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A comparative analysis of biopolymer production by microbial and bioelectrochemical technologies†

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Production of biopolymers from renewable carbon sources provides a path towards a circular economy. This review compares several existing and emerging approaches for polyhydroxyalkanoate (PHA) production from soluble organic and gaseous carbon sources and considers technologies based on pure and mixed microbial cultures. While bioplastics are most often produced from soluble sources of organic carbon, the use of carbon dioxide (CO_2) as the carbon source for PHA production is emerging as a sustainable approach that combines CO_2 sequestration with the production of a value-added product. Techno-economic analysis suggests that the emerging approach of CO_2 conversion to carboxylic acids by microbial electrosynthesis followed by microbial PHA production could lead to a novel cost-efficient technology for production of green biopolymers.

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1. Introduction

Bio-based materials produced from renewable sources of organic carbon instead of petroleum hydrocarbons can play an important role in reducing consumption of fossil fuels and moving our society towards a circular economy. Polyhydroxyalkanoates (PHAs), which are produced for storage of carbon and energy by a large number of microorganisms within the bacterial and archaea domains, are often used for bioplastic production. The insoluble PHA granules inside the microorganisms can make up to 90% of the dry weight of the cell mass.[1] More than 150 types of PHAs have been identified. The most common form of PHA is polyhydroxybutyrate (PHB). Depending on the composition and properties of the PHA, applications can range from use in biodegradable packaging, to use as chemical additives, to usage in the fields of medicine, agriculture, wastewater treatment, and cosmetics.[2–4]

In spite of multiple benefits of using biopolymers, their commercialization continues to be problematic due to high biopolymer production costs compared to polymers produced from conventional feedstock. Indeed, the price of polypropylene and polyethylene is about US $1.25–2.53 per kg,[5] while that for PHAs has been reported to be up to 16 times higher than the major petroleum-derived polymers.[6] According to a study of the global PHA market for 2018,[7] the average PHA price was US $8.0 per kg. This price varied according to the target application and quality of the PHAs. In the same study, the average price of PHAs destined for packaging and food services was calculated to be US $7.6 per kg with an estimated market size of US $25 000 000, while for biomedical applications the average price was US $11.9 per kg with a market size of US $15 000 000. In 2019, the global PHA market was estimated to be US $57 000 000 with a projected compound annual growth rate of 11.2%.[7]

Integration of biopolymers into the global market can be facilitated through a thorough cost analysis and identification of technologies capable of reducing production costs, while minimizing environmental impact. Energy consumption, PHA yield, and the efficiency of the downstream processing are the most important parameters determining the cost of production.[8] Carbon sources used in pure culture microbial fermentations contribute significantly to overall environmental impact and production costs. Therefore, the use of mixed microbial communities capable of PHA production from liquid and gaseous waste streams, such as food wastes,[9] agricultural wastes,[10,11] landfill gas, carbon dioxide (CO_2), wastewater,[12] polystyrene waste,[13] and glycerol[14,15] is seen as a sustainable approach for bioplastics production. Biopolymers produced from CO_2 are of particular interest, as this approach also provides a sustainable method for utilization of CO_2 captured from industrial off-gases and from air.[16]

In this review, PHA production technologies are described based on the type of carbon source (liquid or gaseous) used. Also, single-stage and two-stage production processes are considered. Typically, a single-stage production can be accomplished if a well-defined liquid carbon source is used, either with pure or mixed microbial cultures. If more complex carbon sources such as agro-industrial wastes or flue gases are used, the production of PHAs must be carried out in two stages, where

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the conversion of complex carbon sources into simple sugars or carboxylic acids is followed by a stage of PHA production. Finally, a novel approach of biopolymers production from CO_2 and electrons in a microbial electrosynthesis (MES) system is reviewed. An overview of these technologies is followed by the review of techno-economic assessments evaluating the costs of PHA production, including PHA production from CO_2 through MES. By combining a detailed review of PHA production technologies with a review of published techno-economic assessments, this study helps to select promising cost-efficient technologies for producing biopolymers from renewable carbon sources, including waste biomass and CO_2.

2. PHA production technologies

Microorganisms promote their survival by production of PHAs to store carbon and energy. Typically PHA production is induced by stress caused by a lack of nutrients.[17] Often microbial growth can be followed by PHA production under nutrient-limited conditions. Microbial production of PHAs has been achieved from multiple sources of carbon, including well-defined substrates such as glucose, agro-industrial wastes containing complex carbon sources, gas mixtures (*e.g.*, syngas, biogas) and, more recently, CO_2. In the following discussion we will use PHA as a term to include PHB, unless the article we are citing focuses on PHB.

2.1 PHA production from liquid carbon sources

A number of Gram-positive and Gram-negative bacterial strains are capable of producing PHAs, although most of the PHA producing bacteria are Gram-negative. Table 1 summarizes PHA production from well-defined dissolved (liquid) carbon sources using different fermentation systems. Most of the biopolymer production studies cited in this table are batch processes, which are easier to implement in a laboratory, but also represent disadvantages such as variability in the quality of the product and downtime for the preparation of the bioreactor equipment for the next batch. To resolve this limitation, Atlić *et al.*[18] evaluated PHB production using a mineral medium with glucose in a multi-reactor system consisting of five continuous stirred tank reactors, which approximates a continuous tubular plug flow reactor. The first reactor was used for balanced bacterial growth using *Cupriavidus necator* (also known as *Ralstonia eutropha*), while PHB accumulation was achieved in the subsequent reactors under nitrogen-limited conditions. This approach demonstrated a specific volumetric productivity of 1.97 g $(L\ h)^{-1}$ and a polymer content of 77% w w^{-1}.

PHB production from acetate and valerate using a pure culture of *C. necator* was evaluated by Garcia-Gonzalez *et al.*[19] After 118 h of fermentation, 60 g L^{-1} cell dry matter (CDM) with 72% PHB content was obtained, demonstrating the feasibility of producing PHB from acetate. In another study, PHB production by *Bacillus cereus SPV* from glucose in batch and fed-batch bioreactors was evaluated.[20] The two main differences between the batch and fed-batch fermentations were the time required for maximum PHB accumulation, which was reduced from 48 to 32 h in the fed-batch fermentation, and in which the final PHB yield increased from 29% DCW to 38%. In a recent study of Gahlawat *et al.*[21] PHB productivity was improved from 0.17 g $(L\ h)^{-1}$ in the batch process to 0.6 g $(L\ h)^{-1}$ in the fed-batch process. Also, the PHB content increased from 51 to 75%. Attempts to increase PHA production include the work of Chakraborty *et al.*,[11] which used a condensed corn medium (by-product of ethanol production from corn) for cultivation of *C. necator* at high cell density. Furthermore, this work suggests that butyric and propanoic acids provided the best results in terms of PHA production and optimal levels of these volatile fatty acids (VFAs) were determined. Overall, these studies demonstrated the advantages of using continuous or, at least, fed-batch bioprocesses for reducing cultivation times and maximizing production rates.

The effect of different conditions of nutrient deficiency on some PHB producing cultures have been also explored. For example, it is known that *Alcaligenes latus* produces PHBs even under nutrient-sufficient conditions. Nevertheless, the process of recovering PHBs is more expensive under these conditions and the resulting PHB content was less than 50%.[22] A study conducted by Wang and Lee[23] demonstrated the effect of nitrogen limitation on the production of PHBs by *A. latus* in batch cultures using sucrose as a carbon source. Nitrogen limitation was applied after 12 h with a sucrose concentration between 5 and 20 g L^{-1}. After 8 h of nitrogen limitation, the cell concentration, PHB concentration and PHB content reached 111.7 g (dry cell weight) L^{-1}, 98.7 g L^{-1} and 88%, respectively, resulting in a productivity of 4.94 g PHB per (L h).

In these studies, *C. necator* stands out for its unique physiology. This facultative chemolithoautotrophic microorganism is capable of producing PHBs in the range of 60 to 90% of CDM (cell dry matter) from a broad range of carbon sources. The biosynthesis of PHB in this bacterium can switch between heterotrophic and autotrophic modes for growth and production of PHBs, respectively.[24] In the heterotrophic growth mode, this microorganism can use organic compounds such as sugars, organic acids, VFAs and vegetable oils, while under autotrophic conditions it uses H_2 and CO_2 as energy and carbon sources, respectively, where CO_2 is fixed by the Calvin–Benson–Bassham cycle.[24]

Production of PHAs using mixed microbial consortia (MMC) is a more attractive practical approach since it can reduce operating costs. With MMC, non-sterile conditions are used and the microorganisms are adapted to various carbon sources, including waste effluents. Furthermore, the microorganisms capable of accumulating biopolymers are selected by the operational conditions, *i.e.* the ecosystem is designed instead of the strains.[9] The production of PHA by mixed microbial cultures occurs under transient conditions of carbon or oxygen availability, known respectively as dynamic aerobic feeding and anaerobic/aerobic process.[25] There are two main groups of bacteria responsible for the accumulation of PHAs under these conditions, polyphosphate (PAOs) and glycogen accumulating organisms (GAOs). Under anaerobic conditions, carbon substrates are consumed, PAOs release phosphate, gaining energy for the PHA accumulation process, while GAOs gain

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Table 1 PHA production using well defined and complex dissolved organic carbon sources

Carbon source	Limiting nutrient	Culture	Fermentation system	PHA polymer	Cell dry weight, g L^{-1}	PHA content,%	PHA productivity g $(L\ h)^{-1}$	PHA yield g g^{-1}	Reference
Glucose	Nitrogen	*C. necator DSM 545*	Fed-batch	PHB	81	77 ± 7.5	1.97 ± 0.56	—	18
Acetic acid	Nitrogen	*C. necator DSM 545*	Fed-batch	PHB, PHBV	60–65	PHB – 72 PHBV – 74	PHB – 0.37; PHBV – 0.41	—	19
Glucose	Glucose	*Bacillus cereus SPV*	Fed-batch	PHB	3	38	—	—	20
Sucrose	None	*Alcaligenes latus DSM1124*	Fed-batch pH-stat	PHB	143	50	3.97	0.3	22
Sucrose	Nitrogen	*Alcaligenes latus DSM1123*	Fed-batch	PHB	112	88	4.94	—	23
Sucrose	Sucrose, nitrogen	*Alcaligenes latus DSM1124*	Fed-batch	PHB	39	75	0.6	—	21
Glucose	Nitrogen	*A. eutrophus NCIMB 11 599*	Fed-batch	PHB	164	76	2.42	0.17	76
Condensed corn medium	Nitrogen	*C. necator H16* ATCC 17699	Batch	PHB	6–15	29–41	0.02–0.04	—	11
Acetate	Acetate, nitrogen	Sludge-GAO[c] enriched culture	SBR	P(3HB/3HV)	41% DW	41	—	0.3–0.4[b]	32
Paper mill effluent	Phosphorus	GAO[c] enriched culture	SBR	P(3HB/3HV/3HMV) 33 : 51 : 16	—	42	0.093[a,e]	0.34[b]	33
Acetate	None	PAO[c] enriched culture	SBR	PHB	29%	50	0.2 g $(g\ h)^{-1}$	0.6 (mol mol^{-1})	36
Wastewater	Substrate	Activated sludge	SBR	P(3HB/3HV) 50 : 50	—	53	0.23[a,d]	0.9[b]	34
Sugar cane molasses	Nitrogen	Mixed culture	Fed-batch, CSTR	P(3HB/3HV) 48 : 52	—	56	0.37	0.9[b]	35
Fermented cheese whey	Nitrogen			P(3HB/3HV) 81 : 19	—	65	0.56	0.7[b]	
Molasses and cheese whey	Nitrogen			P(3HB/3HV) 77 : 23	—	40	0.15	0.6[b]	
Wastewater	Nitrogen, phosphorus	Sludge	SBR	—	—	8–18	—	—	80

[a] Units: the amount of PHA produced (g SCOD) per the amount of active biomass (g) per hour. [b] Storage yield is defined as the amount of PHA produced in g COD per amount of carbon source consumed (g COD). [c] Notations: SCOD – soluble chemical oxygen demand; SBR – sequencing batch reactor; CSTR continuously stirred tank reactor; GAO – glycogen accumulating organisms; PAO – polyphosphate-accumulating organisms. [d] Estimated based on available information. [e] Estimated in g_{PHA} $(g_{VSS}\ h)^{-1}$.

energy only from the glycolysis of glycogen. Under aerobic/anoxic conditions, both microbial groups use the stored PHA for growth, maintenance and replenishment of the glycogen reserve.[26]

Depending on the type of substrate used as feedstock, the production of PHA using mixed cultures can takes place in either one or two stages, as illustrated in Fig. 1. In the single-stage process, the growth of PHA-accumulating organisms (under aerobic or anaerobic/aerobic conditions) and the ensuing accumulation of PHAs occurs in the same bioreactor (Fig. 1A). This approach has been mainly applied when organic acids are used as feedstock.[27] The two-stage process shown in Fig. 1B often uses waste-based raw materials, many of which are carbohydrate-rich, and must include a pre-fermentation stage. An additional stage is required to transform the carbohydrates into VFAs, which can then be used for production and accumulation of PHA.[28] This is primarily because mixed cultures subjected to feast and famine conditions are often incapable of storing biopolymers from sugar-based compounds.[29] The culture selection step is performed to enrich for microorganisms that are not only capable of storing the polymers but also have a high storage capacity. Selection of cultures with complex substrates can be performed by implementing anaerobic/aerobic (AN/AE) and aerobic dynamic feeding (ADF) regimes.

The source of carbon for PHA production plays a crucial role, as it can represent more than half the overall production costs.[30,31] For this reason, many studies have focused on finding inexpensive sources of carbon, such as agro-industrial wastes, including whey, lignocellulosic materials, glycerol (obtained from biodiesel production) and other organic waste compounds. Table 1 includes studies that describe production of PHAs from different carbon sources using mixed cultures. In one such study,[32] acetate was used to produce PHBs, poly-3-hydroxyvalerate (PHV) and poly-3-hydroxy-2-methyl-valerate (PHMV), using glycogen accumulating organisms (GAOs). The culture selection stage was carried out for the enrichment of the GAO culture by alternating anaerobic and aerobic conditions with acetate as feedstock. Bengtsson *et al.*[33] investigated the feasibility of using paper mill effluent to produce PHAs (PHB,

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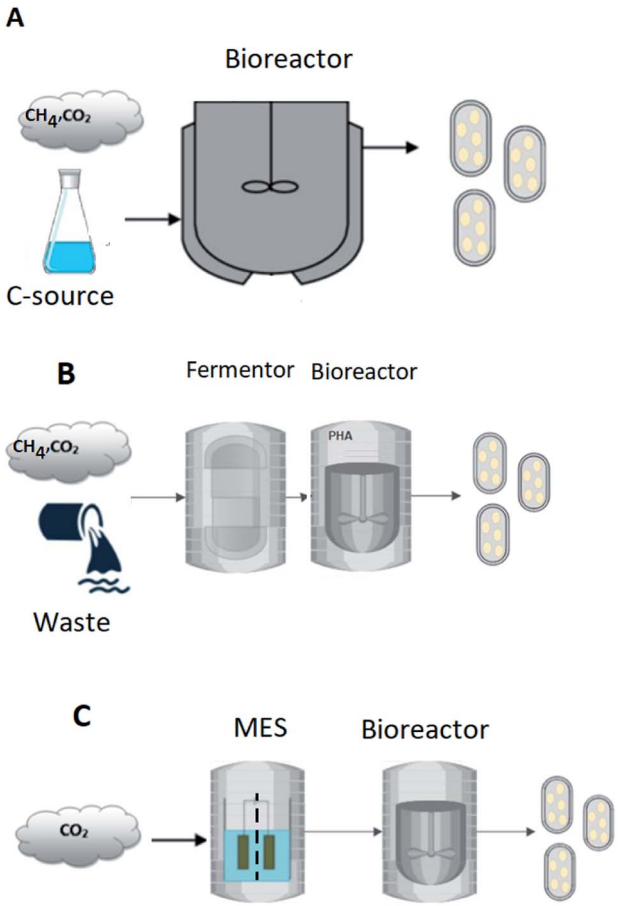

Fig. 1 PHA production from liquid and gaseous sources of carbon in (A) single stage fermentation, (B) two stage fermentation, and (C) two stage microbial electrosynthesis process.

PHV and PHMV) using GAO-enriched activated sludge. A two-phase process comprising acidic fermentation and culture enrichment/batch accumulation of PHAs based on the anaerobic/aerobic cycle was used. It was observed that the highest PHA content was obtained by applying aerobic conditions, in which PHA and glycogen were produced, followed by anaerobic conditions, in which stored glycogen was used for further production and accumulation of PHA. The highest PHA content achieved was 42%, and total yield in the two-stage process was 0.10 kg of PHA per kg of soluble chemical oxygen demand (COD). In another study,[34] a mixed microbial consortium was used to produce PHAs using primary solids fermenter liquor from municipal wastewater as feedstock under AN/AE and aerobic conditions. This resulted in 10 to 25% w w^{-1} PHA content. It was observed that when the batch aerobic reactor was seeded from the anaerobic/aerobic reactor and fed fermenter liquor, approximately 53% PHAs w w^{-1} was obtained. In another study,[35] cheese whey (CW) and sugar cane molasses (SCM) were used as carbon sources in a two-phase PHA production process using mixed microbial cultures. Acetate and butyrate were the main products from CW, while valerate was the dominant intermediate compound from SCM. Maximum PHA contents of 56% and 65% were obtained with fermentation on SCM and CW, respectively, in the second stage. Also, PHB production from synthetic wastewater, mainly composed of acetate, by a PAO-enriched activated sludge showed a PHB content of up to 29%.[36]

2.2 PHA production from gaseous carbon sources

Production of biopolymers from gaseous carbon sources such as CH_4 and CO_2 is gaining attention since these are greenhouse gases (GHG). In particular, several recent studies were focused on the use of CO_2 for the production of PHAs. CO_2 represents about 81% of total greenhouse gas emissions,[37] so it is broadly available and does not compete with the food supply chain. Production of biopolymers has been also explored using syngas and exhaust gases containing carbon monoxide (CO). Table 2 summarizes several studies on the production of biopolymers from gaseous substrates. As can be seen from this table, studies for converting gaseous substrates to biopolymers were conducted using either pure or mixed microbial cultures. In pure culture experiments, the model microorganism is once again *C. necator*, since it is metabolically versatile and capable of changing between heterotrophic growth using organic compounds and autotrophic growth using CO_2 as carbon source, H_2 as energy source, and O_2 as electron acceptor. There are two main cultivation methods to directly use CO_2 for the production of PHBs by *C. necator*: autotroph–autotroph, which uses a gaseous mixture of CO_2, H_2 and O_2 for both cell mass growth and PHB accumulation, and heterotroph–autotroph, which uses an organic substrate such as glucose for cell growth followed by autotrophic PHB production. The presence of O_2 in the gas mixture creates significant difficulties for the practical implementation of this approach, as care must be taken to avoid explosion.

An alternative method for PHB production from CO_2 is to use autotrophic–heterotrophic–heterotrophic cultures, which imply co-culturing or using two different microorganisms in different stages. For example, anaerobic acetogenic bacteria can be used to convert H_2 and CO_2 to acetate followed by acetate utilization for biomass growth and PHB production in the presence of O_2.[19] The same study showed that the autotrophic–autotrophic cultivation with CO_2 as feedstock could theoretically consume 2.84 ton-CO_2 per (ton-PHB) and 0.96 kg H_2 per (kg PHB). The autotrophic–heterotrophic–heterotrophic cultivation consumes the same amount of CO_2 per ton of PHB, but requires about 55% less H_2 (0.42 kg H_2 per (kg PHB)). In the heterotrophic–autotrophic cultivation process where CO_2 is used with glucose, only 1.58 tons of CO_2 per (ton of PHB) are consumed. In contrast, in a heterotrophic cultivation using glucose or similar compounds instead of CO_2, 2.81 tons of CO_2 per (ton of PHB) are emitted. While the autotrophic–autotrophic cultivation results in the highest amount of CO_2 removed, the heterotrophic growth is generally faster, thus reducing the overall cultivation time.

The impact of nitrogen limitation on P(3HB) production was studied by Miyahara *et al.*[38] This work used gas mixture with low hydrogen content ($3.6H_2 : 7.6O_2 : 12.3CO_2 : 76.5N_2$) for autotrophic fermentation of a *C. necator* culture. Here, a nitrogen deficient culture medium yielded the highest polymer content

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Table 2 PHA production from gaseous carbon sources

Carbon source	Limiting nutrient	Culture	Fermentation and time	PHA	Cell dry weight g L^{-1}	PHA concentration g L^{-1}	PHA content,%	PHA productivity g $(L\ h)^{-1}$	Reference
Glucose, biorefinery off-gas ($H_2 : O_2 : CO_2 = 84.0 : 2.8 : 13.2$)	Nitrogen	*C. necator*, DSM 545	Autotrophic/ heterotrophic	PHB	21–38	15.3–24	63–73	CO_2–biogas: 0.23, CO_2–bioEtOH: 0.11	16
Gas mixture ($H_2 : O_2 : CO_2 : N_2 = 3.6 : 7.6 : 12.3 : 76.5$)	Nitrogen and/or phosporus	*C. necator*, ATCC 17699, strain 1F2	Flask, autotrophic	P(3HB), PHBV	0.31–0.52	0.02–0.27	Up to 70	0.00013–0.0018[a]	38
Fructose, gas mixture ($CO_2 : O_2 : H_2 = 10 : 20 : 60$), valeric acid	Nitrogen	*C. necator*, B-5786	Heterotrophic/ autotrophic, two-stage batch, 60–70 h	PHBV	18	15	PHA: 76%; P3HB: 37%, P3HV: 63%	0.2	78
Glucose, gas mixture ($H_2 : O_2 : CO_2 = 84.0 : 2.8 : 13.2$)	Nitrogen	*C. necator*, DSM 545	Heterotrophic/ autotrophic, 68 h	PHBV	32	24.7	78	0.87	79
Gas mixture ($H_2 : O_2 : CO_2 = 77 : 11 : 11$), valeric acid	Nitrogen	*C. necator*, ATCC 17697	Autotrophic, 100 h	P(3HB), PHBV	12	P(3HB): 0.81, P(3HV): 0.25	63	0.012–0.005	39

[a] Value estimated based on available information.

of 70% $w \cdot w^{-1}$. Garcia-Gonzalez *et al.*[16] investigated the impact of using CO_2-rich waste gases on the production of PHBs in two-phase fermentation system using glucose as a substrate for heterotrophic growth followed by autotrophic production of biopolymers from industrial waste gases. Bacterial performance was not affected by the use of CO_2-rich exhaust gases, reaching final PHB content and productivity of up to 73% and 0.227 g $(L\ h)^{-1}$, respectively. Park *et al.*[39] determined that a 1% CO_2 concentration in a gas mixture of $H_2 : O_2 : N_2 = 7 : 1 : 91\%$ (v/v) was optimal for *C. necator* growth and PHB accumulation.

The use of CH_4 for PHB production has also been explored with a mixed methanotrophic consortium. The methanotrophs can produce PHBs even under non-sterile conditions, thus reducing operating costs.[40] The methanotrophs responsible for the biodegradation of CH_4 (type II methanotrophs), need specific conditions to divert the flow of carbon associated with the assimilation of CH_4 to synthesize intracellular PHB. A recent study[41] evaluated the effects of temperature and phosphorus on the rate of CH_4 consumption and the potential for PHB accumulation of different methanotroph-enriched inocula. Higher rates of CH_4 consumption for growth were obtained under non-limiting concentrations of phosphorus at temperatures ranging from 25 to 37 °C. Subsequent PHB production occurred under phosphorus-limited conditions, with the highest PHB content ($13.6 \pm 5.6\%$) obtained with the *Sphagnum* – derived inoculum at 25 °C. Luangthongkam *et al.*[42] and Myung *et al.*[43] have demonstrated the feasibility of PHB and PHBV production using mixed methanotrophic cultures dominated by *Methylosinus* sp. and *Methylocystis* sp., respectively.

2.3 PHA production from CO_2 through microbial electrosynthesis

The recently introduced concept of CO_2 reduction in a microbial electrosynthesis (MES) cell utilizes electroactive microorganisms capable of either direct electron uptake from the cathode and production of short chain fatty acids (SCFAs) and/or CH_4, or bioelectrochemical production of H_2, which is then used for microbial CO_2 reduction.[44–49] A typical MES configuration consists of anodic and cathodic chambers separated by a proton exchange membrane (PEM), although membraneless MES systems have also been developed.[50,51] The bioelectrosynthesis is supported by an applied voltage, typically at a level above the threshold for water electrolysis, which results in water splitting at the anode. Notably, the bioelectrochemical system can be also operated with applied voltages below the onset of water electrolysis if a carbon source is provided to the anode as a source of electrons for anodophilic electroactive microorganisms.[47] However, such microbial electrolysis cell (MEC) shows significantly lower current density and feature CO_2 release at the anode.[50] In MES cells, acetogenic microorganisms can reduce CO_2 using the H_2 produced by the electroactive microorganisms, thus a microbial consortium is formed.[52] Typically, the indirect metabolic pathways of acetogens result in acetate as the predominant product, although the formation of other organic compounds such as propionate, butyrate, ethanol, isopropanol, caproate, and caprylate has been reported.[51]

Recently, the use of a consortium of electroactive and acetogenic microorganisms growing in the MES cell cathode was explored for its ability to produce VFAs from CO_2,[53] which can be subsequently used for PHA production,[52] as shown in Fig. 1C. The use of MES to produce VFAs has certain advantages over a more conventional approach of organic wastes fermentation, including a more consistent composition of produced VFAs and better process control. Indeed, organic waste fermentation products depend on many variables such as temperature, pH, inoculum, type of feed, *etc.*[52] Moreover, hydrolysis of complex organic molecules is notoriously slow, while higher VFA

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production can be expected in the MES using a specialized microbial community growing on a single carbon source (CO_2).

The electroactive microorganisms found in MEC and MES cells are generally chemolithoautotrophic.[44] These microorganisms can either form a biofilm or be planktonic. It has been observed that pure cultures were more efficient at CO_2 bioelectrochemical conversion to acetate, typically higher than 80%, while it was around 60% for a mixed community[54] due to formation of other products such as CH_4. Nevertheless, preference has been given to the use of mixed cultures since sterile conditions are difficult to maintain for industrial-scale operations. For this reason, research has focused on enriching mixed consortia by bioaugmentation, which could lead to higher product yields.

Table 3 summarizes studies on using MES to convert CO_2 to organic compounds. Nevin *et al.*[55] conducted one of the first studies to demonstrate the feasibility of CO_2 reduction to acetate and 2-oxobutyrate in a MES. In this work, *Sporomusa ovata* was used and the cathode potential was maintained at −0.4 V *vs.* Ag/AgCl reference electrode. Tremblay *et al.*[56] demonstrated increased CO_2 conversion by microbial electrosynthesis through adaptive evolution of *S. ovata*, which was shown to grow more rapidly autotrophically with methanol as the sole substrate, leading to 6.5 higher rate of acetate production from CO_2. Furthermore, Marshall *et al.*[57] demonstrated the potential of mixed cultures to produce CH_4, acetate, and H_2 at a granular graphite cathode and high rate of acetate production (1330 g m^{-2} d^{-1}) from CO_2 was achieved at pH 6.7 by Jourdin *et al.*[58] using macroporous vitreous carbon cathode. Bajracharya *et al.*[54] compared CO_2 reduction at different electrode potentials using a mixed culture and a pure culture of *Clostridium ljungdahlii*. The reactor with a pure culture achieved higher production of acetate, CH_4 and H_2 at −1.1 V (*vs.* Ag/AgCl electrode). Batlle-Vilanova *et al.*[59] followed the same approach in their study on CO_2 reduction in a tubular bioelectrochemical system. The cathode was inoculated with mixed microbial culture taken from a syngas fermentation reactor dominated by *Clostridium* spp. Production of butyrate and acetate was studied under CO_2 limited conditions and high partial pressure of H_2, which favored butyrate production.

Another recent study[49] determined the optimal potential required to synthesize organic compounds from the CO_2 present in biogas by aerobic sludge in the cathode chamber. Several cathode potentials from −0.6 V to −1.0 V *vs.* a standard hydrogen electrode (SHE), were tested to evaluate their effect on the MES performance. It was observed that as the applied potential increased, the yields of acetate and butyrate also increased. Consequently, production of organic compounds from CO_2 was achieved with a low energy consumption of 9.15 W h at an applied potential of −0.7 V *vs.* SHE.

VFAs produced in a MES can be used for bioplastics production in the second production step, as proposed by Sciarria *et al.*[52] In the first stage of this work, acetate and butyrate were produced from CO_2 in a MES. Then the VFAs were concentrated and fed to a mixed microbial culture in the second bioreactor to produce PHBs. The MES was operated in batch mode and the cathode was initially inoculated with an enriched carboxydotrophic mixed microbial culture dominated by *Clostridium* spp. The CO_2 fixation efficiency was 73%. In the PHB production step, a maximum PHB concentration of 74% was obtained. The system-wide efficiency calculated in terms of carbon conversion was 0.41 kg of carbon in PHB per 1 kg of initial carbon as CO_2.

Interestingly, Srikanth *et al.*[60] studied the use of a biocathode for PHA production in a microbial fuel cell (MFC) under oxygen-limited conditions. In this work, both electrode compartments

Table 3 Comparative overview of different microbial cultures, cathode materials and products formed in MESs

Microbial culture	Cathode potential (*vs.* SHE)	Cathode	Products synthesized	Production rate (mM m^{-2} d^{-1})	References
Sporomusa ovata	−0.4	Graphite	Acetate, 2-oxobutyrate	0.2	55
S.ovata met-T18-2	−0.69	Graphite	Acetate	133.5 866.7	56
S. ovata	−0.4	Carbon cloth Chitosan treated carbon cloth Cyanuric chloride treated carbon cloth Nickel treated carbon cloth CNT–cotton treated carbon cloth	Acetate	30.0 229.0 205.0 136.0 102.0	81
Clostridium ljungdahlii Mixed culture	−0.69	Carbon felt	Acetate, ethanol, H_2 H2, acetate, CH_4	— —	54
Mixed culture	−0.8	Carbon cloth	Acetate Butyrate	34.7 87.5	59
Pond sediments and wastewater treatment plant sludge	−0.8	Nanoweb 3D RVC	Acetate	59	82
Mixed culture from septic tank	−1	Carbon felt	Acetate Butyrate Propionate	50.2 39.8 27.1	49

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(the anaerobic anode and aerobic cathode) were fed with a glucose-based solution resulting in current generation at the anode and heterotrophic accumulation of PHAs of up to 19% of dry cell weight at the cathode after 48 h of cultivation. PHA production required oxygen-limiting (micro-aerobic) conditions at the cathode, although low oxygen concentration also limited current generation. While this MFC setup required glucose supply to the anode to provide a source of electrons and featured low current density due to low oxygen concentration at the cathode, it can be hypothesized that PHA production can be achieved at a VFA-producing MES cathode provided with a limited supply of oxygen. Accordingly, such MES would be able to combine the steps of VFA and PHA production in the MES cathode, thus resulting in a single – step system for biopolymers production from CO_2 and electrons.

3. Cost comparison

Currently, most industrial scale bioplastics production is carried out using one-step production from well-defined carbon sources. The largest producers are Danimer Scientific (USA), producing PHA from canola oil (a production capacity of 17 000 t per year); Shenzhen Ecomann Biotechnology Co. Ltd (China), producing PHA from sugars (a production capacity of 5000 tons per year); TianAn Biological Materials Co. Ltd. (China) producing PHB from dextrose (a production capacity of 2000 t per year); Kaneka Corporation (Japan) producing PHB from plant oils, and several other companies.[7,61] The following review of techno-economic assessments (TEA) is aimed at comparing these technologies with newly emerging approaches for PHA production from alternative carbon sources described in the previous chapter. As explained below, to enable such comparison the calculation methods were unified and applied to the same production capacity.

3.1 Methodology

To compare the bioplastics production technologies reviewed in the previous chapters, the technologies were divided into three groups and then compared based on already published TEA studies. The following groups were considered. The first group of PHA-producing processes is single-stage reactors using well-defined carbon sources, which combines the growth of PHA accumulating microorganisms followed by the accumulation of PHA in the same bioreactor (Fig. 1A). The second group of processes includes PHA production from complex carbon sources such as wastewater or agricultural biomass, which requires a two-stage process, including the pre-treatment of the feedstock by an acidogenic fermentation followed by the growth of PHA producers and PHA accumulation in the second bioreactor (Fig. 1B). Finally, the third group considers PHA production from CO_2 using MES technology, typically in a two-stage process of CO_2 conversion to VFAs followed by the step of PHA production from the VFAs (Fig. 1C). Detailed description of the calculation methods for each process group is provided in ESI.† These calculations were derived from the published TEA studies reviewed in the following discussion.

3.1.1 PHA production from well-defined carbon sources. For single-stage PHA production from a well-defined carbon source, the comparative analysis was performed based on the methodology outlined by Leong *et al.*[62] PHA recovery using surfactants was assumed, as this was shown to be more economical and environmentally friendly. All PHA production calculations were performed for a production capacity of 9000 tons per year. The total operating time was assumed to be 330 days (7920 h) per year with a fermentation time of 42 hours and a turnaround time of 12 hours required for cleaning and refilling the reactor. To recalculate production capacities from different studies to a target capacity of 9000 tons per year, eqn (S19) and (S20) (ESI)† were used. These equations took into consideration the energy costs of the production plant and the production capacity. All calculated costs were adjusted to 2020 value for the U.S. dollar using the producer price index (PPI) for total manufacturing industries.[63]

3.1.2 PHA production from organic wastes. PHA production from organic wastes requires a two-stage process, in which the first stage is for hydrolysis of carbohydrates and fermentation of the hydrolysis products into carboxylic acids. In the second stage, conversion of these carboxylic acids into PHAs is achieved.[28,29] The second stage includes the growth of PHA-producing microorganisms, *e.g.*, by applying the feast/famine conditions, and PHA accumulation. TEA calculations for this process were based on a study of Fernandez-Dacosta.[12] The following conditions and assumptions were used for the TEA calculations. The production capacity of PHA from wastewater as a carbon source was set at 1500 tons per year, which was based on the availability of 6800 tons of COD (chemical oxygen demand) per year from organic wastes. The acidogenic fermentation of organic wastes to obtain carboxylic acids was assumed to have a yield of 0.91 g COD per (g COD) (initial COD = 26.3 g L^{-1}), a solids retention time (SRT) of 24 hours, and a conversion capacity of 50 kg COD per m^3 per day. The selection process was assumed to require a SRT of 1 day and a cycle time of 0.5 days resulting in an intracellular PHA content of 70%. At this stage, the enrichment of PHA-producing bacteria is also carried out with a biomass yield of 0.34 g biomass per (g COD). The maximum biomass concentration reported in the work of Fernandez-Dacosta[12] was 0.5 kg m^{-3} corresponding to a production rate of 0.0139 kg $(m^3\ h)^{-1}$. The PHA yield was calculated to be 0.44 g PHB per (g COD) and a total suspended solid (TSS) concentration of 2.7 kg TSS per m^3 was achieved. Similar to the single-stage process, the PHA-containing microorganisms at the end of the process will undergo downstream processing for biomass separation, and PHA extraction and purification.

3.1.3 PHA production from CO_2 in a MES. The cost of PHA production from CO_2 was evaluated for both a two-stage and a single-stage production process. As described above, the two-stage process consists of first the production of carboxylic acids (mainly acetate) from CO_2 in a MES followed by PHA production from the carboxylic acid in a bioreactor. In the single stage process these two steps occur in one reactor. The calculation of the production cost of acetate was made considering eqn (A9)

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(ESI).† Equipment cost analysis for MES included the cost of the electrodes. The calculation of operating costs for acetate production was carried out considering both fixed and variable costs. Continuous MES operation with biocatalyst (microorganisms) self-regeneration was assumed, therefore the cost of the biocatalyst (microbial inoculum) was considered to be a one-time expense. The conversion of acetate to PHA was analyzed following the methodology developed for the well-defined carbon source described above. More details can be found in ESI.†

3.2 PHA production from well-defined carbon sources

Table 4 summarizes analyses of PHA production costs in a single stage process using three different well-defined carbon sources: glycerol, glucose, and CH_4. All costs are provided in USD. As mentioned above, Leong *et al.*[62] performed a cost analysis of the production of PHB from glycerol using *C. necator H16* and the methodology developed in this study was used for cost analyses of PHA production from glucose and CH_4. Taking into consideration the volume of the proposed bioreactor and the time required to produce the bioplastics, an overall yield of 0.32 kg of PHB per kg of glycerol and a PHB production cost of $6.72 per kg were estimated. Here, the cost of the carbon source represented about 30% of the total operating costs. In another relevant study conducted by Choi and Lee,[64] the results of using different carbon sources and four bacterial strains were compared with respect to the production of PHBs. Overall, the lowest production costs were obtained when using a recombinant *Escherichia coli* culture and glucose as the carbon source. Two polymer recovery techniques were also evaluated. The authors concluded that the method of recovery using surfactant-hypochlorite digestion is the most cost-efficient. The analysis was conducted for an annual production capacity of 2850 ton with an overall yield of 0.29 kg of PHB per kg of glucose, resulting in a production cost of $6.14 per kg of PHB. When the production capacity was scaled up, the costs dropped to $5.11 per kg of PHB. Adjustment of this cost to 2020 values resulted in $7.87 per kg (Table 4).

Another TEA study was performed by Levett *et al.*[65] for PHB production from CH_4 at a scale of 100 000 tons per year. A culture of thermophilic methanotrophs and the acetone–water solvent extraction method for PHB purification were used. Using a carbon source considered to be a waste, costs related to raw material was reduced to 20% of annual operating costs, resulted in a cost of $4.32 per (kg PHB) produced. When the production capacity was adjusted to 9000 ton per (year), the production cost was estimated to be $7.92 per (kg PHB) (Table 4).

A comparison of these three studies shows that when using well defined carbon sources, the overall yield of PHA production is in a range of 0.3–0.5 kg per kilogram of substrate. The key parameters that determine the size of equipment needed and therefore impact the capital and operating costs are the productivity and the fermentation time. All three cases had similar operating costs, which resulted in PHB production costs of $6.7–7.9 per kg.

3.3 PHA production from complex carbon sources

Production of bioplastics using complex carbon sources, such as organic wastes, requires a two-stage process. Table 5 lists three TEA studies on the production of PHAs using different carbon sources. In the first study, Fernandez-Dacosta *et al.*[12] analyzed techno-economic and environmental aspects of PHB production from wastewater. Three recovery methods were also evaluated in this study: two of them were based on chemical treatment with surfactants combined with either alkali or hypochlorite and the third one was based on solvent extraction combined with dichloromethane. The data reported for the purification method using surfactant and sodium hypochlorite digestion was used as the basis for comparison of the three

Table 4 Single step PHB production from well-defined carbon sources. Costs are adjusted to 2020 US dollars using the producer price index for total manufacturing industries

Parameters	Study		
Reference	Leong *et al.*, 2017 (ref. 62)	Choi and Lee, 1997 (ref. 64)	Levett *et al.*, 2016 (ref. 65)
PHB recovery method	Surfactant and sodium hypochlorite digestion	Surfactant and sodium hypochlorite digestion	Acetone–water solvent
Pure strain	*C. necator H16*	Recombinant *E. coli*	Thermophilic methanotrophs
Carbon source	Glycerol	Glucose	Methane
Productivity, g $(L\ h)^{-1}$	4.00	2.18	2.70
Global yield, kg PHB per (kg substrate)	0.32	0.29	0.54
Fed-batch fermentation time, h	42	39	24
Volume per run, m^3	305.30	584.46	434.54
Target PHB production, ton per (year)	9000.00	9000.00	9000.00
Carbon source cost, $ per (kg)	$0.53	$0.77	$0.26
Total carbon source, ton per (year)	27 982.48	10 758.62	18 246
Total direct fixed capital, $	$178 925 342	$103 699 386	$61 614 960
Total annual operating cost, $	$60 488 465	$70 792 016	$71 267 434
PHB production cost, $ per (kg)	**$6.72**	**$7.87**	**$7.92**

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Table 5 PHB production in a two-step process using complex carbon sources

	Mixed microbial culture using municipal wastes		Pure culture using CO and H_2
Parameters	Fernández-Dacosta *et al.*, 2015 (ref. 12)	Mudliar *et al.*, 2008 (ref. 66)	Choi *et al.*, 2010 (ref. 68)
PHB recovery method	Surfactant and sodium hypochlorite digestion	Alkali-surfactant	Surfactant and sodium hypochlorite digestion
Culture	Mixed microbial culture	Activated sludge	*Rhodospirillum rubrum*
Carbon source	Wastewater paper mill or food industry	Wastewater	Switchgrass biomass
Product formed in step 1	VFA	VFA	Syngas (CO and H_2)
Global yield	2.20 kg PHB per m^3	1.42 kg PHB per m^3	0.17 kg PHB per (kg switchgrass)
Target PHB production, ton per year	9000.00	9000.00	9000.00
Total feedstock	4 080 684.92 m^3	6 347 732.09 m^3	518 522.88 ton
Feedstock per run	43 411.54 m^3	77 411.37 m^3	1576.05 ton
Cost carbon source	—	—	$20.58
Total production step 1, ton per run	1038.97	1852.69	3764.88
Total direct fixed capital, US $	—	$43 833 472.13	$119 718 664.20
Credits obtained	Wastewater treatment credits	Wastewater treatment credits	Hydrogen production and sale credits
Operating and maintenance cost – no credits, US $	$15 909 022.98	$39 422 792.79	$84 147 003.25
Operating and maintenance cost – with credits, US $	$11 597 231.71	—	$2 234 292.95
PHB production cost – without credits, US $ per kg	**$1.77**	**$4.38**	**$9.35**
PHB production cost – with credits, US $ per kg	**$1.29**	**—**	**$0.25**

methods. The authors based their evaluation on the production of 1500 ton PHB per year by a mixed microbial culture using wastewater from either paper mills or the food industry. The overall yield obtained in this analysis was 2.2 kg of PHB per m^3 of wastewater. The first stage of the production process was acidogenic fermentation to obtain VFAs from the wastewater, followed by PHB production from the VFAs using a feast/famine regime in the second reactor. The costs associated with wastewater treatment were considered as a credit to offset the costs of bioplastics production. Final costs were calculated to be $1.29 US per (kg PHB) when considering these credits, and $1.77 US per (kg PHB) without the credits. It must be mentioned that such low costs were due to relatively high PHB production rates and low estimations for operating costs as compared to costs reported in Table 4. Table 5 also shows costs for the three methods normalized for an annual PHB production of 9000 tons.

The study conducted by Mudliar *et al.*[66] considered a two-stage PHB production process from organic wastes with a processing capacity of 100 m^3 d^{-1} and a PHB production capacity of 46.20 ton per year. Since not all values required for TEA calculations were provided by the authors, some values were assumed from the publication of Fernandez-Dacosta *et al.*[12] A PHB production cost of $11.8 per kg was estimated following the methodology outlined in ESI† based on a PHB yield of 44%. The work suggests that by increasing this PHB yield to 70% (based on the results published by Tamis *et al.*[67] with this type of carbon source), the costs would be reduced to $5.38 per kg. When normalized to a production volume of 9000 tons per year, the estimated PHB production cost was $4.38 per kg (Table 5). These results suggest that the production of bioplastics using wastewater as substrate could be feasible, although a number of practical hurdles, such as the planning and construction of a production plant with a very high-volume requirement per batch, around 77 500 m^3, remain to be solved.

Cost estimations were also obtained for PHA production using agricultural waste biomass as a carbon source. Choi *et al.*[68] conducted a TEA to investigate the feasibility of simultaneous production of H_2 and PHBs using switchgrass as feedstock in a two-stage process biorefinery. First, the biomass was converted by thermochemical methods into syngas. Then, the syngas was fermented by a *Rhodospirillum rubrum* culture to produce PHBs and H_2. A total daily biorefinery production capacity of 12 tons PHB and 50 tons of H_2 was considered for the TEA. The H_2 production resulted in a credit of $2 per (kg H_2). The cost of PHB production was estimated to be $9.35 per kg. This cost substantially decreased to $0.25 per kg when H_2 production credit was taken into account. Overall, when comparing the estimated costs for PHA production from well-defined and complex carbon sources it is important to note that the production costs could be similar, but these estimations are not yet supported by existing large scale production systems.

3.4 PHA production in a MES

As discussed previously, the production of biopolymers from CO_2 in a MES can be achieved in a two-stage process in which VFAs (mainly acetate) are produced in the MES and then used in the second stage in a conventional bioreactor to produce PHA (Fig. 1C). A single step PHA production can be also considered,

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where CO_2 reduction to VFAs and PHA production are accomplished by a mixed-culture at the MES cathode.[16] CO_2 conversion in a MES is a biological method of carbon sequestration, therefore following other TEA studies listed above, a CO_2 removal credit could be applied in the calculations. International regulatory initiatives have stated that carbon credits should be considered in the range of \$40–80 per (ton CO_2) by 2020 and \$50–100 per (ton CO_2) by 2030 to meet the temperature reduction targets of the Paris Agreement.[69] Therefore, a value of \$60 per (ton CO_2) was used in the following calculations.

Table 6 summarizes the studies of three scenarios for biopolymer (PHB) production in a MES. The first two scenarios consider the two-stage process and assumes PHB production rates adapted from studies, which used either glycerol[62] or acetate[19] as carbon sources. The third scenario considers PHB production in a single stage by heterotrophic–autotrophic fermentation using CO_2 as a carbon source. This scenario was evaluated based on the data reported by Garcia-Gonzalez *et al.*[16]

For Scenarios 1 and 2, the first stage in the production process was evaluated based on the TEA analysis carried out by Christodoulou and Velasquez-Orta[70] for the conversion of CO_2 to acetate. This TEA study modeled a MES plant with a capacity of producing 100 ton acetate per year based on a production rate of 11.4 kg h^{-1} and a global yield of 0.68 kg of acetate produced per kg of CO_2. The cost of acetate production was estimated to be \$1.88 per kg. The second stage corresponds to the production of PHB from acetate. Considering a broad range of reported PHB production rates on well-defined carbon sources such as glycerol and acetate, the rate of PHB production from glycerol provided by Leong *et al.*[62] (Table 4) was used for this calculation. In this scenario, 27 983 tons of acetate per year are required to achieve the target annual production of 100 ton of PHB. The calculations reported by Christodoulou and Velasquez-Orta[70] were scaled to match this production capacity and a production cost of \$0.67 per kg of acetate was estimated. By applying the CO_2 conversion credits, this cost was further reduced to \$0.57 per (kg). The second step was the fermentation using a pure culture of *C. necator* for the production of PHB from the acetate

Table 6 Comparison of PHB production from CO_2 in a two stage and single stage MES-based bioprocess

PHB production from CO_2	Scenario 1 (2 step)	Scenario 2 (2 step)	Scenario 3 (single step)
Step 1:	Production of acetic acid from CO_2 by MES Christodoulou 2016 (ref. 70)		Production of PHB from acetic acid using pure strains Garcia-Gonzalez & De Wever, 2017 (ref. 16)
Acetic acid production, ton per (year)	27 983.00	41 926 091.83	—
Yield, kg acetic acid per (kg CO_2)	0.68	0.68	0.47
Productivity, g $(L\ h)^{-1}$	4.0	0.41	0.23
Amount of CO_2 required, ton per (year)	50 887.43	61 484 844.02	50 887.43
Total direct fixed capital – without credits, US \$	\$22 874 424.99	\$7 940 032 364.45	N/A
Total annual operating cost – without credits, US \$	\$18 700 165.51	\$6 491 088 605.29	N/A
Acetic acid production cost – without credits, US \$ per (kg)	**\$0.67**	**\$0.15**	**N/A**
Acetic acid production cost – with CO_2 credits, US \$ per (kg)	**\$0.57**	**\$0.06**	**N/A**
Step 2: production of PHB from acetic acid using pure strains	Leong *et al.*, 2017 (ref. 62)	Leong *et al.*, 2017 (ref. 62); Garcia-Gonzalez 2018 (ref. 19)	**N/A**
PHB recovery method	Surfactant and sodium hypochlorite digestion	Surfactant and sodium hypochlorite digestion	Surfactant and sodium hypochlorite digestion
Culture	*C. necator H16*	*C. necator H16*	*C. necator, DSM 545*
Carbon source	Acetate	Acetate	CO_2
Global yield, kg PHB per (kg substrate)	0.32	0.21	0.47
Target PHB production, ton per (year)	9000.00	9000.00	9000.00
Volume per run, m^3	321.44	3121.72	5649.76
Total carbon source quantity, ton per (year)	27 982 481.05	41 926 091.83	20 963 045.91
Total direct fixed capital – without credits, US \$	\$178 913 631.74	\$1 674 522 989.85	\$3 144 685 727.92
Total annual operating cost – without credits, US \$	\$65 161 199.25	\$376 126 961.87	\$706 277 586.13
PHB total production cost – without credits, US \$ per kg	**\$7.24**	**\$41.79**	**\$6.26–\$78.48**
PHB total production cost – with CO_2 credits, US \$ per kg	**\$6.82**	**\$41.18**	**\$5.71–\$78.37**

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with an overall assumed yield of 0.21 kg PHB per (kg acetate) and a productivity of 4.0 g (L h)$^{-1}$. Consequently, PHB production costs of \$7.24 per kg without considering CO_2 credits and \$6.82 per kg with such credits were calculated.

Scenario 2 assumed the same two-stage technology, but with a much lower rate of PHB production. This assumption was based on the results reported in the study of Garcia-Gonzalez *et al.*,[19] which estimated an overall yield of 0.21 kg of PHB per kg of acetate and an estimated productivity of 0.4 g (L h)$^{-1}$. Consequently, the required amount of acetate was significantly higher. Annually, 41 926 092 tons of acetate would be needed to achieve the target production of 100 tons PHB. The estimated cost of acetate production was lower at \$0.15 per kg due to the increased production volume. The production cost decreased to \$0.06 per kg when CO_2 credits were considered. Nevertheless, the overall cost of PHB production was substantially higher due to the low rate of PHB production reported by Garcia-Gonzalez *et al.*[19] Accordingly, PHB production cost of \$41.18 per kg and \$41.79 per kg with and without CO_2 credits, respectively were estimated. The lack of experimental results corresponding to PHA production from VFAs in a MES leads to a broad range of cost estimations for the two scenarios in Table 6.

From these calculations, it can be seen that the productivity and global yield are the two parameters significantly affecting PHA production costs in Scenarios 1 and 2. Low productivity and yield lead to large bioreactor volumes, about 10 times higher in Scenario 2 as compared to Scenario 1. Because it is possible to produce a range of carboxylic acids and CH_4 (ref. 71) from CO_2 through MES technology, significant productivity improvements might be expected. Indeed, recent advances in optimizing operating conditions and developing new cathode materials resulted in CH_4 and VFA production rates that were significantly greater than previously reported.[58,72,73] Furthermore, a broader range of carboxylic acids can be produced by changing operating conditions,[74] which would potentially result in improved productivity and yields during the PHA production phase. Clearly, more experimental work is needed to optimize both phases of this novel CO_2 conversion process.

Calculations for hypothetical single step Scenario 3 to estimate costs used results obtained by Garcia-Gonzalez *et al.*[16] In this study, heterotrophic biomass growth on glucose was followed by autotrophic production of biopolymers in the same reactor using a gas mixture of H_2, O_2, and CO_2 in a ratio of 84 : 2.8 : 13.2, respectively. A productivity of 0.227 g PHB per (L h) and an overall yield of 0.47 g PHB per g CO_2 were reported. Production of PHBs using this methodology leads to a mitigation potential of 1.58 ton CO_2 per (ton PHB). The productivity of 4 g PHB per (L h) used by Leong *et al.*, 2017 (ref. 62) was used for the calculation of expected costs. Also, it was assumed that PHB production rather than CO_2 conversion to VFAs is the rate-limiting step of these biotransformations. The resulting calculations for Scenario 3 are given in Table 6. A broad range of production costs from \$5.7 to \$78.4 per kg was obtained, depending on the value used for productivity. Once again, such broad range is attributed to a lack of experimental results and is expected to be narrowed with the emergence of new experimental studies. Further discussion of the productivity impact on PHA production costs is provided below.

It should be emphasized that while the estimations presented in Tables 4 and 5 are based on published experimental results and TEA calculations, calculations for Scenario 3 in Table 6 represent a hypothetical process and only provide preliminary estimations. Once experimental results are available, a thorough TEA should be carried out to update cost estimations.

3.5 Cost comparison

As can be seen from the estimations presented above, the costs of PHA production are strongly dependent on the rate of PHA productivity and the PHA yield. The effects of these parameters on the PHA production costs are illustrated in Fig. 2. The PHA costs shown in Fig. 2 were calculated for a single-stage process with a production capacity of 9000 tons per year and assuming a carbon source cost of \$0.5 per kg, which would be similar to the cost of well-defined carbon sources such as glucose, sucrose, and acetate.

Fig. 2A suggests that a cost-efficient PHA production requires a productivity of 0.8–1.0 g (L h)$^{-1}$ or higher. Lower productivity values result in a steep increase in PHA production costs, especially at PHA productivities below 0.3 g (L h)$^{-1}$. PHA yield is another significant factor (Fig. 2B). To obtain competitive costs of PHA production, yields higher than 0.2 g (g)$^{-1}$ are required. To achieve PHA production costs below the \$10 per kg, productivity and yields of greater than 2 g (L h)$^{-1}$ and 0.4 g g^{-1}, respectively, are required.

An important factor that significantly affects the productivity is the type of carbon source used. Fig. 3 shows the impact of different liquid and gaseous carbon sources on the productivity. The information presented in this figure is from the studies cited in Tables 4 and 5. For liquid carbon sources such as glucose, several studies reported similar productivities and global yields, *i.e.*, 2.2 g (L h)$^{-1}$ and 0.2 kg PHA per (kg substrate), respectively.[64,75,76] With sucrose as a carbon source, experimental results from different studies gave an average productivity of 2.7 g (L h)$^{-1}$.[21–23] With acetate as a carbon source, a lower productivity of 0.4 (L h)$^{-1}$ was obtained based on the results of Garcia-Gonzalez *et al.*[19] The heterotrophic/autotrophic fermentation, which uses a liquid carbon source for growth and then CO_2 for PHA production is also included in this comparison. Two studies report similar productivities when using glucose (0.23 g (L h)$^{-1}$)[77] or fructose (0.19 g (L h)$^{-1}$)[78] as a carbon source for growth, while a recent study reported a higher productivity of 0.87 g (L h^{-1}) when using glucose and a gas mixture.[79] This comparison suggests that to decrease the cost of PHA production from gaseous carbon sources, such as CO_2, formation of intermediates other than acetate is desirable, as it might lead to increased rate of PHA formation.

4. Conclusion

This review provided an overview of experimental and TEA studies of bioplastics production from various carbon sources.

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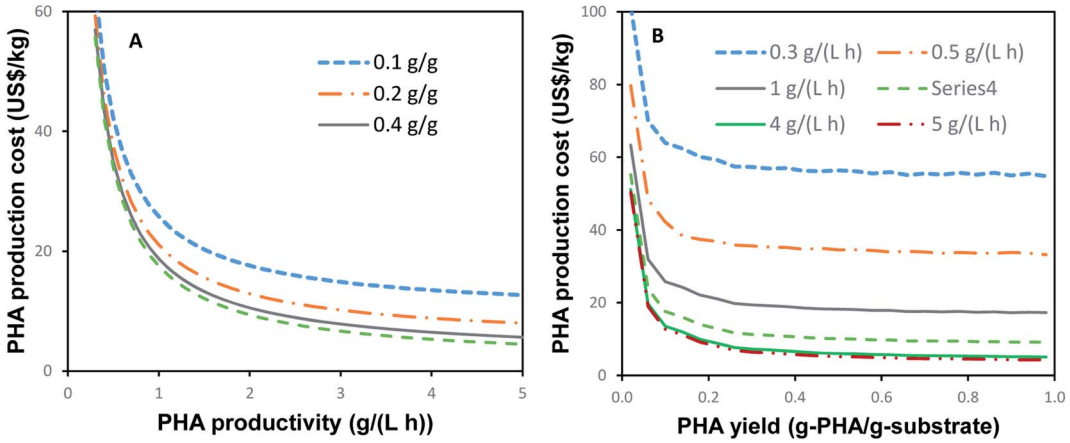

Fig. 2 (A) Effect of productivity at different values of PHA yield on PHA production costs and (B) effect of PHA yield at different productivity values on the cost of PHA production. A production capacity of 9000 tons per year, an overall yield of 0.24 g PHA per (g substrate), and an overall cost of the carbon source of $0.5 per kg were assumed.

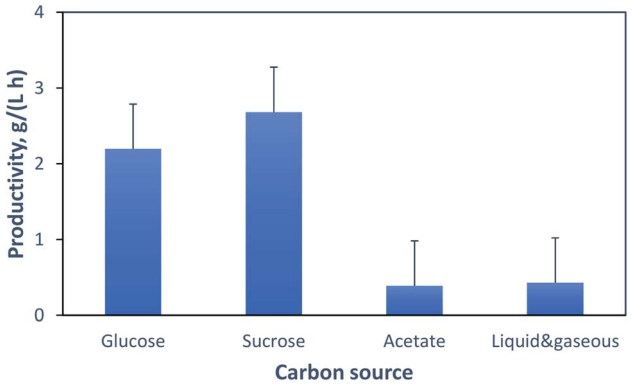

Fig. 3 Dependence of productivity on the carbon source used for PHA production. In heterotrophic–autotrophic fermentations either fructose or glucose was used for biomass growth, while a gas mixture consisting of CO_2, H_2 and O_2 was used for PHA production.

Such comparison of results in available literature showed that the technical and economic feasibility of bioplastics production depends on multiple factors including carbon source, process design, operating conditions, *etc.* However, the most prominent factors affecting production costs are PHA yield, productivity, and the type of carbon source selected for PHA production. In fact, carbon source selection appears to be the most important, as it affects both the productivity and the overall PHA yield per unit of carbon source consumed.

By comparing different TEA studies, it was shown that by using well-established carbon sources, such as glucose and glycerol and assuming a production capacity of 9000 tons per year, PHA production costs fall within the range of $6.9–$7.5 per kg. Also, several TEA studies suggest that when using more complex carbon sources such as wastewater or agricultural biomass, in which an additional carbon source preparation step is necessary, the unit cost of PHA production can range from $5.2 to $11.0 per kg. This range of costs is similar to that obtained when using well-established carbon sources, since feedstock costs are considered to be zero, and credits can be applied to reduce production costs.

Interestingly, our estimation of production costs using the emerging approach of bioplastics production through microbial electrosynthesis from CO_2 suggests that this approach could provide a feasible alternative to traditional carbon sources, such as glucose. Although a broad range of production costs was obtained ($5.71–$78) due to the uncertainties of this novel process, future studies are expected to result in significant improvements in the observed process yield and productivity. A detailed TEA study is needed to evaluate the feasibility of direct CO_2 conversion to bioplastics.

Conflicts of interest

There are no conflicts to declare.

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References

1 J. M. G. Alcantara, F. Distante, G. Storti, D. Moscatelli, M. Morbidelli and M. Sponchioni, *Biotechnol. Adv.*, 2020, **42**, 107582.
2 G. Y. A. Tan, C. L. Chen, L. Li, L. Ge, L. Wang, I. M. N. Razaad, Y. Li, L. Zhao, Y. Mo and J. Y. Wang, *Polymers*, 2014, **6**, 706–754.
3 R. Rai, T. Keshavarz, J. A. Roether, A. R. Boccaccini and I. Roy, *Mater. Sci. Eng., R*, 2011, **72**, 29–47.
4 G. de Roo, M. B. Kellerhals, Q. Ren, B. Witholt and B. Kessler, *Biotechnol. Bioeng.*, 2002, **77**, 717–722.
5 P. Technology, *Prices Up for All Commodity Resins*, **https://www.ptonline.com/blog/post/prices-up-for-all-commodity-resins**, accessed, 5 October 2020.

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6 L. R. Castilho, D. A. Mitchell and D. M. Freire, *Bioresour. Technol.*, 2009, **100**, 5996–6009.
7 *Polyhydroxyalkanoate (PHA) Market by Type (Short Chain Length, Medium Chain Length), Production Method (Sugar Fermentation, Vegetable Oil Fermentation, Methane Fermentation), Application, and Region – Global Forecast to 2024*, **https://www.marketsandmarkets.com/Market-Reports/pha-market-395.html**, accessed, 06 October 2020.
8 C. S. K. Reddy, R. Ghai, Rashmi and V. C. Kalia, *Bioresour. Technol.*, 2003, **87**, 137–146.
9 C. S. S. Oliveira, C. E. Silva, G. Carvalho and M. A. Reis, *New Biotechnol.*, 2017, **37**, 69–79.
10 B. B. Salgaonkar and J. M. Braganca, *Bioengineering*, 2017, **4**, 50.
11 P. Chakraborty, W. Gibbons and K. Muthukumarappan, *J. Appl. Microbiol.*, 2009, **106**, 1996–2005.
12 C. Fernández-Dacosta, J. A. Posada, R. Kleerebezem, M. C. Cuellar and A. Ramirez, *Bioresour. Technol.*, 2015, **185**, 368–377.
13 B. Johnston, I. Radecka, D. Hill, E. Chiellini, V. I. Ilieva, W. Sikorska, M. Musiol, M. Zieba, A. A. Marek, D. Keddie, B. Mendrek, S. Darbar, G. Adamus and M. Kowalczuk, *Polymers*, 2018, **10**(9), 957.
14 J. M. Naranjo, J. A. Posada, J. C. Higuita and C. A. Cardona, *Bioresour. Technol.*, 2013, **133**, 38–44.
15 J. A. Posada, J. M. Naranjo, J. A. Lopez, J. C. Higuita and C. A. Cardona, *Process Biochem.*, 2011, **46**, 310–317.
16 L. Garcia-Gonzalez and H. De Wever, *FEMS Microbiol. Lett.*, 2017, **364**, fnx196.
17 L. J. Vandi, C. M. Chan, A. Werker, D. Richardson, B. Laycock and S. Pratt, *Polymers*, 2018, **10**(7), 751.
18 A. Atlić, M. Koller, D. Scherzer, C. Kutschera, E. Grillo-Fernandes, P. Horvat, E. Chiellini and G. Braunegg, *Appl. Microbiol. Biotechnol.*, 2011, **91**, 295–304.
19 L. Garcia-Gonzalez and H. De Wever, *Appl. Sci.*, 2018, **8**(9), 1416.
20 S. P. Valappil, S. K. Misra, A. R. Boccaccini, I. Keshavarz, C. Bucke and I. Roy, *J. Biotechnol.*, 2007, **132**, 251–258.
21 G. Gahlawat and A. K. Srivastava, *Appl. Biochem. Biotechnol.*, 2017, **183**, 530–542.
22 T. Yamane, M. Fukunaga and Y. W. Lee, *Biotechnol. Bioeng.*, 1996, **50**, 197–202.
23 F. L. Wang and S. Y. Lee, *Appl. Environ. Microbiol.*, 1997, **63**, 3703–3706.
24 R. Shimizu, K. Chou, I. Orita, Y. Suzuki, S. Nakamura and T. Fukui, *BMC Microbiol.*, 2013, **13**, 169.
25 L. S. Serafim, P. C. Lemos, M. G. E. Albuquerque and M. A. M. Reis, *Appl. Microbiol. Biotechnol.*, 2008, **81**, 615–628.
26 H. Pereira, P. C. Lemos, M. A. Reis, J. P. Crespo, M. J. Carrondo and H. Santos, *Water Res.*, 1996, **30**, 2128–2138.
27 M. G. E. Albuquerque, M. Eiroa, C. Torres, B. R. Nunes and M. A. M. Reis, *J. Biotechnol.*, 2007, **130**, 411–421.
28 D. Dionisi, M. Beccari, S. Di Gregorio, M. Majone, M. P. Papini and G. Vallini, *J. Chem. Technol. Biotechnol.*, 2005, **80**, 1306–1318.
29 F. Carta, J. J. Beun, M. C. M. Van Loosdrecht and J. J. Heijnen, *Water Res.*, 2001, **35**, 2693–2701.
30 J. Choi and S. Y. Lee, *Appl. Microbiol. Biotechnol.*, 1999, **51**, 13–21.
31 R. Sirohi, J. P. Pandey, V. K. Gaur, E. Gnansounou and R. Sindhu, *Bioresour. Technol.*, 2020, 123536.
32 Y. Dai, Z. G. Yuan, K. Jack and J. Keller, *J. Biotechnol.*, 2007, **129**, 489–497.
33 S. Bengtsson, A. Werker and T. Welander, *Water Sci. Technol.*, 2008, **58**, 323–330.
34 E. R. Coats, F. J. Loge, M. P. Wolcott, K. Englund and A. G. McDonald, *Water Environ. Res.*, 2007, **79**, 2396–2403.
35 A. F. Duque, C. S. S. Oliveira, I. T. D. Carmo, A. R. Gouveia, F. Pardelha, A. M. Ramos and M. A. M. Reis, *New Biotechnol.*, 2014, **31**, 276–288.
36 M. Rodgers and G. Wu, *Bioresour. Technol.*, 2010, **101**, 1049–1053.
37 U. S. E. P. A. EPA, *Greenhouse Gas Emissions*, **https://www.epa.gov/ghgemissions/overview-greenhouse-gases**, accessed, 01 November 2020, 2018.
38 Y. Miyahara, M. Yamamoto, R. Thorbecke, S. Mizuno and T. Tsuge, *Biotechnol. Lett.*, 2020, **42**, 1655–1662.
39 I. Park, E. H. Jho and K. Nam, *J. Polym. Environ.*, 2014, **22**, 244–251.
40 K. D. Wendlandt, U. Stottmeister, J. Helm, B. Soltmann, M. Jechorek and M. Beck, *Eng. Life Sci.*, 2010, **10**, 87–102.
41 R. Perez, J. Casal, R. Munoz and R. Lebrero, *Sci. Total Environ.*, 2019, **688**, 684–690.
42 P. Luangthongkam, P. J. Strong, S. N. S. Mahamud, P. Evans, P. Jensen, G. Tyson, B. Laycock, P. A. Lant and S. Pratt, *Bioresour. Bioprocess.*, 2019, **6**, 50.
43 J. Myung, W. M. Galega, J. D. Van Nostrand, T. Yuan, J. Z. Zhou and C. S. Criddle, *Bioresour. Technol.*, 2015, **198**, 811–818.
44 B. Bian, S. Bajracharya, J. Xu, D. Pant and P. E. Saikaly, *Bioresour. Technol.*, 2020, **302**, 122863.
45 S. Das, S. Das, I. Das and M. M. Ghangrekar, *Mater. Sci. Energy Technol.*, 2019, **2**, 687–696.
46 P. Dessi, L. Rovira-Alsina, C. Sanchez, G. K. Dinesh, W. Tong, P. Chatterjee, M. Tedesco, P. Farras, H. M. V. Hamelers and S. Puig, *Biotechnol. Adv.*, 2021, **46**, 107675.
47 T. Hua, S. Li, F. Li, Q. Zhou and B. S. Ondon, *J. Chem. Technol. Biotechnol.*, 2019, **94**, 1697–1711.
48 A. Prevoteau, J. M. Carvajal-Arroyo, R. Ganique and K. Rabaey, *Curr. Opin. Biotechnol.*, 2020, **62**, 48–57.
49 S. Das, I. Das and M. M. Ghangrekar, *J. Environ. Chem. Eng.*, 2020, **8**(4), 104028.
50 S. A. Patil, S. Gildemyn, D. Pant, K. Zengler, B. E. Logan and K. Rabaey, *Biotechnol. Adv.*, 2015, **33**, 736–744.
51 S. Das, L. Diels, D. Pant, S. A. Patil and M. M. Ghangrekar, *J. Electrochem. Soc.*, 2020, **167**(15), 155510.
52 T. P. Sciarria, P. Batlle-Vilanova, B. Colombo, B. Scaglia, M. D. Balaguer, J. Colprim, S. Puig and F. Adani, *Green Chem.*, 2018, **20**, 4058–4066.
53 K. Rabaey and R. A. Rozendal, *Nat. Rev. Microbiol.*, 2010, **8**, 706–716.

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54 S. Bajracharya, A. ter Heijne, X. D. Benetton, K. Vanbroekhoven, C. J. N. Buisman, D. Strik and D. Pant, *Bioresour. Technol.*, 2015, **195**, 14–24.
55 K. P. Nevin, T. L. Woodard, A. E. Franks, Z. M. Summers and D. R. Lovley, *Mbio*, 2010, **1**(2), e00103-10.
56 P. L. Tremblay, D. Hoglund, A. Koza, I. Bonde and T. Zhang, *Sci. Rep.*, 2015, **5**(1), 1–11.
57 C. W. Marshall, D. E. Ross, E. B. Fichot, R. S. Norman and H. D. May, *Appl. Environ. Microbiol.*, 2012, **78**, 8412–8420.
58 L. Jourdin, S. Freguia, V. Flexer and J. Keller, *Environ. Sci. Technol*, 2014, **50**, 1982–1989.
59 P. Batlle-Vilanova, R. Ganigue, S. Ramio-Pujol, L. Baneras, G. Jimenez, M. Hidalgo, M. Dolors Balaguer, J. Colprim and S. Puig, *Bioelectrochemistry*, 2017, **117**, 57–64.
60 S. Srikanth, M. V. Reddy and S. V. Mohan, *Bioresour. Technol.*, 2012, **125**, 291–299.
61 C. Kourmentza, J. Placido, N. Venetsaneas, A. Burniol-Figols, C. Varrone, H. N. Gavala and M. A. M. Reis, *Bioengineering*, 2017, **4**, 55.
62 Y. K. Leong, P. L. Show, J. C. W. Lan, H. S. Loh, H. L. Lam and T. C. Ling, *Clean Technol. Environ. Policy*, 2017, **19**, 1941–1953.
63 U. S. B. o. L. Statistics, *PPI industry group data for Total manufacturing industries, not seasonally adjusted*, **https://data.bls.gov/cgi-bin/surveymost**, accessed, 12 March 2021.
64 J. I. Choi and S. Y. Lee, *Bioprocess Eng.*, 1997, **17**, 335–342.
65 I. Levett, G. Birkett, N. Davies, A. Bell, A. Langford, B. Laycock, P. Lant and S. Pratt, *J. Environ. Chem. Eng.*, 2016, **4**, 3724–3733.
66 S. Mudliar, A. Vaidya, M. S. Kumar, S. Dahikar and T. Chakrabarti, *Clean Technol. Environ. Policy*, 2008, **10**, 255.
67 J. Tamis, K. Luzkov, Y. Jiang, M. C. M. van Loosdrecht and R. Kleerebezem, *J. Biotechnol.*, 2014, **192**, 161–169.
68 D. Choi, D. C. Chipman, S. C. Bents and R. C. Brown, *Appl. Biochem. Biotechnol.*, 2010, **160**, 1032–1046.
69 J. E. Stiglitz, N. Stern, M. Duan, O. Edenhofer, G. Giraud, G. M. Heal, E. L. La Rovere, A. Morris, E. Moyer and M. Pangestu, *Report of the High-Level Commission on Carbon Prices*, International Bank for Reconstruction and Development and International Development Association/The World Bank, 2017.
70 X. Christodoulou and S. B. Velasquez-Orta, *Environ. Sci. Technol.*, 2016, **50**, 11234–11242.
71 P. Dessì, L. Rovira-Alsina, C. Sánchez, G. K. Dinesh, W. Tong, P. Chatterjee, M. Tedesco, P. Farràs, H. M. Hamelers and S. Puig, *Biotechnol. Adv.*, 2020, 107675.
72 F. Geppert, D. Liu, E. Weidner and A. ter Heijne, *Int. J. Hydrog. Energy*, 2019, **44**, 21464–21469.
73 D. Liu, M. Roca-Puigros, F. Geppert, L. Caizan-Juanarena, S. P. Ayudthaya, C. Buisman and A. ter Heijne, *Front. Bioeng. Biotechnol.*, 2018, **6**, 78.
74 P. Dessi, C. ánchez, S. Mills, F. Giuseppe Cocco, M. Isipato, U. Z. Ijaz, G. Collins and P. N. L. Lens, *Bioelectrochemistry*, 2021, **137**, 107686.
75 A. Atlić, M. Koller, D. Scherzer, C. Kutschera, E. Grillo-Fernandes, P. Horvat, E. Chiellini and G. Braunegg, *Appl. Microbiol. Biotechnol.*, 2011, **91**, 295–304.
76 B. S. Kim, S. C. Lee, S. Y. Lee, H. N. Chang, Y. K. Chang and S. I. Woo, *Biotechnol. Bioeng.*, 1994, **43**, 892–898.
77 L. Garcia-Gonzalez and H. De Wever, *FEMS Microbiol. Lett.*, 2017, **364**, 20.
78 T. G. Volova and G. S. Kalacheva, *Microbiology*, 2005, **74**, 54–59.
79 S. Ghysels, M. S. I. Mozumder, H. De Wever, E. I. P. Volcke and L. Garcia-Gonzalez, *Bioresour. Technol.*, 2018, **249**, 858–868.
80 C. Chan, A. Guisasola and J. A. Baeza, *J. Clean. Prod.*, 2020, **242**, 118495.
81 T. Zhang, H. R. Nie, T. S. Bain, H. Y. Lu, M. M. Cui, O. L. Snoeyenbos-West, A. E. Franks, K. P. Nevin, T. P. Russell and D. R. Lovley, *Energy Environ. Sci.*, 2013, **6**, 217–224.
82 L. Jourdin, S. Freguia, B. C. Donose, J. Chen, G. G. Wallace, J. Keller and V. Flexer, *J. Mater. Chem. A*, 2014, **2**, 13093–13102.

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