in BC × Requirement of extra carbon and nutrient sources for microalgal growth and bioenergy production	[60]
	Iron-rich biochar pyrolysed at 600 °C (specific surface area of 15.6939 m^2/g, pore diameter of 12.8384 nm)	Organic glass column-based up-flow anaerobic reactor	Synthetic wastewater with C:N:P ratio of 300:5:1	432	76	✓ Excellent COD removal efficiency (98.68%) and methane production (1230 mL/d) ✓ High enrichment of hydrogenotrophic methanogens, syntrophic bacteria, and iron-reducing bacteria favouring methane production ✓ Benefits of iron ions leached from the biochar, e.g., reduction of sludge generated during gas production, enhancement in sludge settleability, stability of anaerobic reactors, decreased requirement of additional COD caused by sludge loss × Clogging of biochar during long-term experiments × High energy consumption during the synthesis of biochar	[38]

[a] CA, calcium alginate; CG, carrageenan; GG, guar gum; SA, sodium alginate; [b] AC, algal chamber; BC, bacterial chamber; FBBR, fluidised-bed reactor; iMBR, immobilized microalgae membrane bioreactor; PBBR, Packed-bed reactor; PBR, photobioreactor; RBC, rotating biological contactors; SBBR, sequencing batch biofilm reactor; [c] COD, chemical oxygen demand; TOC, total organic carbon; TKN, total Kjeldahl nitrogen; TN, total nitrogen; TP, total phosphorus; [d] HRT, hydraulic retention time; N.G., not given.

These lab-scale systems effectively remove a variety of pollutants and recover bioenergy. However, only very few of current articles assessed the energy consumption and carbon emissions of the whole systems. A latest study conducted by Javed et al. [60] created a two-stage immobilized bioreactor to encourage biohydrogen production. This bioreactor had an algal chamber (AC, which was also the immobilized microalgal chamber) and a bacterial chamber (BC, which contained immobilized domestic wastewater activated sludge under anaerobic conditions). There was continuous flow between these two chambers, and glucose served as an external carbon source. The pH in BC dropped due to anaerobic digestion in the dark, but the flow from AC raised the pH to between 7.0 and 10.0 in BC as microalgae growth under light conditions increased the pH. This pH range promoted anaerobic activity in BC and hydrogenase activity in AC for biohydrogen production. At the same time, the oxygen produced by microalgal growth in AC moved to BC. This transfer inhibited methanogenesis and decreased methanogenic activity in BC, which supported biohydrogen production through glucose fermentation. Zhang et al. [38] developed an up flow anaerobic reactor containing iron rich biochar derived from a mixture of residual sludge from municipal wastewater and food waste treatment plants and Fenton iron sludge. The iron rich biochar had a specific surface area of 15.69 m^2/g and an average pore diameter of 12.84 nm. It significantly increased methane production compared with biochar derived only from residual sludge, which had a specific surface area of 9.94 m^2/g and an average pore diameter of 17.29 nm. This improvement may be related to the properties of biochar discussed in Section 2 and the additional functions of iron rich biochar. During oxygen limited pyrolysis, iron-rich biochar promoted the formation of magnetite on its surface. This increased the electrical conductivity of the biochar and enhanced electron transfer. It also enriched iron-reducing bacteria, such as *Desulfomicrobium*, which promoted the formation of an electroactive biofilm together with hydrogenotrophic methanogens and syntrophic bacteria. As a result, direct electron transfer and direct interspecies electron transfer were enhanced. In addition, iron-rich biochar showed stronger expression of electroactive functional genes and methanogenesis related enzymes.

At present, only a few studies have evaluated the economics of green materials in biological wastewater treatment. Based on the available information, the addition of SA gel carriers to a sequencing batch biofilm reactor (SBBR) reduced the aeration time when an intermittent aeration and instantaneous water flow strategy was used for synthetic wastewater treatment. The operating strategy included instantaneous water inflow, 2 h without aeration, 2 h with aeration, 2 h without aeration, and instantaneous water outflow after another 2 h. Under these conditions, power consumption was reduced by about 50%. The cost of the SA carriers was about ¥ 1.5×10^3 per m^3 wastewater per day, which is approximately $221.5 per m^3 wastewater per day [18]. In comparison, pig manure derived biochar cost only $0.07 per m^3 wastewater per day. When biochar was applied in a fluidized bed reactor to treat real phenol containing wastewater, the economic benefit reached up to $6826 based on the energy and gas prices in China in 2023 [32].

6. Sustainability Implications and Challenges

Green materials come from various biomaterials, such as alginate, carrageenan, guar gum, fungi, lignocellulosic materials, biowastes, and agricultural wastes. They are eco-friendly and cost-effective. These materials are excellent for immobilizing microorganisms or enzymes, and for supplying nutrients and carbon sources for microbial growth. In wastewater treatment, green materials help through adsorption and biodegradation, enzymatic transformation, and co-metabolism driven by microorganisms or enzymes, ensuring a high removal rate of both emerging and traditional pollutants. However, some toxic chemicals are used to create these green materials. These include $CaCl_2$ solution, boric acid solution, NaH_2PO_4 solution, and N, N-methylene bisacrylamide, which are needed for making SA- or CA-based carriers. Furthermore, few studies have investigated the leaching of chemicals from green materials or chemicals bound to the materials. Additionally, residual pollutants and intermediate products may remain on the green materials after biodegradation. This situation requires extra steps to separate or properly dispose of the pollutants. The reusability of green materials is limited. Their surfaces can become saturated, and the immobilized microorganisms may not tolerate highly toxic pollutants. This can reduce microbial activity and cause microbial death or detachment. Most recent studies have evaluated green materials in batch experiments or laboratory scale reactors for the treatment of synthetic wastewater or diluted raw wastewater. They commonly remove pollutants like pharmaceutical micropollutants, phenol, dyes, TOC, COD, NH_4^+-N, TN, PO_4^{3-}-P. In most of biofilm systems, green materials have been effective for more than 25 days (as shown in Table 4). However, other applications, such as algal-based systems, and algal-bacterial systems, typically operate for less than 30 days [14,25,50,59]. Only a few studies have looked at techno-economic factors and greenhouse gas emissions [18,32].

Therefore, there is a need for more research to

(1) create green materials made with less toxic chemicals that can effectively eliminate various pollutants while being reusable and enhancing bioenergy production within the circular economy framework;

(2) Develop more advanced systems by integrating green materials into anaerobic digestion-based systems to enhance bioenergy production. Future studies should also investigate microbial enrichment, microbial community succession, and the mechanisms of bioenergy production.

(3) Integrate green materials with nutrient recovery technologies, particularly algal bacterial systems, to improve resource recovery.

(4) Apply green materials in carbon neutral wastewater treatment plants and evaluate their effects on carbon neutrality, bioenergy production, electricity savings, and power consumption.

(5) Explore the effectiveness of green materials for the biological treatment of wastewater containing a wide range of emerging pollutants, such as nanoplastics, microplastics, antibiotic resistance genes, endocrine disrupting compounds, and per and polyfluoroalkyl substances.

(6) Examine the long-term application of green materials in pilot scale or full-scale systems for treating different types of real wastewater. The evaluation should include treatment performance, bioenergy production, operation and maintenance costs, the stability and reproducibility of green materials, and regulatory acceptance [14,25,50,59].

(7) Assess the economic and environmental sustainability of green materials and the overall systems using life cycle assessment (LCA) and techno economic analysis (TEA). For LCA, the system boundaries should include both the construction and operational stages. These stages should cover feedstocks used to prepare green materials, chemical inputs, energy consumption, gas emissions, biomass conversion, treated effluent, and sludge production. Data from both stages should then be collected for the life cycle inventory. Different scenarios should also be evaluated, including influent and effluent conditions, especially the concentrations of target pollutants, the capacities of system components, construction materials, and operation and maintenance parameters, such as biogas upgrading, gas emissions, electricity and energy consumption, and effluent quality. The assessment should then include key impact categories, such as global warming potential, freshwater and marine ecotoxicity, human toxicity, and ozone depletion potential, followed by uncertainty analysis. For TEA, capital expenditure should include the costs of system design and construction, together with feedstocks used to prepare green materials. Operational expenditure should include the costs of electricity, regeneration and replacement of green materials, labour, chemicals required for operation and maintenance, disposal of spent green materials, and effluent monitoring;

(8) explore the end-of-life management, resource recovery potential and recyclability of green materials in the circular economy perspective;

(9) develop artificial intelligence-based models to optimise the properties of green materials, operational conditions and system configurations for achieving excellent treatment performance, maximising bioenergy recovery and/or production, and realising carbon-neutral wastewater treatment processes.

Figure 3 presents the sustainable benefits/challenges of current biofilm carriers and green materials.

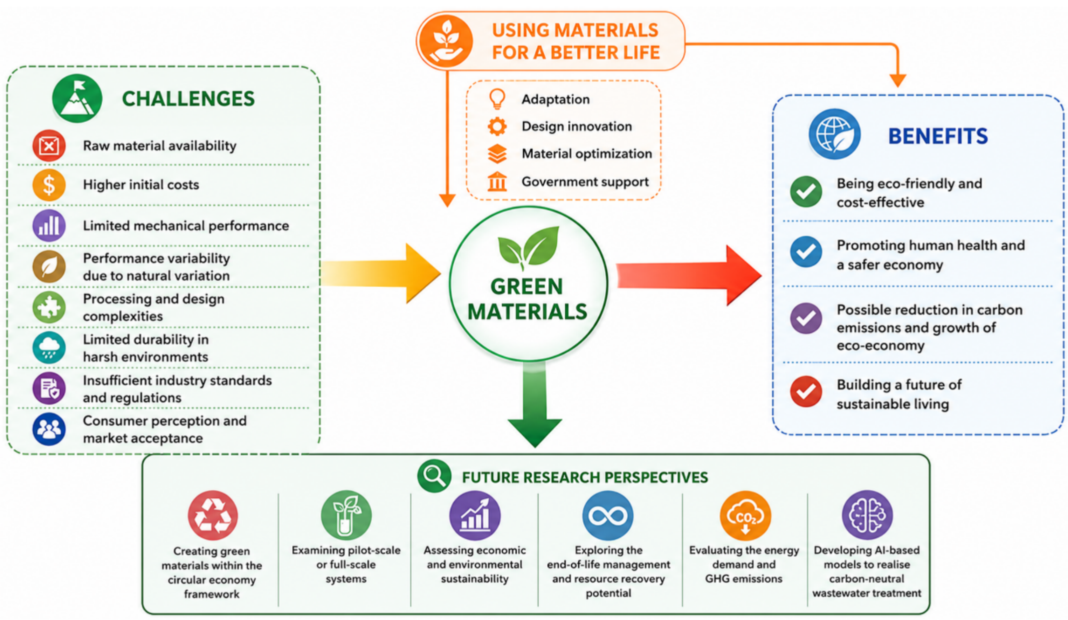

Figure 3. Sustainability benefits/challenges of current biofilm carriers and green materials.

7. Conclusions and Outlook

Natural materials and materials derived from waste can provide biological stimulants to maintain microbial activity. After adsorption reaches equilibrium, various pollutants can be effectively biodegraded by immobilized microorganisms or enzymes. Natural materials generally show better reusability, with up to 19 cycles for LECA, than biologically derived carriers, with up to 12 cycles for SA- and CA-based carriers. In contrast, fungal pellets are difficult to reuse because of their limited structural stability. Applications of green materials have shown promising results for the removal of emerging pollutants, such as almost 100% SMX removal in an algal and bacterial system, and for biogas production, such as methane production of 1230 mL/d in an up flow anaerobic reactor. Biochar has a much lower cost, at $0.07/m^3 of wastewater per day, than biologically derived materials, such as SA based carriers at $221.5/m^3 of wastewater per day.

Future research should move beyond laboratory scale studies towards large scale applications. Integrated systems should also be developed to combine the removal of a wide range of emerging pollutants with resource recovery and bioenergy production. In addition, comprehensive techno-economic analysis, life cycle assessment, and carbon footprint evaluation are needed to confirm the environmental and economic feasibility of these technologies. Future progress also depends on developing durable and reusable green materials, improving interactions between microorganisms and materials, integrating digital monitoring and control technologies, and establishing supportive regulations and commercialization pathways to accelerate large scale application in practical wastewater treatment.

Author Contributions

L.D.: data curation, formal analysis, investigation, validation, writing—original draft preparation; W.G.: data curation, formal analysis, supervision, writing—reviewing and editing; H.Z.: formal analysis, investigation, resources, writing—reviewing and editing; S.W.C.: funding acquisition, resources, writing—reviewing and editing; D.D.N.: formal analysis, validation, writing—reviewing and editing; H.H.N.: funding acquisition, conceptualization, supervision, validation, writing—reviewing and editing. All authors have read and agreed to the published version of the manuscript.

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The data supporting the findings of this study are available within this article. Additional data are available from the corresponding author upon reasonable request.

Conflicts of Interest

The authors declare no competing financial or non-financial interests.

Use of AI and AI-Assisted Technologies

During the preparation of this work, the authors used ChatGPT and Grammarly for limited language editing and clarity improvement. After using these tools, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

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