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Bioprocesses for the recovery of bioenergy and value-added products from wastewater: A review

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ABSTRACT

Wastewater and activated sludge present a major challenge worldwide. Wastewater generated from large and small-scale industries, laundries, human residential areas and other sources is emerging as a main problem in sanitation and maintenance of smart/green cities. During the last decade, different technologies and processes have been developed to recycle and purify the wastewater. Currently, identification and fundamental consideration of development of more advanced microbial-based technologies that enable wastewater treatment and simultaneous resource recovery to produce bioenergy, biofuels and other value-added compounds (organic acids, fatty acids, bioplastics, bio-pesticides, bio-surfactants and bio-flocculants etc.) became an emerging topic. In the last several decades, significant development of bioprocesses and techniques for the extraction and recovery of mentioned valuable molecules and compounds from wastewater, waste biomass or sludge has been made. This review presents different microbial-based process routes related to resource recovery and wastewater application for the production of value-added products and bioenergy. Current process limitations and insights for future research to promote more efficient and sustainable routes for this under-utilized and continually growing waste stream are also discussed.

1. Introduction

The concept of wastewater management broadly encompasses the efficient treatment and appropriate reutilization of different types of wastewaters in a sustainable and ecologically friendly manner (Meena et al., 2019). It is one of the cornerstones of a circular economy and a major focus of research in the present scenario of mitigating water scarcity (Hossain et al., 2020; Nagarajan et al., 2020). Recycled water can be exploited in gardening, agriculture and land conditioning. In addition, wastewater is often a rich source of numerous commercially important organic compounds which are present in free or combined form, albeit in small quantities (Rajasulochana and Preethy, 2016). Valuable resource present in wastewater is becoming increasingly prevalent and this can be recovered using microbes. During biological wastewater treatment two important aspects are combined. Firstly, the pollutants are removed, and, secondly, the economically important compounds are produced (Fig. 1). An integrated system involving wastewater treatment and CO_2 sequestration for the retrieval of value-added compounds and biofuel production is the new aspiration of wastewater management (Bhardwaj et al., 2021; Verma et al., 2020). Wastewater generated from the different process can be treated in common treatment plants, where the sludge obtained from the treatment process can be utilized as feedstock for the anaerobic digestion (AD) plant (da Silva Vilar et al., 2021). The effluent from AD can be further used for the cultivation of microalgae. Further, the microalgal biomass can be sequentially utilized as a feedstock for the production of bioplastics (biopolymers), biofuels and broad a range of different bioactive compounds (Cinar et al., 2020; Mehariya et al., 2021). Recently, photocatalysis appeared as one of the emerging technologies

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Abbreviations

AD –	Anaerobic digestion
COD –	Chemical oxygen demand
CW –	Coffee wastewater
DCMFC –	Dual chamber microbial fuel cell
DSW –	Distillery spent wash
DW –	Domestic wastewater
DWT –	Domestic wastewater treatment
FAME –	Fatty acid methyl ester
FFAs –	Free fatty acids
HY –	Hydrogen yield
HPR –	Hydrogen production rate
HRT –	Hydraulic retention time
MFCs –	Microbial fuel cells
MWW –	Municipal wastewater
OLR –	Organic loading rate
OMW –	Oil mill wastewater
Pd_{max} –	Power density
PEM –	Proton exchange membrane
PHA –	Polyhydroxyalkanoate
POME –	Palm oil mill effluent
PS –	Primary sludge
SCMFC –	Single chamber microbial fuel cell
SWW –	Sewage wastewater
SwWW –	Swine wastewater
VS –	Volatile solids
WAS –	Waste activated sludge
WCO –	Waste cooking oil
VFAs –	Volatile fatty acids
WW –	Wastewater
WWTP –	Wastewater treatment plants

to treat the wastewater and remove the various hazardous contaminants from the effluents (Bharath et al., 2021; Rana et al., 2021; Sharma et al., 2021a; Soni et al., 2021; Tran et al., 2021; Wang et al., 2021). Thus, the wastewater treatment and resource recovery using microorganisms can help to control waste stream and close material loops. Despite of these obvious advantages, the processes and technology used for this purpose should be very carefully selected with respect to the specialized achievability, straightforwardness, financial considerations, cultural requirements and political needs. Scrupulous planning and research should be undertaken to adapt the process specifically to the origin and components of the wastewater, the bioavailability of the organic compounds, and the oxygen and nutrient requirements of the preferred microbial species for the production of economically important products. Huge amount of different wastewaters leads to the development of different processes combining wastewater management and production of industrially-relevant products that are corresponding to circular bioeconomy rules. The wastewater management through microbial processes and efficient production of bioenergy, biosurfactants, organic acids or bioplastics remain as an emerging research topic. Recent trends in the research work related to wastewater treatment and bioprocess development for the recovery of value-added products have been intensively analyzed (Fig. 2). Table 1 summarizes the key topics presented in the related literature reviews that were published in the past two years. However, many of the presented reviews focus on one topic (for example, only on the microalgae application in wastewater management; strategies to obtain biohydrogen or biomethane; strategies to produce bioenergy, etc.). Meanwhile, the present review provides: a) a comprehensive and detailed summary of bioprocesses leading to simultaneous different wastewater bioremediation; b) production of bioenergy as well as industrially relevant, value-added compounds such as organic acids, some pharmaceuticals, biosurfactants, biopesticides and bioplastics. Better understanding and improvement of bioprocesses leading to different wastewater bioremediation and synthesis of bioenergy and valuable compounds could lead to the development of advanced bio-circular economy processes working at a large scale and leading towards sustainable development of the world.

2. Process developments in bioenergy production from wastewater

2.1. Bioelectricity

Bioelectricity is a renewable and sustainable form of electricity produced from biomass waste or wastewater coupled with biological activity of microorganism. One of the promising technologies to produce bioelectricity from wastewater is Microbial Fuel Cells (MFCs). The

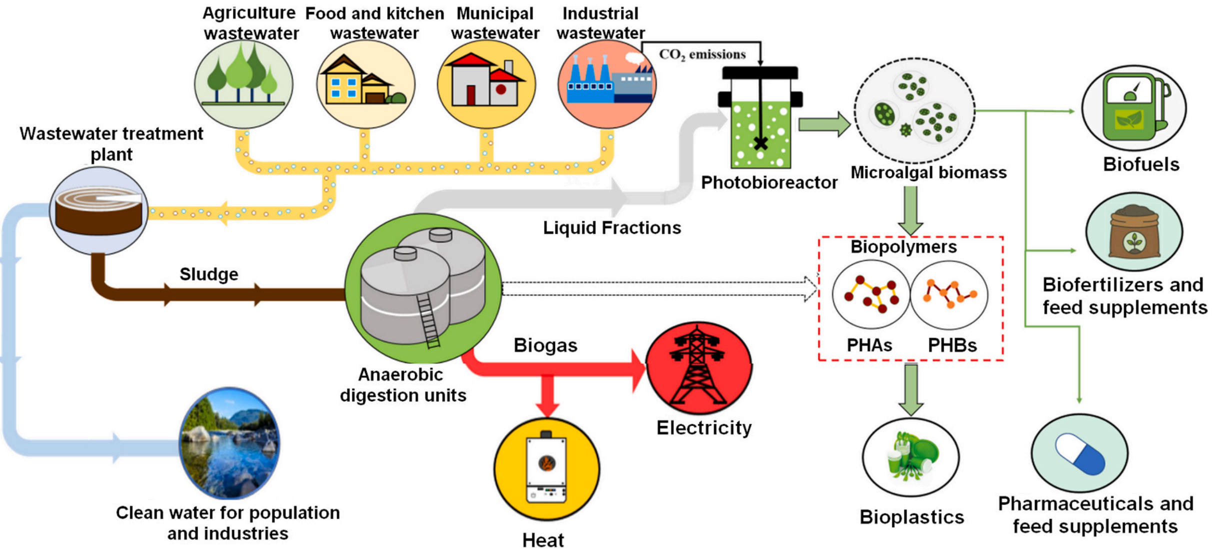

Fig. 1. Integrated bio-refinery strategies involved in multistage recovery of value-added products from wastewater treatment process.

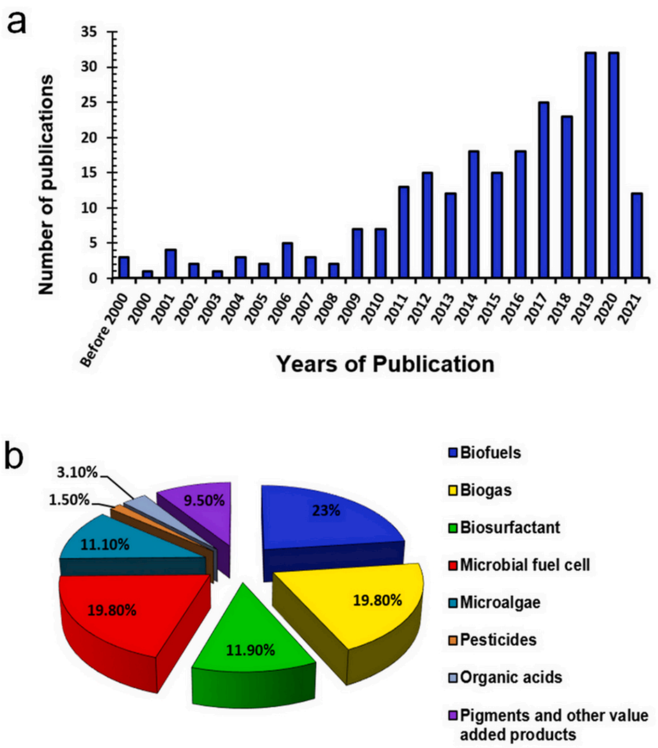

Fig. 2. The contributions at various periods starting from 2000 to 2021 related to wastewater management (a) and publications describing the various value added products recovery combined with wastewater treatment (b).

Table 1
Summary of key topics of the published literature surveys in the past two years related to the present review.

Key topic of review	References
Bioelectricity and biofuels production from wastewater	
Summary of technologies for the conversion of waste organic matters into sustainable bioenergy; characterization of waste organic matters from different sources.	Srivastava et al. (2020)
Wastewater sources, microalgae cultivation methods and microalgae application for resource recovery from wastewater and added-value compounds production.	Bhatia et al. (2021)
Design variability of MFCs; factors affecting the effectiveness of MFCs; application of MFCs; challenges of MFCs.	Gul et al. (2021)
Wastewater treatment strategies, microalgae cultivation and application for wastewater treatment; resource recovery from wastewater and bioenergy.	Mehariya et al. (2021)
MFC design/model, types of wastewater and their use in MFC; the role of microorganisms; electricity generation.	Munoz-Cupa et al. (2021)
MFC reactions, possible substrates for microorganisms, factors affecting the performances of MFCs.	Obileke et al. (2021)
Application of microbial electrochemical technologies for the treatment of petrochemical wastewater; role of different operating parameters on the performance of microbial electrochemical technologies.	Priyadarshini et al. (2021)
Cyanobacteria in the conversion of wastewater to biofuels.	Sadvakasova et al. (2021)
Biohydrogen production from wastewater	
Summary of the modern developments and enhancement strategies for improving the biorefinery route of industrial wastewater to biohydrogen.	Banu et al. (2020)
Biohydrogen production from waste, challenges, production and improvement strategies.	Chandrasekhar et al. (2020)
Biohydrogen production strategies; factors affecting biohydrogen production, microorganisms involved in biohydrogen production, challenges of biohydrogen production.	Mona et al. (2020)
Hydrogen production strategies and technological level; advantages and disadvantages of different hydrogen/biohydrogen production strategies.	Aydin et al. (2021)
Hydrogen production by dark-fermentation: microorganisms and they metabolism and biocatalysts involved in biohydrogen production; strategies to improve production of biohydrogen.	Dahiya et al. (2021)
Bio-based management of different wastewater and value-added compounds production	
Production of PHA by cyanobacteria grown in wastewater.	Arias and Uggetti (2020)
Microalgae cultivation strategies, wastewater treatment and bioremediation by using microalgae.	Nagarajan et al. (2020)
Microalgae for bio-products production in circular economy context.	Catone et al. (2021)
PHA production from waste.	Khatami et al. (2021)
Mechanisms of microalgae on removing micropollutants from wastewater.	Liu et al. (2021a)
Pollution prevention and waste management by using algal-based wastewater treatment technologies.	Leong et al. (2021a)
Different waste and wastewater biorefinery towards a sustainable circular bioeconomy.	Leong et al. (2021b)
Swine wastewater management using different microalgae species.	L’opez-Pacheco et al. (2021)
Distillery wastewater management and added-value compounds production.	Ratna et al. (2021)
Microalgae cultivation in wastewaters and resource recovery.	Ummalyma et al. (2021)

process involves simultaneous metabolizing the organic material and converting the chemical energy stored in this substrate into electricity, using microbial metabolic activities as biocatalysts (Aiyer, 2020; Do et al., 2018; Wang et al., 2020). In general, MFC is made up of three important modules: the anode, the cathode and a proton exchange membrane (PEM) separating them (Fig. 3). The anaerobic microorganisms present in the anodic compartment efficiently degrade the organic materials into electrons and protons with CO_2 as an end product. The electrons are moved from the anode towards the cathode via an electric circuit. Thus, the current and positively charged protons generated can move to maintain electric neutrality. Besides the electricity generation, MFC technology could be actively involved in the conversion of organic materials from wastewater resulting in wastewater treatment not requiring separation and purification (Gul et al., 2021; Mathuriya and Sharma, 2009; Mathuriya, 2014; Munoz-Cupa et al., 2021; Obileke et al., 2021). It can be used to treat diverse types of wastewaters with no pretreatment, generation of less sludge, insensitivity to external stimuli in addition to being energy saving (He et al., 2017; Kondaveeti et al., 2019). Many researchers (Choudhary et al., 2020; Feng et al., 2014; Herrero-Hernandez et al., 2013; Jayashree et al., 2016; Lai et al., 2018; Liang et al., 2018; Mansoorian et al., 2016; Xiao et al., 2017; Yazdi et al., 2015) have explored the treatment and simultaneous power generation from treating wastewaters using MFC (Table S1).

The technical innovations to design effective MFC are reported in several studies. Mahdi Mardanpour et al. (2012) utilized a novel annular single-chamber MFC (ASMFC) with spiral stainless steel mesh anode which produced significantly higher power density (20.2 Wm^{-3}) from dairy wastewater than it was reported in the earlier studies. Besides, the power generation, this novel ASMFC achieved higher COD removal (91%) from the dairy industry wastewater. Jiang et al. (2013) developed a membrane-less MFC coupled photobioreactor for domestic wastewater treatment (DWT) and production of electricity along with microalgal biomass production. The developed system could generate power density (Pd_{max}) of 481 mWm^{-3} (maximum) and was involved in the efficient removal of chemical oxygen demand (COD) (77.9%), phosphorus (23.5%) and nitrogen (97.6%) from DW. Further, the electricity generation from DW was improved by using wastewater from olive oil mill wastewater (OMW) along with DW. The mixture of DW with OMW (14:1 ratio w/w) improved Pd_{max} of 124.6 mWm^{-2} and significantly reduced the total oxygen demand by up to 65%. Ahn et al. (2014) evaluated the efficiency of Multi-anode MFCs – separator electrode assembly (SEA) and closely spaced electrodes (SPA) for generation of electricity and DW treatment. SEA-MFC was able to generate Pd_{max} of 328 ± 11 mWm^{-2}

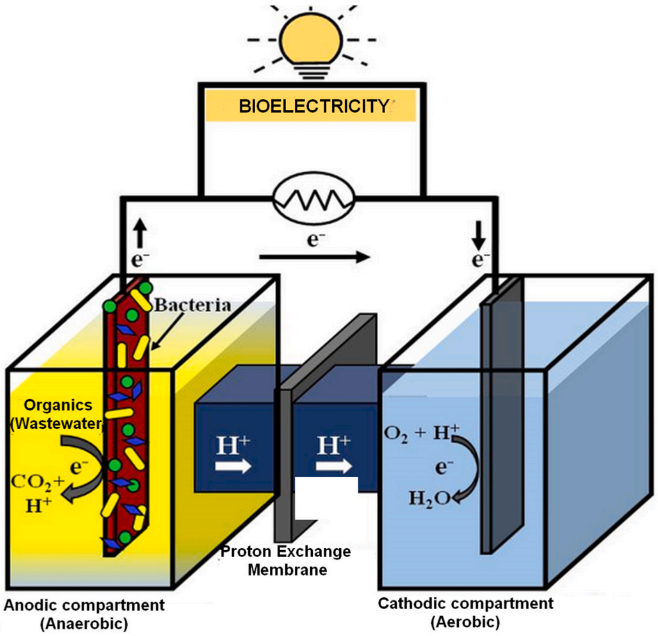

Fig. 3. Microbial fuel cells (MFCs). The MFC is made up of three important modules: the anodic, the cathodic compartments are separated by a proton exchange membrane (PEM).

which was 16% higher than SPA-MFC (282 ± 29 mWm^{-2}). Compared to SPA-MFC, SEA-MFC mediated DW treatment aided in 94% of COD removal and considerable decrease in the substrate diffusion. Later Nayak and Ghosh (2018) developed an H-type MFC to treat the mixture of distillery spent wash (DSW) diluted with sewage wastewater (SWW) in an anaerobic environment for power generation. At 50:50 ratio of DSW and SWW, the maximum power density of 836.81 mWm^{-2} was attained along with 39.66% removal of total dissolved solids and 97% of total suspended solids. Firdous et al. (2018) investigated the electricity generation of vegetable oil industry wastewater using two-chambered MFC. The efficiency of power generation was enhanced with increase in time and temperature. The maximum voltage of 5839 mV was obtained at 35 °C with COD removal of 90% (Firdous et al., 2018). Gao et al. (2020) presented a novel trickling MFCs for electricity generation using a brewery waste stream as carbon source. An average power density of 0.27 W/m^{-2} was generated with ~60% COD removal at hydraulic retention time (HRT) of 1.6 h. Mohamed et al. (2020) presented MFC based on two photosynthetic microorganisms *Synechococcus* sp. And *Chlorococcum* sp. As biocatholyte for bioelectricity generation, CO_2 sequestration and biomass production using kitchen wastewater. *Synechococcus* sp.-based MFC was also tested by Ratna e al. (2021) and power density of 41.5 ± 1.2 mW/m^2 was observed. Wu et al. (2020) presented a novel combined dual MFC system for the continuous removal of Victoria Blue R (VBR) and electricity generation from textile wastewater using anaerobic and aerobic VBR-degrading bacteria, *Shewanella putrefaciens* and *Acinetobacter calcoaceticus*. The VBR removal efficiency was 98.7%. Moreover, the decreased toxicity of the effluent was also detected. Subsequently, the use of waste substrate present in wastewater for electricity generation and simultaneously microbial growth will help to eradicate the harmful toxic compounds turning them into non-toxic counterparts. Moreover, the management of wood-based panel industry wastewater through MFCs was suggested by Toczyłowska-Maminska (2020). Overall, these studies indicated that MFCs technology is favorable strategy to simultaneously reduce BOD and COD values in wastewater and recover bioelectricity. However, more emphasis should be given to improve the efficiency of MFC electrodes and to provide controlled physio-chemical conditions for the growth of microbes during large scale operations.

2.2. Biohydrogen

Among the different gaseous biofuels, biohydrogen possesses higher energy density (141.9 MJ kg^{-1}) and it yields only water as a combustible by-product. Hence, it is considered the “clean biofuel”, which is also easy to be stored and transported (Shanmugam et al., 2020). Currently, most of the hydrogen production depends on non-renewable fossil-fuel-based resources, which simultaneously release harmful greenhouse gases (GHGs) (Lee et al., 2019a). Hence, considerable attention is directed towards the production of hydrogen from renewable feedstocks, which has several advantages such as, reduction/elimination of GHG production, and utilization of cheap renewable materials which eventually improve the economy of production (Dahiya et al., 2021; Hosseini and Wahid, 2016). Biologically mediated hydrogen production is broadly classified into light-mediated and light-independent reactions. Biological hydrogen production involves biophotolysis, photofermentation, dark fermentation and microbial electrolysis (Phan et al., 2020; Rupprecht et al., 2006). An overview of the metabolic reactions involved in the biohydrogen production along with the reported Gibbs free energy is presented in Table 2. Among all, dark fermentation was found to be a cost-effective and high-yield method due to its potential for the utilization of diverse feedstocks, minimal energy requirements and industrial applicability (Chong et al., 2009). In dark fermentation, anaerobic and facultative anaerobic bacteria are predominantly involved in the utilization of complex hydrocarbons from different feedstocks for pyruvate biosynthesis and subsequent hydrogen production via the acidogenic pathway of glycolysis. Further, it can produce up to four mols of hydrogen from one mol of glucose in a quick manner (Sivagurunatha et al., 2017). However, the carbohydrate source or the substrate is the major factor which determines the microbial production and yield of biohydrogen from these pathways. Hence, the selection of an appropriate substrate with high carbohydrate content is the vital parameter in biohydrogen production (Hernández-Mendoza et al., 2014; Kapoor et al., 2020). In recent decades, the conversion of organic matter-rich wastewater into value-added products gained much attention due to its great economic benefit: low cost, suitable carbohydrate content for microorganism’s growth and biodegradability with simultaneous reduction and stabilization of wastes (Kamyab et al., 2019; Preethi et al., 2019). Some of the sources of wastewater exploited as the substrates for the production of biohydrogen includes dairy (da Silva et al., 2019), cassava (Amorim et al., 2014), brewery (Pachiega et al., 2019), palm oil mill effluent (POME) (Taifor et al., 2017), pulp and paper mill (Vaez et al., 2017), pharmaceuticals (Sivaramakrishna et al., 2009) and textile (Li et al., 2012) industry, which are summarized in Table 3 and the major findings are described below.

Sivagurunathan and Lin (2016) demonstrated the feasibility to use brewery wastewater as a substrate and enriched microbiota in continuous stirred tank reactor, resulting in peak hydrogen production rate (HPR) of 37.5 L H_2 L^{-1} with the maximum yield (HY) of 1.62 mol H_2 mol^{-1} hexose at 6 h HRT. They also determined that the substrate concentration at the organic loading rate (OLR) 16.6 g L^{-1} in brewery wastewater has a huge impact on hydrogen production (Sivagurunathan and Lin, 2020). Murugan et al. (2020) utilized rice mill wastewater, food wastewater and sugar wastewater for both the production of biohydrogen and COD removal, among which, food wastewater has shown highest cumulative HY of 1.8 mol H_2 mol^{-1} glucose with concomitant 79.9% COD removal. Vaez et al. (2017) investigated the efficacy of paper mill effluent as an organic feedstock for biohydrogen production through dark fermentation and the highest HY of 55.4 mL g^{-1} COD was obtained along with 569 mL g^{-1} COD of methane from the substrate concentration of 5 g COD L^{-1}. The exploitation of activated carbon and cation exchange resin-pretreated textile industry wastewater as the feedstock for dark fermentation was also evaluated by Li et al. (2012). The pretreated-feedstock devoid of bio-toxic inhibitors, with total sugar concentration of 20 g L^{-1} produced a higher HY of 1.37 mol H_2 mol^{-1} reducing sugars. Further, Lin et al. (2017) utilized coagulation

Table 2
Overview of the metabolic reactions involved in the production of bioenergy and other bio-compounds. NA: Not reported; scl – short acyl chain length compounds; mcl – medium acyl chain length compounds.

Bioenergy and value-added compounds	Process	Metabolic reactions	Gibbs free energy (ΔG^o) (kJ mol^{-1})	Reference
Biohydrogen	*Biophotolysis*			
	Direct biophotolysis	H_2O + Light energy$\rightarrow H_2 + \frac{1}{2} O_2$	+374.5	Benemann et al. (1973)
	Indirect biophotolysis	$6H_2O + 6CO_2$ + Light energy$\rightarrow C_6H_{12}O_6 + 6O_2$ $6H_2O + C_6H_{12}O_6$+ Light energy$\rightarrow 12H_2 + 6CO_2$	NA	Benemann (1996)
	Photofermentation	$CH_3COOH + 2H_2O$ + Light energy$\rightarrow 4H_2 + 2CO_2$	NA	Sarangi and Nanda (2020)
	Dark fermentation	$4H_2O + C_6H_{12}O_6 \rightarrow 2CH_3COO^- + 2HCO_3^- + 4H^+ + 4H_2$ (Acetic acid pathway)	−206.3	Thauer et al. (1977)
		$2H_2O + C_6H_{12}O_6 \rightarrow 2CH_3CH_2CH_2COO^- + 2H_3CO_3^- + 3H^+ + 2H_2$ (Butyric acid pathway)	−254.8	Gottschalk (1986)
		$2HCOOH + 2NAD^+ \rightarrow 2CO_2 + 2NADH + 2H^+$ (Enterobacterial pathway)	−209.1	Stickland (1929)
	Microbial electric cells	$CH_3COO^- + 4H_2O \rightarrow 2HCO_3^- + H^+ + 4H_2$	+104.6	Kadier et al. (2019)
Biomethane	*H_2 - interspecies electron transfer*	$CH_3CH_2CH_2COO^- + 2H_2O \rightarrow 2CH_3COO^- + H^+ + 2H_2$	+48.32	Zhang et al. (2018)
		$4H_2 + H^+ + HCO_3^- \rightarrow CH_4 + 3H_2O$	−135.58	
	Formate - interspecies electron transfer	$CH_3CH_2CH_2COO^- + 2HCO_3 - \rightarrow 2CH_3COO^- + H^+ + 2HCOO^-$	+45.60	
		$4HCOO^- + H_2O + H^+ \rightarrow CH_4 + 3HCO_3^-$	−111.00	
	Aceticlastic methanogenesis	$CH_3COO^- + H_2O \rightarrow HCO_3^- + CH_4$	31.02	
	Methanogenesis (total reaction)	$2CH_3CH_2CH_2COO^- + H_2O + HCO_3^- \rightarrow 4CH_3COO^- + H^+ + CH_4$	−38.94	
Biodiesel	*Transesetrification*	$RCOOR^1 + R^2OH \longleftrightarrow R^1OH + RCOOR^2$	+92.71–171.16	Ferrão-Gonzales et al. (2011); Nautiyal et al. (2014)
Bioethanol	*Fermentation*	$C_6H_{12}O_6 \rightarrow 2CH_3CH_2OH + CO_2$	+94–98 depending on reaction temperature	Arshad et al. (2014)
Biobutanol	*Acetone-butanol-ethanol (ABE) fermentation performed by C. acetobutylicum.* *Solventogenesis*	$C_6H_{12}O_6 \rightarrow \rightarrow \rightarrow \rightarrow$Acetyl-CoA→Acetoacetyl-CoA →3-hydroxybutyryl-CoA→Crotonyl-CoA→Butyryl-CoA→butylaldehide→ butanol	NA	Cheng et al. (2019)
PHA production	*Pathway I (predominantly used in PHB-producing organisms)*	$C_6H_{12}O_6 \rightarrow$2 Acetyl-CoA → Acetyl-CoA →(R)-3-Hydroxybutyryl-CoA → scl-PHA (PHB)	NA	McAdamet al. (2020)
	Pathway II (present in mcl-PHA producing pseudomonads)	Fatty acids →Acyl-CoA→ fatty acid β-oxidation reaction ((S)-3-hydroxyacyl-CoA + (R)-3-hydroxyacyl-CoA) → mcl-PHA	NA	Kniewel et al. (2019)
	Pathway III (present in mcl-PHA producing pseudomonads)	Acetyl-CoA→ malonyl-CoA → 3-ketoacyl-ACP → fatty acid biosynthesis reactions ((R)-3-hydroxyacyl-ACP → (R)-3-hydroxyfatty acid → (R)-3-hydroxyacyl-CoA → mcl-PHA	NA	

pretreated-textile desizing water as substrate yielding HY of 1.52 mol mol^{-1} hexose and the HPR of 3.9 L L^{-d}. Coagulation pretreatment aided in both removals of toxic compounds and increased hydrogen production ability up to 120%. It has been reported that OLR influenced biohydrogen production. The reduced OLR by using dairy industry effluents resulted in increase biohydrogen (da Silva et al., 2019). Also, the use of ferrous ions as enhancers (100 mg L^{-1}) improved the biohydrogen production with an HPR of 5.729 mL $L^{-1}h^{-1}$ in 24 HRT (Paul et al., 2014). The effect of HRT and solid retention time (SRT) on biohydrogen production using POME as substrate was examined using anaerobic sequencing batch reactor (ASBR) and *Thermoanaerobacterium* spp. (Maaroff et al., 2018). It was reported that *Thermoanaerobacterium* spp. Produced hydrogen with HPR of 8.54 mmol H_2 $L^{-1}h^{-1}$ as a dominant species involved in biohydrogen production. The solubilized CO_2 present in the wastewater can also be reduced to carbonates using recently reported strains of *Corynebacterium flavescens* (Sharma and Kumar 2020, 2021). It can be summarized that biohydrogen production can be strongly associated with management of different wastewaters, but the process control, optimization and improvement of technical issues (reliability, durability, etc.) remain as a main challenge for the research and industrial sectors. Biohydrogen production combined with wastewater management on the industrial scale is still in a stage of development.

2.3. Biomethane

Europe's population is projected to grow by 1.7% between year 2016 and 2080, indicating an increase of 8.5 million people and thus a rise in wastewater generation (Colzi Lopes et al., 2018; Commission European, 2017). Wastewater management is an essential part of environmental protection and good public health preservation for a country (Chua et al., 2013; Kamyab et al., 2017a, b; Roudia et al., 2020). The growth of human population promotes the consumption of fossil fuels and global warming. The latter requires great attention and development of renewable and environment friendly alternative fuels and energy sources (Phowan and Danvirutai, 2014). Regarding environmental protection, production of biomethane and biohydrogen are among the most favorable solutions over the fossil fuels or the existing physical-chemical methods of methane synthesis (Arizzi et al., 2016). The combination of wastewater management with production of gaseous fuels, electricity and especially hydrogen (H_2) and methane (CH_4) is an advanced solution to reduce both demand of fossil fuels and wastewater amount. The biomethane production and wastewater management include several debatable aspects: a) the generation of biogas from microalgae cultivated in wastewater; and b) the conversion of sewage sludge from wastewater treatment plants (WWTP) into biomethane. The use of different wastewater sources for microalgae grown to further produce biomethane has been already discussed in detail (Bohutskyi et al., 2018; Chen et al., 2015; Craggs et al., 2012; Hidaka et al., 2014; Kinnunen and Rintala, 2016; Morales-Amaral et al., 2015; Passos et al., 2013; Polishchuk et al., 2015; Prajapati et al., 2013; Quiroz Arita et al., 2015). Due to the ability of microalgae to assimilate nutrients and eliminate it from the wastewater, microalgae cultivation technologies for wastewater treatment holds promise as an alternative to the traditional wastewater management (Bohutskyi et al., 2015, 2016, 2019, 2016; Catone et al., 2021; Kamyab et al., 2017a; Park et al., 2011; Passos et al., 2013). The use of wastewaters for biomass production can help minimize freshwater demand; carbon, nutrients cost; improve phosphorus and nitrogen

Table 3
Biohydrogen production from different industrial wastewaters. WW - Wastewater; ASBR - Anaerobic sequencing batch reactor; CSTR - Continuous stirred tank reactor; UASBR - Upflow anaerobic sludge blanket reactor; STRD - Stirred tank reactor digester; COD - chemical oxygen demand; POME - Palm oil mill effluent; HRT - hydraulic retention time; VS – volatile solids;^{a} g hexose equivalent L^{-1};^{b} g total sugar L^{-1};^{b} g total sugar L^{-1};^{c} mL L^{-1} h^{-1} at 24 h HRT;^{d} mL H_2 g^{-1} VS (sucrose);^{e} mmol H_2 g^{-1} COD;^{f} mmol g^{-1} COD initial;^{g} mL g^{-1} COD.

Type of industrial wastewater	Reactor	Inoculum	Operating conditions			Hydrogen Production (mol H_2 mol^{-1} carbohydrate)	Reference(s)
			Substrate concentration (g COD L^{-1})	pH	Temperature		
Dairy industry WW	Batch	Bacteria B1 and B4	NA*	5.0	70 °C	5.729^{c}	Paul et al. (2014)
	AFBR	Biomass from fermentation	15.44	4.3	30 °C	2.56	da Silva et al. (2019)
Cassava WW	Batch	*Clostridium acetobutylicum*	5.0	7.5	36 °C	2.41	Cappelletti et al. (2011)
	AFBR	Sludge from Swine wastewater treatment	5.0	4.0	28 °C	1.91	Amorim et al. (2014)
Food WW	Batch	*Acinetobacter junii*-AH4	1.3	7.5	37 °C	1.8	Murugan et al. (2018)
Sugar WW			4.8	7.5	37 °C	1.1	
Food WW	Batch	*Klebsiella pneumoniae*-FA2	NA*	5.5	37 °C	2.85	Ramu et al. (2020)
Potato WW	CSTRs	Sludge from WW treatment plant	10.0	5.5	35 °C	320 ^{d}	Salem et al. (2018)
Beverage WW	CSTR	Mixed culture	20.0	6.3	37 °C	1.62	Sivagurunathan and Lin (2016)
	Batch	Anaerobic granulated mixed consortium	2.0	5.5	37 °C	1.5	Pachiega et al. (2019)
	Batch	Mixed culture	20.0	5.5	37 °C	3.76	Sivagurunathan and Lin (2020)
POME	Batch	Anaerobic mixed microflora	NA*	5.5	35 °C	0.41^{e}	Mohammadi et al. (2011)
	Two stage fermentation reactors	Anaerobic sludge	76.5	5.5	55 °C	49.22^{e}	Krishnan et al. (2016)
	Batch	Engineered *E. coli* BW25113	5.4	6.5	37 °C	0.66	Taifor et al. (2017)
	Two stage fermentation reactors	Mixed culture	20.0^{a}	6.5	55 °C	2.99	Maaroff et al. (2018)
Paper mill WW	UASBR	Mixed culture	2.3	5.0	35 °C	5.29^{f}	Farghaly et al. (2015)
Pulp and paper mill WW	Batch	Anaerobic sludge	5	5.0	37 °C	55.4 ^{g}	Vaez et al. (2017)
Probiotic WW	Batch	Sludge from slaughter house	5.0 ^{b}	5.5	37 °C	1.8	Sivaramakrishna et al. (2009)
Rice mill WW	Batch	*Enterobacter aerogenes*	10.2 ^{b}	7.0	33 °C	1.74	Ramprakash and Muthukumar (2014)
	Glass reactor	*Enterobacter aerogenes* RM08	NA*	7.0	33 °C	1.92	Ramprakash and Muthukumar (2016)
	Batch	*Acinetobacter junii*-AH4	2.6	7.5	37 °C	1.4	Murugan et al. (2018)
Textile WW	Batch	*Clostridium butyricum, Klebsiella oxytoca*	20.0	7.0	37 °C	1.37	Li et al. (2012)
Textile desizing WW	STRD	Granular sludge	15 ^{b}	6.8	35 °C	1.52	Lin et al. (2017)

removal and together produce value-added compounds. Such technology is one of the promising applications of circular bioeconomy (Ummalyma et al., 2021). Moreover, it is also suggested to use wastewater alternatively as a conventional fertilizer to cultivate algae (Chen et al., 2015). Despite the ability to apply wastewater for growth of microalgae, the maintenance of a population of a single species is still a challenge. However, mixed species of algae can also be used for wastewater utilization (Chen et al., 2015). Further, the obtained microalgal biomass can be supplied for anaerobic digestion to produce biomethane (Arashiro et al., 2020; Bohutskyi et al., 2018; Perazzoli et al., 2016; Shchegolkova et al., 2018; Thorin et al., 2017). Arashiro et al. (2020) combined wastewater treatment with microalgae cultivation for pigments (phycobiliproteins) production and biomethane generation. After microalgae cultivation, up to 52% of COD, 86% of NH_4^{+}-N, and 100% of phosphorus reduction were detected. The final biomethane yields ranged from 159 to 199 mL CH_4 g^{-1} VS. The latter example clearly indicated beneficial relations between wastewater management and biomethane production corresponding to the main principle of a circular bioeconomy.

The second major discussed aspect is the conversion of sewage sludge from WWTP into biomethane (Caballero et al., 2020; Chua et al., 2013; Ebrahimi-Nik et al., 2018; Kaluža et al., 2014; Paolini et al., 2018). Sewage sludge is obtained during the water cleaning process. Anaerobic digestion (AD) is a promising way for sewage water treatment that enables the generation of renewable energy from the same process (Caballero et al., 2020). During the AD, the bacteria break down organic substances with concomitant production of biogas containing CH_4 and CO_2, which in turn can be applied to produce biofuel, heat and electricity (Table 2). Anaerobic sludge treatment offers an alternative solution with a various advantage such as reduction of organic content to ~50%, and further process to produce biogas; 2) generation of renewable energy; 3) the possibility of independent supply of energy to the sewage treatment plant; 4) lowered production costs; 5) climate protection by improving the balance of CO_2 emitted and waste treatment facilities (Makisha and Semenova, 2018). The combination of different wastewater management and biomethane production possibilities and cases were discussed in detail previously (Berktay and Nas, 2007; Chaiprasert et al., 2017; Chou and Su, 2019; Colzi Lopes et al., 2018;

Kiselev et al., 2019; Michailos et al., 2020; Papadias et al., 2012; Yun et al., 2016). Recent reports suggested CH_4 production from four times diluted piggery wastewater by using purple phototrophic bacteria and algal-bacterial consortia. CH_4 concentrations of 90.8% were obtained (Marín et al., 2019). The use of purple phototrophic bacteria and consortium of algae and bacteria supported the CH_4 concentration up to 93.3 and 73.6%, respectively. Digestion of the shrimp processing wastewater yielded high levels of methane in the biogas at over 70% (v/v) (Zappi et al., 2019). Petrochemical wastewater (Tan et al., 2020), recycled pulp and paper wastewater (Bakraoui et al., 2019), shrimp processing (Zappi et al., 2019), and saline fish wastewaters (Letelier-Gordo et al., 2020) have also gained attention to be applied in biomethane production. It was presented that countries such Australia, Denmark, France, Germany, USA, etc. Successfully combined wastewater treatment with biogas production (Nguyen et al., 2021). Polish researchers presented good perspectives of circular bioeconomy by combining biogas production and wastewater treatment (Kaszycki et al., 2021). Nevertheless, the development of a most suitable and effective system for biogas production with much higher CH_4 content and wastewater detoxification remains the subject of investigation.

2.4. Biofuels

In 2019, world liquid fuel consumption reached ~100 million barrels per day and is predicted to grow (Hacquard et al., 2019). Increasing the consumption of liquid fuels reduces fossil fuel reserves and acts as a driving force to look for alternatives. One of such alternatives is biofuel which usually refers to nowadays industrially important liquid fuels such as bioalcohols and biodiesel that can be used as a replacement for conventional transportation fuels (Ganesan et al., 2020). Different types of wastewater treatment coupled with simultaneous biofuel and production of other bio-products using microorganisms such as microalgae, cyanobacteria, bacteria, yeast and fungi and achievement of their successful exploitation in large-scale plants have been extensively studied (de Souza Candeo et al., 2020; Unc et al., 2017). The most explored microorganisms for the simultaneous wastewater and/or sewage sludge treatment and biofuel production are microalgae. Due to their good ability to grow in different types of wastewaters utilizing nutrients and accumulating lipids, *Chlorella* sp. Such as *C. vulgaris*, *C. minutissima, C. sorokiniana,* and *Scenedesmus* sp. Are widely explored, and commonly used as well (Abu Jayyab and Al-Zuhair, 2020; Ryu et al., 2014). Also, by growing microalgal biomass in different wastewaters or properly pretreated (by anaerobic fermentation or pyrolysis) sewage sludge as an alternative growth media is a promising and ecologically conscious solution to overcome wastewater treatment issues and obtain green energy for biofuel production from specific biomolecules (mostly carbohydrates and triacylglycerols) (El-Dalatony et al., 2019; Kadir et al., 2018). Cells of microalgae growing and assimilating different organic and inorganic wastes present in wastewater accumulate carbohydrates, lipids (as an energy reserve mostly in the stationary phase of the growth) and other molecules such as proteins which can be used for the production of biofuels as follows: carbohydrates for the production of bio-alcohols such as bioethanol, biobuthanol, biogas, biohydrogen, etc. And oil droplets or lipids, most notably $C_{14\text{-}18}$ triacylglycerols (TAGs), - for biodiesel production (Hawrot-Paw et al., 2019; Muller et al., 2014). Microalgae that accumulate proteins in their cells are underexplored. Nevertheless, proteins as by-products from different sources could be used for biofuel and other value-added products production. It could be done through hydrolyzing proteins to amino acids which in turn can be enzymatically biotransformed by engineered microbes to keto acids which in turn can be converted to fuels (ethanol, 1-propanol, etc.) and even pharmaceutical molecule precursors (Huo et al., 2011). However, currently, protein utilization as a source for biofuel production is in the early stage of its development (Li et al., 2018; Santos et al., 2020). Carbohydrates-derived biofuel production is obtained via fermentation, and lipid-derived biodiesel is usually made through transesterification reactions resulting in the formation of fatty acid alkyl esters (biodiesel) (Table 2). Transesterification is preferred to be catalyzed by enzymes, in particular microbial lipases are under great demand for the process, making the technology even more sustainable and ecologically friendly (Sharma et al., 2019). The relations between wastewater management and third/fourth generation biofuel production utilizing microalgae and other microorganisms are presented in Fig. 4.

Bioalcohols (bioethanol and biobutanol) and biodiesel production using microalgae biomass gained tremendous attention in the recent years and has already been comprehensively discussed in previous studies (Ambat et al., 2016; Ganesan et al., 2020; Mata et al., 2010). The circular bioeconomy-based strategy by combining wastewater treatment and microalgae cultivation can be successfully applied to produce third or fourth generation biofuel (Bošnjakovic and Sinaga, 2020; Ganesan et al., 2020; Jeong and Jang, 2020). The latter is produced employing genetically modified microorganisms. Third and fourth generation biodiesel that is produced using different microorganisms has many advantages over first and second generations biodiesel (Cea et al., 2015; Raimondi et al., 2014). In the case of microalgae that are most commonly used for the production of biofuels, their advantages as a feedstock include quick growth and easy cultivation not requiring agricultural lands, production of biodegradable and non-toxic materials during growth, CO_2 sequestration or greenhouse gas fixation and of course wastewater treatment and resources recovery: N recovery, P recovery, heavy metals recovery, etc. (Alaswad et al., 2015; Peter et al., 2021; Sharma et al., 2018). The main disadvantages of first- and second-generation biodiesel are very high raw material costs, sometimes constituting ~80% of the total biofuel production cost (Singh et al., 2011) and notably low effect of diminishing greenhouse gas emissions. Nevertheless, efficient generation of biofuels exploiting microalgae often encounters issues related to major developmental challenges. The latter are usually associated with costs of supply that need to be reduced by ten times to gain its competitiveness (at least in the US) and scaling-up. Some genetically engineered and biotechnologically tailored microalgae strains that are able to maintain suitable growth rate in usually stressful wastewater conditions and accumulate high lipid yields can be a promising solution to lower the production costs. Such engineered strains can help to remarkably improve the technology in the future (Bhatt et al., 2014; Sadatshojaei et al., 2020 Sharma et al., 2018). Different microalgae cultivation systems that could result in the obtainment of high yield of microalgae biomass and recover feed resource from wastewater are now under development. Some of the preferable and mainly discussed strategies are high-rate algal ponds, algal turf scrubber (ATS), open and closed photobioreactor and hybrid systems. The pollution prevention and resource recovering by algal-based wastewater treatment technologies has been in detail discussed by Bhatia et al. (2021), de Assis et al. (2020), Leong et al. (2021a), L'opez-Pacheco et al. (2021), Mehariya et al. (2021). For the successful realization of microalgae for the simultaneous wastewater treatment and biofuel production, it is important to identify the major genetic processes that have shaped their genomes and what impact these processes had on their phenotypes, however, it is a challenging task since both are linked and highly plastic (Brodie et al., 2017). Significant third and fourth generation biofuel production improvements should be also made not only in cultivation, but also in harvesting, drying, extraction, conversion and other stages of the microalgal biofuel production in large-scale for them to become economically feasible and advantageous (Chu et al., 2021; Georgianna and Mayfield, 2012; Javed et al., 2019; Larkum et al., 2012; Sadatshojaei et al., 2020).

The most recent studies of coupling wastewater treatment and biofuel production that have been conducted in the past two to three years for the technology implementation were largely associated with application of *Chlorella* sp. Studies have shown that *Chlorella* sp. Are capable of successfully generating biomass rich in biological macromolecules suitable for green energy production from wastewaters of kitchen, sewage, aquaculture, brewery, tannery, sugar-cane industry and even

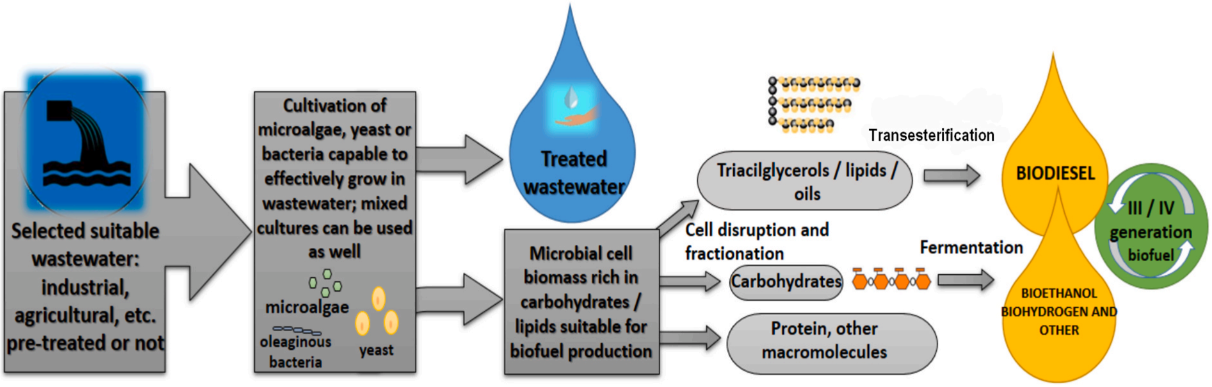

Fig. 4. Wastewater management and microalgae exploitation for industrial prosperity.

pharmaceutical (Amenorfenyo et al., 2019; Hawrot-Paw et al., 2019; Kumar et al., 2019a; Nagi et al., 2020; Nayak and Ghosh, 2020; Zewdie et al., 2020). Abu Jayyab and Al-Zuhair (2020) evaluated the usability of microalgae for the removal of phenolic compounds from wastewater and as a sustainable oil source for biodiesel production. Wastewater pre-treatment such as physical pretreatment (centrifugation, filtration, etc.) removing solid rough materials, dilution, autoclavation, UV-radiation, etc. Done prior microalgal cultivation is a common practice diminishing stressful conditions, promoting better algae growth and generation of biomass that will be converted into biofuel (Amenorfenyo et al., 2019; Salama et al., 2017). Some studies have also identified that biological pretreatment using active sludge or wastewater with indigenous bacteria might be as well cost-efficient and favorable for algal growth and wastewater nutrient removal/conversion (Lee et al., 2019b; Zahmatkesh and Pirouzi, 2020). Ryu et al. (2014) studied *C. vulgaris* for the bioremediation of municipal wastewater (MWW) and joint biofuel production. Study confirmed that the most active growth of the *C. vulgaris* took place in untreated wastewater compared to its growth in filtered and sterilized wastewaters. Toyama et al. (2018) experimentally determined that *Chlamydomonas reinhardtii*, *C. vulgaris* and *Euglena gracilis* grew better in two MWW and one swine wastewater (SwWW) effluent with indigenous bacteria than without it. Biomass of the studied microalgae increased in the range of 1.5- to 2.8-fold depending on the species of microalgae. However, certain bacterial species my cause lysis of microalgae or inhibition of their growth (Covarrubias et al., 2012; Toyama et al., 2018), therefore, the exploration of what bacteria promote the growth of different microalgae species should be done (Wirth et al., 2020).

It was also demonstrated that some oleaginous yeast can be successfully employed to produce biofuels growing in livestock wastewaters (Chung et al., 2016), sewage, domestic, household, industrial (Kanakdande et al., 2019) and sago processing wastewater with *Candida tropicalis* being used mostly (Thangavelu et al., 2020). Furthermore, some indigenous wastewater bacteria have been also shown to be able to accumulate lipids in wastewater treatment plants. A few of the representative microorganisms notable in biological wastewater treatment plants (BWWT) are bacteria *Microthrix parvicella* (long-chain fatty acid accumulating organism) and *Accumulibacter phosphatis* (polyhydroxyalkanoate (PHA) accumulating organism) (Muller et al., 2014). However, studies regarding yeast and bacteria utilization for wastewater treatment and biodiesel production are scarcer than those involving the use of microalgae. Simultaneous microalgae-bacteria or microalgae-yeast cultivation in wastewater coupled with biofuel production is being studied as well, giving some promising results regarding the promotion and improvement of cost-efficiency (the main challenge) of the technology (Gomez et al., 2016; Liu et al., 2018; Rakesh and Karthikeyan, 2019).

Another important point related to wastewater management and biofuel production is employment of sewage sludge. Industrial and MWW treatment generates the sewage sludge, which is estimated to make up ~1–2% of the total volume of treated wastewater (Cea et al., 2015; Godoy et al., 2018). It was indicated that converting wastewater sludge to lipid is considered as one of the best strategies of sludge management (Chen et al., 2020; Liu et al., 2021b; Yi et al., 2018). It has been reported that primary sludge (PS) and waste activated sludge (WAS), has many advantages as a biodiesel feedstock. The main benefits of PS and WAS are low cost and abundantly rich in lipid content (Choi et al., 2019). Melero et al. (2015) suggested that PS and WAS can contain 5–36% and 2–20% free fatty acids (FFAs), respectively, which further can be used for biodiesel and other fatty acid esters production. The sewage sludge containing fatty substances – biodiesel precursors, can be converted into this biofuel via transesterification reactions (Kumar et al., 2020; Sandoval et al., 2017). Transesterification reactions are catalyzed by chemical (acid- and/or base-catalyzed) or, as it was already mentioned, more sustainable and eco-logically friendly catalysts – enzymes, such as lipases and esterases (Table 2). Mondala et al. (2009) generated FAMEs (fatty acid methyl esters) from PS and secondary sludge via chemical transesterification reaction. Authors of the study estimated yield of 10% FAMEs per dry weight of used sludge and estimated production cost of $3.23 per gallon that is lower than usual petroleum-based diesel. Such studies demonstrate that exploitation of oleaginous biomass feedstock could be an important source of wastewater-derived alternative fuel. Understanding different characteristics of a biodiesel feedstock is important for the development and optimization of the technology and achievement of commercial feasibility. Despite the fact that determinations of the best method of collection and treatment of different sewage sludge fractions to achieve the highest lipid extraction yield are challenging, biodiesel generation from wastewater can become an important source of renewable energy in the future.

Another rather new method of sewage disposal is production of bio-oil by pyrolysis. The resulting liquid has a high-heating value and potential for electricity generation. However, this process has only been moderately successful since the complexity of bio-oil complicates the chemical processes needed to refine it (Arazo et al., 2017a, 2017b, 2017b; Dufreche et al., 2007; Supaporn et al., 2018). Thus, different wastewaters can be useful as a feedstock for biofuel and bioenergy production while reducing wastewater contamination load and making it a sustainable, environmentally friendly approach but not always a cheaper mean of biofuel production.

3. Resource recovery from wastewater to produce value-added biomolecules

The bioenergy production by combining different wastewater management and resource recovery gained huge attention. The wastewater can be attractive feedstock for cultivation bacteria, microalgae, and yeast to produce organic acids, bioplastics, biosurfactants or pharmaceutical biomolecules. Nevertheless, the suitable wastewater composition, low toxicity, and technical solutions to combine wastewater treatment with production of value-added compounds in cascade reactions in large industrial scale remains a challenge.

3.1. Biosurfactants

The most promising sector where production of high value compounds is combined with detoxification and utilization of different wastewater is production of biosurfactants. Biosurfactants are naturally occurring molecules that have hydrophilic and hydrophobic chemical moieties. This characteristic enables these molecules to reduce the surface tension and form micelles (Yanez-Ocampo et al., 2017). Surfactants are part of the most versatile group of chemicals potentially used in paints, detergents, pharmaceuticals, paper products, cosmetics, petroleum, food, and water recycling (Akbari et al., 2018). Due to their diversity, biosurfactants are considered as a potential candidate for the environmental cleanup of pollutants (Vijayakumar and Saravanan, 2015). Moreover, biosurfactants are characterized by better biodegradability, environmental compatibility, and lower toxicity than chemical compounds used for the same purpose (Pi et al., 2017). Biosurfactant-producing microorganisms are capable of facilitating the biodegradation of polycyclic aromatic hydrocarbons (PAHs) known as toxic pollutants (Sun et al., 2019). Addition of 0.2%–0.6% (w/w) of biosurfactants makes the PAH removal two-times more efficient (Bezza and Chirwa, 2017). These characteristics have been increasingly attracting the attention of the scientific and industrial community on the biosurfactants (Banat et al., 2010) and their cheaper and easier production. Several benefits of combining use of wastewater for biosurfactants production were suggested: 1) the hazardous pollutant becomes the medium; 2) new beneficial industrial products are made; 3) the pollutant is turned less toxic or hazardous (Veena-Kumara-Adi and Savitri, 2019). The choice of a suitable low-cost raw material can account for 10–30% of the overall cost (Nitschke et al., 2005). The major problem is to find a waste with adequate carbohydrate and lipid composition to support the optimal growth of microorganisms and maximum production of biosurfactants (Makkar and Cameotra, 1999). The application of different agro-culture and oil processing wastes as cheaper/renewable substrates for biosurfactants production has been discussed previously (Banat et al., 2014; Makkar and Cameotra, 2002; Pele et al., 2019; Santos et al., 2016; Sekhon et al., 2012; Vandana and Singh, 2018), but application of wastewater as nutrient source for production of microbial biosurfactants has not been clearly summarized. The low-cost feed stocks as substrate for biosurfactant production has been already used by Nitschke and Pastore (2006). Surfactant produced by *Bacillus subtilis* LB5a which was cultivated on cassava wastewater as a substrate resulted in the formation of stable emulsions with different hydrocarbons. Achieved concentration of surfactant in 48 h reached 3.0 g L^{-1}. The most popular industrial wastewater source is cooking wastewater (Waste Cooking Oil (WCO)) (Table 4). The main advantages of cooking wastewater as most preferable feedstock are high levels of hydrophobic organics (oil, quinoline, pyridine) providing the needed environment for successful growth of biosurfactant-producing microorganisms (Zhang et al., 2012). The use of WCO as a raw material resulted in 3.7 g L^{-1} (Yanez-Ocampo et al., 2017) and 5.2 g L^{-1} (Rocha e Silva et al., 2013) yields of suitable biosurfactants. The similar yield of biosurfactant (4.9 g L^{-1}) was detected using *Pseudomonas aeruginosa* and sunflower oil refining wastes as carbon source (Benincasa and Accorsin, 2008). Colak and Kahraman (2013) reached 9.6 and 13.3 g L^{-1} production of rhamnolipid in 72 h using *P. aeruginosa* and its recombinant strain, respectively. The second most promising feedstock for cultivation of biosurfactant-producing microorganisms is the wastewater obtained after depulping and demucilage of coffee fruits (known as coffee wastewater (CW)). CW is rich in proteins, carbohydrates (mannose, galactose, glucose, arabinose) and minerals (potassium, phosphorus, magnesium, calcium, iron, etc.) (Bonilla et al., 2014; Murthy and Naidu,

Table 4
Wastewater as raw material for biosurfactants production. ND – data not showed.

Biosurfactants – producing microorganism	Wastewater use as the carbon source	Yield (g L^{-1}) and surface tension (mN m^{-1})		Reference(s)
		Yield	Surface tension	
Pseudomonas aeruginosa strain BS2	Distillery and curd whey wastes	0.97	Reduction from 57 to 27 mN m^{-1}	Dubey and Juwarkar (2001)
Bacillus sp.	Cassava flour wastewater	3.0	26.59	Nitschke and Pastore (2003), 2006; Nitschke et al. (2004)
Bacillus subtilis LB5a	Cassava wastewater	2.4	Reduction from 51 to 27 mN m^{-1}	Cavalcante Barros et al. (2008)
Nevskia ramosa NA3	Palm oil mill effluent	ND	27.2	Chooklin et al. (2013)
Six biosurfactant-producing strains; strain S2	Oilfield wastewater	ND	25.7	Liu et al. (2013)
Rhamno lipid production by recombinant *Pseudomonas aeruginosa*	Raw cheese whey and olive oil mill wastewater	13.3	ND	Colak and Kahraman (2013)
Pseudozyma tsukubaensis	Cassava wastewater	ND	26.87	Cavalcante Fai et al. (2015)
Rhamnolipid production using Pseudomonas SWP-4	Waste Cooking Oil	13.93	Reduction from 71.8 to 24.1 mN m^{-1}	Lan et al. (2015)
Sophorolipid from Starmerella bombicola MTCC 1910	Synthetic dairy waste water and waste oil	2.8 g/ 100 mL	ND	Vidhya et al. (2015)
Bacillus subtilis	Industrial waste containing high rate of glucose	ND	Reduction from 70 to 44 mN m^{-1}	Secato et al. (2016)
Bacterial isolates from the Mexican state of Chiapas	Two agro-industrial wastes -Waste Cooking Oil (WCO) and Coffee Wastewater (CW)	3.7	Reduction from 50 to 29–30 mN m^{-1}	Yanez-Ocampo et al. (2017)
Bacillus licheniformis and Bacillus clausis	Pharmaceutical effluents	ND	ND	Akintokun et al. (2017)
Surfactin produced by Bacillus subtilis LB5a	Cassava wastewater	ND	ND	Cosmann et al. (2017)
Bacillus subtilis UCP 0146	Cassava wastewater	2.69	39	Maia et al. (2018)
Pseudomonas aeruginosa S5	Cooking wastewater	ND	Reduction from 72 to 30 mN m^{-1}	Sun et al. (2019)
Bacillus subtilis MTCC 1427	Spent wash	ND	Reduction from 72 to 29 mN m^{-1}	Veena-Kumara-Adi and Savitri (2019)
Serratia marcescens UCP 1549	Cassava flour wastewater and corn waste oil	ND	25.92	Araújo et al. (2019)

2012; Mussatto et al., 2011). More examples of biosurfactants production using different wastewater as a substrate are presented in Table 4. The different wastewater (industrial wastewater, WCO and CW wastewater, cassava wastewater, etc.) can be cheap and attractive source of nutrients for microorganisms to produce value-added products like biosurfactants. However, optimization of their addition to nutrient media, cultivation parameters such as temperature, duration, agitation speed, pH in addition to refining both upstream and downstream processes for product recovery remains important challenges for biosurfactants production from wastewater in industrial scale.

3.2. Bioplastics

Traditional plastics pose a problem for the planet, whether it is due to the traditional fuels used to produce them or the detrimental chemicals released during their exposure in environment and during slow degradation. Bioplastics are regarded as the most promising alternative to traditional plastics and a way to reduce environmental damage. The main bioplastic which production is combined with the utilization of different waste sources is polyhydroxyalkanoates (PHAs). PHAs are the non-toxic, insoluble in aqueous media, moldable biopolymer, which can be used for production of packing materials, films, bottles, and in biomedical applications such as tissue engineering (Mayeli et al., 2015). The main metabolic pathway related with PHA production are presented in Table 2. Due to the high demand for application, the reduction of production costs of PHAs is very important. A cost reduction involved several aspects: 1) cheap substrates; 2) improved novel fermentative strategies; 3) improved recovery and purification steps and 4) use of microorganisms accumulating PHAs to high level (Khatami et al., 2021). Earlier reviews and research reports focused on bioplastics production using food wastes as additional nutrient component for microorganisms (Bengtsson et al., 2008; Bosco and Chiampo, 2010; Chakravarty et al., 2010; Khardenavis et al., 2007; Mohanty et al., 2021; Nielsen et al., 2017; Tsang et al., 2019). However, the production of bioplastics using wastewater from other sources has not been given enough attention. Ceyhan and Ozdemir (2011) produced poly-β-hydroxybutyrate (PHB) from domestic wastewater using *Enterobacter aerogenes* 12Bi strain. The use of the domestic wastewater (DWW) as carbon source resulted in the achievement of PHB that made up more than 90% of the dry cell weight. Ben et al. (2011) reported bioplastic production using wood mill effluents as feedstock and Mayeli et al. (2015) applied petrochemical wastewater as carbon source for production of PHB by *Bacillus axaraqunsis*. The PHA yield was 6.33 g L^{-1} concentration, corresponding to 66% of cell dry weight. In 2015 Morgan-Sagastume et al. proposed a pilot-scale process: it was operated over 22 months at the Brussels North Wastewater Treatment Plant (WWTP) in order to evaluate PHA production integration with services of MWW and sludge management. The biomass production with 0.5 gPHA $gVSS^{-1}$ mentioned as realistically achievable within the typically available carbon flows at municipal waste management facilities.

The newest research analyzed application of MWW for production of PHA and possible combination of production of these bioplastics with wastewater treatment plants (WWTPs) in Germany (Pittmann and Steinmetz, 2017). The authors described detoxification of the MWW coupled with production of value-added products. However, food industry wastewater including pickle industry wastewater with high organic content are among the most inexpensive sources for PHA production (Guventurk et al., 2020). The authors use two laboratory scale sequencing batch reactors for cultivation enriched microbial culture under aerobic feeding conditions. PHA content was 1.820 mg COD L^{-1} (44% in the biomass). It can be summarized that the combination of microorganisms grown on wastewater and bioplastics production can be suitable strategy for both wastewater management and economic important biopolymers production. However, acid production during PHA polymerization requires control since acid composition can affect the composition and mechanical characteristics of PHA. Engineering of microorganisms and successful screening for enhanced PHA production may be helpful to improve PHA yield.

3.3. Organic acids

Production of organic acids and wastewater management are related in two ways: 1) the wastewater can be a source of recovery of essential organic acids and 2) wastewater can serve as carbon and other nutrient sources for cultivation of microorganisms to produce organic acids. A natural/organic acid is a natural compound with acidic properties. The most widely recognized natural acids are carboxylic acids, whose causticity is related with their carboxyl gathering. Natural acids basically contain hydrogen and carbon with another component. The carboxylic acids are known as volatile fatty acids (VFAs) or short-chain unsaturated fats (SCFAs). The VFAs come in close vicinity of bacteria during their random and regular movement in AD digestor. Hence, VFAs are broadly present in actuated slop, landfill leachates and wastewater. The detachment of natural buildups from wastewater discharged from enterprises is significant for contamination control and recuperation of valuable materials. Squander waters contain huge amounts of natural acids such as acetic, propionic and formic acids perceived as a huge cost to the business. The removal of organic acids from wastewater and recycling use could be an ideal option for wastewater management. Chantarasukon et al. (2016) suggested that most common acids present in wastewater are acetic, formic, propionic, isobutyric, caproic and valeric acids. These organic acids are important in many industrially sectors (food industry, textile, tanning, rubber processing manufacture of pharmaceuticals, etc.). Thus, highly efficient methods are required for the inexpensive and bioeconomic production of organic acids. Various advanced techniques including electrodialysis, electrometathesis, electrodeionization, electro-membrane processes, bipolar membrane have been used for the extraction of organic acids from broth medium (Handojo et al., 2019).

The second important topic regarding organic acids is related with utilization of sewage sludge to produce biogas. During the second phase of biomethane production (Table 2), anaerobic acidification results in the production of VFAs including acetic, propionic, butyric and valeric acid (Khan et al., 2016; Lim and Vadivelu, 2019; Moestedt et al., 2019). Darwin et al. (2019) used the sugar-containing wastewater for anaerobic acidification to produce organic acids and ethanol. Earlier, Hwang et al. (2001) applied swine wastewater (SwWW) to maximize acetic acid production in partial acidogenesis. The production and extraction of VFAs in this process depends on several factors: a) the used reactor type; b) substrate composition and product spectrum; c) temperature and pH; d) retention time; e) organic loading rate (Darwin et al., 2019; Gracia et al., 2020; Khan et al., 2016; Strazzera et al., 2018). Thus, the wastewater is promising feedstock to produce value-added compounds such as different organic acids. The development of strategies corresponding the circular bioeconomy principles and combining wastewater management and organic acids production remain an important future research topic and industrial challenge.

3.4. Biopesticides

The wastewater can be promising nutrient source for cultivation of *Bacillus thuringiensis* (Bt) which is a well-known producer of biopesticides. Biopesticides derived from *B. thuringiensis* (Bt) are the most prominent biological agents for selective control of pest insects The feedstock used for the production of Bt-based biopesticides makes up a substantial part (35–59% approximately) of the overall production cost (Stanbury et al., 1995). Therefore, a cheap, simple and widely available raw material is needed for the effective commercial production of Bt toxin (Yezza et al., 2006). Wastewater and wastewater sludge have been extensively explored for the Bt biopesticide as a novel value-addition approach (Hoa et al., 2014; Montiel et al., 2001; Vidyarthi et al., 2001; Yezza et al., 2004, 2005). Yezza et al. (2006) used starch industry wastewater (SWW), slaughterhouse wastewater (SHWW) and secondary sludges from three different wastewater treatment as feedstock to produce Bt-based biopesticides. The authors found that secondary sludges and SWW act as good raw materials for the production Bt biopesticide with higher yield. Ndao et al. (2017) and Kumar et al. (2019b) reported the production of Bt-based biopesticide formulation using SWW as substrate. The unit production cost was estimated to be \$2.54 L^{-1} for 5 million L plant capacity. The results were obtained at pilot plant (2000L capacity fermenter). These data suggested reliable production of BT-based toxins by using wastewaters as feedstock. The recent study (Keskes et al., 2020, Keskes et al., 2021) proposed enhanced production of *Photorhabdus temperate* biopesticides using Tunisian wastewater and wastewater from food industry as a raw material. These several studies clearly showed that wastewater can be applied to produce biopesticides. However, the application of this combination in industrial scale is still limited by costly pretreatment and low yield of entomotoxicity. The improvement of entomotoxicity level is the next goal.

3.5. Pharmaceutical biomolecules

The production of vitamins, antibiotics and anticancer drug precursors from wastewater is a topic of discussion and a field of research and challenge for both academic an industrial sector worldwide. Would it be really feasible to extract such high value compounds or not? Very limited number of reports are currently available regarding this matter. Some findings of the possibilities of the extractions of Vitamin B12 are shortly presented in the present review.

One potential wastewater source which has been successfully exploited for production of vitamin B12 is tofu wastewater (Yu et al., 2015). Tofu is a traditional oriental food and its production results in a generation of substantial amounts of wastewater (Yonezawa et al., 2012). The tofu wastewater displays COD values ranging from 17 to 26 g L^{-1} and BOD from 5.8 to 7.9 g L^{-1}, respectively (Belén et al., 2012). Tofu wastewater has a high content of proteins, oligosaccharides, and isoflavones that can be used as important ingredients in purified form for functional food products. However, for now tofu wastewater is not properly utilized. Despite the development of isolation and fermentation technology for vitamin B12, the low yield and impurities such as other pharmaceutical biomolecules still prevent the production of B12 by using tofu wastewater in the large scale.

4. Practical applications and future research prospects

The practical applications of wastewater treatment and recovery of value-added products solely depend on the cost and efficiency of the process. A great success has been achieved in the bioelectricity generation using fuel cells, other bioenergy products like biomethane, biohydrogen and biogas production are also currently intensively investigated. However, generation and accumulation of different wastewaters is a major challenge that needs to be overcome to achieve successful circular bioeconomy because the wastewater cannot be stored for longer time due to various detrimental effects on the living system. Different research data revealed that the production or recovery of organic acids and other chemical or complex biological molecules from wastewater remain challenging with high cost of recovery process. Overall, the use of microorganisms for the generation of bioelectricity using MFCs and other biogases seems to be more practical and feasible as compared to the recovery of complex molecules. In bioenergy generation microbes utilize most of the soluble impurities/pollutants from the wastewater for their growth and simultaneously generate the energy. Such approach can help to create cleaner environment and lead towards a sustainable circular bioeconomy. Nonetheless, microbial consortia generally need a balanced nutrient composition, ambient temperature and pH range. These are the challenges in the wastewater biorefinery always encountered by the researchers at large scale. Moreover, in order to avoid this limitation integrated system of bio-refinery to extract the feedstock, nutrient, energy rich compounds with direct application in the bioenergy generation should be implemented. Advanced techniques are also required to remove the newly emerging contaminants such as plastic waste, nanoscale particles, and heavy metal ion impurities to maintain the standard of water quality to an appropriate level. Presently, wastewater management and biorefineries aimed to extract the value-added products are playing a crucial role in the economic growth. The circular economy application with ecosystem protection should be considered while recovering the products of commercial values. Maximum utilization of natural bioconversion machinery, including plants, algae, and microorganisms could help to reduce the overall operational cost of wastewater biorefinery. However, the design of bioresource recovery should emphasize on lowering the demand of treatment operations with maximum extraction of value-added products using inexpensive steps.

5. Conclusion

This review presents the bio-based production of various high-value and bioenergy-based products by using wastewater resource. Resource recovery processes and emerging techniques from wastewater have been explored for their future possibilities and sustainability. However, integrated system of bio-refinery to effectively extract the feedstock, nutrient, energy rich compounds with direct application in the bioenergy generation should be implemented. Thus, maximum utilization of natural bioconversion machinery, including plants, algae, and microorganisms could help to reduce the overall operational cost of wastewater biorefinery. As far as time of treatment and recovery is an important parameter in wastewater remediation, a more emphasis should be given to transfer the chemical and physical methods from lab to field scale operation to improve the sustainability. The social and public perceptions of these products are equally necessary especially in the case of feedstocks or food nutrients.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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