Critical Reviews in Biotechnology

ISSN: 0738-8551 (Print) 1549-7801 (Online) Journal homepage: https://www.tandfonline.com/loi/ibty20

Recent development of advanced biotechnology for wastewater treatment

Huan Yi, Minfang Li, Xiuqin Huo, Guangming Zeng, Cui Lai, Danlian Huang, Ziwen An, Lei Qin, Xigui Liu, Bisheng Li, Shiyu Liu, Yukui Fu & Mingming Zhang

To cite this article: Huan Yi, Minfang Li, Xiuqin Huo, Guangming Zeng, Cui Lai, Danlian Huang, Ziwen An, Lei Qin, Xigui Liu, Bisheng Li, Shiyu Liu, Yukui Fu & Mingming Zhang (2019): Recent development of advanced biotechnology for wastewater treatment, Critical Reviews in Biotechnology, DOI: 10.1080/07388551.2019.1682964

To link to this article: https://doi.org/10.1080/07388551.2019.1682964

Published online: 05 Nov 2019.

CRITICAL REVIEWS IN BIOTECHNOLOGY
https://doi.org/10.1080/07388551.2019.1682964

REVIEW ARTICLE

Recent development of advanced biotechnology for wastewater treatment

Huan Yi[a,b], Minfang Li[a,b], Xiuqin Huo[a,b], Guangming Zeng[a,b], Cui Lai[a,b], Danlian Huang[a,b], Ziwen An[a,b], Lei Qin[a,b], Xigui Liu[a,b], Bisheng Li[a,b], Shiyu Liu[a,b], Yukui Fu[a,b] and Mingming Zhang[a,b]

[a]College of Environmental Science and Engineering, Hunan University, Changsha, Hunan, China; [b]Key Laboratory of Environmental Biology and Pollution Control, Hunan University, Ministry of Education, Changsha, Hunan, China

ABSTRACT

The importance of highly efficient wastewater treatment is evident from aggravated water crises. With the development of green technology, wastewater treatment is required in an eco-friendly manner. Biotechnology is a promising solution to address this problem, including treatment and monitoring processes. The main directions and differences in biotreatment process are related to the surrounding environmental conditions, biological processes, and the type of microorganisms. It is significant to find suitable biotreatment methods to meet the specific requirements for practical situations. In this review, we first provide a comprehensive overview of optimized biotreatment processes for treating wastewater during different conditions. Both the advantages and disadvantages of these biotechnologies are discussed at length, along with their application scope. Then, we elaborated on recent developments of advanced biosensors (i.e. optical, electrochemical, and other biosensors) for monitoring processes. Finally, we discuss the limitations and perspectives of biological methods and biosensors applied in wastewater treatment. Overall, this review aims to project a rapid developmental path showing a broad vision of recent biotechnologies, applications, challenges, and opportunities for scholars in biotechnological fields for "green" wastewater treatment.

ARTICLE HISTORY

Received 12 June 2018
Revised 11 July 2019
Accepted 29 August 2019

KEYWORDS

Biotreatment; immobilized biomass reactor; advanced oxidation process; integrated biotechnology; biosensor; wastewater treatment

Introduction

Highly efficient materials and technologies for wastewater treatment are urgently needed due to the aggravation of the water crisis in the green twenty-first century [1,2]. Until now, great achievements have been made in this area. Wastewater treatment methods are mainly classified to three categories: physical technology, chemical technology, and biotechnology [3,4]. Physical technology includes: membrane separation [5], adsorption [6], coagulation treatment [7], etc. Electrolysis [8], chemical oxidation [9], and chemical reduction [10] are commonly used chemical technology. Physical treatment processes are usually time-consuming and inefficient for most pollutants removal from wastewater. Chemical technology is widely used to reduce the turbidity and chroma and remove high molecular substances, but likely to destroy the ecosystem owing to the generation of oxidizing substances and the change in environmental pH, temperature, or oxygen concentration. Biotechnology makes use of microorganisms' resources for pollutant removal from wastewater, hardly changing the surroundings. Compared with physical and chemical technology, biotechnology costs lower for actual wastewater treatment and more suitable for current eco-friendly development [11,12].

Today's wastewater treatment plants commonly utilize conventional biotechnologies to achieve convincing treatment results, but are simultaneously facing some challenges, such as the large land area required, large sludge production, and inefficient for many wastewater pollutants (e.g. pharmaceuticals and personal care products, azo dyes, and endocrine disrupting compounds). To overcome these challenges, emerging improved biological methods and various integrated biotreatments have been proposed. The primary biotechnologies used in wastewater treatment involve biofilm methods, anaerobic ammonia oxidation (anammox) systems, and denitrifying anaerobic methane oxidation (DAMO) process. Physical or chemical treatments (e.g. Fenton's oxidation [13], ozonation [14], photocatalytic process [15], and activated carbon adsorption [16]) are often used as pretreatment or post-treatment to

CONTACT Guangming Zeng zgming@hnu.edu.cn; Cui Lai laicui@hnu.edu.cn College of Environmental Science and Engineering, Hunan University, Changsha, Hunan, 410082, China

promote biotreatment processes. The combined process generally can achieve higher treatment efficiency than a single biotreatment process.

Until now, many researchers have tried to take full advantage of suitable biological technology for wastewater treatment. For example, Escapa et al. [8] highlighted the microbial electrolysis cells used for actual wastewater treatment in the pilot plant. Nancharaiah and Reddy [17] focused on aerobic granular sludge technology for compact and cost-effective treatment of high-strength real wastewater. Wang et al. [18] described the technologies for reducing sludge production in wastewater treatment plants. Alvarino et al. [19] analyzed the sorption and biotransformation in biological wastewater treatment processes, which have a great impact on organic micro pollutants removal efficiency. These papers focus on only one method or one aspect of the biotechnology. For actual wastewater treatment, each biotechnology has its own benefits and limitations depending on the applied fields. It is significant to uncover the most suitable methods to meet the specific requirements in practical situations. A comprehensive summary of optimum biotechnological processes for treating wastewater in different conditions is needed.

This review provides a comprehensive overview of biotechnologies from three aspects: (i) single biotreatment processes, (ii) combined diverse biotreatments, and (iii) biosensors. To show the readers efficient biotreatment processes, we focus on the characteristics of each method applied in pollutant removal from wastewater. Biosensors basically meet the requirements of simple, rapid, automatic, and continuous determination of many biochemical indicators in monitoring, which plays an important role in wastewater treatment process. The applications and limitations of biosensors in wastewater monitoring are discussed. We are confident that this review on wastewater biotreatment processes will project a rapid developmental way to provide a full view of advanced biology technologies, applications, challenges, and opportunities.

Emerging biological methods

Moving bed biofilm reactor

The activated sludge method is used as the major way during wastewater treatment, and microorganisms play a crucial role [20]. When aeration is continued in the wastewater, aerobic microorganisms propagate and gradually form suspension flocs, finally depositing them at the pool bottom in order to obtain activated sludge. It is necessary to recycle the activated sludge to ensure

Figure 1. Technological scheme of MBBR. Adapted from Ref. [25].

the presence of enough microorganisms in the reactor [21]. However, the active sludge may fail to settle on the tank bottom if flocculation is inadequate [22], and the activated sludge process will not be able to effectively remove micro-organic pollutants [23,24]. Based on above questions, researchers have turned to a study of a new biotreatment system, a moving bed biofilm reactor (MBBR) (Figure 1) [25].

MBBR is a biological unit with a biofilm growing on the carrier surface. The biofilm is formed by an aerobic outer layer and the anoxic inner layer contains a high concentration of active biomass. Therefore, organic carbonaceous matter can be degraded, and nitrification and denitrification can co-occur in the MBBR. Additionally, the carrier can move all around the tank via mixing or aeration, which improves the oxygen utilization and benefits the contact of the biofilm with the pollutants to improve overall wastewater treatment. The physical properties of the carrier – including shape, size, dimensionality, voidage, and protected surface area – and hydraulic efficiency (HE) play an important role in the whole process from startup to treatment of the pollutants [26]. Generally, spherical carrier media show larger voidage, faster biofilm formation rates with the reduced amounts of dead or stagnant zones, contributing much to removing chemical oxygen demand (COD) and ammonia. The protected surface area weakly correlated with oxygen mass transfer and biofilm formation rates for COD and ammonia removal. Dimensionality, voidage, and HE present good correlations with oxygen mass transfer and biofilm formation rates for heterotrophic and nitrifying bacteria [27,28]. Knowing the impact of the media's physical properties well benefits the commissioning of MBBR and will achieve a higher efficient treatment process.

As a compact, simple, flexible, and reliable process, MBBR has been well applied in developing countries that possess limited land resources, which can be transformed on the basis of existing wastewater treatment plants. MBBR shows high performance in treating urban wastewater, industrial wastewater, and wastewater containing micro-pollutants systems [29,30]. For example, MBBR inoculated with activated sludge and appropriate microorganisms can remove 67.79% of COD, 61.12% of soluble COD (sCOD), 76.26% of particulate COD, and 56.97% total ammonia nitrogen (TAN) from primary settled wastewater under steady-state conditions with organic and hydraulic shock loadings [31]. Virgin polyethylene carriers have been used in MBBRs with a 70% filling fraction. The parameters for primary settled wastewater contain 225 mg L^{-1} of COD, 133 mg L^{-1} of sCOD, 15 mg L^{-1} of TAN, and 102 mg L^{-1} of total suspended solids (SSs) at pH 7.2 [31]. Bester et al. [32] reported that staged MBBR shows high degradation efficiency on pharmaceutical hospital wastewater that coincided with both COD and nitrogen removal. A three staged MBBR was used to treat a fraction of hospital wastewater. The three identical 3 L reactors in MBBR were filled with 50% 500 AnoxKaldnes™ K5 carriers. The raw hospital wastewater was first filtered via an 80 μm filter. The filtered wastewater was collected in a 100 L reactor and maintained at 15–18 °C to equalize the flow. In MBBR, X-ray contrast media were degraded more efficiently than in activated sludge treatment. Also, the degradation half-life for diclofenac removal is only 2.1 h, which is much faster than in other wastewater bioreactors [32]. A summary of the performance of MBBR in wastewater treatment is presented in Table 1.

MBBR is also remarkable for its stability during harsh conditions, such as high or low load and extreme temperatures [34]. Shortening the startup time can increase the economic competitiveness of the MBBR process [21]. A carrier with good performance is helpful to shorten the startup time [27]. Additionally, it is also supposed that the implementation of an attachment enhancing strategy, for example, increasing the attachment of anammox bacteria (AnAOB) to the carrier media, can decrease the startup time [33]. Moreover, compared with a single-stage MBBR, the startup time of an MBBR enriched with granular sludge reduced the processing time from 90 days to 50 days, because granular sludge can produce a biofilm more rapidly [33].

Overall, MBBR exhibits five benefits for wastewater treatment: (i) short sludge retention time, (ii) high biological concentration, (iii) less space required, (iv) no removal of activated sludge, and (v) stability during harsh conditions.

Anaerobic ammonia oxidation process

A traditional anaerobic–anoxic–oxic (A^2/O) process applied in the denitrification and dephosphorization of wastewater has some shortcomings, such as high energy consumption, low processing efficiency, and considerable residual sludge [35,36]. Especially, the effluent is far above the discharge standards under the high ammonia nitrogen concentrations [36]. Thus, the anammox process, showing great promise for self-sustaining biological dephosphorization and denitrification in low C/N wastewater, has attracted wide attention [37,38]. Compared with the A^2/O process, the anammox process has some improvements in wastewater treatment, such as lower operational costs, no requirement for organic carbon, less greenhouse gas emissions, and reduced sludge production [39,40]. Also, the anammox process removed nitrogen without producing N_2O [41]. Conclusions, summarizing the performance of the anammox process are provided in Table 2.

In the anammox process, when there is enough O_2, the anammox biochemical process with three consecutive conversations of the substrates is divided into three steps (Figure 2) [45]: (i) nitrite reductase from denitrifying microorganisms catalyzes the one-electron reduction of nitrite to NO, (ii) NO reacts with ammonium to generate hydrazine, with the hydrazine synthase enzyme as the catalyst, meanwhile three electrons are the input, and (iii) hydrazine is finally oxidized to be N_2 with the release of four electrons in order to drive steps (i) and (ii) in the anammox biochemical process. When there is little O_2, nitrite is used as the electron acceptor and ammonia functions as the electron donor to form N_2 via AnAOB. Adding Mn-oxides can enhance the granule-based anammox process [41]. Mn-oxides are superior to denitrification as a sink for nitrate. In mainstream wastewater treatment plants (800,000 m^3/d of wastewater, 28–32 °C, 5 d of total sludge retention time), anammox showed high performance during biological nitrogen removal with an efficiency of 37.5% [46]. The organics in the treatment system are alternately used to produce methane in order to improve the energy recovery from wastewater [47].

The anammox process has shortcomings when used in actual applications. The growth rate of AnAOB is extremely slow, this results in high sensitivity to a variety of environmental conditions (e.g. different pHs, temperature, and wastewater composition) [48]. Additionally, high salinity content is a great inhibitor of the anammox process for wastewater treatment. Recently, several effective solutions for these shortcomings have been proposed: adjusting the environmental

Table 1. Performance of MBBR in wastewater treatment.

			Specification of carrier			Operational condition							
Target	MBBR volume (L)	Configuration	Carrier	SSA ($m^2\ m^{-3}$)	Materials	HRT (h)	MFF (%)	pH	T (°C)	DO/aeration rate	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Ozonation products of pharmaceuticals	0.2	Aerobic-MBBR	AnoxKaldnes™ K5 carriers	–	Polypropylene	–	4	–	–	–	Diclofenac: >83%	Intensified carbon management and intermittently feeding	[20]
Sludge digester liquor	10,000	SNAD-MBBR	BioMTM carriers	700–750	Polyethylene	48	30	6.9–7.3	27–30	0.3–0.5 mg L^{-1}	TN: >70%	Combination with SNAD process	[21]
Laundry wastewater	260	Two-stage MBBR	Kaldnes K5	800	Polyethylene	6.24	50	>8	25–35	2–4 mg L^{-1}	COD: 89–94%; BOD_5: 95–98%	Kaldnes K5 based carrier	[25]
Wastewater containing benzotriazoles	4.5	MBBR-AS	Kaldnes K3	500	Polyethylene	10.8 ± 1.2	30	7.0–8.0	Room T	4 mg L^{-1}	COD: 86% NH_4^+-N: 93–95%	Specific microorganisms	[26]
Real wastewater	2000	Aerobic-MBBR	Biofil	112	Polypropylene	144	60	7.6 ± 0.1	15.1 ± 2.2	3.6–18.7 $m^3\ m^{-2}\ h^{-1}$	COD: 88 ± 4%; Ammonia: 50 ± 13%	Spherical carrier media	[27]
Pharmaceuticals in conventionally treated wastewater	3	Two-stage MBBR	Anoxkaldnes™ K5 carriers	–	Polypropylene	1	50	7.3–8.5	23 ± 2	6–9 mgL^{-1}	Diclofenac: >50%; Atenolol: >50%; Ammonia: >95%	Intermittently feeding	[29]
Benzotriazoles	3	Two-stage MBBR	Kaldnes K3	500	Polyethylene	12.4 ± 0.6	30	7.0 ± 0.2	Room T	>4 mg L^{-1}	4-Methyl-1H-benzotriazole: 41% 2-Hydroxybenzothiazole: 88%	Combination with activated sludge	[30]
Water from wastewater treatment plants	3	MBBR-EM	Virgin polyethylene carriers	850	Polyethylene	6	70	7.2	23 ± 2	~4 mg L^{-1}	COD: 76.26%	Effective microorganisms	[31]
Pharmaceuticals in hospital wastewater	3	Three-stage MBBR	AnoxKaldnes™ K5 carriers	–	Polypropylene	6	50	7.0–8.0	15–18	0.50 L h^{-1}	Diclofenac: ~30%; Trimethoprim: ~45%; Clindamycin: ~90%	Staged MBBR	[32]
Synthetic wastewater	16	MBBR	Kaldnes K3	500	Polyethylene	24	40	7.6 ± 0.2	33 ± 1	Anaerobic conditions	TN: >80% (with 50 days startup time)	Seeded with granular biomass	[33]

AS: Activated sludge; DO: Dissolved oxygen; EM: Effective microorganisms; HRT: Hydraulic retention time; MFF: Media filling fraction; SSA: Specific surface area; T: Temperature.

Table 2. Performance of anammox in wastewater treatment.

				Influent concentration		Operational condition								
Target	Reactor	Volume (L)	Configuration	NH_4^+-N (mg-N L^{-1})	NO_2^--N (mg-N L^{-1})	HRT (h)	pH	*T* (°C)	DO/aeration rate	Biomass	BC (gVSS L^{-1})	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Low-strength wastewater	UB	3	Anammox-up-UB	320	320	24–2h	–	28–30	Anaerobic conditions	Biofilm	23.91	TIN: 76.3%	High QS communities	[37]
Landfill leachate	USB	27.5	Anammox-USB	460–500	0.5–10	–	7.2–8.3	35–38	0.3–1.5	Suspended sludge	3–4	TN: 95%	Partial-denitrification for post-anammox	[38]
Cd (II)	CB	1	Anammox	~50	~50	3 h	8	25	Anaerobic conditions	Biofilm	–	Cd (II): 99%; TN: 84.2%	Acute and persistent suppression by Cd (II)	[39]
Municipal wastewater	SBR	2.8	Anammox-SBR	199 ± 4.5	221 ± 5.6	4 h	–	32 ± 2	–	Suspended sludge	–	NH_4^+-N: 92 ± 0.6%; NO_2^--N: 94 ± 0.8%	Simultaneous anammox and denitrification system	[40]
Synthetic wastewater	UASB	3.5	Anammox-UASB	280	280	0.96	7.5–7.6	35 ± 1	Anaerobic conditions;	Granular	27.8	TN: 90–93%	Introduction of MnO_2	[41]
Municipal wastewater	SBR	1	Anammox-SBR	0.907	0.006	–	7.5	30	Anaerobic conditions	Suspended sludge	1.0 ± 0.2	Specific anammox activity: 0.490 ± 0.029 gN $gVSS^{-1}$ d^{-1}	Correction and adequate control of pH	[42]
Synthetic wastewater	UFNA	66	Anammox	150	150	24 h	8–8.4	32 ± 1	0.1–0.2 mg L^{-1}	Suspended sludge	3.44	Nitrogen removal rate: >44%	Addition of trace elements	[43]
High salinity wastewater	UASB	1	Anammox-UASB	210–350	210–350	0.94 h	–	35 ± 2	–	Granule	1.03	Nitrogen removal rate: 1.34 ± 0.12 kg N m^{-3} d^{-1}	Addition of calcium	[44]

BC: Biomass concentration; CB: Cylindrical biofilter; DO: Dissolved oxygen; HRT: Hydraulic retention time; QS: Quorum sensing; *T*: Temperature; UB: Up-flow biofilm; UFNA: Up-flow nitritation-anammox.

pH (generally optimized at 7.0–8.0, and the range narrows with a decreased temperature). [42] Also, supplementing the external field energy and micronutrients [43] can enhance the activities of AnAOB. As for the impact of a high salinity content in wastewater, at low temperature (9–25 °C) or high sodium levels (10–15 g L^{-1}), high concentration of calcium can reduce the inhibition caused by the high salinity content [44]. Furthermore, the anammox process is often combined with nitrification and partial denitrification to ensure there is a source of nitrite.

Denitrifying anaerobic methane oxidation

The DAMO process plays an important role in the nitrogen and carbon cycle [49,50]. The performance of the DAMO process in wastewater treatment is summarized in Table 3. The DAMO process uses methane as the carbon source for denitrification, reducing methane emission and nitrogen levels without requiring costly electron donors [54]. The DAMO process is proposed to have two different reaction pathways: inter-aerobic denitrification executed by *M. oxyfera* and reverse methanogenesis conducted by *M. nitroreducens* [55,56]. In anaerobic conditions, methane can also be oxidized and provides electrons for the conversion of nitrate to nitrite via reverse methanogenesis executed by *M. nitroreducens* [57]. Acetyl-CoA synthetase has been found in *M. nitroreducens*, supposing that a full reductive acetyl-CoA pathway might occur to produce acetate in order to achieve carbon fixation [58]. Nitrite cannot be reduced further via *M. nitroreducens*, because the genes for nitrite reduction have not been determined in *M. nitroreducens*. Nitrite reduction during anaerobic conditions is carried out via an inter-aerobic denitrification route. Nitrite is reduced to nitric oxide, and then nitric oxide is bioconverted to nitrogen and oxygen. The bioconverted oxygen can be utilized by a methane oxidation process to generate methanol via the particulate methane monooxygenase. Methanol is subsequently metabolized to formaldehyde via methanol dehydrogenase, and the formaldehyde is oxidized to formate via methylene-H_4F dehydrogenase or methylene-H_4MPT dehydrogenase. Finally, formate dehydrogenase converts the formate to CO_2. The generated CO_2 can participate in the Calvin–Benson–Bassham cycle as the carbon source for *M. oxyfera* growth.

Three mathematical models for DAMO the process have been proposed: model A, B, and C. These are based on the basics of enrichment of: DAMO bacteria, DAMO bacteria and AnAOB, and DAMO bacteria-DAMO archaea-AnAOB, respectively. Different models show a

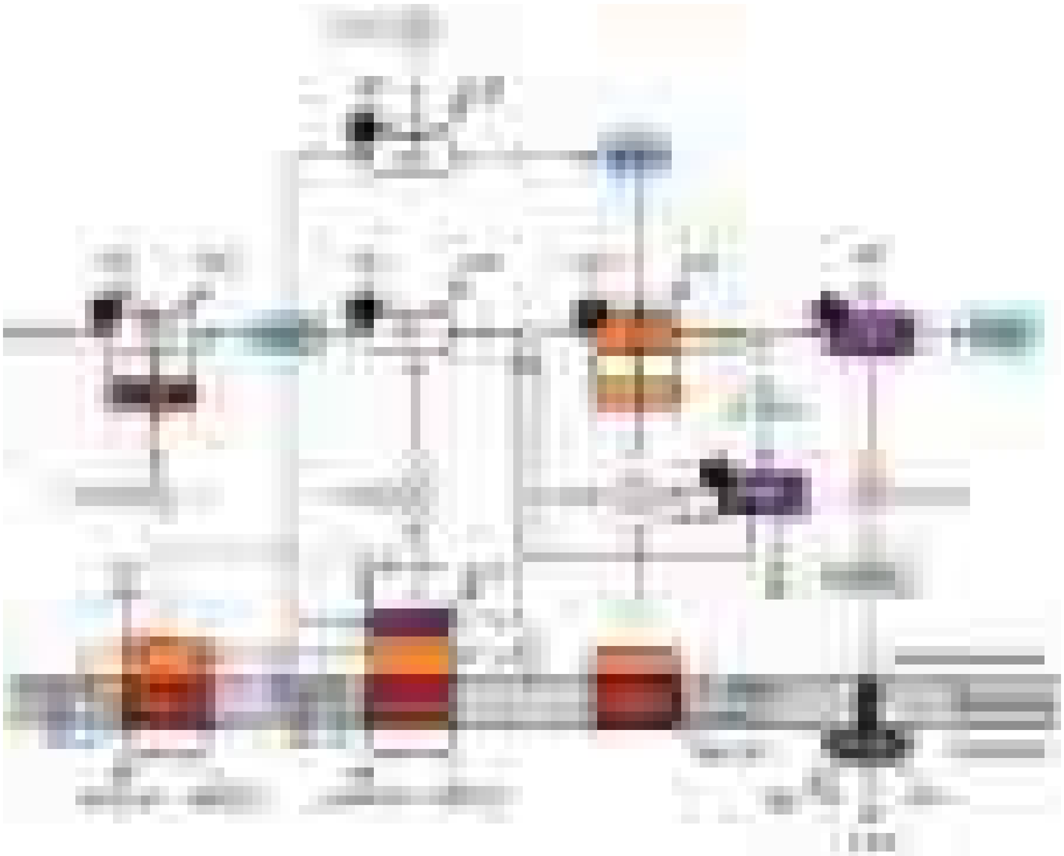

Figure 2. Proposed process of nitrogen formation and ATP generation from ammonium and nitrite with nitric oxide (NO) and hydrazine (N_2H_2) as intermediates under the catalysis of various enzymes. 1: Nir; nitrite reductase; 2: HZS; hydrazine synthase; 3: HDH; hydrazine dehydrogenase; 4: HOX; hydroxylamine oxidase; 5: NXR; nitrite: nitrate oxidoreductase; 6: Nrf; nitrite reductase. Adapted from Ref. [45].

variety of microbial pathways. Models A and B apply Monod kinetics, and model B needs to consider nitrate inhibition. Model C applies a one-dimensional biofilm model. The whole DAMO process and mechanisms are presented in Figure 3 [57]. It was found that substrate concentrations (e.g. methane and nitrite), environmental oxygen concentration, temperature, and pH also have an impact on the DAMO process. The influencing factors are also summarized in Figure 3. Equations (1) and (2) exhibit the denitrification process:

$$CH_4 + 4NO_3 \rightarrow CO_2 + 4NO_2^- + 2H_2O_2 \qquad (1)$$

$$3CH_4 + 8NO_2^- + 8H^+ \rightarrow 3CO_2 + 4N_2 + 10H_2O \qquad (2)$$

According to the tests on a bench-scale, the DAMO process shows high performance in nitrate and nitrite removal during wastewater treatment [51,52]. For example, Zeng et al. [53] proposed coupled anammox and DAMO microorganisms in a hollow-fiber member biofilm reactor (DAMO-anammox HfMBR) for nitrogen removal. The bench experimental results showed that both NH_4^+ and NO_3^- treated by DAMO-anammox HfMBR were undetectable with $\sim$180 mg N L^{-1} of NH_4^+ and $\sim$350 mg N L^{-1} of NO_3^- occurring in the influent. NH_4^+ and NO_2^- are bio-converted to dinitrogen gas and NO_3^- via AnAOB. Methane is oxidized via DAMO and provides electrons for denitrification. DAMO *M. nitroreducens* utilizes NO_3^- produced by AnAOB, meanwhile AnAOB and DAMO *M. oxyfera* utilize NO_2^- generated from reverse methanogenesis executed by *M. nitroreducens* [53].

Although some progresses have been made, DAMO technology is still at an early stage, which has not been widely used in full-scale application. Scaling up the systems and guaranteeing wastewater treatment efficiency are significant for expanding the application of DAMO technology in practical industrial engineering. Moreover, the gases (e.g. CO_2, hydrogen, and H_2S) generated from the sludge digestion might have an impact on DAMO performance, which needs further exploration. The other challenge for DAMO is the slow growth of the biofilm. Efficient methods for accelerating the growth rate needs development.

Table 3. Performance of DAMO in wastewater treatment.

Target	Reactor	Volume (L)	Configuration	Influent concentration: NH_4^+-N (mg-N L^{-1})	Influent concentration: NO_2^--N (mg-N L^{-1})	Operational condition: HRT (h)	pH	T (°C)	DO/aeration rate	Biomass	BC (gVSS L^{-1})	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Nitrate-contaminated water	MBR	0.8	DAMO-MBR	50	50	120–16.8	7.0–8.0	22 ± 2	7–9 mg L^{-1}	Biofilm	–	Removal rate: 0.045 kg N m^{-3} d^{-1}	MBR	[51]
Synthetic wastewater	MBR	2.358	DAMO-MBR	470	560	192–24	7.0–8.0	35	Anaerobic conditions	Suspended sludge	3	Removal rate: >1 kg N m^{-3} d^{-1}	Combined with anammox	[52]
Synthetic wastewater	HfMBR	5.02	Anammox-DAMO-HfMBR	180	20	120–2.4	7.8 ± 0.2	35 ± 1	Anaerobic conditions	Biofilm	–	NH_4^+-N: 46.28 mg L^{-1} day^{-1} NO_3^--N: 63.95 mg L^{-1} day^{-1}	Combined with anammox	[53]

BC: Biomass concentration; DO: Dissolved oxygen; HfMBR: Hollow-fiber membrane biofilm reactor; HRT: Hydraulic retention time; *T*: Temperature.

Integrated biotreatment processes

Simultaneous partial nitrification, anammox, and denitrification process in integrated fixed film activated sludge

A single treatment method is often insufficient for a wastewater system, thus the combination of two or more methods has become a research priority. This strategy, used for wastewater treatment, is more cost efficient and environmental-friendly owing to the less energy consumption, such as no oxygen supply required and the low sludge production [53]. Simultaneous nitrification, anammox, and denitrification (SNAD) process is a currently an important technology applied in wastewater treatment. However, some shortcomings, such as high oxygen consumption, high temperature, low C/N requirement, and the slow growth rate of AnAOB, exist when SNAD is used in practical applications. A strategy for the SNAD process in an integrated fixed film activated sludge (SNAD-IFAS) and was presented by Yang et al. [59] to overcome some of the above shortcomings and achieve mainstream nitrogen removal. IFAS is an efficient technology for wastewater with high C/N at room temperature, and is widely used during denitrification processes [60]. Ammonium oxidizing bacteria (AOB) and AnAOB can grow well in IFAS, and AOB mainly adhere to activated sludge while AnAOB are primarily located in the biofilm. AOB and AnAOB considerably contribute to the improved denitrification rate and their operational stability.

The relationship of the pivotal functional nitrogen removal guilds in the SNAD-IFAS process is proposed in Figure 4 [59]. AnAOB (*Candidatus* Kuenenia) utilized ammonium from wastewater and nitrite from AOB (*Nitrosomonas*) and denitrifying bacteria (DNB, *Denitratisoma*) to generate nitrate and soluble microbial production. Subsequently, the generated nitrate and microbial production were used as the substrate for heterotrophic bacteria (*Limnobacter*, *Bryobacter*, and uncultured Anaerolineaceae). Meanwhile, heterotrophic bacteria as the main force of the microbial floc provided favorable environment for AnAOB to prevent inhibition of adverse conditions (oxygen and organic matter). *Denitratisoma* utilized the carbon source from the wastewater and the degradation products of *Lewinella*.

The SNAD-IFAS system exhibits high efficiency for mainstream nitrogen removal in wastewater treatment plants with an optimum COC/N ratio at ~1.2 [59]. The SNAD-IFAS system only requires 40–80 mg L^{-1} of organic matter and 0.4 mg L^{-1} of oxygen. Compared with the 150–200 mg L^{-1} of organic matter demand

Figure 3. Microorganisms responsible, relevant mechanisms, mathematical models and influencing factors of DAMO process. Adapted from Ref. [57].

and 7–9 $mg\,L^{-1}$ of oxygen demand in denitratation/anammox or 1.5 $mg\,L^{-1}$ of oxygen demand in nitritation/anammox process, SNAD-IFAS is more cost-effective and can save the considerable energy consumption used for aeration.

There are two pathways for nitrogen removal in the SNAD-IFAS reactor [59]. Nitrogen removal from the suspended sludge was started from the conversion of ammonium to generate nitrite via ammonia-oxidizing bacteria. The residual ammonium and generated nitrite were bioconverted to nitrogen gas via AnAOB. Moreover, nitrite-oxidizing bacteria (NOB) converted a part of nitrite to nitrate, while DNB bio-converted nitrate and the carbon source to nitrogen gas. In biofilms, DNB bioconverted the nitrate from NOB products and the anammox process, and then provide nitrite for AnAOB under low COD concentration. Anammox showed significant cooccurrence with some heterotrophic bacteria. AnAOB provided substrate for heterotrophic bacteria, while heterotrophic bacteria protected AnAOB from the inhibition of hostile environments, leading to the high survivability of AnAOB in an organic matter environment.

Combined up-flow anaerobic sludge blanket-biological aerated filter

Apart from SNAD-IFAS process already discussed, up-flow anaerobic sludge blanket (UASB) coupled with a biological aerated filter (BAF) was also a good choice for wastewater treatment [61]. Table 4 summarizes the experimental parameters and conditions of SNAD-IFAS and UASB-BAF in wastewater treatments. UASB acts as a high-speed anaerobic reactor [63], and the existing granular sludge improves the treatment efficiency [64]. A single UASB reactor obtained great removal of nitrogen from wastewater but also undesirable phosphorous removal [65].

Considering the advantages of immobilized microorganism technology, such as high hydraulic loading rate, low sludge production, ensured biomass amounts, and undamaged microorganism [66], the solution to

Figure 4. Proposed interactions between key functional nitrogen removal guilds in SNAD-IFAS process. Adapted from Ref. [59].

obtain desirable phosphorous removal and develop integrated USAB-immobilized microorganism technology [67]. BAF, attractive immobilized microorganism technology, is filled with micropores and macropores carriers, which acts as the support for biomass attachment and SS filtration. This system has the advantages of a small occupation room, short hydraulic retention time, no treatment for excess sludge, and no requirement for a clarifier [68].

UASB-BAF showed high performance during microcrystalline cellulose (MCC) wastewater treatment [61]. The MCC removal efficiency achieved 83.5% under 2000 mg L^{-1} of the MCC load. Cellulolytic and noncellulytic microorganisms play the major role in the first stage (full-value cellulose) and the second stage (low-value cellulose) of the UASB and BAF system, respectively. Figure 5 illustrates the whole process of UASB-BAF, including two-stage UASB (U1 and U2) and two-stage BAF (B1 and B2) [61]. The water inlets are set at the bottom and the outlets are set at the top in both U1 and U2. B1 and B2 filled with microporous and macroporous carriers (porous polyurethane foam, 98% porosity) operate at room temperature with aeration [61]. Macropores ensured a good three-phase mixture of air, wastewater, and a carrier, which enhanced mass transfer propulsion. Micropores played an important role in the immobilization of microorganisms via active chemical groups, including –OH, $–NH_2$, –COOH, $–CH_2$, $–CHOCH_2$, etc. [66].

A great success was also obtained on non-biodegradable organic pollutant treatment via the UASB-BAF process [62,68]. Bae et al. [68] employed the combined UASB-BAF process to promote removal of non-biodegradable compounds from wastewater. In Tong's work [62], the UASB-BAF process was used to

Table 4. Performance of SNAD-IFAS and UASB-BAF in wastewater treatment.

				Operational condition								
Target	Reactor	Volume (L)	Configuration	HRT (h)	pH	T (°C)	DO (mg L^{-1})	Biomass	BC (gVSS L^{-1})	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Mainstream wastewater	IFAS reactor	8	SNAD-IFAS	18	7.2 ± 0.2	25 ± 2	0.4 ± 0.1	Sludge and biofilm	–	TN: 72 ± 2%; COD: 88%	Co-existence of flocculent sludge, granular sludge and biofilm	[59]
Nitrogen removal	IFAS reactor	500	SNAD-IFAS	24	8.15–8.38	25–28.5	0.3–1.0	Sludge and biofilm	9.5	Removal rate: 1.2 kg N m^{-3} d^{-1} (removal efficiency: 80%)	Co-existence of flocculent sludge, granular sludge and biofilm	[60]
MCC wastewater	UASB-BAF	11.63	UASB-BAF	18.5	7.2–7.4	35 ± 1	–	Granular	–	MCC: 83.5%	First stage: cellulolytic microorganisms, second stage: non-cellulolytic microorganisms	[61]
Heavy oil wastewater	UASB-BAF	4600	UASB-BAF	12 h	7.8–8.3	14.6–44.2	2–4	Suspended solid	16–24	COD: 74%; NH_4^+-N: 94%; SS: 98%	Combined biotreatment system	[62]

BC: Biomass concentration; DO: Dissolved oxygen; HRT: Hydraulic retention time; MCC: Microcrystalline cellulose; T: Temperature.

Figure 5. Technological scheme of combined UASB/BAF. Adapted from Ref. [61].

treat heavy oil wastewater with high concentrations of insoluble organic compounds and a low content of N and P nutrients, and notably, the removal efficiency of COD, ammonia nitrogen (NH_3-N) and SSs obtained 74%, 94%, and 98%, respectively.

Combined advanced oxidation-biotreatment process

Fenton's reaction coupled with a biotreatment process

Fenton's oxidation is a widely used advanced oxidation process (AOP) due to its low cost and no secondary pollution [69,70]. Traditional Fenton's reagents mainly include ferrous irons and H_2O_2. In wastewater treatment process, a strong oxidation hydroxyl radical is produced by the decomposition of hydrogen peroxide catalyzed by ferrous ion, which enables the oxidation of the refractory organic matter (e.g. antibiotics and other pharmaceutical reagents) into H_2O and CO_2 or easily degradable intermediate products [71]. Fenton's reaction can be summarized into Equations (3) and (4):

$$H_2O_2 + Fe^{2+} \rightarrow \cdot OH + OH^- + Fe^{3+} \tag{3}$$

$$H_2O_2 + Fe^{3+} \rightarrow H^+ + Fe^{2+} + \cdot OOH \tag{4}$$

To achieve the higher efficiency of the Fenton reaction in wastewater treatment, there are three requirements: (i) an acid environment with a pH of 2–3, (ii) moderate temperature and pressure, and (iii) low ratio of biochemical oxygen demand (BOD)/COD (<0.3) [72–74].

An aerobic sequencing batch reactor (SBR) process with activated sludge is characterized by its flexibility and low costs, containing five steps: fill, react, settle, draw, and idle [75]. In aerobic SBR, microorganisms in the activated sludge utilize the degradable intermediate products as nutrients to participate in the metabolization process, enhancing the pollutant removal in wastewater. In this way, the aerobic SBR process can act as a platform to provide a satisfactory reaction environment for Fenton's oxidation [76]. Part of excess sludge from aerobic SBR can be reduced via the Fenton reaction, decreasing the cost of subsequent sludge disposal [77].

The addition of a catalyst, hydrogen peroxide feed concentration, temperature, and contact time all have effects on the treatment process. The treatment efficiency can be improved by adjusting these parameters. For example, Bae et al. [78] found that utilizing activated carbon as a catalyst in the Fenton-SBR process can improve the treatment effect on acrylic dyeing wastewater under the following influent conditions: pH 10–12, 40 °C, $\sim$1150 mg L^{-1} of total COD, $\sim$1100 mg L^{-1} of sCOD, and $\sim$1180 ADMI units of color. The Fenton process tends to just break some bonds in the organics not degrade them. However, the soluble chemical oxygen demand can be greatly reduced. Biotreatment eliminated the degradable organics, while the following Fenton's process decreased the recalcitrant organics and ferric iron can coagulate those residual particulate matters after biological and chemical oxidation. Additionally, it is worth noting that the biodegradability of wastewater can be greatly improved by a combined Fenton-aerobic SBR process according to Madeira's studies [71]. Fenton's oxidation was used as a pretreatment, aiming to degrade organic materials to some smaller molecules. Subsequently, SBR can remove the smaller biodegraded organic molecules. After treatment, the BOD_5:COD ratio and specific oxygen uptake rate reached 0.27 from <0.001 and 11.1 from <0.2 mg $O_2/(g_{VSS} \cdot h)$, respectively. Also, the toxicity of the acrylic dyeing wastewater was greatly reduced according to the decreased inhibition of *Vibrio fischeri* (from 92.1% down to 6.9%). It was shown that the Fenton-aerobic BAF process was economically attractive.

Furthermore, to avoid the large production of sludge containing ferric ions owing to the precipitation of ferric ions in the traditional Fenton's oxidation, combination of photo-Fenton, electro-Fenton, Fenton-like, or photo-electro-Fenton reaction and biotreatment has attracted much attention in recent years [79–81]. The photo-Fenton reaction shows two characteristics [82]: (i) more production of hydroxyl radicals via photolysis

Table 5. Performance of Fenton's oxidation-biotreatment in wastewater treatment.

Target	Reactor	Volume (L)	Configuration	Operational condition HRT (h)	pH	T (°C)	DO (mg L^{-1})	Fe^{2+}	H_2O_2	Light	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Acrylic dyeing wastewater	CSTR	0.92	Fenton-CSTR-SBR	<1.5	3.0	40	3	0.227 mg L^{-1}	3.52 g L^{-1}	–	COD:78.0%; TOC :80.9%; BOD_5:60.6%, TN:48.4%; TP:55.5%	Combined with the Fenton's process with biological	[71]
Azo dye	Batch reactor	0.4	Fenton-SBR	48	3.0	22 ± 2	3–5	40 mg L^{-1}	0.25 g L^{-1}	–	COD: 91% Phosphorus: 87%; Dye decolorization: 94%	Alternating anaerobic aerobic SBR with external feeding	[76]
Dyeing wastewater	AS	–	Fenton-AS	0.5	3.5	30	3–5	4.2 mM	4.0 mM	–	SCOD: 66% Color: 73%	Combined with AS	[78]
Landfill leachate	Batch reactor	–	Photo-Fenton	1	3.0–3.5	Room T	0.5–2	2 g L^{-1}	5 g L^{-1}	UV light	COD: 86%	Photo-Fenton process	[82]
Pesticide-containing wastewater	Recirculation tank	16	Photo-Fenton-IBR	0.75	2.6–2.9	30	0.5–2	140 mg L^{-1}	16 mM	UV light	An abatement higher than 86% for 18 pesticides[a]	Biological oxidation process, photo-Fenton process with CPCs, and biological oxidation process	[83]

AS: Activated sludge; CPCs: Compound parabolic collectors; CSTR: Continuous stirred tank reactor; DO: Dissolved oxygen; HRT: Hydraulic retention time; SCOD: Soluble chemical oxygen demand; *T*: Temperature.
[a]Eighteen pesticides: S-metolachlor, 2,4-D, MCPA, imidacloprid, alachlor, terbuthylazine, isoproturon, bentazone, tebuconazole, atrazine, linuro, metobromuron, dimethoate, diuron, metribuzin, metalaxyl, chlorotoluron, and simazine.

(Equations (5) and (6)) and (ii) photo-decarboxylation of ferric carboxylates (Equations (7) and (8)).

$$[Fe-OH]^{2+} + hv \rightarrow Fe^{2+} + \cdot OH \tag{5}$$

$$H_2O_2 + UV \rightarrow \cdot OH + H^+ \tag{6}$$

$$Fe(III)RHCO_2 + hv \rightarrow Fe^{2+} + CO_2 + \cdot RH \tag{7}$$

$$\cdot RH + O_2 \rightarrow \cdot RHO_2 \rightarrow \text{products} \tag{8}$$

Vilar et al. [83] presented a photo-Fenton process combined with biological oxidation reaction for pesticide-containing wastewater treatment, including three steps: biological oxidation process was utilized to decompose the biodegradable organic carbon, and then the photo-Fenton process degraded pesticides to low-molecular-weight carboxylate anions to improve the biodegradability of wastewater. Finally, the biological oxidation process was utilized again to decompose the residual low-molecular organic carbon fraction. In this photo-Fenton (62 L of bio-treated wastewater volume, 2.08 m^2 of compound parabolic collectors)-biological oxidation (55 L of bio-photo-treated wastewater volume) process for actual pesticide-containing wastewater treatment, the initial iron dose depends on the phosphates concentration in wastewater because of the negative effect caused by the $FePO_4$ formation. Based on the initial phosphate concentration (~30 mg PO_4^{3-} L^{-1}) in actual wastewater, 70 mg Fe^{2+} L^{-1} was likely to be enough to ensure the efficiency of the photo-Fenton-biological oxidation process. After treatment, the pesticide was totally removed. A final COD below 150 mg O_2 L^{-1} was achieved. The aforementioned Fenton's oxidation-biotreatment processes are summarized in Table 5.

Ozonation coupled with biotreatment process

For the combined AOPs/biotreatment process, ozonation is also applied as an integrated method to improve wastewater treatment. For example, the oil sands process affected water treatment using only MBBR. It removed 18.3% of the acid-extractable fraction (AEF) and 34.8% naphthenic acids (NAs), while MBBR with ozone pretreatment under the same operation conditions showed 41.0% and 78.8% on AEF and NAs removal, respectively [84]. Several ozonation-biotreatment integrated methods in wastewater treatment are summarized in Table 6. Ozonation increased the biodegradability of these organic matters and can reduce the toxicity of wastewater toward microorganisms [85,86]. In wastewater treatment, the ozone oxidation agent mainly contains ozone molecules and fleshly generated oxygen and hydroxyl radicals [87]. Generally, catalysts are used to accelerate ozone decomposition in

Table 6. Performance of ozonation-biotreatment in wastewater treatment.

Target	Reactor	Volume (L)	Configuration	Operational condition HRT (h)	pH	T (°C)	O_3 (mg L^{-1})	OT (min)	Treatment performance (removal)	Proposed reason for high treatment performance	Ref.
Coking wastewater	Cylindrical glass column	7.5	BAF-ozonation	2.4	7.1–8.3	20–32	60	50	COD: 49.7%, BOD_5: 58.8%, NH_4^+-N: 75.8%, color: 91.1%, UV_{254}: 82.3%	Combined ozonation and biological aerobic filter	[85]
Coal gasification wastewater	Cylindrical water-jacketed reactor	1	Catalytic ozonation	–	6.8–7.2	22	2	15	Quinoline: 100%	Catalysis via nano MgO	[86]
Ibuprofen	Double jacket glass reactor	1.1	Ozonation	4	3.0	25	3–6	60	Ibuprofen: 93%	Ozonation study in a semi-batch reactor	[87]
Petrochemical secondary effluent	Bench-scale reactor	4.2	Ozonation	–	7.3 ± 0.4	–	36	60	Organic micro-pollutants: 87.96%	Filtration removal of particulate matter	[88]
Petrochemical secondary effluent	Ozonation tower	316	Fe^{2+}-BAF-ozonation	24	7.02–7.49	–	25	60	TP: 78%, TSS: 99%	Combination of Fe^{2+}-BAF and ozonation	[89]
Fluorescent dissolved organic matters in textile wastewater	Cylindric glass reactor	1	BAF-ozonation	2.4	8.5–9.0	25–35	30–40	25	Color: 96%, UV_{254}: 62.4%, COD: 43.8%	The combination of ozonation and BAF process	[90]
Pharmaceutical wastewater	Borosilicate glass bubble column	1	Ozonation	–	10	Room T	18	180	Antibiotic amoxicillin: 99%, COD: 98%, toxicity: 90%	Multistage ozone and biotreatment system	[91]

HRT: Hydraulic retention time; OT: Ozonation time; T: Temperature.

order to obtain more hydroxyl radicals [86]. The initial pH has a great impact on the ozonation reaction pathway because it influences the self-decomposition of ozone. During the ozonation process, alkaline conditions benefit the generation of hydroxyl radicals, and the organic matters are oxidized via indirect radical reaction. The indirect radical reaction can enhance the mineralization of organics [88]. While under acid conditions, ozone tends to attack the organic matters directly, especially those with an unsaturated structure (e.g. aromatic rings and double bond), to form small molecules [92]. To combine ozonation and biotreatment well, the environmental pH for ozonation can be set at ~7.5, which reveals weak alkalinity and is suitable for the biotreatment process. However, ozonation cannot achieve low COD concentrations.

Adding a BAF after ozonation (ozonation-BAF) process is an efficient method to obtain a lower COD [89,90]. Due to the sterilization effects of ozone, two separated containers for ozonation and BAF were established. BAF promotes the COD removal of the ozonation process. In return, a low ozone concentration in the ozonation effluent benefits the biofilm performance in the BAF. The positive influence of ozonation on BAF performance is shown in Figure 6 [93]: (i) ozonation decomposes the refractory organics to small molecules to enhance the biodegradability for the BAF; (ii) ozonation benefits the mass transfer of the BAF biofilm owing to the increased specific surface area of filter media and the decreased thickness of the attached biofilm; (iii) the biomass and microbial population varies with the ozonation process, which might be beneficial for BAF in order to remove specific organic pollutants. In practical use, the integrated ozonation-BAF process applies very well to the textile wastewater and medical wastewater. Above 90% of most refractory pollutants (e.g. polyvinyl alcohol and antibiotic amoxicillin) can be removed [62,91].

The performance of the integrated ozonation-BAF process depends mainly on the ozone dosage. Enlarging the ozone dosage is the crucial way to promote treatment efficiency for wastewater with high refractory organic matters. However, the residual ozone concentration has an impact on BAF. Low ozone concentration benefits the biofilm performance in BAF. Therefore, how to increase the ozone dosage is a challenge. A solution to overcome this drawback is to improve the reaction rate of ozonation, like catalytic ozonation. Development of various catalysts is necessary owing to the different wastewater [94,95]. Besides, utilizing BAF effluent recycle to lower the ozone concentration may be another way to enlarge the ozone

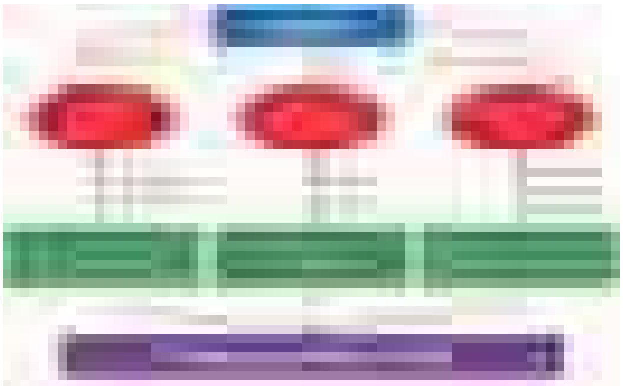

Figure 6. The positive influence of ozonation on the performance of BAF in ozonation-BAF process. Adapted from Ref. [93].

dosage and ensure the performance of BAF, which also needs further exploration.

Biosensors for environmental monitoring

Owing to the capability to address a large array of analytical problems and challenges, biosensors have been widely applied to environmental monitoring. Traditional detection technologies, like atomic absorption spectroscopy [96], high performance liquid chromatography [97], and inductively coupled plasma mass spectrometry [98], are not easy to meet the requirements for high sensitivity and simple operation owing to the necessary for sophisticated instrumentation, long-term sample pretreatment processes and skilled personnel [99–101]. Therefore, numerous laboratory biosensors have been developed to be the technological solution in order to address these challenges.

Biosensors basically satisfy the simple, rapid, automated and continuous determination of many biochemical indicators during environmental monitoring. In this section, we elaborate on three types of biosensors classified according to the transducers, concluding optical, electrochemical, and other biosensors. The recent biosensors mentioned below are presented in Table 7.

i. *Optical biosensor.* Optical biosensors have attracted great interest in environmental monitoring owing to their high sensitivity, quick response, low cost, and simple operation [102,103,113]. Recently, Bidmanova et al. [104] reported on a fluorescence-based optical biosensor for the detection of halogenated aliphatic hydrocarbons pollutants, such as 1,2-dichloroethane, 1,2,3-trichloropropane, and γ-hexachlorocyclohexane with limits of the detection achieved at 2.7 mg L^{-1}, 1.4 mg L^{-1}, and 12.1 mg L^{-1}, respectively. The fluorescence change of the pH indicator can be observed on the basics of the enzymatic reaction with halogenated aliphatic hydrocarbons [104]. Zhang et al. [105] proposed a grating-coupled surface plasmon resonance (SPR) smartphone biosensor with the advantages of portability and simplicity for lipopolysaccharides detection, which showed high selectivity in real samples/matrices detection, and the detection limit of endotoxins achieved 32.5 ng mL^{-1}. The changed signals during the detection were observed via the smartphone's built-in flash light and camera [105].
ii. *Electrochemical biosensor.* The electrochemical biosensor is composed of active sensing biomaterial and electrochemical signal transducers. They have the advantages of simple fabrication, strong specificity, high detection sensitivity, and good stability. With the introduction of new materials and technologies, electrochemical biosensing technology has been greatly developed. In particular, the introduction of functional nanomaterials is available for enhancing the electrochemical sensors performance [114]. For example, integrating graphene for electrochemical biosensors exhibited fast electron transfer and manipulable multi-functionalized surface chemistry, making the sensors more accurate [106]. The layered transition metal disulfide (TMD) nanomaterials also greatly improved the performance of electrochemical biosensors [115]. Additionally, it was found that the autonomous electrochemical biosensor prepared by silicon micro-processing technology can detect herbicides by analyzing photosynthesis and metabolic activities of algae [107]. Ali et al. [108] proposed an all-printed electronic biosensor, which was applied into the fast detection and classification of pathogens, including *Salmonella typhimurium*, the *Escherichia coli strains JM109* and *DH5-α*. A concentration bacterial range from 10^5 CFU mL^{-1} to 10^7 CFU mL^{-1} can be detected accurately. Kuo et al. [109] presented an improved miniaturized label-free electrochemical biosensor for various detections of PoC applications, like the cardiovascular disease biomarker within the range of 10 ng mL^{-1} to 10 μg mL^{-1} can be detected.
iii. *Other biosensors.* Besides optical and electrochemical biosensors, there are many other biosensors also applied in environmental monitoring. For example, Xu et al. [110] proposed a paper-based multi-anode microbial fuel cell combined with power management systems for wastewater, which was a self-support real time biosensor and

Table 7. Summary of recent biosensors.

Biosensors types	Platform	Analytes	Operational conditions			Linear range	Limit of detection	Ref.
			pH	*T* (°C)	Detection time			
Optical biosensor	RuSiNPs-GCE	Ethanol	7.5	Room T	–	10^{-7} to 10^{-2} M	5.0×10^{-8} M	[102]
Optical biosensor	GOD in GFGC-Au/Ag alloy NPs	Glucose	8	Room T	60 s	1.2×10^{-6} to 6.25×10^{-3} M	5.0×10^{-7} M	[103]
Optical biosensor	CF-BSA-CNF-BSA	1,2,3-Trichloropropane	4–10	5–60	60 s	0–40 mg L^{-1}	1.4 mg L^{-1}	[104]
Optical biosensor	SPR-sensor chips-Au diffraction gratings	Lipopolysaccharides	–	–	20 min	10 ng mL^{-1} to 10 μg mL^{-1}	32.5 ng mL^{-1}	[105]
Electrochemical biosensor	DNA-tweezers- FET-SNP	DNA	–	Room T	–	10^{-7} to 10^{-4} M	10^{-9} M	[106]
Electrochemical biosensor	Pt Bl array electrode	Diuron herbicide	–	–	–	0.2–1 μM	0.2 μM	[107]
Electrochemical biosensor	Ag–Ag nanowires-PET	*E. coli*/salmonella	–	37	Real time monitoring	10^5 to 10^7 CFU mL^{-1}	10^4 CFU mL^{-1}	[108]
Electrochemical biosensor	Biochip-PoC	CVD biomarkers	7.4	Room T	–	10 ng mL^{-1} to 10 μg mL^{-1}	10 ng mL^{-1}	[109]
DSS sensor	PMMFC -PMS	Chlorine shocks	–	–	Real time monitoring	100–200 mg L^{-1}	50 mg L^{-1}	[110]
MFC-based biosensor	MFCs-air-cathodes	Formaldehyde	–	20–50	–	0.003–0.075%	0.003%	[111]
REP biosensor	IEMs-ECL	Glucose	6.9	–	25 s	0.5–10 mM	0.5 mM	[112]

BSA: Bovine serine albumin; CF: Fluorescence response of conjugates of 5(6)-carboxyfluorescein; CNF: 5(6)-Carboxynaphthofluorescein; CVD: Cardiovascular disease; DSS: Disposable self-support shock; ECL: Electrochemiluminescence; FET: Graphene field effect transistor; IEMs: Ion-exchange membranes; GCE: Glassy carbon electrode; GFGC: Glutaraldehyde-functionalized glass cell; GOD: Covalent immobilization of glucose oxidase; MFC: Microbial fuel cell; PET: Polyethylene terephthalate; PMMFC: Paper-based multi-anode microbial fuel cell; PMS: Power management system; PoC: Point-of-care; Pt Bl: Platinum black; REP biosensor: Reverse electrodialysis-powered biosensor; RuSiNPs: $Ru(bpy)_3^{2+}$-doped silica nanoparticles; SNP: Single nucleotide polymorphism; *T*: Temperature.

it achieved 28 times data transmission per charging cycle, posing great potentials in *in-situ* monitoring. Reliable, highly sensitive, and practical online monitoring of water quality biosensors based on single-chambered microbial fuel cells was also described by Yang et al. [111] for the estimation of toxic substances. Formaldehyde ranging from 0.003% to 0.075% in media can be detected, and this biosensor can be reusable and show potentials in long-term *in situ* monitoring of water quality [111]. Baek et al. presented a miniaturized reverse electrodialysis-powered biosensor with using electrochemiluminescence, which was widely applicable in electrochemical-sensing. Glucose ranging from 0.5 mM to 10 mM can be detected via this biosensor by observing electrochemiluminescence emissions [112]. Notably, miniaturization applied in these biosensors can offer considerable advantages, like easy integration with other sensors and less damage when used in *in vivo* implantation [116]. Besides, Ali et al. [108] proposed a disposable all-printed electronic biosensor, which was applied in the fast detection and classification of pathogens, including *Salmonella typhimurium, the Escherichia coli* strains JM109 and DH5-α. Concentration of bacterial range from 10^5 CFU mL^{-1} to 10^7 CFU mL^{-1} can be detected accurately.

Summary and perspectives

Summary

Biological methods, as prior wastewater treatment processes, play an important role in pollutant removal from wastewater. With increasingly serious water pollution, some advanced biotechnologies stand out to meet the growing demand for treating processes, including emerging biological methods, integrated biotreatment processes, and high sensitive biosensors.

As an emerging biological method, MBBR has good degradation effects on the refractory pollutants that cannot be removed by a traditional sludge process. Anammox processes show great promise for self-sustaining biological denitrification and dephosphorization in low C/N wastewater. The DAMO process can decrease the nitrogen levels and methane emissions simultaneously during wastewater treatment. Furthermore, to meet the needs of all wastewater treatment systems that cannot be achieved by a single biotreatment process, researchers also paid more attention to the combination of two or more methods.

The SNAD process combined with the IFAS process can not only overcome the disadvantages of high oxygen consumption and the slow growth rate of functional bacteria in the SNAD process, also greatly reduces the operating costs. The UASB-BAF process is a stable and efficient wastewater treatment process, which has good degradation effects on refractory heavy oil wastewater. Also, the addition of AOPs technology provides a new possibility to improve the performance of biotreatment processes. Fenton's oxidation and ozonation processes combined with biotreatment technology also shows high efficiency in wastewater treatment.

In addition to the fast development of the above biotreatments for wastewater, more and more advanced biosensors have been developed in environmental monitoring. Progress about three types of biosensors, optical, electrochemical, and other biosensors, is concluded. Generally, optical biosensors need shorter test time and electrochemical biosensors show higher sensitivity in the actual situation. Other biosensors also have some advantages, like easy miniaturization and great potentials during *in situ* monitoring.

Challenges and opportunities

Although these advanced biotechnologies show numerous advantages for wastewater treatment, there are still some limitations and challenges for further applications. Opportunity coexists with challenges, which are also discussed at length in this section.

Biotreatment

i. Due to the slow growth of functional bacteria, most of the biotreatment processes are highly sensitive to various environmental conditions (including pH value, temperature, and wastewater components), resulting in a too long startup time, and then the economic benefits are reduced and the practical applications are limited.
ii. Despite the advantages of softer requirements of the surrounding conditions in wastewater treatment, integrated biotreatment process shows some shortcomings. For example, UASB combined with BAF seems to be a good choice to substitute AOPs coupled with biotreatment, but it needs a long retention time, and the reaction rate is too slow to use in large or even middle capacity wastewater treatment plants.
iii. The application of biotreatment in special wastewater treatment plants, like petroleum wastewater, is seldom discussed. The composition of petroleum wastewater is more complex. However, there are few studies on biotreatments in this field.
iv. Many new wastewater treatment processes still require large capital and operating costs, which limit the promotion of new treatment processes.

Biosensors

i. The cost of biosensors is seldom mentioned. As we know, when biosensors use immobilized bioactive substances such as catalysts, valuable reagents can be used repeatedly while low-priced materials are easily deactivated and have poor reproducibility. For long-term development, the monitoring must be run in both eco-friendly and cost-effective ways.
ii. Applications under extreme conditions are less known. In view of the sensitivity and specificity of biometrics, biosensors have not been widely applied under extreme conditions.
iii. How to transfer laboratory measurements to practical applications? Although many studies have shown that the stability of biosensors can be improved via some methods in the laboratory, further research on improving the performance of biosensors in actual monitoring is still needed.
iv. Can biological recognition elements be used and reused over long periods? Biosensors mainly monitor toxic substances. Therefore, inevitably, there will be a lack of resistance to some toxic substances, affecting the detection capability of the sensor, resulting in reduced sensitivity.

For further progress in biotreatment and biosensing technology, advanced process with powerful oxidation capacity and non-secondary pollution is the most promising and potential method for the future. The development direction of biotechnology applied in wastewater treatment focuses on efficient, energy savings and automation processes. In addition, research and development of portable biosensors applied in full-scale environmental monitoring is also an inevitable trend for the future, which may be combined with some newly developing materials. We believe that the application of biological methods for pollutant removal and biosensors in pollutant detection for wastewater treatment will have a promising and bright future.

Disclosure statement

No potential conflict of interest was reported by the authors.

Funding

This study was financially supported by the Program for the National Natural Science Foundation of China [51879101, 51579098, 51779090, 51709101, 51521006, 51809090, 51278176, 51378190], the National Program for Support of Top-Notch Young Professionals of China [2014], the Program for Changjiang Scholars and Innovative Research Team in University [IRT-13R17], Hunan Provincial Science and Technology Plan Project [2018SK20410, 2017SK2243, 2016RS3026], and the Fundamental Research Funds for the Central Universities [531119200086, 531118010114, 531107050978].

References

[1] Yi H, Qin L, Huang D. Nano-structured bismuth tungstate with controlled morphology: fabrication, modification, environmental application and mechanism insight. Chem Eng J. 2019;358:480–496.
[2] Yi H, Zeng G, Lai C. Environment-friendly fullerene separation methods. Chem Eng J. 2017;330:134–145.
[3] Wang Y, Wang D, Liu Y, et al. Triclocarban enhances short-chain fatty acids production from anaerobic fermentation of waste activated sludge. Water Res. 2017;127:150–161.
[4] Zhang QH, Jin PK, Ngo HH, et al. Transformation and utilization of slowly biodegradable organic matters in biological sewage treatment of anaerobic anoxic oxic systems. Bioresour Technol. 2016;218:53–61.
[5] Bengani-Lutz P, Zaf RD, Culfaz-Emecen PZ, et al. Extremely fouling resistant zwitterionic copolymer membranes with similar to 1 nm pore size for treating municipal, oily and textile wastewater streams. J Membr Sci. 2017;543:184–194.
[6] Sarkar B, Mandal S, Tsang YF, et al. Designer carbon nanotubes for contaminant removal in water and wastewater: a critical review. Sci Total Environ. 2018;612:561–581.
[7] Wu H, Lai C, Zeng G, et al. The interactions of composting and biochar and their implications for soil amendment and pollution remediation: a review. Crit Rev Biotechnol. 2017;37:754–764.
[8] Escapa A, Mateos R, Martínez EJ, et al. Microbial electrolysis cells: an emerging technology for wastewater treatment and energy recovery. From laboratory to pilot plant and beyond. Renew Sustain Energy Rev. 2016;55:942–956.
[9] Yi H, Yan M, Huang D, et al. Synergistic effect of artificial enzyme and 2D nano-structured Bi_2WO_6 for eco-friendly and efficient biomimetic photocatalysis. Appl Catal B: Environ. 2019;250:52–62.
[10] Yi H, Huang D, Qin L, et al. Selective prepared carbon nanomaterials for advanced photocatalytic application in environmental pollutant treatment and hydrogen production. Appl Catal B Environ. 2018;239:408–424.
[11] Yang C, Qian H, Li X, et al. Simultaneous removal of multicomponent VOCs in biofilters. Trends Biotechnol. 2018;36:673–685.
[12] Huang D, Guo X, Peng Z, et al. White rot fungi and advanced combined biotechnology with nanomaterials: promising tools for endocrine-disrupting compounds biotransformation. Crit Rev Biotechnol. 2018;38:671–689.
[13] Yongrui P, Zheng Z, Bao M, et al. Treatment of partially hydrolyzed polyacrylamide wastewater by combined Fenton oxidation and anaerobic biological processes. Chem Eng J. 2015;273:1–6.
[14] Bourgin M, Beck B, Boehler M, et al. Evaluation of a full-scale wastewater, treatment plant upgraded with ozonation and biological post-treatments: abatement of micropollutants, formation of transformation products and oxidation by-products. Water Res. 2018;129:486–498.
[15] Yi H, Jiang M, Huang D, et al. Advanced photocatalytic Fenton-like process over biomimetic hemin-Bi_2WO_6 with enhanced pH. J Taiwan Inst Chem Eng. 2018;93:184–192.
[16] Macias-Garcia A, Corzo MG, Dominguez MA, et al. Study of the adsorption and electroadsorption process of Cu (II) ions within thermally and chemically modified activated carbon. J Hazard Mater. 2017;328:46–55.
[17] Nancharaiah YV, Reddy G. Aerobic granular sludge technology: mechanisms of granulation and biotechnological applications. Bioresour Technol. 2018;247:1128–1143.
[18] Wang Q, Wei W, Gong Y, et al. Technologies for reducing sludge production in wastewater treatment plants: state of the art. Sci Total Environ. 2017;587–588:510–521.
[19] Alvarino T, Suarez S, Lema J, et al. Understanding the sorption and biotransformation of organic micropollutants in innovative biological wastewater treatment technologies. Sci Total Environ. 2018;615:297–306.
[20] El-Taliawy H, Casas ME, Bester K. Removal of ozonation products of pharmaceuticals in laboratory moving bed biofilm reactors (MBBRs). J Hazard Mater. 2018;347:288–298.
[21] Xu XC, Wang G, Zhou L, et al. Start-up of a full-scale SNAD-MBBR process for treating sludge digester liquor. Chem Eng J. 2018;343:477–483.
[22] Abu Bakar SNH, Abu Hasan H, Mohammad AW, et al. A review of moving-bed biofilm reactor technology for palm oil mill effluent treatment. J Clean Prod. 2018;171:1532–1545.
[23] Gaur N, Narasimhulu K, PydiSetty Y. Recent advances in the bio-remediation of persistent organic pollutants and its effect on environment. J Clean Prod. 2018;198:1602–1631.
[24] Alvarino T, Lema J, Omil F, et al. Trends in organic micropollutants removal in secondary treatment of sewage. Rev Environ Sci Biotechnol. 2018;17:447–469.
[25] Bering S, Mazur J, Tarnowski K, et al. The application of moving bed bio-reactor (MBBR) in commercial laundry wastewater treatment. Sci Total Environ. 2018;627:1638–1643.
[26] Mazioti AA, Stasinakis AS, Pantazi Y, et al. Biodegradation of benzotriazoles and

hydroxy-benzothiazole in wastewater by activated sludge and moving bed biofilm reactor systems. Bioresour Technol. 2015;192:627–635.

[27] Dias J, Bellingham M, Hassan J, et al. Influence of carrier media physical properties on start-up of moving attached growth systems. Bioresour Technol. 2018;266:463–471.

[28] Dias J, Bellingham M, Hassan J, et al. Impact of carrier media on oxygen transfer and wastewater hydrodynamics on a moving attached growth system. Chem Eng J. 2018;351:399–408.

[29] Tang K, Ooi GTH, Litty K, et al. Removal of pharmaceuticals in conventionally treated wastewater by a polishing moving bed biofilm reactor (MBBR) with intermittent feeding. Bioresour Technol. 2017;236: 77–86.

[30] Mazioti AA, Stasinakis AS, Psoma AK, et al. Hybrid moving bed biofilm reactor for the biodegradation of benzotriazoles and hydroxy-benzothiazole in wastewater. J Hazard Mater. 2017;323:299–310.

[31] Safwat SM. Performance of moving bed biofilm reactor using effective microorganisms. J Clean Prod. 2018;185:723–731.

[32] Casas ME, Chhetri RK, Ooi G, et al. Biodegradation of pharmaceuticals in hospital wastewater by staged moving bed biofilm reactors (MBBR). Water Res. 2015;83:293–302.

[33] Kowalski MS, Devlin TR, Oleszkiewicz JA. Start-up and long-term performance of anammox moving bed biofilm reactor seeded with granular biomass. Chemosphere. 2018;200:481–486.

[34] Bian W, Zhang S, Zhang Y, et al. Achieving nitritation in a continuous moving bed biofilm reactor at different temperatures through ratio control. Bioresour Technol. 2017;226:73–79.

[35] Ashfaq M, Li Y, Wang Y, et al. Monitoring and mass balance analysis of endocrine disrupting compounds and their transformation products in an anaerobic–anoxic–oxic wastewater treatment system in Xiamen, China. Chemosphere. 2018;204:170–177.

[36] Gong BZ, Wang YM, Wang JL, et al. Intensified nitrogen and phosphorus removal by embedding electrolysis in an anaerobic–anoxic–oxic reactor treating low carbon/nitrogen wastewater. Bioresour Technol. 2018;256:562–565.

[37] Sun YP, Guan YT, Zeng DF, et al. Metagenomics-based interpretation of AHLs-mediated quorum sensing in anammox biofilm reactors for low-strength wastewater treatment. Chem Eng J. 2018;344:42–52.

[38] Wu L, Li Z, Zhao C, et al. A novel partial-denitrification strategy for post-anammox to effectively remove nitrogen from landfill leachate. Sci Total Environ. 2018;633:745–751.

[39] Zhang X, Chen Z, Ma Y, et al. Acute and persistent toxicity of Cd(II) to the microbial community of anammox process. Bioresour Technol. 2018;261: 453–457.

[40] Shu DT, Yue H, He YL, et al. Divergent assemblage patterns of abundant and rare microbial sub-communities in response to inorganic carbon stresses in a simultaneous anammox and denitrification (SAD) system. Bioresour Technol. 2018;257:249–259.

[41] Xu J-J, Zhu X-L, Zhang Q-Q, et al. Roles of MnO_2 on performance, sludge characteristics and microbial community in anammox system. Sci Total Environ. 2018;633:848–856.

[42] Tomaszewski M, Cema G, Ziembińska-Buczyńska A. Significance of pH control in anammox process performance at low temperature. Chemosphere. 2017; 185:439–444.

[43] Li H, Yao H, Zhang D, et al. Short- and long-term effects of manganese, zinc and copper ions on nitrogen removal in nitritation-anammox process. Chemosphere. 2018;193:479–488.

[44] Xing B-S, Guo Q, Yang G-F, et al. The influences of temperature, salt and calcium concentration on the performance of anaerobic ammonium oxidation (anammox) process. Chem Eng J. 2015;265:58–66.

[45] Kartal B, Keltjens JT. Anammox biochemistry: a tale of heme c proteins. Trends Biochem Sci. 2016;41: 998–1011.

[46] Cao Y, Kwok Bee H, Yan Z, et al. Mainstream partial nitritation/anammox nitrogen removal process in the largest water reclamation plant in Singapore. J Beijing Univ Technol (China). 2015;41:1441–1447.

[47] Ma B, Wang S, Cao S, et al. Biological nitrogen removal from sewage via anammox: recent advances. Bioresour Technol. 2016;200:981–990.

[48] Tomaszewski M, Cema G, Ziembińska-Buczyńska A. Influence of temperature and pH on the anammox process: a review and meta-analysis. Chemosphere. 2017;182:203–214.

[49] Wang D, Wang Y, Liu Y, et al. Is denitrifying anaerobic methane oxidation-centered technologies a solution for the sustainable operation of wastewater treatment plants? Bioresour Technol. 2017;234: 456–465.

[50] Luo JH, Chen H, Hu SH, et al. Microbial selenate reduction driven by a denitrifying anaerobic methane oxidation biofilm. Environ Sci Technol. 2018;52:4006–4012.

[51] Luo JH, Chen H, Yuan ZG, et al. Methane-supported nitrate removal from groundwater in a membrane biofilm reactor. Water Res. 2018;132:71–78.

[52] Xie GJ, Cai C, Hu SH, et al. Complete nitrogen removal from synthetic anaerobic sludge digestion liquor through integrating anammox and denitrifying anaerobic methane oxidation in a membrane biofilm reactor. Environ Sci Technol. 2017;51:819–827.

[53] Lu Y-Z, Li N, Ding Z-W, et al. Tracking the activity of the anammox-DAMO process using excitation–emission matrix (EEM) fluorescence spectroscopy. Water Res. 2017;122:624–632.

[54] Ding J, Lu Y-Z, Fu L, et al. Decoupling of DAMO archaea from DAMO bacteria in a methane-driven microbial fuel cell. Water Res. 2017;110:112–119.

[55] Haroon MF, Hu SH, Shi Y, et al. Anaerobic oxidation of methane coupled to nitrate reduction in a novel archaeal lineage. Nature. 2013;500:567.

[56] Ettwig KF, Butler MK, Le Paslier D, et al. Nitrite-driven anaerobic methane oxidation by oxygenic bacteria. Nature. 2010;464:543.

[57] Wang Y, Wang D, Yang Q, et al. Wastewater opportunities for denitrifying anaerobic methane oxidation. Trends Biotechnol. 2017;35:799–802.
[58] Cui M, Ma A, Qi H, et al. Anaerobic oxidation of methane: an "active" microbial process. Microbiol Open. 2015;4:1–11.
[59] Wang C, Liu S, Xu X, et al. Achieving mainstream nitrogen removal through simultaneous partial nitrification, anammox and denitrification process in an integrated fixed film activated sludge reactor. Chemosphere. 2018;203:457–466.
[60] Zhang L, Liu M, Zhang S, et al. Integrated fixed-biofilm activated sludge reactor as a powerful tool to enrich anammox biofilm and granular sludge. Chemosphere. 2015;140:114–118.
[61] Ji G, Wu Y, Wang C. Analysis of microbial characterization in an upflow anaerobic sludge bed/biological aerated filter system for treating microcrystalline cellulose wastewater. Bioresour Technol. 2012;120: 60–69.
[62] Liu G-h, Ye Z, Tong K, et al. Biotreatment of heavy oil wastewater by combined upflow anaerobic sludge blanket and immobilized biological aerated filter in a pilot-scale test. Biochem Eng J. 2013;72: 48–53.
[63] Wu HS, Tu Y, Xu M, et al. Pilot-scale study on pretreatment of printing and dyeing wastewater by UASB. Environ Sci Technol. 2010;33:146–149.
[64] Shen DS, Liu XW, He YH. Studies on adsorption, desorption and biodegradation of pentachlorophenol by the anaerobic granular sludge in an upflow anaerobic sludge blanket (UASB) reactor. J Hazard Mater. 2005;125:231–236.
[65] Khorsandi H, Movahedyan H, Bina B, et al. Innovative anaerobic upflow sludge blanket filtration combined bioreactor for nitrogen removal from municipal wastewater. Int J Environ Sci Technol. 2011;8: 417–424.
[66] Tong K, Zhang YH, Liu GH, et al. Treatment of heavy oil wastewater by a conventional activated sludge process coupled with an immobilized biological filter. Int Biodeterior Biodegrad. 2013;84:65–71.
[67] Yang B, Wang JZ, Wang JF, et al. Correlating microbial community structure with operational conditions in biological aerated filter reactor for efficient nitrogen removal of municipal wastewater. Bioresour Technol. 2018;250:374–381.
[68] Bae W, Han D, Kim E, et al. Enhanced bioremoval of refractory compounds from dyeing wastewater using optimized sequential anaerobic/aerobic process. Int J Environ Sci Technol. 2016;13:1675–1684.
[69] Usman M, Hanna K, Haderlein S. Fenton oxidation to remediate PAHs in contaminated soils: a critical review of major limitations and counter-strategies. Sci Total Environ. 2016;569:179–190.
[70] Venny Gan S, Ng HK. Modified Fenton oxidation of polycyclic aromatic hydrocarbon (PAH)-contaminated soils and the potential of bioremediation as post-treatment. Sci Total Environ. 2012;419:240–249.
[71] Esteves BM, Rodrigues CSD, Boaventura RAR, et al. Coupling of acrylic dyeing wastewater treatment by heterogeneous Fenton oxidation in a continuous stirred tank reactor with biological degradation in a sequential batch reactor. J Environ Manage. 2016; 166:193–203.
[72] Qian XF, Ren M, Fang MY, et al. Hydrophilic mesoporous carbon as iron(III)/(II) electron shuttle for visible light enhanced Fenton-like degradation of organic pollutants. Appl Catal B: Environ. 2018;231:108–114.
[73] Zhang AY, Huang NH, Zhang C, et al. Heterogeneous Fenton decontamination of organoarsenicals and simultaneous adsorption of released arsenic with reduced secondary pollution. Chem Eng J. 2018;344: 1–11.
[74] Espinosa JC, Catala C, Navalon S, et al. Iron oxide nanoparticles supported on diamond nanoparticles as efficient and stable catalyst for the visible light assisted Fenton reaction. Appl Catal B: Environ. 2018; 226:242–251.
[75] Yusoff N, Ong S-A, Ho L-N, et al. Evaluation of biodegradation process: comparative study between suspended and hybrid microorganism growth system in sequencing batch reactor (SBR) for removal of phenol. Biochem Eng J. 2016;115:14–22.
[76] Azizi A, Moghaddam MRA, Maknoon R, et al. Comparison of three combined sequencing batch reactor followed by enhanced Fenton process for an azo dye degradation: bio-decolorization kinetics study. J Hazard Mater. 2015;299:343–350.
[77] Mo R, Huang S, Dai W, et al. A rapid Fenton treatment technique for sewage sludge dewatering. Chem Eng J. 2015;269:391–398.
[78] Bae W, Won H, Hwang B, et al. Characterization of refractory matters in dyeing wastewater during a full-scale Fenton process following pure-oxygen activated sludge treatment. J Hazard Mater. 2015;287: 421–428.
[79] Coledam DAC, Sanchez-Montes I, Silva BF, et al. On the performance of HOCl/Fe^{2+}, HOCl/Fe^{2+}/UVA, and HOCl/UVC processes using in situ electrogenerated active chlorine to mineralize the herbicide picloram. Appl Catal B Environ. 2018;227:170–177.
[80] Yue DT, Qian XF, Kan M, et al. A metal-free visible light active photo-electro-Fenton-like cell for organic pollutants degradation. Appl Catal B Environ. 2018; 229:211–217.
[81] Cheng M, Zeng G, Huang D, et al. Degradation of atrazine by a novel Fenton-like process and assessment the influence on the treated soil. J Hazard Mater. 2016;312:184–191.
[82] Umar M, Aziz HA, Yusoff MS. Trends in the use of Fenton, electro-Fenton and photo-Fenton for the treatment of landfill leachate. Waste Manage. 2010; 30:2113–2121.
[83] Vilar VJP, Moreira FC, Ferreira ACC, et al. Biodegradability enhancement of a pesticide-containing bio-treated wastewater using a solar photo-Fenton treatment step followed by a biological oxidation process. Water Res. 2012;46:4599–4613.
[84] Shi Y, Huang C, Rocha KC, et al. Treatment of oil sands process-affected water using moving bed biofilm reactors: with and without ozone pretreatment. Bioresour Technol. 2015;192:219–227.

[85] Zhang S, Zheng J, Chen Z. Combination of ozonation and biological aerated filter (BAF) for bio-treated coking wastewater. Sep Purif Technol. 2014;132: 610–615.

[86] Zhu H, Ma W, Han H, et al. Catalytic ozonation of quinoline using nano-MgO: efficacy, pathways, mechanisms and its application to real biologically pretreated coal gasification wastewater. Chem Eng J. 2017;327:91–99.

[87] Saeid S, Tolvanen P, Kumar N, et al. Advanced oxidation process for the removal of ibuprofen from aqueous solution: a non-catalytic and catalytic ozonation study in a semi-batch reactor. Appl Catal B: Environ. 2018;230:77–90.

[88] Zhang SY, Wu CY, Zhou YX, et al. Effect of wastewater particles on catalytic ozonation in the advanced treatment of petrochemical secondary effluent. Chem Eng J. 2018;345:280–289.

[89] Wu C, Zhou Y, Wang Y, et al. Innovative combination of Fe^{2+}-BAF and ozonation for enhancing phosphorus and organic micropollutants removal treating petrochemical secondary effluent. J Hazard Mater. 2017;323:654–662.

[90] Wu Q, Li W-T, Yu W-H, et al. Removal of fluorescent dissolved organic matter in biologically treated textile wastewater by ozonation-biological aerated filter. J Taiwan Inst Chem Eng. 2016;59:359–364.

[91] Marcelino RBP, Leao MMD, Lago RM, et al. Multistage ozone and biological treatment system for real wastewater containing antibiotics. J Environ Manage. 2017;195:110–116.

[92] Bu F, Gao B, Shen X, et al. The combination of coagulation and ozonation as a pre-treatment of ultrafiltration in water treatment. Chemosphere. 2019;231:349–356.

[93] Wu C, Zhou Y, Sun X, et al. The recent development of advanced wastewater treatment by ozone and biological aerated filter. Environ Sci Pollut Res. 2018; 25:8315–8329.

[94] Camera-Roda G, Loddo V, Palmisano L, et al. Photocatalytic ozonation for a sustainable aquaculture: a long-term test in a seawater aquarium. Appl Catal B Environ. 2019;253:69–76.

[95] Malik SN, Khan SM, Ghosh PC, et al. Treatment of pharmaceutical industrial wastewater by nano-catalyzed ozonation in a semi-batch reactor for improved biodegradability. Sci Total Environ. 2019;678: 114–122.

[96] Merten J, Johnson B. Laser continuum source atomic absorption spectroscopy: measuring the ground state with nanosecond resolution in laser-induced plasmas. Spectrochim Acta B. 2018;139:38–43.

[97] Sahu PK, Ramisetti NR, Cecchi T, et al. An overview of experimental designs in HPLC method development and validation. J Pharm Biomed Anal. 2018; 147:590–611.

[98] Cajka T, Fiehn O. Toward merging untargeted and targeted methods in mass spectrometry-based metabolomics and lipidomics. Anal Chem. 2016;88: 524–545.

[99] Lai C, Liu X, Qin L, et al. Chitosan-wrapped gold nanoparticles for hydrogen-bonding recognition and colorimetric determination of the antibiotic kanamycin. Microchim Acta. 2017;184:2097–2105.

[100] Qin L, Zeng G, Lai C, et al. A visual application of gold nanoparticles: simple, reliable and sensitive detection of kanamycin based on hydrogen-bonding recognition. Sens Actuators B Chem. 2017;243: 946–954.

[101] Xie S, Tang Y, Tang D, et al. Highly sensitive impedimetric biosensor for Hg^{2+} detection based on manganese porphyrin-decorated DNA network for precipitation polymerization. Anal Chim Acta. 2018; 1023:22–28.

[102] Jia TT, Cai ZM, Chen XM, et al. Electrogenerated chemiluminescence ethanol biosensor based on alcohol dehydrogenase functionalized $Ru(bpy)_3^{2+}$ doped silica nanoparticles. Biosens Bioelectron. 2009;25: 263–267.

[103] Chaichi MJ, Alijanpour SO. Glucose chemiluminescence biosensor based on covalent immobilization of enzyme in glutaraldehyde-functionalized glass cell and direct coupling of chitosan-induced Au/Ag alloy nanoparticles. J Anal Chem. 2016;71:163–171.

[104] Bidmanova S, Kotlanova M, Rataj T, et al. Fluorescence-based biosensor for monitoring of environmental pollutants: from concept to field application. Biosens Bioelectron. 2016;84:97–105.

[105] Zhang J, Khan I, Zhang Q, et al. Lipopolysaccharides detection on a grating-coupled surface plasmon resonance smartphone biosensor. Biosens Bioelectron. 2018;99:312–317.

[106] Hwang MT, Wang ZJ, Ping JL, et al. DNA nanotweezers and graphene transistor enable label-free genotyping. Adv Mater. 2018;30:e1802440.

[107] Tsopela A, Lale A, Vanhove E, et al. Integrated electrochemical biosensor based on algal metabolism for water toxicity analysis. Biosens Bioelectron. 2014;61: 290–297.

[108] Ali S, Hassan A, Hassan G, et al. Disposable all-printed electronic biosensor for instantaneous detection and classification of pathogens. Sci Rep. 2018;8: 5920.

[109] Kuo Y-C, Lee C-K, Lin C-T. Improving sensitivity of a miniaturized label-free electrochemical biosensor using zigzag electrodes. Biosens Bioelectron. 2018; 103:130–137.

[110] Xu Z, Liu Y, Williams I, et al. Disposable self-support paper-based multi-anode microbial fuel cell (PMMFC) integrated with power management system (PMS) as the real time "shock" biosensor for wastewater. Biosens Bioelectron. 2016;85:232–239.

[111] Yang W, Wei X, Choi S. A dual-channel, interference-free, bacteria-based biosensor for highly sensitive water quality monitoring. IEEE Sens J. 2016;16: 8672–8677.

[112] Baek S, Kwon S-R, Yeon SY, et al. Miniaturized reverse electrodialysis-powered biosensor using electrochemiluminescence on bipolar electrode. Anal Chem. 2018;90:4749–4755.

[113] Urrutia A, Goicoechea J, Rivero PJ, et al. Optical fiber sensors based on gold nanorods embedded in polymeric thin films. Sens Actuators B Chem. 2018;255: 2105–2112.

[114] Reta N, Saint CP, Michelmore A, et al. Nanostructured electrochemical biosensors for label-free detection of water- and food-borne pathogens. ACS Appl Mater Interfaces. 2018;10:6055–6072.

[115] Wang YH, Huang KJ, Wu X. Recent advances in transition-metal dichalcogenides based electrochemical biosensors: a review. Biosens Bioelectron. 2017;97:305–316.

[116] Ammam M. Electrochemical and electrophoretic deposition of enzymes: principles, differences and application in miniaturized biosensor and biofuel cell electrodes. Biosens Bioelectron. 2014;58:121–131.