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REVIEW ARTICLE

Examining current trends and future outlook of bio-electrochemical systems (BES) for nutrient conversion and recovery: an overview

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Abstract

Nutrient-rich waste streams from domestic and industrial sources and the increasing application of synthetic fertilizers have resulted in a huge-scale influx of reactive nitrogen and phosphorus in the environment. The higher concentrations of these pollutants induce eutrophication and foster degradation of aquatic biodiversity. Besides, phosphorus being non-renewable resource is under the risk of rapid depletion. Hence, recovery and reuse of the phosphorus and nitrogen are necessary. Over the years, nutrient recovery, low-carbon energy, and sustainable bioremediation of wastewater have received significant interest. The conventional wastewater treatment technologies have higher energy demand and nutrient removal entails a major cost in the treatment process. For these issues, bio-electrochemical system (BES) has been considered as sustainable and environment friendly wastewater treatment technologies that utilize the energy contained in the wastewater so as to recovery nutrients and purify wastewater. Therefore, this article comprehensively focuses and critically analyzes the potential sources of nutrients, working mechanism of BES, and different nutrient recovery strategies to unlock the upscaling opportunities. Also, economic analysis was done to understand the technical feasibility and potential market value of recovered nutrients. Hence, this review article will be useful in establishing waste management policies and framework along with development of advanced configurations with major emphasis on nutrient recovery rather than removal from the waste stream.

Keywords Bioelectrochemical system · Nutrient · Nitrogen removal · Phosphorus recovery · Struvite · Vivianite · Hybrid configurations · Economic analysis

Abbreviations

WWTPs	Wastewater treatment plants
GHGs	Greenhouse gasses
DO	Dissolved oxygen
BES	Bioelectrochemical system
MFC	Microbial fuel cell
EAB	Electroactive bacteria
FAO	Food and agriculture organization
TKN	Total kjeldahl nitrogen
MDC	Microbial desalination cell
MES	Microbial electrosynthesis cell
MEC	Microbial electrolysis cell
NOB	Nitrite oxidizing bacteria
AOB	Ammonium oxidizing bacteria
EET	Extracellular electron transfer
OLR	Organic loading rate
SND	Simultaneous nitrification and denitrification
ANAMMOX	Anaerobic ammonium oxidation
CNT	Carbon nanotube
TOC	Total organic carbon
SEM	Scanning electron microscopy
CB	Carbon brush
GF	Graphite felt
TN	Total nitrogen
TP	Total phosphorus
IEM	Ion exchange membrane
COD	Chemical oxygen demand
HRT	Hydraulic retention time
DNRA	Dissimilatory nitrate reduction to ammonium
ADB	Aerobic denitrification bacterium
MBER	Membrane bioelectrochemical reactor
EMP	Electrochemically mediated precipitation
HAP	Hydroxyapatite

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PAFC	Poly aluminum ferric chloride
ORR	Oxygen reduction reaction
GAC	Granular activated carbon
PMFC	Photoautotrophic microbial fuel cell
MFC-IFAC	Microbial fuel cell integrated fixed film activated sludge
FO	Forward osmosis
AAFO-MFC	Anaerobic acidification and FO membrane microbial fuel cell
OsMFC	Osmotic microbial fuel cell
C/N	Carbon/nitrogen
MS	Microbial separator
ACMFC	Air cathode microbial fuel cell
MA-MFC	Multi anode microbial fuel cell
FA-MFC	Flat panel microbial fuel cell
ABMFC	Algae biofilm microbial fuel cell
SPEEK	Sulfonated polyether ketone
SMDC	Submersible microbial desalination cell
DC	Direct current
AC	Alternating current

Introduction

The surge in the nutrient concentration in the aquatic bodies, attributable to anthropogenic activities, has posed an environmental concern (Fallahi et al. 2021). In water, nitrogen, and phosphorus exist as ammonium and phosphate ions (Ye et al. 2019). Excessive nutrient concentration induces eutrophication that causes degradation of the freshwater ecosystem due to algal bloom. The decomposition and decay of algal biomass result in decrease in oxygen levels in the water bodies that leads to loss of macro and micro-organism biodiversity. Furthermore, the production of toxins by cyanobacteria and blue-green algae also deteriorates water quality (Rahimi et al. 2020). Besides, excess of nitrate-nitrogen (N O_3^--N) species in drinking water can cause blue baby syndrome (methemoglobinemia) in infants, miscarriages and can damage the thyroid gland by impeding the uptake of iodine (Rout et al. 2021) whereas osteoporosis and kidney damage are associated with excessive phosphate uptake (Bhambri and Karn 2020). The major dominating source of nitrogen and phosphorus are effluents from agricultural fields, discharge of domestic and industrial wastewater, livestock farming, aquaculture and atmospheric deposition (Bhambri and Karn 2020; Rout et al. 2021). Globally, the major industries contributing to higher nutrient loading in water bodies include dairy industries, paper and pulp industries, food and beverages industries, and slaughterhouses (Bhambri and Karn 2020). Correspondingly, the contribution of nitrogen loading into groundwater and surface water by municipal wastewater treatment plants (WWTPs) is approximately 80,000 tonnes/year (Rahimi et al. 2020). Therefore, nutrient removal/recovery from these waste streams is indispensable to protect the natural aquatic ecosystem and to avoid the pernicious impact on the environment.

In fact, across the world, approximately half of the food production depends on the usage of synthetic fertilizer (Paucar and Sato 2021). Ammonia-based fertilizer are generally employed in large scale to boost the crop yield as well as to ensure food security (Bruning et al. 2012; Paucar and Sato 2021). At present, the industrial Haber–Bosch process is used for the manufacturing of fertilizer in which atmospheric nitrogen (N_2) is converted to ammonia (NH_3) (Paucar and Sato 2021). The ammonia production consumes around 35–50 MJ/kg.N which holds 2% of global energy (Yan et al. 2018). This manufacturing process is cost-intensive as the reaction mandates high pressure and temperature (Paucar and Sato 2021). In addition, the Haber process is unsustainable as it significantly emits greenhouse gasses (GHGs) of around 1.5 t CO_2eq/t NH_3 which is around 1.2% of global GHG emissions (El-Qelish and Mahmoud 2022). The huge energy requirement of this process can be saved and release of GHG emissions can be prevented by direct recovery of N H_3 from the nutrient-rich wastewater (Bruning et al. 2012; Tang et al. 2023).

On the other hand, the major source for the extraction of phosphorus is from the non-renewable phosphate rock deposits which are present only in a few countries (China, Morocco, Syria, Algeria, and Irag) and are limited in quantity (Nancharaiah et al. 2016; El-Qelish and Mahmoud 2022). For meeting the internal phosphorus demand, India and Western Europe are completely relying on imports (Nancharaiah et al. 2016). Furthermore, phosphorus is vital for crop yields as well as for all living entities and there is no alternative for it (Paucar and Sato 2021). Studies have predicted that by 2050, the world population can rise to 9 billion, therefore ensuring a phosphorus supply will be pivotal for future food security. The WWTPs being an untapped source of phosphorus has a global potential of 3 million tons of phosphate annually. Around 15–20% of the global demand for phosphorus can be met if the total phosphorus existing in the sewage is completely recovered (Paucar and Sato 2021). Therefore, the prevailing crisis demands urgent investigation of the nutrient management options and for scrutinizing the nutrient recovery feasibility from organic matter waste streams to meet the nutrient demand.

At present, conventional phosphorus removal processes, from wastewater, via biological uptake and chemical precipitation whereas ammonium removal via ammonia stripping, nitrification, and denitrification, requires an exorbitant amount of energy and chemicals (Paucar and Sato 2021). Moreover, conventional nitrogen removal process requires internal circulation from aerobic to anoxic tank which leads to increase in pumping cost as well as sub-optimal

denitrification performance because of inadequate availability of substrate due to competition between the anaerobic and aerobic microorganism for substrate oxidation. Consequently, additional carbon dosing such as molasses, ethanol, or methanol are added to maintain ideal C/N ratio to ensure complete denitrification. Thus, treatment cost increases as well as safety issues pertaining to storage and transportation of chemicals became a major concern. In fact, higher dissolved oxygen (DO) level in the recirculated water from aerobic tank builds an unfavorable environment for the denitrification performance at the anoxic tank (Nguyen and Babel 2022; Virdis et al. 2011). In the biological phosphorus removal method, excess sludge production is the major drawback (Yan et al. 2018; Ye et al. 2019). In addition, membrane application is ineffective and only 10% of total nutrients are removed and complexities like membrane fouling are also involved (Rout et al. 2021). In ion exchange or electrodialysis, nutrient recovery is a major challenge owning to opposite ionic charges between nitrate/nitrite and ammonium (Li et al. 2020). The higher capital, operational, and maintenance costs allied with these technologies impose financial constraints for wider application in developing countries (Rout et al. 2021). It was reported that in the United States, an extra 4% of electricity is required for nitrogen removal in WWTPs (Yan et al. 2018). The study revealed that an energy supply of 45 MJ/N·kg is utilized for nitrogen removal from wastewater. Also, the removal of nutrients may exacerbate global warming due to the release of 0.9 kg CO_2/m^3 as a repercussion of these processes. Therefore, recovery of the nutrient is valuable than removal of nutrient as it (1) can ensure food security by producing nutrient-based fertilizer (2) can reduce the environmental footprints of WWTPs by lowering excess sludge production and minimizing eutrophication potential (Yan et al. 2018; Ye et al. 2019).

Thus, it is imperative to devise a novel sustainable method for nutrient recovery specifically nitrogen and phosphorus from agricultural sources and WWTPs to strengthen the idea of a circular bioeconomy (Fallahi et al. 2021; Rout et al. 2021). Furthermore, limiting the environmental contamination arising due to the release of nutrients is a challenging task and is a subject of national importance (Rout et al. 2021). A bio-electrochemical system (BES) is an economical and sustainable technology that not only obtains higher nutrient recovery, but also reduces the chemical and energy requirement associated with nutrient recovery (El-Qelish and Mahmoud 2022). Microbial fuel cell (MFC), a type of BES, involves the application of electroactive bacteria (EAB) as a biocatalyst for electricity generation which impels the ionic migration through ion exchange membrane and leads to wastewater purification and valuable product recovery with a minimum carbon footprint. In addition, the recovered energy from the MFCs can be utilized for its maintenance and operation (Ye et al. 2019). Additionally, in MFC, the separation of electrolyte and bacteria in two chambers facilitates decrease in oxidation of organic carbon by aerobic microorganisms as the competition between anaerobic and aerobic microorganisms for substrate oxidation is minimized. Therefore, the requirement for carbon/nitrogen (C/N) ratio is lowered as compared to traditional nitrogen removal technologies (Nguyen and Babel 2022; Virdis et al. 2011). Moreover, several other advantages of BES application for nutrient removal and recovery includes lower sludge production, valuable resource recovery and electricity generation whereas major drawback that restricts it large scale application is higher operational expense, lower and unstable power density as well as membrane fouling. These associated drawbacks can be solved by incorporating economical reactor components or by integrating MFC with other treatment technologies (Baby and Ahammed 2022; Deng et al. 2023).

The growing environmental concern along with broader scope and significant rise in research interest in this domain makes it important to timely review the ongoing advancements in BES technology for nutrient recovery. This review is categorized into several sections, originating with brief introduction in the "Introduction" section which explicate the detrimental effect of increased nutrient loadings on the ecosystem, drawbacks of conventional technologies and need for nutrient management technology. The "Assessing regional nutrient demand and quantifying nutrients from potential sources" section elucidates the sources and quantification of nutrients followed by concise description of working mechanism and advancement of BES in the "Principles and advancement of BES" section; the "Function of diverse microbial consortia" section discusses the functions and enrichment strategies of biocatalyst whereas different mechanisms of nutrient recovery and their governing factors along with performance evaluation of different reactor configurations are included in the "Nutrient recovery" section; the "Nutrient removal/recovery using hybrid configurations" section explains the importance and requirement of different hybrid configuration of MFC for nutrient recovery. At the end, economic feasibility of BES for nutrient recovery is covered in the "Economic analysis" section. This study comprehensively recapitulates the fundamental aspects of BES, influential factors and investigates the critical concepts in nutrient recovery/removal using BES.

Despite the number of published articles on the BES, there still exists a broader gap in terms of performance assessment of the different configurations of reactors and their nutrient removal mechanisms. Also, global nutrient demand and in-depth detailing of the role of governing factors were not focused in the past literature. Therefore, the present review systematically summarizes the recent innovations, assesses nutrient demand and economic viability as well as highlights the future research scope to fulfill

the lacunas existing in the past studies. Furthermore, this review article will provide a generic roadmap for developing a robust BES system and different integrated configurations for optimizing and boosting nutrient recovery, bio-electricity generation, and wastewater treatment.

Assessing regional nutrient demand and quantifying nutrients from potential sources

The expeditious growth in the population throughout the world has triggered a rise in demand for nutrient-based fertilizers of around 1.8% per year for the production of food (Yan et al. 2018). Figure 1a and b demonstrates the statistics of global nutrient demand and potential balances which was reported in the Food and Agriculture Organization (FAO) outlook to 2022 (FAO 2019). Among all, Asia exhibits higher nitrogen and phosphorus demand, i.e., 61.976 and 25.099 million tonnes, respectively against its supply capability of 67.893 and 23.007 million tonnes respectively. Besides, America displays second largest nutrient demand, i.e., 25 million tonnes nitrogen and 12 million tonnes phosphorus against the limited supply capability of 17.319 million tonnes nitrogen and 6.683 million tonnes phosphorus, thus resulting in negative nutrient balance. Furthermore, in the list of top countries on the basis of percentage share in nitrogen consumption for agriculture from 2001 to 2020, China (39.21%) holds first position followed by India (21.59%), USA (15.98%), Brazil (4.78%), Pakistan (4.23%), Indonesia (3.79%), and Canada (3.06%) and in case of percentage share in phosphorus consumption, the order remains same except Brazil (13.15%) come ahead of USA (12.72%) as shown in Fig. 1c.

In order to meet the fertilizer demand, there is a need to explore the sustainable potential sources for extraction of nutrients. Globally the wastewater generation is projected to rise by 24% and 51% in 2030 and 2050, respectively (Qadir et al. 2020). Wastewater is considered to be a rich source of nutrients (Batstone et al. 2015) and these nutrients find their pathway to natural water bodies from different point and non-point sources. Table 1 provides the insights into the potential of different types of wastewaters as nutrient source (N and P). For domestic wastewater, the nutrients concentration varies from region to region depending on socio-economic factors, climate etc. On an average, the TN and TP concentration in domestic wastewater ranges from 35 to 100 and 18 to 29 mg/L, respectively (Rout et al. 2021).

Studies have reported that the nutrient contribution of human urine to municipal wastewater is around 40–50% of phosphorus, 54–90% of potassium, and 75–87% of total nitrogen (Kumar and Pal 2015). Out of the total wastewater entering a treatment plant, human urine accounts for only 1%

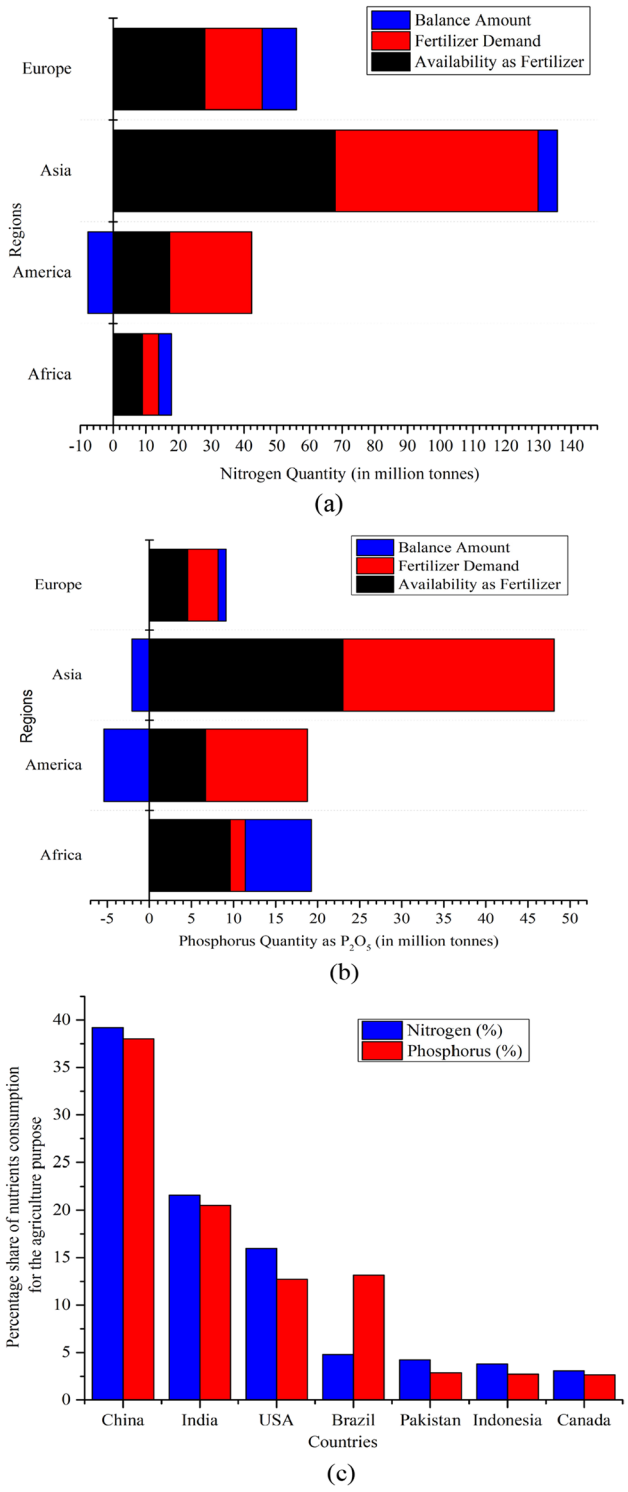


Fig. 1 Forecast of global requirement and availability of **a** nitrogen, **b** phosphorus as P_2O_5 of year 2022 (FAO, 2019), and **c** percentage share of nutrient consumption by top countries for agriculture purpose from 2001–2020 (FAOSTAT, 2022)

Table 1 Nutrient concentration in different waste stream

Category of the wastewater	Details	Total nitrogen (TN), mg/L	Total phosphorus (TP), mg/L	Reference
Municipal wastewater	Sewage	19.7–78.4	5–20	(Minhas et al. 2022; Cai et al. 2013)
	Urine	4000–13,900	350–2500	(Kundu et al. 2022)
	Domestic wastewater	35–100	18–29	(Rout et al. 2021)
	Landfill Leachate	100–500	1–10	(Drexler et al. 2014)
Industrial wastewater	Paper Mill	1.1–10.9	0.6–5.8	(Cai et al. 2013)
	Textile	10–20	< 10	(Yaseen and Scholz 2019)
	Tannery	545	0.4–21	(Alemu et al. 2019; Carrillo et al. 2020)
	Olive Mill	131–2900*	64–350	(Carrillo et al. 2020)
Animal wastewater	Dairy	65–3305	12–226	(Carrillo et al. 2020)
	Poultry	98–1825	15–446	
	Swine	2350–3570	194–780	

**TKN* total kjeldahl nitrogen

and is the largest source of nutrient loading to the wastewater. Nutrient recovery from urine is more feasible if it is separated from other wastewater streams in domestic household such as shower and kitchen wastewater. Therefore, source separation of urine is considered to be an effective method to get a nutrient-rich source of wastewater (Kundu et al. 2022; Kumar and Pal 2015). Studies have reported that 22% of global phosphorus demand can be met through phosphorus recovery from human waste, i.e., feces and urine (Kundu et al. 2022). Furthermore, leachate, arising from the water percolation through the municipal solid waste landfill contains higher concentrations of P O_4^{3-}, N H_4^+, organic matter and salt (Kumar and Pal 2015).

The wastewater characteristics varies based on the source of its origin (Drexler et al. 2014; Rout et al. 2021). The effluents from animal and plant product based industries contains greater total nitrogen and phosphorus concentration (Carrillo et al. 2020; Kumar and Pal 2015). Agricultural wastewater, mainly originating from livestock farming is also rich in nutrients. In case of dairy, beef feedlot and swine wastewater, the nitrogen to phosphorus ratio (N/P) ranges from 2 to 8 and is recognized as one of the major contamination sources around the livestock farms, that stimulates eutrophication of aquatic bodies (Cai et al. 2013; Kumar and Pal 2015).

Presently, nutrient recovery does not exist in the majority of WWTPs across the globe as they mainly focus on nutrient removal to reduce the environmental impact. Therefore, recovery of resources from the wastewater is under-explored, and at present, paradigm shift is under progress from an outlook that contemplate wastewater as undesirable streams to be treated and disposed of, to rising interest in wastewater for recovery of nutrient and energy to promote circular bioeconomy and for improving water quality by reducing nutrient pollution (Qadir et al. 2020; Kundu et al. 2022). Currently, for nutrient recovery and removal from domestic and industrial wastewater, BES based technology has received significant attention. In order to achieve maximum nutrient recovery from the aforementioned potential sources, a thorough understanding of BES is of paramount importance. Therefore, the next section briefly elucidates the working mechanism and advancement of BES.

Principles and advancement of BES

Bio-electrochemical systems (BES) can be defined as an electrochemical cell that utilizes electroactive microorganisms (EAMs) as biocatalyst on either or both the electrodes (Bajracharya et al. 2016). These electroactive microorganisms tend to develop biofilm over the surface of the electrodes to facilitate the transmission of electrons (Cabrera et al. 2021). BES converts chemical energy stored in the organic waste into electrical energy and vice versa (Bajracharya et al. 2016). BES comprises of anode, cathode and electrolytes. In case of dual chamber BES, the anodic and cathodic chamber are separated by a membrane either cation exchange membrane or proton exchange membrane. At anode, microorganisms oxidizes the organic substrate and induces generation of electrons, protons and other metabolic products (Cabrera et al. 2021). The liberated proton migrates towards cathodic chamber through ion exchange membrane to maintain electroneutrality while the generated electron moves via external circuit towards cathode and facilitates reduction of electron acceptors such as oxygen and nitrate. This flow of electron generates an electric current and power when load is applied to the external circuit (Bajracharya et al. 2016). In BES, either reductive or oxidative current can be generated. In case of oxidative current, EAM forms biofilm at anode, i.e., positive potential and nutrients present in the solution are oxidized, thus leads to production

of oxidative current, with electrode being terminal electron acceptor whereas in the latter case, when microorganism are grown at cathode, i.e., negative potential, the electrode acts as reducing agent and generates reductive current (Doyle and Marsili 2015).

BES offers various opportunities for clean and efficient generation of valuable chemicals and fuels using microbial consortia (Bajracharya et al. 2016). In BES, treatment efficiency varies depending on various factors such as strength and characteristics of wastewater, temperature, reactor configuration, and hydraulic retention time. BESs can attain higher organic matter removal efficiency in case of low-strength wastewater or in presence of simpler organic compounds while removal efficiency declines when handling complex organic substrates and high-strength wastewater. The nutrients removal mainly nitrogen and phosphorus are difficult in anode chamber as their removal requires different transformation process, i.e., nitrification and denitrification. Therefore, anode chamber effluent is partially treated, which still comprises of residual matter such as nutrients and organic matter. Studies have reported that the nutrients and organic matter removal efficiency enhances with cathodic treatment which is categorized into cathode-stimulated and cathode-supported treatment (Jain and He 2018).

Cathode-stimulated treatment is a reduction reaction process and is feasible in presence of biological or abiotic catalyst which can lower the activation energy and thus, facilitates transfer of electron from cathode to terminal electron acceptor. At cathode, nitrate reduction to nitrite was first demonstrated in BES using *Geobacter metallireducens* as pure culture (Jain and He 2018). Basically, for nitrate removal, two different types of streams have been studied in BES cathode. Firstly, domestic wastewater in which ammonia is dominant, initially undergoes nitrification in external nitrifying reactor and subsequently denitrification in cathodic chamber (Jain and He 2018). Secondly, nitrate contamination in groundwater is prominent and therefore bio-electrochemical denitrification can be performed directly in the cathode chamber. In general, BES cathode exhibits a major role in eliminating emerging contaminants and biorefractory organic compounds (Jain and He 2018).

Cathodic supported treatment takes benefit of physical space of cathodic chamber and does not involve direct electron acceptance from the cathode electrode. The various examples of such treatment technique are loop operation, membrane integration, advanced oxidation and algal cultivation. In case of loop operation, anodic effluent is sent to cathodic chamber to solve the issue of inadequate organic matter removal in anode chamber. Under advanced oxidation process, production of reactive radicals and strong oxidants can remove recalcitrant compounds. Apart from these, algal cultivation in cathodic chamber can perform dual role (1) provide dissolved oxygen for reduction reaction (2) nutrient removal due to nutrients uptake by algae for algal growth. Furthermore, membrane/biocathode integration can eradicate broad range of contaminants and produce clear effluents (Jain and He 2018). In general various configurations of BES exists such as microbial desalination cell (MDC), microbial fuel cell (MFC), microbial electrolysis cell (MEC), and microbial electrosynthesis cell (MES) (Cabrera et al. 2021). In all these microbial electrochemical technologies, the microbial-electrode interaction is critical for the system performance (Chiranjeevi and Patil 2020). Therefore, next section explicates the function and enrichment strategies of microbial consortia.

Function of diverse microbial consortia

In BES, microbes play a prominent role in oxidizing the organic contaminants into anions and cations at anode (Ivase et al. 2020). Additionally, at cathode, the inorganic carbon can be electrochemically reduced by bacteria to valuable products and fuels such as volatile fatty acids, alcohol etc. Some of these chemical reactions occurs spontaneously or requires application of external energy (Chatterjee et al. 2019; Logan 2019). In addition, diverse variety of microorganisms (bacteria, eukaryotic, and archaeal) can generate electric current as well as can transfer and receiving electrons to and from the anode and cathode respectively. The former is referred as exoelectrogens and the latter is electrotrophs (Logan 2019). Out of total bacterial abundance, the exo-electrogens accounts only for 0.26–7.70% in the environmental samples. In the initial stage of biofilm formation, planktonic exoelectrogens attach on to the electrode to differentiate themselves from the non-exoelectrogens. Furthermore, the community dominance is strengthened by continuous substrate supply followed by the maturation phase (Yan et al. 2020).

In both anodic and cathodic chamber, *Proteobacteria* have been found to be dominant bacterial population. In biofilm anode, β-proteobacteria is spotted frequently. This subcategory includes various bacteria which are efficient in organic matter removal. Moreover, it was reported that thauera genera accounts for 70.1% of total β-proteobacteria. Besides, *Desulfovibrio and Geobacter* come under the sub category δ-proteobacteria, while *Shewanella* and *Pseudomonas* belongs to subcategory ϒ-proteobacteria. The common denitrifiers are *Pseudomonas* and *Thauera* genera which have potential to reduce nitrate under aerobic environment. In general, most aerobic denitrifiers are heterotrophic in nature, but *Pseudomonas* can exhibit autotrophic denitrification using CO_2 and H_2 as carbon source and electron donor respectively (Nguyen and Babel 2022).

At the cathode, nitrite-oxidizing bacteria (NOB) and ammonium-oxidizing bacteria (AOB) genetically falls

under the subcategory α-proteobacteria and β-proteobacteria respectively. Under AOB, the most frequently detected microorganism is *Nitrosomonas* genera and it is an autotrophic denitrifier that can carry out reduction of nitrite to nitrogen gas. In addition, *Nitrosopira*, *Phycisphaera*, *Nitrosococcus*, *Nitrosolobus*, *Nitrosovibrio*, *Truepera*, and *Aquamicrobium* were also discovered to be AOB. On the other hand, NOB comprises of *Nitrocystis*, *Nitrococcus*, *Nitrobacter*, *Nitrospina*, and *Nitrospira*. On increasing the dissolved oxygen concentration in the cathode chamber, the ratio of AOB to total bacteria increases. The list of microorganisms and their functions in BES reactor are reported in Fig. 2. Several denitrifying bacteria can also produce electricity in the system such as *Rhodopseudomonas*, *Comamonas*, and *Pseudomonas*. For the majority of denitrifying bacteria, anoxic condition is suitable but, some can achieve complete denitrification at higher DO level as well such as *Thauera*, *Paracoccus*, *Limnobacter*, *Diaphorobacter*, and *Comamonas* (Nguyen and Babel 2022).

In summary, microbial community demonstrate a crucial role in the BES reactor as they are responsible for electricity generation, contaminant removal, and valuable product formation. Genetic engineering has been extensively applied in BES to target specific genes so as to enhance the metabolic capacity and extracellular electron transfer (EET) rate of microbes. It is indispensable to determine the effective gene modification target based on the application of BES and type of microorganisms. Correspondingly, future research should investigate the difference in metabolic process at the interior and exterior of the biofilm layer under varied environmental and operational conditions. Besides, studies should also focus on the coupling mechanism of various electron transfer pathways in case of mixed culture and should explore the syntrophic interaction between the genetically engineered microorganisms. Furthermore, upcoming studies should also compare the electrochemical behavior of exoelectrogens in lab-scale and industrial-scale systems. To ameliorate the performance of BES in terms of nutrient recovery/removal, in-depth understanding of different strategies for microorganism enrichment is vital and is therefore briefly reviewed in the next subsection.

Enrichment of electroactive microorganism

The effective operation of BES is directly allied with the effective enrichment of EAMs at anode (Yousaf et al. 2017). Therefore, several strategies were developed to enrich EAMs during BES reactor onset and operation (Chatterjee et al. 2019).

Effect of external resistance and anodic potential

The appropriate selection of external resistor can indirectly control the anodic potential under steady cathodic conditions. The anodic potential increases by decreasing the external resistance, thereby facilitating biofilm formation with higher biomass density and greater electron flow (Chatterjee et al. 2019; Cheng et al. 2008). The external resistance governs the anode role as electron acceptor and current generation. A higher external resistance leads to higher voltage and lower current generation as it impedes the electron flow from anode to cathode, because of which, anode remains "charged up" due to electron transfer by the bacteria, thus causing low anodic potential (Cheng et al. 2008). A higher external resistance tends to achieve open circuit condition (Koók et al. 2021). On the other hand, lower external resistance induces more current and less voltage due to reduced kinetic and mass transfer limitations (Khan et al. 2017). At lower external resistance, a more positive anode potential is developed which allows bacteria to gain more energy, and stimulates enrichment of EAMs because of which electricity production increases (Jung and Pandit 2018; Jadhav et al. 2019).

In BES, reactor performance depends on the microorganism interaction with the anode electrode. Being negatively charged by nature, most of the microorganisms are

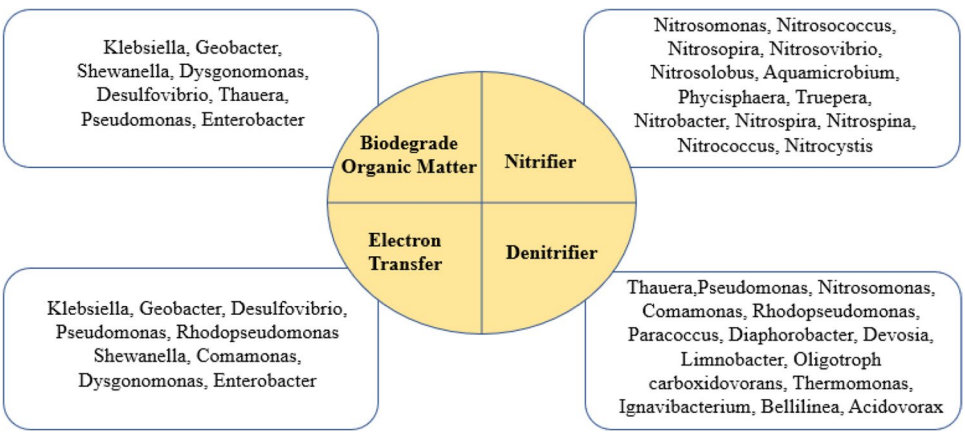


Fig. 2 Schematic illustration of microorganisms and their functions in BES (Nguyen and Babel, 2022)

attracted towards solid surface of positive charge (Mahadevan et al. 2014). Studies have reported that in the anode chamber, the growth of EAM increases at low external resistance whereas enhanced growth of anaerobic microorganisms were observed at higher external resistance due to lower anodic potential (González Del Campo et al. 2016). It is also important to mention that external resistance can also affect the rate of cathodic reaction as it depends on the rate of electron transfer to the cathode (Koók et al. 2021). However, Rismani-Yazdi et al. (2011) observed, that the influence of external resistance on cathodic potential is minor as compared to its impact on anodic potential.

The microorganism, for their survival, utilizes Gibbs free energy during the oxidation of organic matter (González Del Campo et al. 2016). The Gibbs free energy is proportional to the quantity of electrons transferred to the anode and the potential difference between anodic potential and redox potential of the organic substrate (Eq. 1) (Jung and Pandit 2018; González Del Campo et al. 2016). The microorganism energy gain (basically a loss of electrical energy) is necessary and vital for the growth and maintenance of bacterial cells to ensure efficient electron transfer and sustainable long-term operation of fuel cell (Schröder 2007). As shown in Fig. 3, out of the total Gibbs free energy, i.e., $\Delta G^{0'}{}_{total}$, EAMs utilizes a part of energy, i.e., $\Delta G^{0'}{}_{biol}$, while the remaining Gibbs free energy is utilized for the electricity generation (Schröder 2007). The equation of Gibbs free energy is shown below (Milner et al. 2016).

$$\Delta G^{0'} = -nF\Delta E^{0'} \quad (1)$$

where $\Delta E^{0'} = E^{0'}$ *(electron acceptor)* $- E^{0'}$ *(electron donor)*,

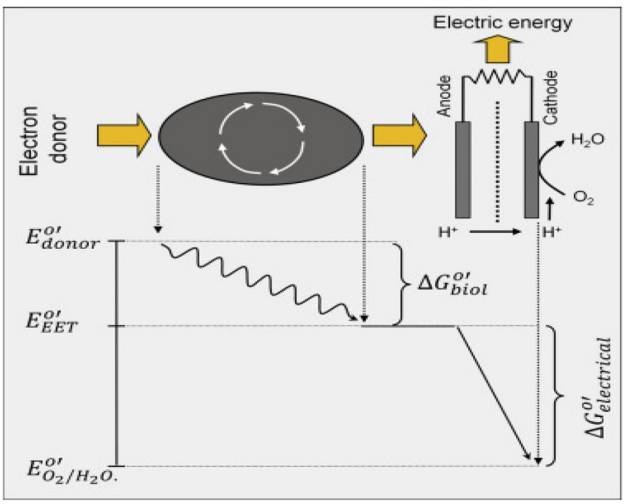


Fig. 3 Schematic illustration of bacterial energy gain and electric energy generation (Schröder and Harnisch 2017)

$\Delta G^{0'}$ indicates change in Gibbs free energy; $\Delta E^{0'}$ denotes difference in cell potential between electron acceptor and donor, $\Delta G^{0'}$ denotes change in Gibbs free energy at pH value 7; *n* refers to number of transferred electrons; F denotes Faraday constant (i.e., 9.64853 X 10^4 C/mol).

$$\Delta G^{0'}{}_{total} = \Delta G^{0'}{}_{biol} + \Delta G^{0'}{}_{electrical} \quad (2)$$

$$\Delta G^{0'}{}_{biol} = -nF(E^{0'}{}_{EET} - E^{0'}{}_{donor}) \quad (3)$$

$$\Delta G^{0'}{}_{electrical} = -nF(E^{0'}{}_{O2/H2O} - E^{0'}{}_{EET}) \quad (4)$$

The Gibbs free energy is positive in case of non-spontaneous reaction and negative for spontaneous reaction (Khan et al. 2017). For a reaction to be thermodynamically favorable in BES, the anode should have higher potential than the endogenous or exogenous mediator or terminal protein in the bacterial cell's electron transfer chain (Wagner et al. 2010). The microorganism diversity, metabolic activity and population varies with varying external resistance because microbial colonization at anode is dependent on the external resistance (Malekmohammadi and Mirbagheri 2021). It has been reported, that electron acceptor, i.e., anode at more positive redox potential acts as favorable electron acceptor and thereby allows the bacteria to effectively oxidize the electron donor as it imparts higher amount of Gibbs free energy (Cheng et al. 2008; Stoll et al. 2016). In other words, the bacterial energy gain can be increased by establishing higher redox potential of anode (i.e., more positive) than redox potential of electron donor (Chatterjee et al. 2019). When energy gain by microorganism is extremely high, the electricity generation will become infinitesimal, and formation of unwanted biomass and stronger cell growth may occur (Schröder 2007).

Despite the benefits associated with positive anodic potential, there exists lack of agreement regarding the selection of either positive or negative anodic potential to facilitate the biofilm growth for higher power density. Jung and Pandit (2018) reported current generation is higher at positive anodic potential while Logan (2009) and González Del Campo et al. (2016) reported that anodic potential should be as lower (i.e., negative) and cathodic potential should be as higher (i.e., positive) as possible to maximize the current generation because more negative anodic potential will reduce the microorganism energy gain and increase the electrical energy output in BES. The higher electricity generation at different anodic potential depicts that electroactive microorganisms modulate their extra-cellular electron transfer chain to adjust to varying anodic potentials (Zhu et al. 2014). Moreover, Zhang et al. (2017) and Malekmohammadi and Mirbagheri (2021) reported higher external resistance accelerates the MFC start up time process and gradual shift

to lower external resistance results in generation of higher current.

Furthermore, Rismani-Yazdi et al. (2011) reported that current generation increases with decrease in external resistance from 1000 Ω to 20 Ω. Besides, Zhang et al. (2017) witnessed increase in current density with increase in external resistance from 10 to 50 Ω due to higher active biomass and beyond further increase in external resistance till 1000Ω, the current density declines. Therefore, based on the aforementioned literatures, it is clear that external resistance significantly influences the anodic potential, electricity generation, microorganism diversity and growth. Therefore, to enhance the performance of BES, selection of optimal external resistance is of prime importance.

Effect of organic substrate

The concentration and type of substrate significantly influences the operational effectiveness of BES (Chatterjee et al. 2019; Prathiba et al. 2022). The substrates acts as an electron donor and thus facilitates the growth of EAMs (Yousaf et al. 2017; Chatterjee et al. 2019). Generally, substrates are classified into fermentable substrate (xylose, fructose, glucose, sucrose), non-fermentable substrate (acetate, butyrate) and complex substrate (domestic and industrial wastewater). In fact, it is also classified as simple and complex substrate (Prathiba et al. 2022). All the substrates in the anodic chamber demonstrates varying efficiencies to get oxidized and among all, higher conversion efficiency of 72% were observed in case of acetate (Mohyudin et al. 2022). In most of the scientific literature, for the preliminary analysis and optimization, single substrate like glucose, sucrose and acetate are used in BES reactor. In addition, other substrate such as urine, leachate etc. have been used recently in MFC for the treatment (Khandaker et al. 2021). The complex substrate give rise to intricate microbial profile and requires diverse microbial consortia to promote degradation whereas simple non-fermentable substrate namely acetate stimulates higher current densities (Michie et al. 2013). The major limitation while selecting substrate is its biodegradability and nature. The complex wastewater comprises of higher organic carbon content, which includes large amount of C ring structures and their degradation into simpler molecule is difficult whereas simpler substrates are readily degraded by the biocatalysts (Venkata Mohan et al. 2014). The major limitation associated with the BES inoculated with mixed culture is the competition between different microbes for the organic substrate. As a mitigation strategy, pre-enrichment of EAM on non-fermentable substrates can lead to selective EAMs enrichment in the biofilm at anode. At anode, the electron donors influence the morphology of microbial community more specifically the nanowires formation that contributes in enhancing electrical connections between the anode surface and bacterial cells (Yousaf et al. 2017).

A medium containing fermentable substrate such as sucrose or glucose induces the growth of different microbial community, including EAMs whereas non-fermentable substrate, i.e., acetate and lactate promote selective growth of EAMs specifically *Geobacter* and *Shewanella*, respectively (Chatterjee et al. 2019; Doyle and Marsili 2015). In general, complex or fermentable substrate might entail syntrophic interaction between the non-electroactive and electroactive microorganisms, whereas acetate is converted to electricity directly by EAM. Besides, the low energy requirement of EAMs makes them capable to grow under substrate depleted condition. Therefore, start-up of BES reactor at lower substrate loading rate encourages the growth of EAMs (Chatterjee et al. 2019). Furthermore, addition of poorly soluble iron (Fe^{3+}) forms in the medium favors the growth of EAMs such as *Acidithiobacillus spp*., *Shewanella*, and *Geobacter* at the anode during the enrichment stage (Chiranjeevi and Patil 2020). In addition, it was observed that addition of heavy metals Cd^{2+} or Cu^{2+} in the medium resulted in strong adherence of *Shewanella* cells on the electrode surface and increased EET owning to excessive production of riboflavin in cells (Xu et al. 2016).

In real wastewater, electrical conductivity is low and wastewater composition varies. The real wastewater are complex in nature and certain compounds present in it can be harmful to microorganism. Also, electrogenic community in the BES are likely to get affected by the indigenous microorganisms present in the wastewater as well as higher COD level can lower the BES performance. The microbial inhibition can be prevented by setting up an adaptation period in which wastewater flow is raised gradually to increase the organic loading rate (OLR). Furthermore, introduction of simple substrate and electron acceptors in the anodic chamber can expedite the electrogenic biofilm formation and thereby escalates the power production in the long run. Generally, this method is adopted for the start-up of the lab scale BES reactor operated with the real wastewater (Chatterjee et al. 2019).

Nutrient recovery

Nitrogen

The excessive release of nitrogen-rich effluents into the natural water bodies has posed a serious environmental concern. Biological nitrogen removal is an effective and promising treatment approach that can achieve complete reduction of nitrate to harmless nitrogen gas. Correspondingly, many researchers are focussing on the discovery of effective technology for utilizing energy from waste and BES is one

such technology (Vijay et al. 2022). In fact, various literatures have reported successful removal of nitrogen in BES (Nguyen and Babel 2022). Therefore, this section briefly explains the different mechanisms involved in nitrogen removal along with existing and recent studies using BES.

Sequential nitrification and denitrification

Conventionally, nitrification followed by denitrification are adopted to remove nitrogen from wastewater (Ali and Okabe 2015). In the nitrification method, at first, ammonium-oxidizing bacteria (AOB) oxidizes ammonium to nitrite and subsequently nitrite-oxidizing bacteria oxidizes nitrite to nitrate under aerobic conditions. Later in the denitrification step, under anoxic conditions, the nitrate is reduced to nitrogen gas (Ali and Okabe 2015). The denitrification process can also occur at BES cathode where nitrogen oxides act as electron acceptors to promote a reduction reaction to produce nitrogen gas (Nguyen and Babel 2022). Table 2 represents the denitrification reactions at the cathode and their corresponding redox potentials.

The denitrification process at the cathode is categorized into two modes namely conventional denitrification (i.e., heterotrophic denitrification (HD)) and bio-electrochemical denitrification (i.e., autotrophic denitrification (AD)) (Nguyen and Babel 2022). Autotrophic denitrification occurs by utilizing cathode electrode as an electron donor in MFC and thus exhibits a prominent role in lowering the requirement for carbon sources for nitrate reduction (Nguyen and Babel 2022) whereas organic carbon (methanol, acetic acid, glucose, glycerol, starch, ethanol) is utilized as an electron donor by the heterotrophic denitrifiers (Vijay et al. 2022) (Fig. 4). The advantages associated with autotrophic denitrification include a lesser requirement for organic matter and lower biomass yield (Nguyen and Babel 2022). However, compared to heterotrophs, the autotrophs have slower growth rates as certain portion of electron and energy is spent on reducing CO_2 to organic carbon. Still, in the autotrophic process, the power output is higher than the heterotrophic process owing to significant difference in redox potential across anode and cathode, although the denitrification rate is comparatively lower because fraction of electrons is utilized in cell synthesis, thus less number of electrons are available for nitrate reduction. In the heterotrophic process, the coulombic efficiency and power output declines because at anode, nitrate competes for the electron with electrode or existence of organic substrate in the cathode chamber lowers the potential difference (Vijay et al. 2022). Vijay et al. (2019) performed comparative performance assessment of heterotrophic and autotrophic denitrification in MFC. Autotrophic system demonstrated a higher power output (4.45 W/m^3) and lower nitrate nitrogen (NO_3^--N) removal rate (0.118 kg/

Table 2 Reduction reactions and theoretical standard redox potential in MFC (Nguyen and Babel 2022; Vijay et al. 2022)

Electron acceptor	$E_{cathode}$ (V vs. standard hydrogen Electrode)	Reduction reaction
Nitrate	+0.433	$NO_3^- + 2H^+ + 2e^- \rightarrow NO_2^- + H_2O$
	+0.749	$2NO_3^- + 12H^+ + 10e^- \rightarrow N_2 + 6H_2O$
Nitrite	+0.350	$NO_2^- + 2H^+ + 2e^- \rightarrow NO^- + H_2O$
Nitric oxide	+1.175	$NO^- + H^+ + e^- \rightarrow 0.5N_2O + 0.5H_2O$
Nitrous oxide	+1.355	$0.5N_2O + H^+ + e^- \rightarrow 0.5N_2 + 0.5H_2O$

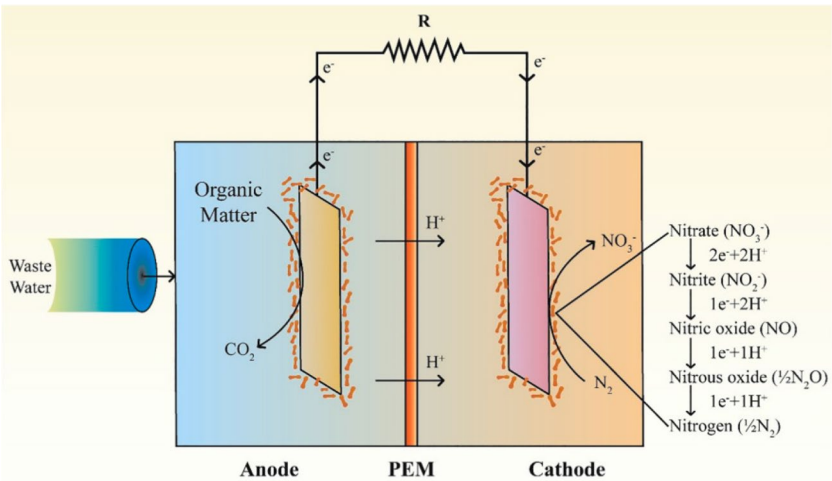


Fig. 4 Schematic diagram of denitrification process in MFC (Vijay et al. 2022)

m^3d) whereas lower power output (3.02 W/m^3) and higher NO_3^--N removal rate (2.06 kg/m^3d) were observed in the heterotrophic cathode. Heterotrophic and autotrophic cathodes achieved total nitrate removal rate of 96% and 86.08%, respectively.

The integration of biological nitrogen removal in BES solves the existing problems such as higher sludge generation, higher energy requirement for aeration and in-sufficient nitrogen removal efficiency for wastewater with lower C/N ratio. The benefits allied with the nitrogen removal/recovery using MFC technology has motivated the scientific communities and in fact, many researchers have developed different configurations of MFC to enhance the power generation and nitrogen removal/recovery efficiency (Nguyen and Babel 2022). Therefore, Table 3 elaborates the specification, removal mechanism and performance of different configuration of MFCs for nitrogen removal.

Simultaneous nitrification and denitrification (SND)

In traditional wastewater treatment processes, aerobic nitrification and anaerobic denitrification are carried out in two separate steps, as microorganism behavior and operational conditions are different. While in MFC, both the processes can occur simultaneously as it integrates the biofilm process with the suspended biomass. The electrodes in MFC provides a higher surface area for the formation of biofilm, with deeper layers near to the electrode surface performs denitrification whereas ammonium oxidation is carried out in the outer layers because of the existence of oxygen gradient within the cathodic biofilm. In the SND process, at first, nitrifiers oxidizes ammonium to nitrate. This is achieved either by separate nitrifying reactor supplied with oxygen or a stratified layer of aerobic/anaerobic biofilm at cathode for SND. Subsequently in the cathode chamber, the produced nitrate is reduced to nitrogen gas (N_2) (Vijay et al. 2022). Feng et al. (2015) studied the nitrogen removal in dual chamber MFC by feeding synthetic wastewater containing 230 mg/L NH_4^+-N and different concentration of phenol (0–1400 mg/L) in the aerobic cathode chamber. They found no inhibitory effect of phenol upto 600 mg/L on nitrification reaction but lower inhibitory effect was observed when its concentration was increased upto 1400 mg/L, thus resulting in longer reaction time for removal of ammonium. Bacterial analysis revealed SND process and anaerobic denitrification in the cathode and anode chamber respectively is responsible for the complete nitrate removal. Also, under different operational conditions, Zhu et al. (2016) evaluated the performance of double-chamber MFC fed with decomposed cyanobacteria solution as anodic and cathodic substrate. An efficient SND process with a TN removal rate of 0.064 kg/m^3/day was achieved at dissolved oxygen (DO) concentration of 5 mg/L in cathodic chamber, but with further increase in DO concentration upto 6 mg/L, denitrification process was inhibited in the cathode chamber. Furthermore, SND process was enhanced in the cathodic chamber at closed-circuit condition than open circuit condition because of pH stability and electric field in the closed-circuit condition boosted the metabolic activities and escalated the cofactor regeneration of both denitrifiers and nitrifiers that caused increased nitrogen removal efficiency of MFC system. Correspondingly, Sotres et al. (2016) found that intermittent aeration in the cathodic chamber facilitates growth of simultaneous nitrifying and denitrifying microbial population. The short intermittent aeration in cathodic chamber leads to nitrate removal of 17.8% whereas long intermittent aeration achieves nitrate removal of 8.3% without addition of acetate and 41.2% with addition of acetate. Also, outside the MFC, denitrification batch assays carried out using cathode effluent in anoxic conditions showed that acetate addition to cathodic effluent foster denitrification, resulting in higher NO_3^--N removal upto 99.7%.

Shortcut nitrification and denitrification

This process aims to bypass the formation of nitrate and facilitate the direct trasnformation of nitrite to nitrogen gas. The shortcut pathway involves the nitrification step where ammonia is partially oxidized to nitrite and in the denitrification step, the nitrite is reduced to nitrogen gas. This process is also referred to as anaerobic ammonium oxidation (ANAMMOX). Compared to conventional biological nitrogen removal, the benefit associated with this method includes lower retention time, reduced oxygen demand, and lower requirement for carbon substrate. It is an autotrophic process and, in this process, ammonium is converted to nitrogen gas without the presence of organic matter, due to which organic matter can be completely utilized by exo-electrogenic bacteria for energy production in MFC. Researchers are targeting nitrogen removal using integrated approach of BES-denitrification-ANAMMOX to achieve simultaneous nitrogen removal and power generation (Bavasso et al. 2018; Zekker et al. 2020). This process can save 25% oxygen and 40% carbon required for nitrification denitrification respectively as compared to nitrogen removal through nitrate transformation. Besides, compared to nitrate reduction, the reduction rate of nitrite is 2 times faster and produces 40% lesser sludge in the process of nitritation and denitritation (Nguyen and Babel 2022).

In the batch mode, Li et al. (2016) studied the functioning of shortcut nitrification and autotrophic denitrification MFC (SNAD-MFC) as shown in Fig. 5 by varying the operational parameters. Results showed that oxidation of ammonium to nitrite occurs at a DO level lower than 3.5 mg/L in the cathodic chamber.and a total nitrogen removal efficiency of 99.9% with a removal rate of 0.0125 kg/m^3/d was achieved.

Table 3 Specifications, operational conditions and performances of different configurations of MFC for organic matter and nutrient removal

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
A) Nitrification—denitrification											
1	Synthetic wastewater	O-MFC cathode: Mixed nitrifying sludge; A-MFC cathode: Mixed denitrifying sludge	Sequential Nitrification and Denitrification	Three Chamber coupled MFC system; Continuous Mode	A: Graphite Felt; C: Graphite Felt	Vol: 5L each; Cation Exchange Membrane; External Resistance: 20 Ω (O-MFC), 20 Ω -5 Ω (A-MFC)	COD: 950.2–982.6 mg/L; N H_4^+-N: 29.7–31.8 mg/L; TN: 29.3–31.8 mg/L	O-MFC: 15.1 W/m^3; A-MFC: 6.8 W/m^3	O-MFC: 19.4 A/m^3; A-MFC: 26.1 A/m^3	COD: 98.6–98.8%; N H_4^+ –N: 88.5–95%; TN: 56.4–87.1%; CE: 103.9–116.8	(Xie et al. 2011)
2	Synthetic wastewater	Mixed Activated and Digested Sludge (1:1)	Sequential Nitrification and Denitrification	Dual Cathode MFC	A: Carbon Cloth Outer C: Carbon Cloth coated with platinum catalyst (0.3 mg Pt/cm^2) Inner C: Carbon brush	Temp: 20 ℃; Anion Exchange Membrane; Cation Exchange Membrane; Resistance:1 Ω (between anode and inner cathode), 60 Ω (between anode and outer cathode); Anodic HRT: 10.4 h; Anode Vol.: 250 mL; Inner Cathode Vol.: 550 mL; Outer Cathode Vol.: 2L	COD: 0.59–2.46 kg/m^3/day; TN: 0.07–0.21 kg N/m^3 day	2.9 W/m^3	3.5–5.2 mA	COD: 84.9–98.9%; TN: 66.7–89.6%; N H_4^+ –N>96%; NO_3^- –N: 67.8–91.9%; CE: 48–100%	(Zhang and He 2012)
3	Synthetic Wastewater	Anoxically cultivated sludge	Denitrification	Single Chamber MFC; Batch Mode	A: Rectangular Graphite; C: Rectangular Graphite	Temp: 30 ± 2℃; Vol.:1L; HRT: 24 h;	NO_3^-: 30 mg/L	–	9.1 mA	NO_3^-: 1.09 g/m^3/h; TN: 0.97 g/m^3/h; CE: 0.958 −26.45%	(Huang et al. 2013)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
4	Synthetic Wastewater	Activated sludge	Denitrification	Double Chamber MFC;	A: Rectangular Graphite Felt; C: Rectangular Graphite Felt;	Anode Vol.: 350 mL; Cathode Vol.: 350 mL; Temp: 25 ℃; Proton Exchange Membrane; External Resistor: 1000Ω	NO_3^-: 30 mg/L	–	–	NO_3^-: 78%	(Nguyen et al. 2015)
5	Synthetic Wastewater	Anaerobic mixed culture sludge	Denitrification	Double Chamber MFC; Continuous Mode	A: Activated Carbon Flakes; C: Activated Carbon Flakes	Anodic Vol.: 0.67 L; Cathodic Vol.: 0.67 L; HRT: 24 h; Proton Exchange Membrane; External Resistor: 1000 Ω	COD: 1142 mg/L;NO_3^-:141 mg/L	669 mW/m^3; 370 mV	3487 mA/m^3	NO_3^-: 88%	(Oon et al. 2016)
6	Pharmaceutical Wastewater	Anaerobic consortium	Denitrification	Single Chamber MFC; Batch Mode	A: Graphite; C: Graphite and Stainless Steel	Total Vol. 2.6 L; HRT: 48 h	OLR: 3–15 g/L	–	–	NO_3^-: 45–50%	(Modestra et al. 2016)
7	Synthetic Wastewater	Denitrifying Microorganisms	Denitrification	Single Chamber	Cylindrical carbon cloth and Stainless steel mesh	Amplitude:0.1–9.6 V_{p-p}; Frequency: 10–60 Hz; Effective Vol.: 5L; Temp.: 25 ℃	NO_3: 600 mg/L; C/N:2.5–5	–	–	NO_3^-: 74.4–95.5%	(Hoseinzadeh et al. 2017)
8	Synthetic Wastewater	Denitrifying Microorganisms	Denitrification	Single Chamber	Cylindrical carbon cloth and Stainless steel mesh	Amplitude:8V_{p-p}; Frequency: 10 Hz; Effective Vol.: 5L; Temp.: 25℃	NO_3: 50–600 mg/L; C/N:4.2	–	–	NO_3^-: > 95%	(Hoseinzadeh et al. 2018)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
9	Synthetic Wastewater	A: Cow manure C: Acclimatized denitrifying microorganism present in cow manure and soil;	Denitrification	Double Chamber MFC; Batch Mode	A: Graphite Felt C: Graphite Felt	Temp.: 30 ℃; Vol.: 100 mL; External Resistance: 100 Ω; HRT: 4 days; Proton Exchange Membrane	KNO_3: 2.5 g/L	3.02–4.45 W/m^3;310–585 mV	29.7 A/m^3–40.5 A/m^3	NO_3^-: 86.08–96%	(Vijay et al. 2019)
10	Synthetic Wastewater	A: Anaerobic culture	Nitrification	Double Chamber MFC; Fed-batch Mode	A: Carbon Felt C: Biofilm covered platinum coated carbon cloth (0.5 mg Pt/cm^2)	Temp.: 30 ℃; Anodic Vol.: 100 mL; Cathodic Vol.: 100 mL; External Resistance: 1000 Ω; Proton Exchange Membrane	N H_4^+–N: 80 mg/L	0.97 W/m^2	–	COD: 95.5%;NH_4^+–N: 99%; TN: 89%; CE: 56.1%	(Jin et al. 2020)
11	Synthetic Wastewater	Anaerobic activated sludge	Denitrification	Single chamber Air Cathode MFC	A: Carbon Brush C: N-doped graphene oxide coated carbon cloth	Temp.: 35 ℃; Vol.:100 mL; External Resistance:1000 Ω;	KNO_3: 0.361 g/L	1800 W/m^3	–	NO_3^-–N: 87.4% TN: 64.5%;	(Li et al. 2023)
B) Simultaneous nitrification—denitrification											
12	Synthetic Wastewater	A: Treated coking wastewater C: Aerobic Activated Sludge	SND	Double Chamber MFC; Batch Mode	A: Graphite Felt C: Graphite Felt	Temp: 30 ℃; Anode Vol.: 250 mL; Cathode Vol.: 250 mL; Anion Exchange Membrane	NH_4^+-N: 230 mg/L	–	–	NO_3^- – N: 100%	(Feng et al. 2015)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
13	Diluted Decomposed Cyanobacteria solution	A: Mesophilic anaerobic sludge; C: Mixed nitrification and denitrification sludge	SND	Double Chamber MFC; Batch Mode	A: Graphite Felt C: Graphite Felt	Anode Vol.: 4.1 L; Cathode Vol.: 4.1L; Proton Exchange Membrane; External Resistor: 20 Ω	COD: 800–1000 mg/L; NH_4^+-N: 60–100 mg/L	–	–	COD: 60%NH_4^+–N: 0.063 kg/m^3/day TN: 0.064 kg/m^3/day	(Zhu et al. 2016)
14	Synthetic wastewater	A: Anaerobic Digestate of slaughterhouse waste	SND	Double Chamber MFC; Continuous Mode	A: Granular Graphite Rod C: Stainless steel mesh	Temp: 23 ℃; Anode Vol.: 165 mL; Cathode Vol.: 250 mL; External Resistance: 500 Ω; HRT: 6.3 h at anode and 9.4 h at cathode chamber; Cation exchange membrane	COD: 9.5 g/L/d; NH_4^+-N: 4.2 g/L/d	767.2–806.8 mW/m^3; 193.90 mV	2.32 A/m^3	NO_3^- – N: 8.3–41.2%	(Sotres et al. 2016)
15	Synthetic Wastewater	-	SND	Single Chamber MFC with air cathode	A: Carbon Brush C: Pt-CNT, N-CNT and pristine CNT with stainless steel mesh	Vol.: 28 mL; HRT: 0.5–2 days	TOC: 202–404 mg/L;NH_4^+-N: 31 mg/L	31–408 mW/m^3; 86.6–338.1 mV	2.36 A/m^2	TOC:58.3–95.2%; TN: 29.4–975%;NH_4^+–N: 91.5–97.7%; CE:10.6–55.6%;	(Zuo et al. 2016)
16	Domestic Wastewater	A: Activated Sludge	SND	Flat panel air cathode MFC	A: Graphite Felt; C: Carbon cloth with a platinum catalyst	Temp:10–25 ℃; Anode Vol.: 150 mL; External Resistance:50 Ω; HRT: 2.5 h	COD: 144 mg/L; TN: 28.6 mg/L	8.9 W/m^3	–	COD: 85%; TN: 94%	(Park et al. 2017)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
17	Synthetic High Concentration Ammonia Wastewater	Aerobic Sludge	SND	Double chamber MFC; Continuous mode	A: Carbon Brush C: Carbon Brush	Temp:31 ± 1 ℃; Anode Vol.: 0.452 L; Cathode Vol.: 0.452 L; Proton exchange membrane; External Resistance: 50–100 Ω HRT:10.4 h	COD: 300 mg/L;NH_4^+–N: 480 mg/L	–	–	TN:8.5–14.6 mg/L/d	(Zhao et al. 2017)
18	Synthetic Wastewater	A: Anaerobic Activated Sludge; C: Activated Sludge Membrane Zone: Activated Sludge	SND	Membrane bioelectrochemical reactor (MBER), i.e., Two MFCs with aerobic membrane zone; Continuous Flow mode	A: Carbon Fiber Brush C: Carbon Fiber Brush	Anode Vol.: 250 mL; Cathode Vol.: 302 mL; Cation Exchange Membrane; Hollow Fiber microfiltration membrane; External Resistance: 50, 100 and 500 Ω; HRT: 4–10 h	COD: 192–492 mg/L; NH_4^+-N: 51.1–53.2 mg/L	1.8 W/m^3	8.5 A/m^3	COD: 92.3%;NH_4^+–N: 99.6%; TN: 84.3%	(Li et al. 2017)
19	Synthetic Wastewater	Aerobic denitrifying sludge	SND	Air cathode single chambered MFC; Batch Mode	A: Carbon Felt; C: Manganese based catalyzed carbon air cathode	Effective Vol.: 100 mL; External Resistance: 1kΩ	COD: 240 mg/L; NH_4^+-N: 130 mg/L	1270 mW/m^2	0.48 mA/cm^2	COD: 90%;NH_4^+–N: 98%; TN: 95%; CE: 60–70%	(Yang et al. 2019)
20	High-salt nitrogenous wastewater	Effluent from the primary sedimentation tank	SND	Three phase single chamber MFC with air cathode	A: Graphite Carbon Fiber Brush	Temp: 30 ℃; Vol.: 28 mL; External Resistance:1000 Ω;	–	439.3 mV	–	TN: 63.4%	(Zeng et al. 2020)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
19	Synthetic Wastewater	A: Anaerobic Sludge C: Mixed Anoxic and SND Sludge	SND	Multi-Anode Microbial Fuel Cell (MA-MFC); Batch Mode	A: Graphite Felt C: Graphite Felt	Temp: 25 ± 3 ℃; Total Vol.: 2.8 L; External Resistance: 1000 Ω; Proton Exchange Membrane; HRT: 7 days	–	236.7 mW/m^3	–	COD: 94.90%; TN: 71.1%	(Huang et al. 2020)
	Organic Acid Industrial Wastewater	A: Electrogenesis Microorganism	SND and anaerobic ammonia oxidation process	Double Chamber MFC; Continuous Mode	A: Carbon Rod; C: MnO_2/TiO_2/g-C_3N_4@ GAC cathode	Proton Exchange Membrane; External Resistance: 1000 Ω; HRT: 8–12 h;	COD: 2700–9000 mg/L;NH_4^+-N: 37–92 mg/L	1176.47 mW/m^3	6.95 A/m^3	COD: 98%; NH_4^+–N: 99%;NO_3^-–N: 99%	(Zhang and Liu 2020)
20	Freshwater and Seawater	Halomonas Strains	SND	Single Chamber MFC; Batch Mode	A: Graphite Fiber Brush C: Air Cathode coated with activated carbon catalyst layer	Temp: 30 ± 2 ℃; Vol: 28 cm^3; Reactor Length: 4 cm; External Resistance:1000 Ω;	C/N: 1–10; NH_4^+-N: 500–1000 mg/L; NaCl: 30–150 mg/L	0.61 W/m^3; 214 mV	–	NH_4^+–N: 93.8%	(Liu and Wu 2021)
21	Synthetic Wastewater	A: Anaerobic Sludge C: Mixed aerobic and SND sludge	SND	Multi-Anode Microbial Fuel Cell (MA-MFC); Batch Mode	A: Graphite Felt C: Graphite Felt	Temp: 25 ± 1 ℃; Anode Vol.: 2.8 L; Cathode Vol.: 2.8 L; External Resistance: 145–1000 Ω; Proton Exchange Membrane; HRT: 6 days	COD: 65–1123 mg/L; N H_4^+-N: 61.7–64.2 mg/L; TN: 60–64 mg/L COD/N: 0–4.5	241.8 mW/m^3	-	COD: 78.4–97.9%; N H_4^+–N: 85.4–91.7%; TN: 30.3–81.4%; CE: 0.86–1.58%	(Zhu et al. 2021)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
22	Synthetic Wastewater	Aerobic Denitrifying mixture	SND	Single Chamber Air Cathode Microbial Fuel Cell (ACMFCs); Continuous mode	A: Carbon Graphite Felt C: Air cathode laminated with MnO_2 catalyst layer and porous Teflon film	Temp: 30 ± 2 ℃; Vol.: 0.10L	COD: 0–434 mg/L Ammonia Nitrogen: 0–534 mg/L; COD/N: 0–8.7 mg/L	1400 mW/m^3	0.5–12.5 mA	COD: 65–94%; TN: 11.15–93.5%; NH_4^+ − N:11.15–92%; CE: 2.70–69.2%	(Yang et al. 2021)
23	Raw wastewater	–	SND	Microbial Electrochemical System (MES) allied Microbial Separator	–	HRT: 4.5–18 h	TN: 40 and 140 mg/L	–	–	TN: 0.08–0.13 kg/m^3/day; N H_4^+ −N: 0.05–0.09 kg/m^3/day	(Li et al. 2022)
24	Synthetic Wastewater	A: Anaerobic Sludge; C: Mixed Sludge comprising anoxic sludge	SND	Multi-anode MFC; Batch Mode	A: Graphite Felt; C: Graphite Felt and Carbon Brush	Cation Exchange Membrane; Proton Exchange Membrane; Volume: 2.8 L; HRT: 160 h; Temp: 25 ± 3 ℃; DO:0.7 mg/L	COD:1000 mg/L(anode) and 225 mg/L (cathode);NH_4^+-N: 64 mg/L	12.28–46.11 mWh	-	COD: > 96%; TN: 34.9–68%; N H_4^+ −N: 84.5–93%	(Huang et al. 2021)
C) Shortcut nitrification and denitrification											
25	Raw wastewater	A: Influent of WWTP; C: Sludge from aeration tank of WWTP	Shortcut Nitrification/Denitrification	Three Chambered MFC; Batch Mode	A: Carbon Brush C: Carbon Cloth loaded with Pt (0.5 mg/cm^2)	Anode Vol.: 300 mL; Cathode Vol.: 300 mL; Cation Exchange Membrane;	N H_4^+: 50 mg/L	59.6–294.9 mW/m^2	0.158 mA/cm^2	TN: 0.0125 kg/m^3/day; CE: 2.41%	(Li et al. 2016)

Table 3 (continued)

S. No	Substrate	Innoculum	Removal mechanism	Reactor configuration and operation mode	Electrode specification (A: Anode, C: Cathode)	Reactor operation and specification	Initial concentration	Maximum power density/voltage	Current/Current density	Removal efficiency/rate and coulombic efficiency (CE)	Reference
26	Synthetic Wastewater	Supernatant liquid of the agricultural waste and cow manure digestate	Shortcut Nitrification/Denitrification	Double Chambered MFC; Batch Mode	A: Carbon Paper C: Carbon Paper	Anode Vol.: 300 mL; Cathode Vol.: 300 mL; Cation Exchange Membrane	NH_4^+: 100 mg/L;	–	–	NH_4^+: 63–76%; NO_2^-: 22.40–80.3%	(Bavasso et al. 2018)

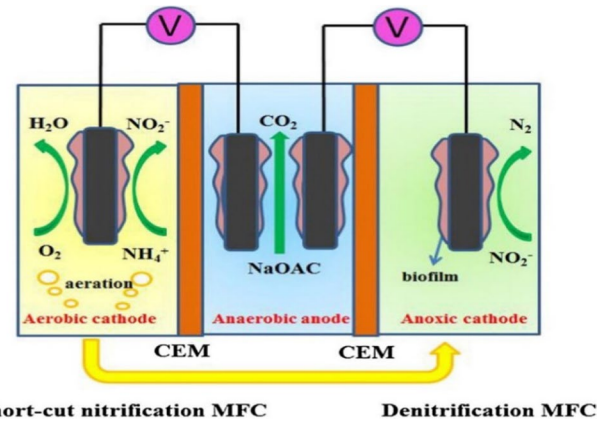


Fig. 5 Schematic illustration of integrated shortcut nitrification and autotrophic denitrification MFC (SNAD-MFC) (Li et al. 2016)

A maximum power generation and current density were found to be 294.9 mW/m^2 and 0.158 mA/cm^2 respectively at DO level of 3.5 mg/L in the shortcut nitrification. Moreover, platinum-coated cathode exhibits faster reduction of nitrite and nitrate than Pt-free plain cathode in the anoxic cathode. Besides, based on carbon substrate saving, electric power production, and aeration saving, an energy-positive operation with the net energy balance of 0.0066–0.007 kWh/m^3 was achieved and therefore SNAD-MFC systems were considered as beneficial over energy-negative MFC systems and carbon-intensive traditional biological nutrients removal methods. In another study, Zekker et al. (2020) observed that compared with heat-treated anaerobic inoculum, the MFC inoculated with ANAMMOX consortia achieved higher power density of 9.5 W/m^3 and 6 W/m^3 as well as TN removal efficiency upto 89% and 73% during the first and last 30 cycles of operational period respectively. In addition, COD removal efficiency was also higher in the case of MFC with ANAMMOX consortia than compared with heat-treated anaerobic inoculum, thereby depicting it as a potential solution to ensure higher organic carbon and nutrient removal efficiency at lower operation cost and maintenance to produce reusable treated water.

Influencing factors controlling nitrogen removal

Nitrogen removal in the MFC system depends on various factors such as inoculum source, pH, electrode material, internal and external resistance, carbon/nitrogen ratio, and organic matter concentration as shown in Fig. 6 (Vijay et al. 2022). All these factors influences the microbial activity in terms of nitrogen removal and rate of generation and transfer of electrons (Sun et al. 2020).

Electrode material, configuration, and application of catalyst The appropriate selection of cathodic electrode material

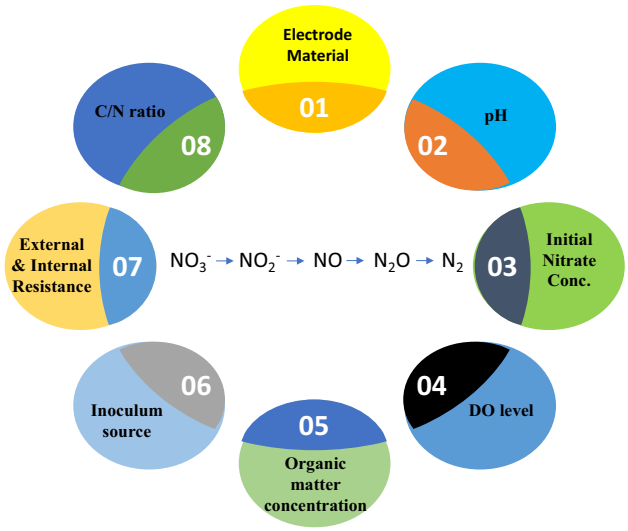


Fig. 6 Schematic representation of the factors affecting the denitrification reaction in the MFC system

is vital for the microbial interaction, electricity generation, and optimization of MFCs (Mier et al. 2021; Qiu et al. 2021). The cathodic material should be chemically stable, efficient, and economical (Rusli et al. 2019). The stability of cathode is imperative during MFC operation when they are exposed to the bacteria, composite organic matter, and water (Qiu et al. 2021). Table 3 demonstrates different types of electrodes used by researchers for nitrogen removal.

In general, the role of biocathode MFC in wastewater treatment is usually nutrients and COD removal (Mook et al. 2013; Rusli et al. 2019). The effect of different cathodic material was studied by Modestra et al. (2016) for the degradation of high strength pharmaceutical wastewater. They found that BES with graphite as cathode material exhibits higher nitrate (50%) and phosphate removal efficiency (58%) as compared with stainless steel (SS) material (45% and 48%) due to development of high cathode potential and electron acceptance conditions which depicts graphite is an effective cathode material over SS for pharmaceutical wastewater treatment. Researchers have reported that surface modification of electrode during electrode fabrication, through application of catalytic nanoparticles, strengthens the bacterial adhesion and electron transfer rates (Guo et al. 2020; Wei et al. 2011). In single chamber MFC, Zuo et al. (2016) used nitrogen doped carbon nanotube membrane (N-CNT), Pt-coated (CNT) membrane and pristine CNT membrane as filtration air cathode for treatment of synthetic wastewater. Among the three types of CNT membrane, N-CNT filtration MFC demonstrated higher TOC (95.2%) and N H_4^+-N (97.7%) removal efficiency as well as maximum current density and power density of 2.36 A/m^2 and 408 mW/m^2, respectively during continuous operation of 39 days due to rich N functional groups, micropore structure and higher specific surface area. In another study, using manganese based catalyzed carbon air cathode and aerobic culture of *Thauera* dominated denitrifiers in single chamber MFC, Yang et al. (2019) achieved COD, TN and ammonia removal efficiency of 90, 95, and 98%, respectively along with maximum power output of 1270 mW/m^2. Furthermore, in dual chamber MFC, Jin et al. (2020) fabricated a composite cathode by in-situ cultivation of biofilm over platinum-coated cathode (BPC) and found BPC-MFC shown higher voltage output along with higher N H_4^+-N (99%) and COD removal efficiency (95.5%) than platinum-coated MFCs (PC-MFCs). In fact, BPC-MFC demonstrated lower charge transfer resistance, i.e., 35.5 Ω and a maximum power density of 0.97 W/m^2. The superior performance of BPC-MFC is due to bifunctional cathode that facilitates coupled reaction involving ORR and biological nitrification as shown in Fig. 7a which indicates integration of biotic and abiotic catalysts in composite electrode is effective in wastewater treatment. In another study, Zhang et al. (2020a, b) prepared SnCu-Pd/CFC catalytic cathode and observed maximum TN removal efficiency of 96.3% and nitrogen removal rate of 1.69 kg.N/m^3d when operated with nitrogen loading rate of 1.74 kg.N/m^3d, thereby inferring excellent performance of SnCu-Pd/CFC cathode in reduction of nitrate in single chamber MFC. Also, the effect of manganese dioxide/titanium dioxide/graphitic carbon nitride @ granular activated carbon (MnO_2/TiO_2/g-C_3N_4@GAC) electrode as MFC cathode was investigated by Zhang and Liu (2020) for the treatment of organic acid industrial wastewater. They found effective removal of both N H_4^+-N and NO_3^--N upto 99% and COD removal efficiency was more than 98% with maximum removal capacity of 17.77 kg/m^3d. Besides, MnO_2/TiO_2/g-C_3N_4@GAC showed 65.29% higher power density as compared with GAC cathode due to higher energy recovery efficiency because of lower internal resistance of the system. Thus, the results infer that MnO_2/TiO_2/g-C_3N_4@GAC electrode MFC system is effective, economical, and technically feasible in treatment of high strength industrial wastewater treatment.

Recently, under long term operation of 300 days, Huang et al. (2021) studied the influence of different cathode electrode material and proton exchange membrane on the power generation and nitrogen removal performance of MFC from low C/N ratio wastewater. Compared to carbon brush (CB), MFC with graphite felt (GF) as cathode showed higher electricity production and better denitrification performance. The lower electrical performance of CB was due to (a) greater charge transfer impedance, (b) lower electrical conductivity, and (c) low rate of utilization of outer space of CB electrode for microbial adhesion owning to radial shape.

Li et al. (2023) investigated the performance of air cathode MFC (AMFC) with graphene-based cathode catalyst for nitrogen removal and found that AMFC with nitrogen doped graphene oxide-based cathode achieved higher

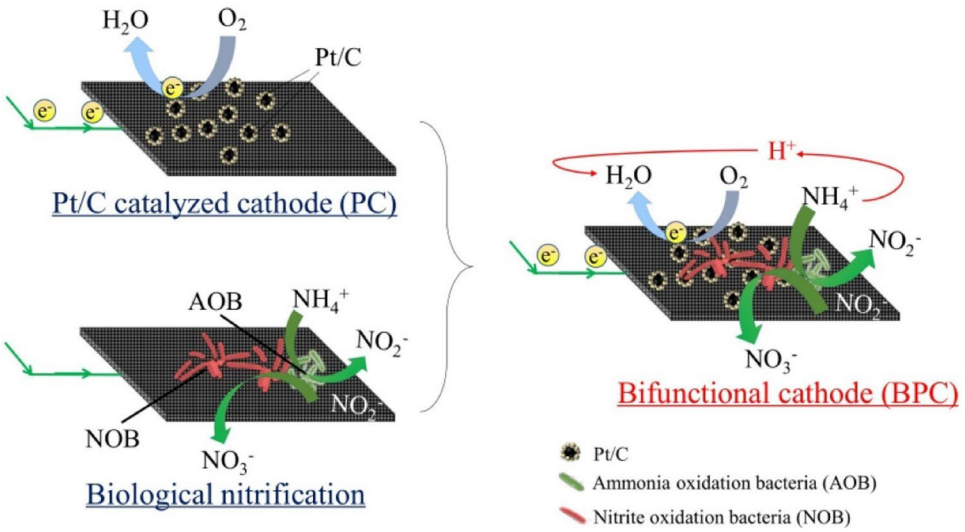


Fig. 7 Schematic illustration of **a** coupled mechanism between ORR at platinum coated carbon cathode and biological denitrification by NOB/AOB (Jin et al. 2020) **b** air cathode MFCs with tridimensional electrode configuration (Yang et al. 2023)

denitrification and TN removal efficiency of 87.4% and 64.5%, respectively along with higher power density of 1800 mW/m^3 as compared with other catalyst. The higher catalytic sites of NGO_{10} due to porous structure resulted in higher oxygen reduction reaction performance while increased abundance of denitrifying bacteria led to enhanced denitrification activity.

Apart from electrode material, the geometrical electrode arrangement can also have a significant influence on nutrients removal and power output. The impact of different tridimensional anode configuration namely flat carbon felt, rolled carbon felt and spiral stair like carbon felt as shown in Fig. 7b was studied by Yang et al. (2023) in air cathode MFC. They noticed that rolled carbon felt exhibits higher total nitrogen removal efficiency (98.6%) than flat carbon felt (94.7%) and spiral carbon felt (96.7%) due to greater external surface area per working volume of reactor (28.2 m^2/m^3), thus facilitating increased active biomass and microbial diversity on the anode for nitrogen degradation. Moreover, power density was also found to be maximum in MFC with rolled carbon felt (1800 mW/m^3) than spiral carbon felt (1535 mW/m^3) and flat carbon felt (315 mW/m^3) due to lower charge transfer resistance indicating that the electrode orientation has remarkable impact on the power generation performance of the reactor.

5.1.4.2 Carbon/nitrogen (C/N) ratio The Carbon/Nitrogen (C/N) ratio is an important factor that affects the biological nitrogen removal process. An appropriate C/N is required for autotrophic denitrification process. At C/N ratio of 2–2.7, nitrite accumulation is significantly declined. A higher C/N ratio facilitates the proliferation of heterotrophic denitrifying bacteria, causing higher generation of electrons, while at low C/N ratio, chemical energy is converted to electrical energy. This means that an increased C/N ratio do not enhance the percentage of autotrophic denitrification process in BES, though it increases the nitrogen removal efficiency. A higher C/N ratio boosts the BES performance in terms of nitrate removal and lowers the accumulation of nitrite (Sun et al. 2020). The optimal C/N ratio in conventional biological process for aerobic denitrification is around 5 and can reach sometimes 9–10. In BES system, electron donor is cathode, therefore C/N ratio required for the denitrification process remains in much broader range than the conventional process. Moreover, BES can effectively treat wastewater with low C/N ratio. In BES, both autotrophic and heterotrophic microorganisms exist at higher C/N ratio and together eradicates the nitrogenous waste, whereas in case of wastewater with low C/N ratio, autotrophic bacteria is dominant and facilitates removal of nitrogen with cathode as electron donor (Sun et al. 2020; Wang et al. 2018).

Bavasso et al. (2018) investigated the shortcut biological nitrogen removal in the anodic chamber using synthetic wastewater. They observed that nitritation depends on the TOC/N ratio and a low TOC/N ratio enhances the conversion of ammonium and nitrite accumulation. At TOC/N ratio < 1, ammonium removal efficiency reached up to 63–76%. This is because at lower organic concentration in the anodic chamber, dissolved oxygen promotes ammonia oxidation. Conversely, the nitrite removal was enhanced with an increase in TOC/N ratio upto 0.35 and with further increase in TOC/N ratio from 0.52 to 1, nitrate removal remains almost constant. A higher nitrite removal efficiency up to 80.3% was achieved at TOC/N ratio of 0.75 in the MFC system, thus indicating positive influence of increase in organic carbon on nitrite reduction. Furthermore, in coupled MFC system, with an increase in COD/N ratio from 5.5 to 10.3, Nguyen and Babel (2023) observed an increase in total nitrogen efficiency from 56.9 to 80.2% and reported complete nitrification in cathode chamber under DO level

of 4 mg/L while at low DO level of 0.6 mg/L, ammonium oxidation upto 95% and higher accumulation of nitrite was observed.

5.1.4.3 Dissolved oxygen The Dissolved Oxygen (DO) level must be properly controlled because at very lower DO concentration, incomplete nitrification occurs, thus leading to N H_4^+ accumulation whereas at higher DO concentration, activity of denitrifying organisms is inhibited, causing decrease in N O_3^- reduction rate (Kelly and He 2014; Sun et al. 2020). Tao et al. (2014) observed that at cathodic DO level of 3.5 and 2.8 mg/L, N H_4^+-N is converted to N O_2^--N but significant decrease in TN was not observed because DO level was too higher for the denitrifying bacteria whereas TN removal was greater than 85% at lower cathodic DO level of 2 and 2.5 mg/L. On the other hand, the voltage output and power density declined from 521 to 303 mV and 530 to 178 mW/m^2, respectively, when DO level reduced from 3.5 to 2 mg/L. Similarly, Zhang et al. (2020a, b) also reported higher rate of nitrosification and nitrification along with increased output voltage with increasing DO concentration in the cathodic chamber. This is because higher oxygen concentration not only enhances the mass transfer efficiency that leads to increased utilization rate of cathode microorganism and electrode but it also increases the cathodic potential due to increased flow of electrons, thus promotes higher electricity generation.

5.1.4.4 pH In BES, the application of ion exchange membranes (IEMs) promotes the migration of anions and cations other than hydroxyls and protons, resulting in pH splitting between anodic and cathodic chamber of BES. The oxidation reaction at anode produces proton which induces localized acidification in the vicinity of anode surface and thus adversely affects the microbial activity, thereby impeding the electrocatalytic activity of the biofilm. Besides, the reduction reaction at cathode consumes the proton and engenders alkalization in the cathodic chamber which ultimately lowers the bacterial activity. The pH split is an unfavorable condition in the BES which negatively influences the long term stability and energy output of the BES reactor (Zeppilli et al. 2021). For the denitrification, the optimal pH must be in the range of 7 ~ 8. The pH increases during the denitrification process because of the alkali formation and this increased pH can partially neutralize the acidic effluent of anodic chamber. Therefore, catholyte pH can be kept stable by looping the acidified effluent of anodic chamber as an influent for cathodic chamber, thereby eliminating the requirement of external chemicals for maintaining the pH (Sun et al. 2020). A study carried out by Li et al. (2013) to investigate the effect of anodic influent pH on MFC performance revealed that at higher anodic pH of 8, maximum power density of 54 W/m^3 was achieved because higher anodic influent pH balances the acidification of anolyte resulting from bacterial metabolism, which assist in the electrocatalytic activity of exo-electrogenic bacteria. Moreover, both nitrate and COD removal rate were lower at anodic pH of 6 as compared with pH values ranging from 6.5 to 8, thereby suggesting anodic influent pH values ranging from 7 to 8.5 is effective for electricity generation and pollutant removal.

5.1.4.5 Inoculum source The inoculum source and its culturing technique affects the composition of electroactive bacteria and performance of BES (Zhou et al. 2022). The pure culture inocula comprises of electrogenic bacteria whereas a certain portion of bacteria are non-electrogenic in case of mixed culture inocula, which consumes the substrate and escalates the electron transfer resistances across the multilayered biofilms, thereby decreases the overall power generation in the MFC (Jiang et al. 2010). Mainly, the mixed culture originates from the activated sludge, soil, sediments, and domestic sewage (Zhou et al. 2022). To boost the power generation of the MFCs, it is important to understand the effect of inoculum on the microbial adhesion and process of electron transfer. Though pure culture proved to be effective, but their performances may not be the same in case of heterogenous medium as compared with the mixed culture. *Geobacter sulfurreducens* can only utilize simple organic acids namely acetate and would rely on the other species to break down the complex molecules. In contrast, the superior attributes of the mixed microbial consortia include nutrient adaptability to utilize wide substrate range and concentrations present in the wastewater and stability towards fluctuations. Thus, using mixed microbial consortia for production of electricity has many practical advantages (Jiang et al. 2010).

In the biocathode, the functional microorganisms, originated from the innocula are the drivers of denitrification reactions. Therefore, the major regulating factor of biocathode denitrification is the development of functional microorganisms. The electrotrophic microbes are the functional microorganisms on the cathodic biofilm. The electron mediators and metabolic pathways differs with different electrotrophic microorganisms. Therefore, it is imperative to select proper inoculum to facilitate the electrotrophic microbial growth on the biocathode (Ding et al. 2018). In another study, Lee et al. (2013) explored the externally powered bio-electrochemical denitrification system (BEDS) by inoculating cathode electrode with anaerobic digested sludge and reported 81% nitrate removal and approximately 70% conversion of nitrate to nitrogen. Furthermore, Ding et al. (2018) investigated the effect of three different inocula, i.e., denitrifying, denitratating, and anammox sludge on MFC performance in terms of electricity generation and biocathode denitrification. They observed that at COD/N ratio of 5, denitratating and denitrifying biocathode demonstrated

higher nitrate removal rate as compared with anammox inoculum because chemoheterotrophic population were the functional microorganisms and organic matter was electron donor. Furthermore, with decrease in COD/N ratio from 5 to 1, the denitratating and denitrifying biocathode conversion efficiency declined whereas anammox biocathode conversion efficiency increased due to growth of mixotrophic population which can simultaneously use both organic matter and electricity as electron donor. On further decrease in COD/N to 0, these mixotrophic populations declined and electrotrophic population emerged because organic matter was replaced completely by electricity as electron donor for denitrification and therefore anammox biocathode shown relatively better denitrification performance than other inoculums. In addition, output power was higher in case of anammox, i.e., 45.93 mW/m^2 than denitratation (14.11 mW/m^2) and denitrifying (24.01 mW/m^2) biocathode system thereby suggesting anammox sludge is the optimal inoculum for both nitrogen removal and electricity generation.

5.1.4.6 Hydraulic retention time Hydraulic retention time (HRT) is also considered as an important aspect that influences the performance of BES. The improper hydrodynamics inside the reactor results in heterogeneous colonization of the cathodic chamber which leads to development of zones with different denitrification capability. For better water flux distribution within the reactor, researchers have attempted to modify the design and operation of the reactor (Pous et al. 2017). The variation in HRT significantly influences the change in water flux (hydrodynamics) and nutrient loading rates which in turn affects the power output, contaminant removal efficiency and microbial population in the bioreactor (Ye et al. 2020a, b; Sun et al. 2020). The effect of HRT on nitrate removal rate investigated by Pous et al. (2017) using tubular BES found that nitrate removal rates increased when HRT was reduced from 10.89 to 0.46 h. This was due to enhanced denitrifying activity because of better flux distribution owning to higher nitrate loading rate (at lower HRT). They also observed that decrease in HRT not only escalated the nitrate removal rate but it also boosted the reduction of denitrification intermediates as nitrite accumulation was not detected, thus suggesting upscaling of denitrifying BES by building and connecting compact reactors (lower volume) in series and operating at low HRTs to achieve higher nitrate removal rate.

Furthermore, the role of HRT in nutrient recovery and power output was also investigated by Ye et al. (2020b) in continuous flow mode two chamber MFC using synthetic municipal wastewater. They observed that with decrease in HRT from 0.69 to 0.35 days, the maximum voltage declined because lower HRT adversely effects the mixing conditions due to high flow rate of feed solution as well as causes loss of anaerobic microbes from anode chamber to a certain amount whereas higher HRT enhances the substrate retention time within the reactor and assist in complete degradation of substrate via electroactive bacteria. As a result, there will be an increase in the generation and transfer of electrons to the anode surface at higher HRT which could improve the power output. Therefore, compared with different HRTs (0.41, 0.52, and 0.35 days), the maximum power density (253.84 mW/m^2) and coulombic efficiency (25.01%) were achieved under HRT of 0.69 day. Moreover, in this study, decrease in HRT from 0.69 to 0.35 days has no significant influence on COD and nutrient removal rates because lowest HRT value is still sufficient for anaerobic microorganisms to effectively remove the organic compounds and uptake nutrients ($N\ H_4^+$ and $P\ O_4^{3-}$). In contrast, several researchers have reported that higher HRT is positively correlated with COD degradation because greater residence time of substrate in anodic chamber allows more contact time with anaerobic microbes for effective organic matter degradation and thereby results in better effluent quality (Ye et al. 2020b).

5.1.4.7 Effect of alternating current In BES, electrostimulation can be used for improving the denitrification efficiency by enhancing the microbial activity. Currently, direct current (DC) is generally used for applying an electric field but it can promote formation of unwanted and toxic byproducts for microbial consortia such as H_2O_2, Cl_2, O_3, and $OH^{\bullet}$ during a chemical reaction. Therefore, compared with DC, alternating current (AC) is favorable as the magnitude of frequency changes periodically and net charge is zero (Hoseinzadeh et al. 2018). Furthermore, alternating current solves the issue of cathode inactivation and anode corrosion as well as exhibits a positive influence on the denitrifying bacteria in terms of bacterial cell movement, metabolic activity, cell morphology, and shape based on the applied electrical characteristics including waveform, frequency, and voltage. The application of an electric field increases the cytoplasmic membrane permeability and improves the transfer of nutrients through the membrane (Hoseinzadeh et al. 2018; Dehghani et al. 2018a). Studies have reported that low voltage low frequency alternating current increase the enzymatic activity and in turn lead to a higher nitrate removal rate whereas at higher voltage and frequency, the nitrate removal rate decreases. This is because at higher voltage, permeabilization through cell membrane becomes irreversible and results in cell death while at higher frequency, due to electrode polarization, repulsive forces increase which lowers the bioavailability of nitrate to electrode biofilm (Hoseinzadeh et al. 2017; Dehghani et al. 2018b). Moreover, Dehghani et al. (2018b) investigated the effect of AC waveform and reported that nitrate removal efficiency was higher in triangular (88.84%) and sinusoidal waveform (76.33%) than as compared with square waveform (63.8%) implying AC

waveform characteristics such as peak voltage, pulse width, rest time induces a vital role in nitrate removal.

Reactor configuration

The application of BES for nutrient conversion and recovery has demonstrated several advantages compared to traditional technologies. However, still there exist several challenges that need to be solved to ensure this technology to be commercially viable, stable, efficient and profitable. Thus, it is important to majorly focus on developing innovative configuration of reactors with high performance capability to enhance the effluent quality and maximize power production (Nguyen and Babel 2022). Therefore, this section briefly discusses the different configuration of BES used for nutrient conversion and recovery to fulfill the gap existing in the current literatures.

5.1.5.1 Single chamber configuration Currently, for high salt nitrogenous wastewater, biological treatment is not effective owning to inhibition of microorganism's metabolic activity and growth. Therefore, to ameliorate the electricity generation and SND efficiency under higher salt condition, Zeng et al. (2020) constructed three phase single chamber MFC (TP-MFC) by integrating *Halomonas Cells* as an immobilized phase in the traditional bipolar MFC. Compared with common MFC, TP-MFC demonstrated a higher nitrogen removal efficiency of 63.4% as well as the higher average voltage, i.e., 439.3 mV at NaCl concentration of 30 mg/L. The microbial analysis discovered that TP-MFC were enriched with *Acinetobacter*, *Pseudomonas*, *Halomonas*, and *Alcaligenes* inferring that secretion of ecotine by immobilized *Halomonas* provided sufficient salt tolerance to the exo-electrogenic microorganisms growing in the high salt environment. Furthermore, Liu and Wu (2021) found that among the seven strains of *Halomonas*, *Halomonas sp* B01exhibits higher power density (24.2 mW/m^2) and nitrogen removal rate (93.8%) on sea water substrate. In fact, under varying NaCl concentration (30 -150 mg/L), the average nitrogen removal rate was found to be 88.9% thereby indicating greater capability of *Halomonas sp* B01 for removal of nitrogen on seawater substrate. Figure 8a depicts the nitrogen removal mechanism where function of *Halomonas sp* B01 biofilm is to perform nitrification at anode and denitrification at cathode.

To treat low C/N ratio wastewater, Yang et al. (2021) operated a series of air–cathode microbial fuel cells (ACMFCs) and achieved maximum TN, NH_4^+-N and COD removal efficiency upto 93.5%, 92%, and 94%, respectively. It was found that an optimally low C/N ratio wastewater (LCNW) facilitated a balance between the nitrification and denitrification that eventually enhanced the SND process. Moreover, a higher coulombic efficiency (69.2%) and maximum power density (1400 mW/m^2) was achieved at a low C/N ratio of 1.7–2.6 thereby inferring that ACMFCs can be used as a potential technology in treatment of low C/N ratio wastewater with higher recovery of energy.

5.1.5.2 Double chamber configuration For optimization of the reactor configuration and to enhance the rate of nitrogen removal, Zhang and He (2012) developed dual-cathode MFC comprising of common anode along with inner cathode for bio-electrochemical denitrification and outer cathode for nitrification as shown in Fig. 8b. They reported that the maximum COD removal occurs at anode while aerobic oxidation of the remaining COD occurs at the outer cathode. The organic removal in outer cathode is crucial for effective functioning of inner cathode because entry of remaining COD in the inner chamber will acts as electron donor which will facilitate the development of heterotrophic denitrifying bacteria, as a consequence coulombic efficiency will decline. Moreover, findings showed that ammonium and nitrate removal rate escalated with decrease in COD/N ratio. At different nitrogen loading rate, ammonium removal was more than 96% whereas nitrate removal efficiency declined at higher nitrogen loading which ultimately lowered the total nitrogen removal efficiency indicating denitrification as the rate limiting process in dual-cathode MFC.

In 2013, Huang et al. studied the influence of C/N ratio (2, 2.7 and 3.5) and found that nitrate and total nitrogen removal rate in BES increased upto 1.09 $g/m^3/h$ and 0.97 $g/m^3/h$ respectively with increase in C/N ratio upto 3.5 because at lower C/N ratio, autotrophic denitrifying bacteria are responsible for nitrate reduction, whereas higher C/N ratio facilitates the growth of heterotrophic denitrifying bacteria, as a result, the total denitrification rate increased. Furthermore, Nguyen et al. (2015) compared the autotrophic denitrification performance at biocathode using either biotic or abiotic anode in double chamber BES. The nitrogen removal efficiency and rates were found to be higher in BES with biotic anode, i.e., 78% as compared with abiotic anode, i.e., 43%. In addition, development of different bacterial consortia in biocathode was observed with varying operational conditions in BES and the developed bacterial consortia exhibits a pivotal role in the denitrification performance.

A continuous operation of dual chamber MFC was investigated by Oon et al. (2016) for 180 days. They witnessed lower nitrate removal in open circuit condition than closed circuit condition due to non-availability of electron at the cathode under open circuit condition. Moreover, in the cathode chamber, increase in the concentration of ammonium ($N H_4^+$) ions was observed and it was hypothesized that ammonium generation can be due to dissimilatory nitrate reduction to ammonium (DNRA) process carried out by DNRA bacteria. Apart from these, both COD and nitrate reduction enhanced when organic loading is increased and

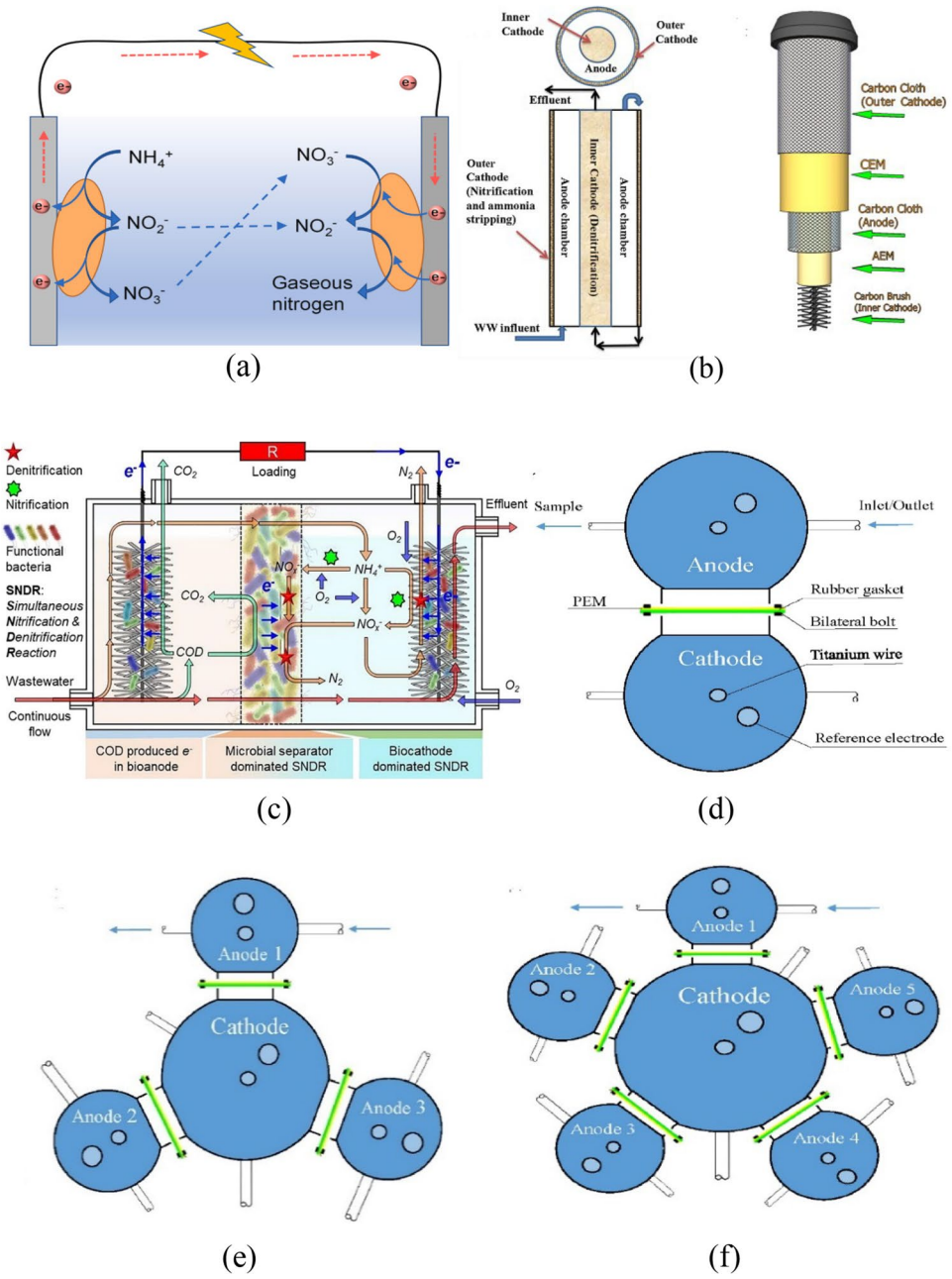


Fig. 8 Schematic illustration of **a** nitrogen removal mechanism of *Halomonas sp*. B01 in single chamber MFCs (Liu and Wu 2021), **b** dual cathode MFC system (Zhang and He 2012), **c** microbial electrochemical system comprising biocathode and microbial separator (MS) (Li et al. 2022); multi-anode (MA) MFC system, **d** 1A-MFC, **e** 3A-MFC, and **f** 5A-MFC (Huang et al. 2020)

thus stimulated higher power output. The nitrate removal was majorly due to autotrophic denitrification and partially conversion to ammonium. In another study, under varying temperatures and dissolved oxygen levels, Zhao et al. (2017) witnessed that increase in temperature from 36 to 48 ℃ within the reactor resulted in higher TN removal due to volatilization of ammonia whereas NO_2^--N concentration in cathodic effluent declined due to inhibition of ammonia oxidizing bacteria (AOB) growth at higher temperature. This is because appropriate temperature for AOB metabolic activity is 20–30 ℃. Besides, at lower DO concentration of 0.5–1 mg/L, traditional heterotrophic denitrification was dominant, thus facilitates greater TN removal whereas at higher DO level of 3–4.2 mg/L, electrode denitrification by aerobic denitrifiers was dominant in the cathode and thus advantageous for attaining higher voltage in MFC. In fact, variation in DO level at higher temperature stimulated change in dominant species from facultative heterotrophic bacteria and thermophilic autotrophic nitrifiers at lower DO level to thermophilic aerobic denitrification bacterium (ADB) at higher DO level.

The microbial separator has emerged as an economical substitute for the costly ion exchange membrane, which lowers the operational cost of MES. Therefore, Li et al. (2022)

constructed microbial electrochemical system comprising biocathode and microbial separator (MS) as shown in Fig. 8c. They observed 1.8 and 1.6 times greater N H_4^+-N and TN removal rates respectively in case of low influent TN concentration (40 mg/L) than high influent concentration (140 mg/L). Microbial analysis revealed higher relative abundance of denitrifiers, i.e., 29.5–34.6% on biofilm of microbial separator which proves effective denitrification role of MS whereas higher relative abundance of SND bacteria (22.5%) and denitrifiers (26.9%) on the biocathode proves nitrification and denitrification function of biocathode.

5.1.5.3 Multiple chamber configuration To examine the impact of multi-anode (MA) system on the nitrogen removal efficiency and power generation Huang et al. (2020) constructed three MFCs with different number of anode electrode (1A, 3A, and 5A) as shown in Fig. 8d–f. The investigation revealed that 3A-MFC shown maximum power density of 236.7 mW/m^3 which was 1.2 and 2.6 times higher than 5A-MFC and 1A-MFC, respectively. Besides, compared with other configurations, a stable voltage was achieved quickly by 3A-MFC because 3A-MFC have more electron output channels as compared with 1A-MFC, while in case of 5A-MFC, due to a greater number of anodes, stable voltage is achieved after a long time as compared with 3A-MFC. In terms of COD removal efficiency, 1A-MFC shown greater performance, i.e., 94.90% than 3A-MFC (94.04%) and 5A-MFC (84.10%). This is because multi anode configurations not only increases the H^+ and e^- migration pathway but also increases the number of O_2 channels from cathodic to anodic chamber and therefore, dissolved oxygen level in the anodic chamber rises. This disturbs the anaerobic environment of microorganisms because of which COD removal efficiency of anodic chamber declines. Besides, 3A-MFC shown higher TN removal efficiency, i.e., 71.1% and simultaneous nitrification and denitrification rate, i.e., 93.5% as compared with other two configurations due to higher electric energy output which enhanced the TN removal efficiency and SND rates. Subsequently, under different operational conditions and configurations, Zhu et al. (2021) studied the performance of multi-anode MFC (MA-MFC) system comprising of three anodes. The NH_4^+-N and TN removal rates were higher in parallel connection than series connection configuration of MA-MFC system because electron migration from anode to cathode occurs through various paths that stimulates higher current generation and thereby boosts the removal rates. Besides in series configuration, increase in cathodic COD/N ratio from 0 to 4.5 resulted in decrease in NH_4^+-N removal efficiency from 91.73 to 85.4% because aerobic heterotrophic bacteria utilized more oxygen as compared with aerobic autotrophic nitrifier bacteria because of higher competitiveness. Furthermore, inhibition of autotrophic denitrification and dominance of heterotrophic denitrifying bacteria with increase in cathodic COD/N ratio resulted in decrease in power density from 241.8 to 151.2 mW/m^3. Moreover, TN removal rates increased by 17.8% in the closed-circuit condition as compared with open circuit condition because of electrochemical denitrification.

5.1.5.4 Integrated configuration For simultaneous carbon and nitrogen removal, Xie et al. (2011) used coupled MFC system comprising of an anoxic biocathode MFC (A-MFC) and oxic biocathode MFC (O-MFC) as shown in Fig. 9a. In this system, to solve the problem associated with ammonium losses due to migration of N H_4^+-N from anodic to cathodic chamber in A-MFC, the effluent of A-MFC was recirculated to cathode chamber of O-MFC at different recirculation ratios for further nitrification. It was reported that A-MFC

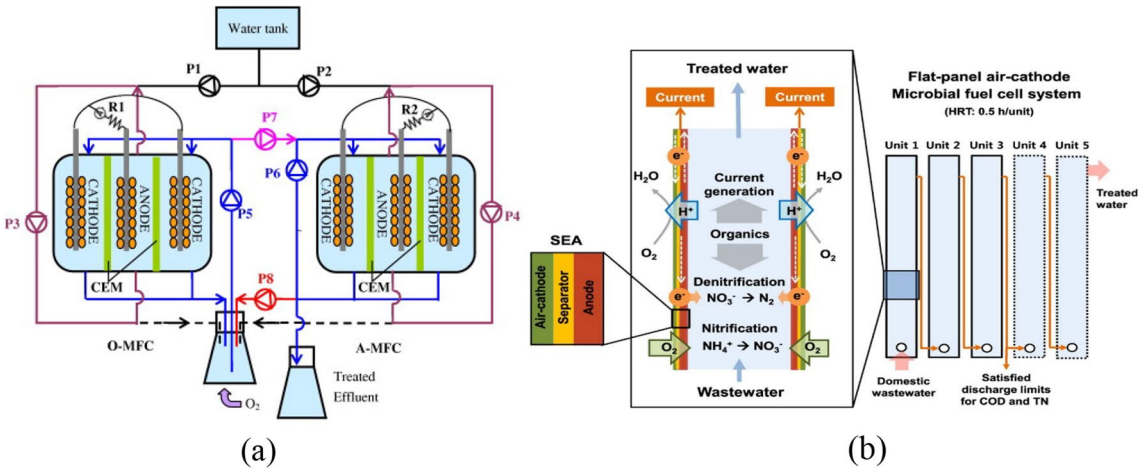


Fig. 9 Schematic illustration of **a** coupled MFC system (Xie et al. 2011), **b** flat panel air cathode MFC (Park et al. 2017), and **c** pilot scale cylindrical reactor (Suransh et al. 2023)

demonstrated maximum power density of 6.8 W/m^3 at external resistance of 5 Ω whereas O-MFC shown 15.1W/m^3 at 20 Ω. Also, the total nitrogen removal efficiency increased from 56.4 to 87.1% whereas N H_4^+-N removal efficiency declined from 95 to 88.5% with decrease in external resistance from 20 to 5 Ω in A-MFC. Correspondingly, in A-MFC, power density gradually increased when recirculation ratio is increased because at higher recirculation ratio, more amount of ammonium is oxidized in the O-MFC as a result of which nitrate loading in A-MFC increased thus, resulted in higher power generation. Besides, in coupled MFC system, total COD removal efficiency was greater than 98% and influent flow ratio had a significant influence on the TN removal efficiency.

To ensure the post treatment process after MFC along with SND and energy recovery, Li et al. (2017) constructed membrane bio-electrochemical reactor (MBER) by integrating two MFCs with aerobic MBR. A lower pH gradient in the anodic and cathodic chamber was observed. Results depicted that higher COD loading rate restrained the cathodic reactions and induced lower electricity generation. At higher COD/N ratio, current density declined causing impediment to the electrochemical denitrification in MBER. Still, TN removal efficiency enhanced from 66.5 to 84.3% with surge in COD/N ratio from 3.6 to 9.3 due to the availability of adequate carbon source for heterotrophic denitrification reaction. On the other hand, at higher COD/N ratio, no significant variation in N H_4^+-N removal was noticed because nitrification was suppressed attributable to decrement in dissolved oxygen in the cathodic chamber. Moreover, it was concluded that long HRT and lower external resistance boosts the nutrient removal efficiency and electricity generation in MBER.

5.1.5.5 Stacked configuration Aimed for higher removal of organics and nitrogenous compounds from domestic wastewater to meet the discharge limits, Park et al. (2017) built flat panel air cathode MFC (FA-MFC), consisting of series connection of five MFC units as shown in Fig. 9b. They observed that first unit produced higher current due to availability of higher COD concentration while the subsequent units produced a smaller amount of current due to lesser availability of readily degradable COD in the incoming liquid. As a whole, FA-MFC achieved TN and COD removal efficiency up to 85% and 94% respectively in 8 months of operation under HRT of 2.5 h. Suransh et al. (2023) developed pilot scale reactor by parallely stacking four individual cell for domestic wastewater treatment as shown in Fig. 9c and obtained average removal efficiency of 71.59%, 84.93%, and 93.52% for ammoniacal nitrogen, nitrate and COD, respectively under 92 days of operational period. Furthermore, the reactor attained higher power density and maximum current upto 23.52 mW/m^3 and 43.7 mA respectively, thus proving its capability as cost-effective solution for field scale application.

Phosphorus

The lower phosphorus concentration in wastewater consumes higher energy that makes the recovery process of phosphorus uneconomical. In the recent year, energy induced migration of phosphorus using electroactive bacteria has been reported. Recovery of phosphorus can be performed in single, double and triple chambered BES. In case of higher phosphate concentration, precipitation leads to production of vivianite, struvite and hydroxylapatite ($Ca_5(PO_4)_3OH$). In these methods, electroactive bacteria either act as the catalyst to extract biomass energy for concentrating the phosphate or function as Fe(III) reducer to supply Fe(II) for the synthesis of vivianite (Li et al. 2020). Therefore, this section critically examines the developed methods so far for phosphorus recovery and highlights the multiple factors affecting the recovery process with major stress on the research gaps.

Struvite

The electrochemically mediated precipitation (EMP) of P minerals at the surface of the cathode is attained through pH elevation, despite no direct participation of P minerals in the electrochemical reactions. At the cathodic chamber, reduction of water molecules leads to increase in localized pH due to production of H_2 and OH^-. Meanwhile, cations from the anodic chamber move and accumulate in the cathodic chamber due to electric field. This causes increase in saturation index (SI) of P minerals, thus stimulates the formation and precipitation of calcium phosphate mineral (Ca-P) as shown in Eq. 5. Increased localized pH near the cathode will lower the calcium phosphate mineral (Ca-P) solubility and this way, direct collection of precipitate is possible from the cathodic chamber without requirement of solid–liquid separation process, as needed in traditional methods (Y. Wang et al. 2022a, b). The precipitation of Ca-P mineral can be achieved without external addition of calcium ions as appropriate stoichiometric amount for precipitation already exist in the engineered and natural aquatic system (Lei et al. 2020).

$$5Ca^{2+} + 3HPO_4^{2-} + 4OH^- \leftrightarrow Ca_5\left(PO_4\right)_3OH \downarrow + 3H_2O \quad (5)$$

In the EMP system, Ca-P and struvite are the common products (Wang et al. 2022a, b). Struvite is referred to a crystal with equimolar concentration of the magnesium ammonium phosphate bonded with six water molecules ($MgNH_4PO_4.6H_2O$) (Wang et al. 2022a, b; Rahman et al. 2014; Peng et al. 2018). In the supersaturated solution,

struvite is formed due to chemical reaction between free PO_4^{3-}, NH_4^+, and Mg^{2+} ions as shown in below Eq. (6) (Krishnamoorthy et al. 2021). The struvite formation has the benefit of recovering both phosphorus (P) and nitrogen (N) (Wang et al. 2022a, b).

$$Mg^{2+} + NH_4^+ + PO_4^{3-} + 6H_2O \leftrightarrow MgNH_4PO_4.6H_2O \quad (6)$$

The above reaction is pH dependent. In alkaline condition, precipitation reaction occurs within pH 7 to 11.5. Various parameters affect the process of struvite precipitation which includes pH, degree of saturation, foreign ions, seeding, hydraulic retention time, Mg/P molar ratios, mixing energy, seeding, and feeding sequence. At the cathode, the direct production of struvite is intricate because very high local pH will convert NH_4^+ to NH_3. In addition, high pH environment may result in $Mg(OH)_2$ production instead of struvite. Therefore, OH^- production which depends upon current density needs to be monitored. It was reported that with increase in current density from 5.8 to 17.3 A/m^2, struvite recovery decreased from 91 to 18%. In additional, extra Mg^{2+} is needed for effective recovery of struvite. When wastewater contains lower concentration of N and Mg^{2+}, then it is apt to recover P in the form of Ca-P (Eq. 5) dueue to existence of abundant Ca^{2+} in wastewater (20–120 mg/L) (Wang et al. 2022a, b).

Influencing factors affecting struvite precipitation

Effect of pH, current density and initial phosphorus concentration The pH is a decisive parameter which governs the P minerals precipitation. The term local pH denotes pH at the cathode while bulk pH refers to the pH of the solution. Under different pH condition, the phosphorus exists in different forms. For example, at pH below 7.2, phosphate exists as $H_2PO_4^{2-}$ whereas at pH above 7.2, the predominant form is H PO_4^{2-}. At higher pH values, SI of typical phosphate increases because of deprotonation of phosphate and higher availability of OH^- ions, therefore constituents of hydroxyapatite (HAP) exist at higher pH. The solubility of struvite decreases with increment in pH values, which in turn ameliorates its precipitation potential (Daneshgar et al. 2018). In the electrochemical system, local pH is more critical than bulk pH because water molecules reduction at the cathode increases the local pH and facilitates the nucleation as well as growth of P crystals. Correspondingly, bulk pH also exhibits some effect. It could indirectly influence the phosphate precipitation by regulating the speciation and concentration of bicarbonate in wastewater. In case of acidified wastewater (low pH), the concentration of bicarbonate will decrease, signifying less bicarbonate will compete with phosphate for Ca^{2+} ion. This way bulk pH can eliminate the unfavorable effect of biocarbonate concentration on the phosphate precipitation (Wang et al. 2022a, b). Studies have reported that pH value near to 9 is most suitable for the precipitation of struvite. Moreover, struvite precipitation can be hindered when pH continues to grow beyond 9 due to decrease in availability of ammonium ions owning to its conversion into ammonia gas (Siciliano et al. 2020).

Besides, in the electrochemical systems, the current density also modulates the phosphorus precipitation by governing the rate of formation of O H^- at the cathode. The removal percentage of phosphate increases with increase in the current density because higher current density facilitates the production of O H^- at the cathode, thereby engenders the phosphate precipitation. But when current density increases beyond a particular limit, the phosphate removal rate remains stable (Wang et al. 2022a, b).

Initial phosphorus concentration in the influent also limits the recovery efficiency for struvite. The reaction kinetics for the struvite precipitation are exorbitantly increased by the concentration of phosphorus in the influent. Struvite precipitation can be effectively achieved when phosphate concentration is greater than 100 mg/L in the feed stream (Kundu et al. 2022). A lower phosphorus concentration in the influent decreases the struvite recovery performance and entails a longer induction time. As a result, economic feasibility of struvite recovery process is greatly reduced. The availability of higher phosphorus concentration is challenging because phosphorus concentration typically ranges from 6 to 25 mg/L in the influent wastewater. In the feed stream, when the concentration of ortho-phosphate is low (i.e., lower than 55 mg/L), supplementary phosphate salts (H_3PO_4 and KH_2PO_4) are added for the struvite precipitation. For the effective recovery of phosphate via struvite precipitation, combination of urine and manure is an ideal choice among all waste feedstock due to presence of significant higher phosphate concentration, i.e., 350–2500 mg/L in urine and 370–600 mg/L in manure (Kundu et al. 2022).

Influence of temperature, mixing speed and electrochemical cell configuration The morphology, solubility and the formation of struvite crystals are significantly affected by temperature (Siciliano et al. 2020). At higher temperature, the precipitation of struvite decreases and therefore, optimal temperature range for the precipitation of struvite is around 25–35 ℃ (Kundu et al. 2022).

In addition, solution mixing speed also affects the struvite crystallization process. When the mixing speed of the solution is high, the liberation of CO_2 surges and facilitates an increase in solution pH, leading to struvite crystallization. In addition, the shape and size of the crystal vary with mixing rates. At lower mixing rates, the size of the crystal was smaller owing to less dissipation, causing greater saturation (Krishnamoorthy et al. 2021). While high mixing speed expedites the nucleation rate as well as enhances the

crystal breakage, thereby resulting in smaller-sized crystals (Krishnamoorthy et al. 2021; Kundu et al. 2022). Studies have reported that, compared with non-stirring condition increase in phosphorus removal efficiency was observed from 72.7 to 97.3% under stirring condition (160 rpm). Furthermore, phosphate removal efficiency was more than 97.3% in the initial 5 min of mixing and its reached 99% after 30 min of mixing indicating mixing time is more important than mixing speed for removal of phosphate (Krishnamoorthy et al. 2021).

The precipitation of phosphorus occurs at cathode; therefore, cathode design is vital for the phosphorus removal and recovery. The cathodic surface area can affect the removal efficiency and purity of recovered product. Ca-P precipitation from domestic wastewater increases with higher specific surface area of cathode under low current density (Wang et al. 2022a, b). For large specific surface area, carbonaceous material, i.e., graphite felt are suitable than traditional titanium plates. In addition to surface area, electrode gap is also important as it governs the recombination of anodic-generated H^+ and cathodic-generated OH^- ions and can affect the rate of utilization of OH^- for phosphate precipitation. Apart from these, membrane application for separation of anodic and cathodic chamber can also significantly enhance the phosphorus removal (Wang et al. 2022a, b).

Effect of Mg/P and Ca/Mg molar ratio In the struvite crystallization, Mg is the essential element. It is externally added as nutrient enriched wastewaters are mostly devoid of the stochiometric amount of Mg needed for the struvite precipitation. Theoretically, struvite formation occurs when molar ratios of Mg^{2+}:NH_4^+:PO_4^{3-} is 1:1:1. A higher molar ratio of Mg:P increases the degree of saturation at a particular pH and in turn facilitates the phosphorus removal. Mg:P ratio in the range 1–1.6 was investigated and found effective removal rates were attained at higher Mg:P ratio. Basically, a higher Mg:P ratio have a strong effect on lowering the phosphate concentration (Krishnamoorthy et al. 2021). On the other hand, the cost associated with magnesium dosing is around 75% of total operational cost and therefore various laboratory and pilot scale studies have been conducted with different Mg salts to lower the operational expense as well to ensure the standard quality products. Compared with $Mg(OH)_2$ and MgO, $MgCl_2$ is found to be efficacious due to its higher solubility and lower dissolution time along with its non-corrosive and non-toxic nature, but it is expensive and require supplementation of alkali for improvement of pH. On the basis of the efficiency, the increasing order is found to be $MgCO_3 < Mg(OH)_2 < MgO < MgSO_4 < MgCl_2$. $MgCO_3$ is found to be least effective because of extremely low solubility (Krishnamoorthy et al. 2021). Furthermore, with increase in the concentration of Ca (Ca/Mg of 0–1.8) in the influent wastewater, the efficiency of recovery and precipitation increased due to phosphate precipitation as struvite and calcium phosphate. At molar ratio of Ca/Mg of 1:1, a high amorphous content was formed at all NH_4 concentrations, and it decreased when Mg concentration increased, thus indicating the function of Ca in crystallization. It was reported that formation of HAP delayed when Mg:Ca < 0.2 whereas at Mg:Ca > 2, undesirable effect of Ca gets eliminated during struvite crystallization (Krishnamoorthy et al. 2021).

Effect of organic matter and existence of competitive ions The different types of organic substances are present in the wastewater. These substances lower the rate of struvite formation. A higher Mg/P ratio governs the humic substances and transforms the prismatic structure of struvite crystal to pyramid structure. It was also noticed that both phosphocitrate and citrate were the potential growth inhibitors. This is because they get adsorbed onto the crystal surface and blocks the active sites for crystal growth and thus escalates the induction time of crystallization. Furthermore, they can damage the environment and human health when utilized as fertilizer. Both succinic and humic acid can inhibit the development and growth of struvite crystal whereas no significant effect was observed in case of acetic acid. Conversely, higher glucose concentration in the solution enables structural formation and phosphorus removal efficiencies. Overall, organic substance exhibits a slight influence on the composition of struvite without any impact on the purity (Krishnamoorthy et al. 2021).

Also, it is well known that different wastewater comprises of different ionic species of varied concentrations such as Zn^{2+}, Al^{3+}, Ca^{2+}, Cu^{2+}, SO_4^{2-}, HCO_3^-, and CO_3^{2-} (Siciliano et al. 2020). The presence of ions adversely affects the nutrient recovery efficiency, reaction speed, and morphology. Therefore, the purity is hindered by the metallic ions that compete for phosphate and forms complexes with struvite during precipitation. Moreover, these ions get adsorbed and blocks the active site on the surface of the crystals, thereby affects the crystal growth. It was reported that crystal size reduces by up to 46% in presence of NO_3^-, Ca^{2+}, and Fe^{2+}. These non-participating cations accumulate around the anionic species of struvite and thereby increases the crystal induction time. In case of higher Ca^{2+} concentration, irregular crystals are formed and size of struvite crystal particles declines from 34.2 to 18.4 µm. Also, higher Ca^{2+} concentration tends to prolong the induction time and severely impedes the struvite formation because calcium ions react with PO_4^{3-} to form HAP and apatite (Krishnamoorthy et al. 2021). As a consequence, PO_4^{3-} availability in the solution declines, and in turn reduces the struvite formation (Siciliano et al. 2020; Krishnamoorthy et al. 2021). However, this phenomenon is not much observed in presence of sulfates (SO_4^{2-}), sodium (Na^+) and carbonate (CO_3^{2-}) (Krishnamoorthy et al. 2021).

Furthermore, a higher alkalinity also effects the struvite precipitation. The HCO_3^{2-} and CO_3^{2-} can form bonds with NH_4^+ and Mg^{2+}, thus results in formation of stable aqueous phase of $Mg(HCO_3)_2$, $MgCO_3$ and NH_4HCO_3 and thereby lowers the concentration of two components for the struvite nucleation (Siciliano et al. 2020).

Vivianite

The phosphorus recovery can also occur in the form of vivianite ($Fe_3(PO_4)_2.8H_2O$), i.e., hydrous iron phosphate mineral (Pikaar et al. 2022). The formation of vivianite is favored in anoxic non-sulfide environment enriched with orthophosphate (PO_4^{3-}) and ferrous ions (Fe^{2+}) (Yuan et al. 2021). In general, iron in the wastewater originates from the application of iron containing flocculants namely poly aluminum ferric chloride (PAFC) and ferric chloride ($FeCl_3$) in WWTPs. While phosphorus mainly originates from the anthropogenic sources such as industrial, agriculture, and domestic waste. The formation of vivianite is associated with the (1) dissimilatory reduction of Fe (III) with the help of dissimilating iron reducing bacteria (DIRB) (2) conversion of organic phosphorus to phosphate by anaerobic microorganisms (Yuan et al. 2021). The continuous occurrence of these aforementioned processes leads to an increase in localized concentration of PO_4^{3-} and Fe^{2+} ions. When the concentration of these ions reaches the requirement of solubility product constant ($K_{sp}=10^{-36}$), the vivianite formation begins to occur (Zangarini and Sciarria 2020). The following reaction depicts the vivianite formation.

$$\text{Organic P} \rightarrow PO_4^{3-} \tag{7}$$

$$Fe(OH)_3 + 3H^+ + e^- \rightarrow Fe^{2+} + 3H_2O \tag{8}$$

$$3Fe^{2+} + 2PO_4^{3-} + 8H_2O \rightarrow Fe_3(PO_4)_2.8H_2O \tag{9}$$

Among all, *Shewanella oneidensis* and *Geobacter sulfurreducens* is considered as prominent DIRB (Li et al. 2020; Yuan et al. 2021). The vivianite possess magnetic properties and can be utilized as slow release fertilizers and in manufacturing of lithium ion batteries (Yuan et al. 2021; Pikaar et al. 2022). In the nature, the vivianite formation can be influenced by the various factors such as redox conditions, nutrients conditions, sulfide formation, microbial community composition, pH and temperature (Li et al. 2020). For generation of energy to support the bacterial growth, the electroactive bacteria can perform reduction of metal such as Mn(IV) and Fe(III) (Pikaar et al. 2022). Microorganism can also alter the redox conditions through uptake of NO_3^-, O_2 and various electron acceptors. The crystallization of vivianite occurs in neutral to slight-basic pH range thereby eliminating the requirement of alkaline dosage in wastewater as required in struvite removal. However, large quantity of dissolved iron is required, which exhibits a problem when vivianite crystals are removed from the sludge (Pikaar et al. 2022). For vivianite biosynthesis, a balanced concentration of Fe, S, and P exhibits a vital role. The existence of sulfide can lower the concentration of reactive ferrous iron available for biosynthesis of vivianite. Increased salinity and excessive organic matter promote microbial sulfate reduction to produce sulfide, which competes with phosphate to react with iron, thus restraining the combination of phosphate and iron and impeding the formation of vivianite. An appropriate S:Fe ratio is important for occurrence of vivianite. The molar S:Fe ratio < 1.5 indicates iron supply is more than the sulfide production, which promotes the vivianite formation whereas at molar S:Fe ratio > 1.5, vivianite formation is restricted due to limited supply of iron as majority of iron tends to associate in sulphidic form. A higher concentration of reactive iron is likely to eliminate the interference of sulfide in vivianite formation. Also, studies have reported that under phosphorus limiting condition, vivianite competes phosphorus with the bacterial growth. Primarily, the phosphate is to be utilized for the microorganism's growth and then for the production of vivianite. The vivianite biosynthesis is observed to be more prominent at a molar Fe:P ratio of 1. Therefore, it is obligatory to provide adequate phosphorus supply that can satisfy the phosphorus requirement for biomass growth of DIRB and vivianite formation. The phosphorus recovery as vivianite is more valuable as compared with struvite and hydroxyapatite due to greater economic value, easy accessibility and broader applications (Yuan et al. 2021).

Nutrient removal/recovery using hybrid configurations

MFC-Algae integration

The major limitation associated with the MFC is the oxygen supply as electron acceptor in the cathode chamber because decrease in oxygen availability in the cathode chamber lowers the MFC performance. Moreover, higher aeration cost in the cathodic chamber limits the economic viability of MFC applications. Hence, photosynthetic microalgae have been utilized in the cathodic chamber to achieve self-sustainability (Colombo et al. 2017; Nookwam et al. 2022). The algal based MFC forms a syntrophic relationship between the algal biomass and bacterial population and this system operates with lower net energy input (Saratale et al. 2017). Some of recent studies based on microalgae application in

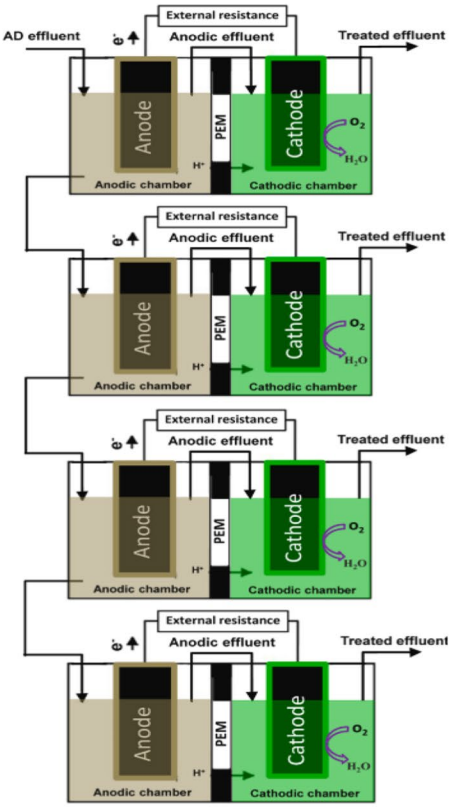
MFC for nutrient and organic matter removal have been summarized in Table S1 (supplementary material).

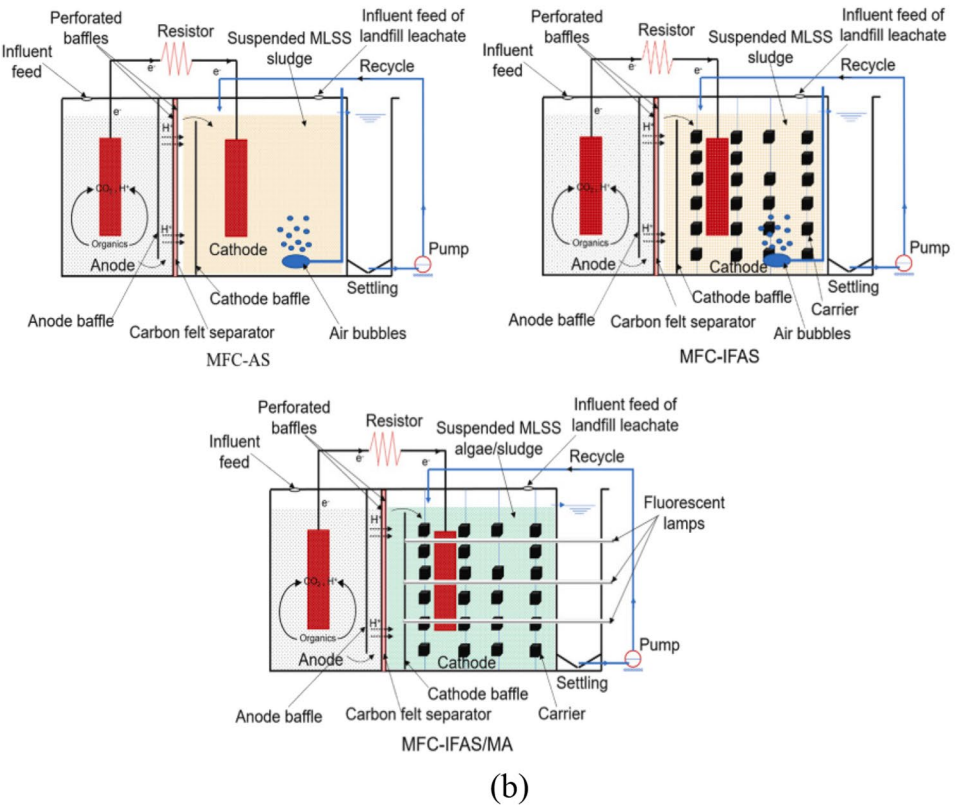
To understand the operational behavior of photosynthetic organisms in MFC, Colombo et al. (2017) built photosynthetic MFC (P-MFC) and compared its performance with air cathode MFC (A-MFC) and water cathode MFC (W-MFC). They observed comparable COD removal efficiency and similar current density upto 5 A/m^2 in case of A-MFC and P-MFC. But power density was slighter higher in A-MFC (0.98 W/m^2) than P-MFC (0.85 W/m^2). On the other side, W-MFC shown lower COD removal, current density and power generation. The varying ORR efficiency due to different DO levels in A-MFC, P-MFC and W-MFC has influenced the MFC performance in terms of organic matter removal. It was witnessed that photosynthesis in P-MFC exhibited a major role in sustaining the oxygen reduction reaction (ORR) at the interface of cathode whereas DO level at interface and bulk phase were extremely lower in case of W-MFC. Besides, 83% and 64% of N H_4^+-N were removed in the anodic compartment of P-MFC and W-MFC respectively due to nitrification reactions. While in the cathodic compartment, N H_4^+-N concentration was found to be undetectable in P-MFC due to algal uptake whereas it was 5.5% of initial N H_4^+-N concentration in case of W-MFC due to electro-osmotic flux.

Traditional MFC underperforms when encountered with wastewater having higher organic/inorganic forms of phosphorus and nitrogen (Nagendranatha Reddy et al. 2019). In A-MFC, algae at the cathode can take up the phosphorus and nitrogen for their growth and development and at the same time can utilize the CO_2 produced from the oxidation of organic matter at the anode for photosynthesis (Nagendranatha Reddy et al. 2019). Moreover, algae in the catholyte can act as buffering agent. The ORR at the cathode results in increase in alkalinity. Due to inorganic carbon demand, algae consume the CO_2 from the medium, thereby causing increase in pH. Also, algae can buffer the pH increase due to proton generation during respiration at night. The increase and decrease in pH during day and night time acts as buffering agent in balancing the pH of the catholyte (Luo et al. 2017). In algae MFC, most of the researchers have focused on enhancing the current output using specific algal strains and by implementing engineering strategies to certain extent (Saratale et al. 2017). Substantial research are being carried out to exploit the full potential of algae in different configurations of MFC to satisfy the growing energy demand and treatment of diverse pollutant (Saratale et al. 2017).

For removal of nutrients and bioenergy generation, Yang et al. (2018) built algae biofilm microbial fuel cell (ABMFC) and found higher removal efficiency for TP (96.4%), TN (95.5%) and COD (81.9%) respectively under continuous mode than batch mode, implying ABMFC has the potential to handle complex, real and variable wastewater continuously. Also, Wang et al. (2019) developed immobilized microalgal based photoautotrophic microbial fuel cell (PMFC) and studied the effects of illumination intensity and inoculated microalgal biomass on nutrients removal and electricity generation. They found that power density of PMFC system increased up to 466.9 mW/m^3 with increase in inoculated microalgal biomass from 0.25 to 0.75 g/L. In addition, nutrient removal efficiency significantly increased at higher inoculated algal biomass in the cathodic chamber indicating, nutrients uptake enhanced with surge in microalgal content. Moreover, optimal illumination for nutrient removal was found to be 10,000 Lux because lower (5000 Lux) and higher (15000 Lux) illumination intensity induces photo-inhibition phenomenon, which restrains the microalgal cells growth. The PMFC system achieved sCOD, TN, N H_4^+-N, and PO_4^{3-}-P removal efficiency upto 93.2%, 95.1%, 95.9%, and 82.7%, respectively. The enhanced TN removal efficiency is due to the symbiotic interactions among nitrification and denitrification bacteria and microalgae in the cathodic chamber, thus concluding the feasibility of this system in energy recovery and wastewater treatment in the economical manner. Moreover, Zhang et al. (2019) studied the swine wastewater treatment using photosynthetic algal MFC (PAMFC) and observed higher removal efficiency for total nitrogen (70.2%), ammonia nitrogen (85.6%), and total organic carbon (93.9%) in anode chamber and ammonia nitrogen (68.7%) in cathode chamber as compared to standalone MFC thus indicating the eminence of PAMFC in terms of nutrient and carbon removal.

As internal resistance of system mounts up with increasing size of the reactor, therefore multiplication of MFC reactors is a viable and practical approach for enhancing the power output with more capacity to treat a huge quantum of wastewater. Additionally, vertical cascade configuration will lower the energy requirement for fluid flow. Therefore, Nookwam et al. (2022) developed vertical cascade dual chamber MFCs with photosynthetic cathode chamber inoculated with *Scendesmus sp.* as shown in Fig. 10a to achieve higher power output level with enhanced capability to treat a large quantity of wastewater. They observed that, at optimized OLR, vertical cascade MFC fed with AD effluent from the rubber industry demonstrated a maximum total power density of 116.9 W/m^3 and which is higher than double chambered MFC (59.96 W/m^3). Furthermore, in a vertical cascade configuration, the first MFC shown highest power density, i.e., 62.68 W/m^3 followed by sequentially downstream MFCs because more nutrients are received by top MFC as compared with downstream MFCs which in turn allows better biofilm development and efficient transport of electrons towards the anode. In addition, the pollutant removal was also enhanced in vertical cascade MFC indicating efficient sequential treatment of wastewater. In another study, effectiveness of tubular photo-MFC reactor


AD effluent
External resistance
Anodic effluent
Treated effluent
Anode
PEM
Cathode
Anodic chamber
Cathodic chamber
(a)


Perforated baffles
Resistor
Suspended MLSS sludge
Influent feed of landfill leachate
Influent feed
Recycle
Cathode
Anode
Pump
Settling
Anode baffle
Cathode baffle
Carbon felt separator
Air bubbles
MFC-AS
MFC-IFAS
Carrier
Suspended MLSS algae/sludge
Fluorescent lamps
MFC-IFAS/MA
(b)

was investigated by Bolognesi et al. (2022) as polishing step for treatment of anodic effluent wastewater. The photo-MFC demonstrated higher COD (94%) and nutrients removal (55% TN and 62% TP) efficiency. Correspondingly, the COD removal rate and electricity generation increased with higher applied flow rate in photo-MFC due to higher OLR and turbulence that resulted in improved biomass composition and substrate diffusion.

Leachate is a complex and high strength wastewater. It leads to soil and groundwater pollution, thus, engenders health risks to humans. Leachate contains higher concentration of ammonia and organic compounds (Wu et al. 2015; Iskander et al. 2016). The leachate treatment using MFC has been explored in which organic matter were effectively removed but there exists knowledge gap regarding coupling effect of microalgae and attached biofilm activated sludge process in MFC for treatment of landfill leachate. Therefore, Elmaadawy et al. (2022) investigated and compared the leachate treatment efficiency and energy production of *Chlorella-Vulgaris* microalgae coupled MFC integrated fixed film activated sludge (MFC-IFAS/MA) system with conventional activated sludge (MFC-AS) and MFC integrated with fixed-film activated sludge (MFC-IFAS) as shown in Fig. 10b. The power density (20.5 mW/m^2), current density (238 mA/m^2) and output voltage (0.26 V) were higher in case of MFC-IFAS/MA than that of MFC-AS and MFC-IFAS systems. Also, compared with other two systems, MFC-IFAS/MA shown excellent N H_4^+-N and TN removal efficiency, i.e., 90.3% and 88.6% respectively due to dual effect, i.e., microalgae biofilm assimilation along with simultaneous nitrification and denitrification reactions on the surface of fixed carriers that resulted in greater ammonia and TN removal. The vigorous growth rate and productivity of microalgae evince the supremacy of MFC-IFAS/MA system in the treatment of landfill leachate and confirms the ability of algal biofilm in treatment process.

In summary, A-MFC, a bifunctional tool demonstrates significant advantages in terms of wastewater treatment and electricity generation in sustainable manner. The power generation in A-MFC can be augmented by optimizing reactor design (electrode spacing, membrane, and reactor geometry), optimizing operational conditions (illumination intensity, HRT, substrate concentration, C/N ratio, etc.), and using modified cathode electrode for enhancement of ORR. Furthermore, the nutrient removal and oxygen production can be enhanced by selecting genetically modified algal species. For upscaling and commercialization, technoeconomic assessment needs to be performed to account the overall cost and to ensure commercial feasibility. Also, integrative/hybrid A-MFC configuration with different processes need to be explored for multipurpose use including wastewater remediation, bioelectricity production and valuable bioproducts formation to attain zero waste discharge.

MFC-anaerobic digestion

Anaerobic digestion is a biological treatment process in which methane gas is produced during organic matter oxidation in absence of oxygen. This method is primarily used for the treatment of high strength organic waste such as poultry and livestock waste. Compared to the aerobic process, this process demonstrates higher COD removal, lower sludge production and higher destruction of complex solids. The integration of MFC with AD has received significant interest recently (Kim et al. 2015). A study was carried out by Kim et al. (2015) using MFC in batch mode for the treatment of AD effluent, initially fed with swine wastewater. They observed that using AD effluent in MFC solves the problem of low COD/TAN ratio due to effective removal of TAN in MFC.

The techno-economic potential of hybrid AD-BES for nutrient recovery, toxicity reduction and enhancement of CH_4 generation in full scale depends upon the type of integration of AD and BES (Fig. 11) (De Vrieze et al. 2018). The BES integration with AD can be implemented at parallel and series mode. In AD-BES series mode, BES can perform additional treatment of AD effluent or function as pre-fermented stage for pre-treatment of substrate whereas in parallel mode of operation, liquid and solid parts of substrate are treated in BES and AD respectively (Wang et al. 2022a, b).

MFC-forward osmosis

The forward osmosis (FO) process is an economical and energy efficient process with low tendency for membrane fouling, as the driving force is osmotic pressure instead of hydraulic pressure. The FO membrane can produce higher water quality due to elimination of broader range of contaminants. Several studies have reported the technical feasibility of integration of MFC with FO for simultaneous energy recovery, waste extraction and wastewater treatment. An investigation was carried out by Liu et al. (2017) by integrating anaerobic acidification and FO membrane with air cathode MFC (AAFO-MFC) and achieved maximum power density of 4.38 W/m^3 during long term operation of 40 days using low strength wastewater. Also, AAFO-MFC demonstrated higher TOC removal efficiency of 97% and complete rejection of phosphorus (> 97%).

In OsMFC, the selection of anolyte and catholyte solution is vital for energy production and water recovery as they produce adequate osmotic gradient across the FO membrane. Basically, in OsMFC, the feed and draw solutions are generally wastewater and NaCl solution respectively. The combination of FO into MFC will facilitate an additional function of better quality of water recovery along with desalination, wastewater treatment,

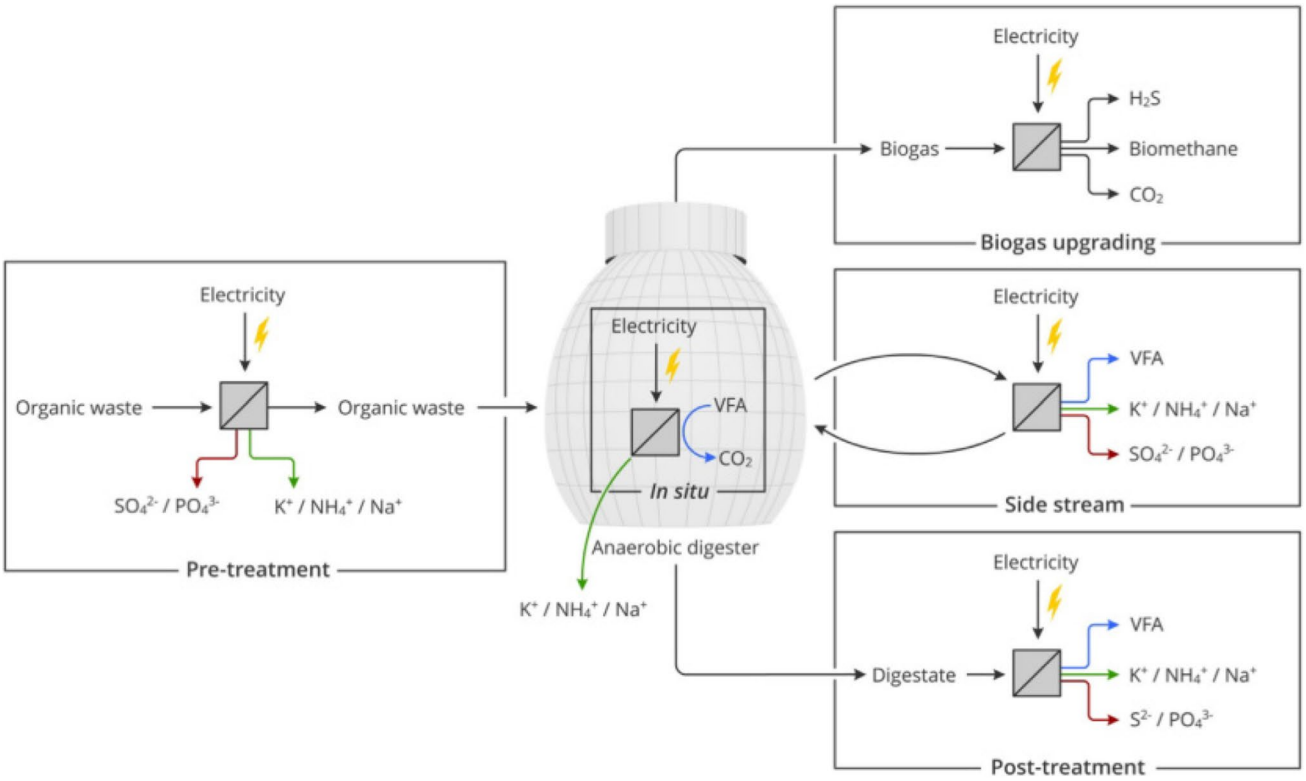


Fig. 11 Schematic illustration of possible combination of hybrid AD-BES in different stages, i.e., pre-treatment, side stream and post-treatment (De Vrieze et al. 2018)

and bioelectricity generation. In addition, lower internal resistance and improved proton diffusion through FO membrane induce lesser pH fluctuations and higher power output, which makes OsMFC more advantageous than MFC. The potential of OsMFC for nutrient concentration, electricity generation and energy production from urine was investigated by Gangadharan et al. (2021) under different experimental conditions. They found that the water recovery in OsMFC is due to the concentration gradient between the NaCl and urine. A higher rejection rate of TOC, P O_4^{3-}, NH_4^+, and TN was observed due to electrostatic repulsion between the negative charge of the membrane surface and existing solutes as well due to the large hydrated size of the solute, signifying its potential in concentrating nutrients from urine. The major challenge that needs to be address in case of OsMFC is the lower power production and, the decrease in water flux with time during water recovery due to dilution of draw solution and concentration of feed solution. This problem can be solved by membrane cleaning and refilling the feed and draw solution periodically. Besides, the lower power production is attributable to inadequate time for microbial growth on the electrode. Therefore, power production can be boosted by increasing the residence time or increasing the conductivity and surface area of anode with usage of nanocomposite materials and ameliorating membrane properties using antifoulant materials.

Economic analysis

The economic analysis deals with how nutrient recovery through MFCs is economically sustainable and feasible. The two factors which constitute the self-sustainability of MFCs are input operational costs and the output in form of product and energy recovery. The macronutrients, i.e., ammonia and phosphate are generally recovered as struvite. The benefit associated with the struvite recovery is that it is a slow-release fertilizer and is safe and can be directly used for land application (Ye et al. 2018). Wastewater treatment plants in Europe expends around €65,000 (≈ US $72,284.55) to eliminate the precipitate of struvite formed in the machinery and pipes and this maintenance cost can be significantly minimized through the recovery of nutrients (Ye et al. 2020a). Studies have reported that the operational costs were found to be reduced by €500,000 (≈ US $583,275)/year when a wastewater treatment plant (WWTP) in Amsterdam West used the method of struvite precipitation for ammonium recovery. Furthermore, compared with traditional processes, a power of around 456 kWh/kg.N

could be saved through this process (Ye et al. 2018). It was also noted that the energy demand for WWTP was reduced and an amount of about €3680 (≈ US$4092.16)/day was saved just by implementing phosphate recovery instead of its removal. Besides, the cost of sludge handling was also reduced to nearly AUD$1.13 (≈ US$0.78) per kg of struvite due to less sludge production (Ye et al. 2020a).

Despite environmental benefits, the economical expense of struvite recovery is high due to the requirement of large quantities of alkaline chemicals to regulate pH along with huge magnesium dosage for struvite formation due to the lack of adequate magnesium sources in wastewater (Ye et al. 2018). The application of BES for nutrient recovery is economical because of electricity generation and elevation in pH induced by cathodic reaction thereby eliminating the requirement for external salts. Figure 12 depicts the energy analysis of different technologies in terms of ammonium recovery. The MFC shows a positive energy balance due to less energy requirement for aeration (Bruning et al. 2012). Conventional ammonium stripping requires a dosage of alkaline chemicals like CaO and NaOH for pH elevation and demands higher input energy without any energy output thus resulting in a negative energy balance (Ye et al. 2018). Moreover, in the case of MEC, out of total energy input, a significant amount is consumed by the external power supply followed by aeration in the cathode and anodic recirculation, therefore net energy yield is negative (Qin and He 2014).

Furthermore, studies have reported that just by increasing phosphate concentration in influent from 50 to 800 mg-P/L could also result in a reduction in total costs for phosphorus recovery from €2800 (≈ US$3113.60) to €520 (≈ US$578.24) per ton of struvite. Correspondingly, a reduction in the cost of ammonia recovery from €10.7 (US$11.90) to

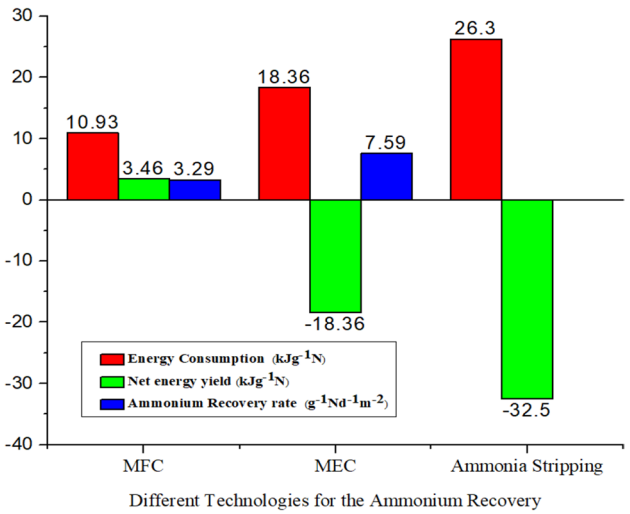


Fig. 12 Analysis of energy consumption and yield of different technologies for ammonium recovery (Ye et al. 2018)

€2.63 (≈ US$2.92)/kg.N is possible by increasing the ammonium concentration from 539 to 2470 mg/L. Therefore, the economic feasibility of a nutrient recovery system can be enhanced through nutrient enrichment (Ye et al. 2020a).

At present, the commercialization of recovered nutrients has not been widely performed; therefore, the market values of recovered products are still unknown. Despite this, some online estimated data do exist. The market value of struvite in Japan and Australia were €220 (US$244.64)/ton and AUS$300–500 (US $207.04–345.07)/ton, respectively. On the other hand, fertilizers based on phosphate had a market value of about €1.9–€3.3 (US $2.11–3.67)/kg.P (Ye et al. 2020a). The existing market price of conventional phosphorus fertilizer, i.e., €1.9–3.3 (≈ US $2.11–3.67)/kg.P is still lower than the recovery-based fertilizers; hence, there is a need to find out more economical methods for recovery to ensure competitive pricing. Moreover, it is also imperative to assess the performance of recovered nutrients. The performance of struvite recovered from swine wastewater was studied and compared with conventional fertilizers by Rahman et al. (2011). They observed that the leaching rate and releasing rate of nutrients from struvite were considerably low during the growth phase of plants in comparison to conventional phosphate fertilizer. The lower release rate of struvite allows a gradual supply of nutrients for a longer period, which in turn enhances the efficiency of fertilizer, and uptake rate and decreases leaching loss. In fact, struvite application will decrease the GHGs emission by reducing the N loss through volatilization and leaching. In another study, Uysal et al. (2014) investigated the effect of struvite on the growth of tomato and maize plants and found that the bioavailability of struvite was more, i.e., plants were able to uptake nutrients from struvite at broader pH conditions and soil types due to slow releasing rate of struvite.

The techno-economic evaluation of BES reactor is also essential to analyze the economic feasibility in commercial upscaling. In terms of reactor architecture, the initial investment cost in the reactor fabrication comprises of cost associated with ion exchange membrane, electrode, reactor maintenance, current collectors, reactor material and electrical connections for large scale application (Jadhav et al. 2021; Selvasembian et al. 2022). Compared to conventional wastewater treatment, the capital cost is 30 times higher in case of MFC even if carbonaceous material based electrode are used (Munoz-cupa et al. 2021). The electrode and membrane are the major contributor of the total cost (Fatehbasharzad et al. 2022; Jadhav et al. 2021). These costs varies depending upon the mode of reactor operation and type of reactor configuration, therefore operational cost and capital cost needs to be balanced to ensure sustainable treatment of wastewater along with electricity generation (Munoz-cupa et al. 2021).

Studies have found that MFC operated in series connection has positive returns due to higher energy recovery

per operation cost, which is two times higher than MFC operated in parallel. Nevertheless, stacking can lead to significant increase in construction and maintenance cost (Selvasembian et al. 2022). Furthermore, operational and design complexities may also arise in stacking and can results in lower power output due to increase in internal resistance with increase in reactor volume. Therefore optimization studies are vital prior commercial or industrial upscaling (Jadhav et al. 2021). Moreover, studies should also focus on constructing low-cost reactors using economical and widely available materials without affecting the performance of reactor in long run and at field scale. For example, for commercial application, modified granular activated carbon which is durable, and bio-compatible can be an economical substitute as bioanode in place of traditional carbon felt and carbon cloth. Besides, biochar has also gained significant attention as a sustainable substitute in terms of environmental aspects, cost and characteristics as compared with traditional electrode (Selvasembian et al. 2022; Kamali et al. 2022).

Furthermore, from an upscaling perspective, membrane such as ultrafiltration membrane, ion exchange membrane and forward osmosis membrane are not cost-effective (Liu and Cheng 2014). The cost of ion exchange membrane can account for 60% of total cost of large-scale reactor. The membrane less reactor, despite being cost effective solution, demonstrate certain limitation such as intermixing of anolyte and catholyte, lower energy generation in long run and possibility of short circuiting. Therefore, selection of cost-effective and stable membrane is essential (Selvasembian et al. 2022). At present, rapid fouling of Nafion membrane is another major barrier in large scale application as it can adversely affect the power generation and coulombic efficiency. The application of polymeric membrane by integrating with charge species or nanoparticle (ZrO_2 and TiO_2) can be an effective solution in preventing the fouling issues (Kamali et al. 2022). Studies have reported that sulfonated polyether ketone (SPEEK) can be an economical substitute in place of Nafion 117. Despite lower power density of MFC with SPEEK than Nafion 117, the cost of SPEEK ($595/W) is nearly two times lower than Nafion 117 ($1134/W). In addition, cost of ceramic membrane ($2.5–3.5/$m^2$) is significantly lower than Nafion 117 ($2300/$m^2$) (Fatehbasharzad et al. 2022). As a whole, to ensure MFC operation to be cost competitive, the focus should be either on using cost effective electrode or membrane materials or power output need to be enhanced (Fatehbasharzad et al. 2022).

In terms of energy consumption, conventional processes such as nitrification and denitrification process in sewage treatment plant consumes 12.5 $kWh/kgN_{removal}$, while it is 4.2 $kWh/kgN_{removal}$ for anammox process. In general, energy consumption of BES is generally compared with the cost of fertilizer production through Haber-Bosh process, which holds 8.5 $kWh/kgN_{recovery}$ whereas MFC shows lower energy consumption of 0.87 $kWh/kgN_{recovery}$ as external energy is only utilized for aeration. On the other hand, energy consumption in MEC operation ranges from 1.17 to 2.7 $kWh/kgN_{recovery}$ and during aeration or recirculation, its consumption increases to nearly 5.7 $kWh/kgN_{recovery}$ (Cerrillo et al. 2023). A study carried out by Zhang and Angelidaki (2015) using submersible microbial desalination cell (SDMC) for ammonia recovery reported net energy recovery of 8.77 $kWh/kgN_{recovery}$ during continuous operation. Therefore, compared with other treatment technologies, energy consumption with respect to nitrogen removal is sustainable in BES.

Conclusion and way forward

In summary, nitrogen and phosphorus are essential macronutrients present in the ecosystem. Their removal/recovery from the diverse waste streams is crucial to prevent the eutrophication of water bodies. BES is a promising technology that employs different mechanisms such as nitrification, denitrification, microbial absorption and chemical precipitation in the nutrient recovery/removal process.

The BES operation is strongly governed by different factors such as temperature, type of membrane, HRT, pH, OLR, light intensity, DO level, external resistance, nutrients concentration, and applied AC current characteristics. Microbial diversity also exhibits a major role in nitrogen removal efficiency and power production. In the BES, mixed microbial culture has diverse organic matter degradation capabilities than pure culture. Furthermore, genetic engineering in the BES can pave the way to enhance the EET rate and metabolic capacity of microbial catalyst.

More intense research is required to enhance the output potential to the theoretical maxima and to achieve higher efficiency along with potential implementation at the field scale which at present is a major challenge in terms of reactor configuration, optimization of parameters, operational expenses, and investment. In fact, future studies should investigate the long-term stability of MFC along with usage of multiple electrode assembly to solve the mass transfer limitations. Correspondingly, there is a need to develop a legitimate multidimensional mathematical model for precise description and fair prediction of performance of BES. Lastly, studies should perform model validation for important parameters by integrating experimental studies with the mathematical models.

This study delineates different reactor configurations and nutrient removal and recovery mechanisms. This will help in developing conceptual understanding and subsequent designing of advanced and efficient BES systems using sustainable materials to achieve higher power density at a lower

operational and maintenance cost. Moreover, the behavior and performance of reactors in different operational conditions have been discussed to provide an overall idea about the electrode kinetics, microbial interactions, and chemical reactions. This can be helpful for scientific community working in the area of environmental engineering, material science, chemical engineering, and chemistry in exploring their implementation for the in-situ treatment of complex pollutants and recovery of nutrients from diverse waste streams. Correspondingly, this review article elucidates recent studies for a cohesive understanding of the performance of hybrid configurations to broaden the vision regarding possible integration with next-generation technologies for shifting towards a green economy.

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Author contribution Ayushman: conceptualization, writing original draft. Pritha Chatterjee: conceptualization, writing, review and editing, Supervision. Shashank: writing.

Data availability The data used in the review paper for the discussion are available within the article and in the supplementary material.

Declarations

Competing interests The authors declare no competing interests.

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