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Production of Value-Added products and Commodities by Electrofermentation and its Integration to Biorefineries

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ABSTRACT. Microbial electrosynthesis cells (*MESynC*) are a new technology that requires biocatalysts and electric power for enhanced production of value-added products (*VAPs*). This process can be coupled with biorefineries. *MESynC* has received great attention in the last years as an alternative to chemical synthesis or conventional fermentation. Yet, the information is still diverse. Thus, the goal of this work was to critically review recent efforts on *MESynC.* The objective of this work is to critically review recent efforts on *MESynC*. The scope of this review includes the following issues: (*i*) Principles of *MESynC* for chemicals´ production; (*ii*) *VAPs* and other products; (*iii*) energy efficiency and mass/energy balances; (*iv*) system analysis of *MESynC*; (*v*) *MESynC* and biorefineries; and *(vi)* Perspectives and conclusion.

*MESynC*s need to overcome large barriers as economic costs, electrode materials, membranes, microorganisms, etc. There are few published works on producing chemical products from microorganisms powered with electricity, considerable further work will be necessary to complete understanding of bacterial-electrode electron flow at a molecular level for a better development of *MESynC*. On the other hand, there is more information on the bioelectrochemical production of methane and hydrogen, although these compounds are more commodities than special *VAPs*.

There are few studies regarding the systemic evaluation of the *MESynC* performance that determine energy efficiency as well as environmental impacts. Life cycle analysis (*LCA*) is an accepted method for determining these aspects. Though in the last years have been published some studies, definitive incorporation of *LCA* and energy balances to *MESynC R&D* is still missing. These gaps, along with that of scarce experience with scale up of the processes, should be filled up in order to demonstrate the potential commercialization of *MESynC*. Finally, this work gives an overview of the advantages and advances of coupling of *MESynC* to biorefineries. This integration may lead to accelerated development of sustainable environmental technologies as well as new paradigms in modern societies.

Keywords: Microbial electrosynthesis cells, life cycle assessment, value-added products, biorefinery.

INTRODUCTION

Bioelectrochemical systems (*BES*) constitute a promising technology which use biocatalysts in the anode, cathode or both. It is well known that *BES* rely on the action of biocatalysts. First, we distinguish two broad categories of *BES,* those driven by enzymes (Durand et al., 2012) and those driven by live microorganisms (Hamelers et al., 2009 and 2010). Both enzymes and live microorganisms are biocatalysts, although the second ones are live biocatalysts.

BES that use microorganisms can, in turn, be subdivided into two main types: microbial fuel cells (*MFCs*) and microbial electrolysis cells (*MEC*). The first type of devices produces electrical energy from organic substrates whereas the second one needs a constant supply of electricity in order to operate (Figure 1). As it was mentioned above, *MFCs* are devices capable of generating electricity via the oxidation of organic compounds, usually

waste biomass (likely wastewaters or effluents, Logan et al., 2006; Chaudhuri & Lovley, 2003) (Figure 2).

There are several special subytpes of *MFC*s such as the microbial desalination cells (*MDCs*, Cao et al., 2009) or microbial remediation cells (*MRCs* or also known as bioelectrochemical soil-slurry reactors or *BECSR*, Blanco-Mendoza et al., 2017, Camacho-Pérez et al., 2013). The main purpose of *MDC* is to remove salts from effluents, whereas the goal of *BECSR* focus on the remediation of soils and sediments and remediate a contaminated effluent or soil respectively, although, can simultaneously provide electrical energy. Microbial Solar Cells (*MSC*) (Figure1) constitute a subtype of *MFC* that oxide photosynthetic biomass instead of waste biomass thus generating bioelectricity (Strik et al., 2011) (Figure 2).

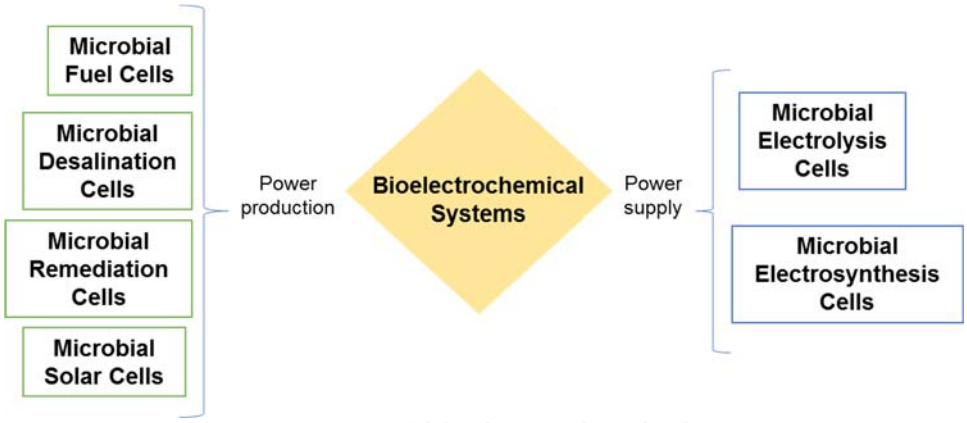

FIGURE 1. Types of bioelectrochemical systems.

On the other hand, some *BES* do not produce bioelectricity, rather they need an additional input of electricity for sound operation. For instance, in more conventional *MEC*s, the organic material fed to the device is degraded to CO_2, protons, and electrons in the anode, whereas the protons migrate inside the cell to reach the cathode where H_2 production occurs (Jeremiasse et al., 2010). The external electric power supply needed to produce H_2 is around 0.6 to 1 V, whereas abiotic electrolysis cells without biological catalysts typically require around 1.8 to 2 V for carrying out the electrohydrogenesis (Logan et al., 2008; Wrana et al., 2010). In other words, the microorganisms decrease the invested electric energy (Rabaey & Rozendal, 2010).

Microbial electrosynthesis cells (*MESynC*) constitute a special subtype of *MEC*s. It is an emerging biotechnology that requires a supply of electric power, with the aim to obtain value-added products (*VAPs*) through bioelectrochemical systems where microorganisms have the function of catalysts. Soluble organic compounds or CO_2 or other carbon sources can be reduced to generate *VAPs*, and this conversion is support partially or completely by electric power (Rabaey & Rozendal, 2010; May et al., 2016; Zhao et al., 2016).

The *MESynC* have received great attention in the last years as an alternative to chemical synthesis or conventional fermentation, due to the potential advantages of this technology, For instance, recognized positive features of *MESynC* are (Harrington et al., 2015; Butler & Lovley, 2016): *(i)* storage of electrical energy in a product, *(ii)* production of a fuel or chemical building blocks (using renewable or waste feedstocks) to replace blocks produced from fossil sources, *(iii)* end metabolite profile in electrofermentation can be modulated or shifted by regulation of the power supply.

MESynC provide a way to reduce problems such as global warming by sequestering atmospheric CO_2, which is the raw material to produce carbon compounds more elaborate. The potential of *MESynC* is multiplied by help in dealing with problems such as storage

and harvesting of electricity in solar, wind and natural gas exploration farms, this because microorganism are responsible for capture this electric power in chemical energy forming carbon-carbon bonds. It is especially important in solar energy since the electricity production does not always coincide with the electricity demand peak requirement (Lewis & Nocera, 2006; Nevin et al., 2011).

The objective of this work is to critically review recent efforts on *MESynC*. The scope of this review includes the following issues: (*i*) Principles of *MESynC* for chemicals´ production; (*ii*) *VAPs* and other products; (*iii*) energy efficiency and mass/energy balances; (*iv*) system analysis of *MESynC*; (*v*) *MESynC* and biorefineries; and *(vi)* Perspectives and conclusion.

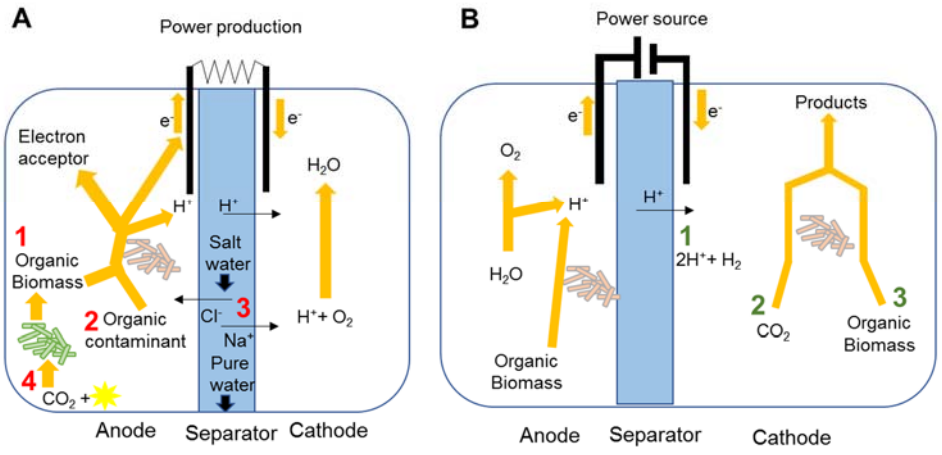

FIGURE 2. (A) Bioelectrochemical systems with power production. Red numbers inside anodic chamber of the figure indicate BES type: 1, Microbial Fuel Cells; 2, Microbial Remediation Cells, 3, Microbial Desalination Cells; 4, Microbial Solar Cells;
(B) Bioelectrochemical systems with power supply. Green numbers inside cathodic chamber of the figure indicate BES type: 1, Microbial electrolysis cell (abiotic cathode); 2, Microbial electrosynthesis cell with VAPs production from inorganic carbon source; 3, Microbial electrosynthesis cell with VAPs production from organic carbon source.
Examples of *VAPs* and/or commodities produced from CO_2 or organic substrate, as well electron transfer mechanism are mentioned in Table 1.

PRINCIPLES OF MICROBIAL ELECTROSYNTHESIS CELLS FOR CHEMICALS PRODUCTION

Microbial electrosynthesis cell process and configuration. A typical *MESynC* comprises of two compartments, the anodic and cathodic chambers fitted with working electrodes (anode and cathode). Chambers are commonly separated by a semipermeable, selective membrane (usually a cation exchange membrane, Gildemyn et al., 2015, Figure 3). In the anode, the oxidation process occurs (typically water is splitted to O_2, protons, and electrons) whereas in the cathode the reduction process takes place (typically the reduction of carbon dioxide or an organic carbon source). The cathode is fitted with a reference electrode that allows to determine the poised potential of the device and performing electrochemical monitoring of the process. Thus, obtaining useful information on resistive and capacitive elements of the device as well as identification of key redox subprocesses and substances (i.e., electrochemical impedance spectroscopy, cyclic voltammetry analysis) (Gildemyin et al. 2015; Marshall et al., 2013).

In most cases, only the cathodic chamber is seeded with biocatalysts; it is sought that these biocatalysts were able to accept electrons from the cathode (electrochemically-active bacteria, *EAB*) and form a biofilm attached to the cathode, although planktonic biocatalysts can also be found and help to overall performance of the device (Zhang et al., 2013; Chen et al., 2014).

In general, from the configuration point of view, *MESynC* can be classified in two-chamber (see Figure 3) and three-chamber devices (not shown, Gildemyn et al. 2015; Table 1). The latter has a distinct advantage: it allows for the separation/concentration/extraction of the target product

Gildemyn et al. (2015) pointed out that there is a disadvantage of the two-chamber device: products cannot be recovered at high concentration. Indeed, high product concentration, in turn, could result in the appearance of concomitant products and inhibition of the main redox process. Also, two-chamber MESynC should be complemented by ancillary separation processes in order to concentrate and acidify the products to recover them from the spent electrolytes (typically the catholyte). An intermediate third chamber is able to separate and concentrate the target compound, thus decreasing costs of post-treatment/separation of the product.

Gildemyn et al. (2015) produced and concentrated acetic acid from CO_2 in a three-chamber *MESynC* (Table 1) and proved the advantage of this approach. The third chamber (here-in after extraction chamber) was between the anodic and cathodic chambers; it was loaded with a saline solution. They used an anion exchange membrane as separator between cathode and the extraction chamber, whereas the separator between anode and extraction chamber was a cation exchange membrane. The electrical current simultaneously drove two processes, namely the reduction of CO_2 into acetate and its extraction into the extraction solution of the intermediate, third chamber. In this way, product inhibition of the cathode synthesis of the product was avoided and up to 13 g L^{-1} of acetic acid was accumulated in the extraction liquid (with none or little organic impurities).

Regarding the geometry of *MESynC*, Table 1 shows that there a predominance of the *H* type device (two-chamber), where the separator is placed in the horizontal tube that connects the two chambers (or flasks). Only 2 out of 14 cases used a different geometry called as flat plate reactor, either 2- or 3-chamber devices (Steinbusch et al., 2010; Gildemyn et al., 2015).

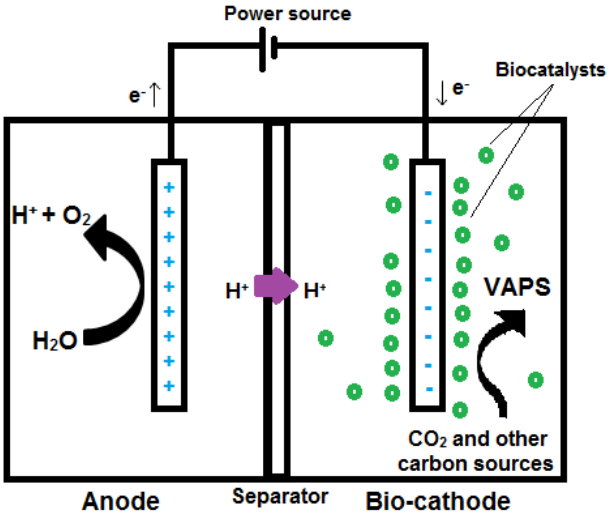

FIGURE 3. A typical two-chamber microbial electrosynthesis cell.

Concerning the electrode materials used in *MESynC*, graphite felt predominates (nearly half of the cases reported in Table 1), and carbon felt in five cases. No chemical catalyst was used in the cathodes.

BES can be operated either in batch, continuous, or fed-batch modes. The first two are the most predominant. The *MESynC* reviewed in our work were operated in batch mode (Table 1).

Mechanisms of Electron Transfer between Cathode and Microorganisms. Microorganisms take advantage from the electron flow for the *VAPs* production, as well as growth and maintenance of the cells. Until now the extracellular electron transfer (*EET*) between cathode-microorganism is not as well studied as anode process and anodic *EET* (Sharma et al*.,* 2014; Jourdin et al., 2015). Nonetheless, modifications have been done in the surface cathode to facilitate the microbe colonization enhancing *MESynC* efficiency, for instance, treating carbon cloth cathode with metal nanoparticles stimulate acetate electrosynthesis or anchoring nickel nanowire to graphite cathode to improve microbe-electrode interaction (Nie et al., 2013; Zhang et al., 2013). Some rather similar theories to anode *EET* have been proposed to describe cathode *EET*. Electrodes can transfer electrons in an indirect or direct way to the microorganisms as reported depth Rabaey and Rozendal (2010).

Below we will discuss the three recognized mechanisms of *EET* that have been reported for *MFC*s, and hopefully, they would be also valid for *MESynC* as well.

Indirect Electron Transfer through Mediators. Mediators are soluble electroactive species that provide redox coupling between the electrode and the redox center in the biological compound, and act as "electron shuttles" (Figure 4) (Fultz & Durst, 1982).

According to Fultz & Durst (1982) and Szentrimay *et al.* (1977), *ideal* mediators should exhibit the following desirable characteristics: *(i)* reasonable solubility in aqueous media in a wide interval of pH or at least at or near pH 7; *(ii)* stability of the oxidized and reduced forms of the mediator; *(iii)* rapid electron transfer; *(iv)* known standard redox potential, $E^{o'}$; *(v)* known electron stoichiometry; *(vi)* no toxic effects to the biocatalsyts; *(vii)* easy separation from the products; *(viii)* no interaction with the

biocatalyst that could impact its redox potential; and *(ix)* lack of optical interference if optical monitoring of either the mediator or the biocomponent is needed.

In our discussion of mediators, any chemical compound that could act as an intermediate electron donor (i.e., H_2, formate, etc.) are not considered to be mediators, rather they are included in a third special mechanism of electron transfer (see below).

Mediators are used mainly to accelerate electron transport from the cathode to organisms. They could influence the metabolism of organisms to some extent, and freeze undesirable microbial processes such as methanogenesis that would compete for electrons with the substrate (Steinbusch et al., 2010). Park et al. (1999) pointed out that microbes harbored in the *MESynC* can obtain free energy and reducing power from the electron driving force generated by the potential difference between the coupled oxidoreduction half-reactions of mediators. Since the electron transfer does not occur between the electrode and the microbes, rather via electrode-mediator-microbe, the mechanism is also known as indirect transfer of electrons.

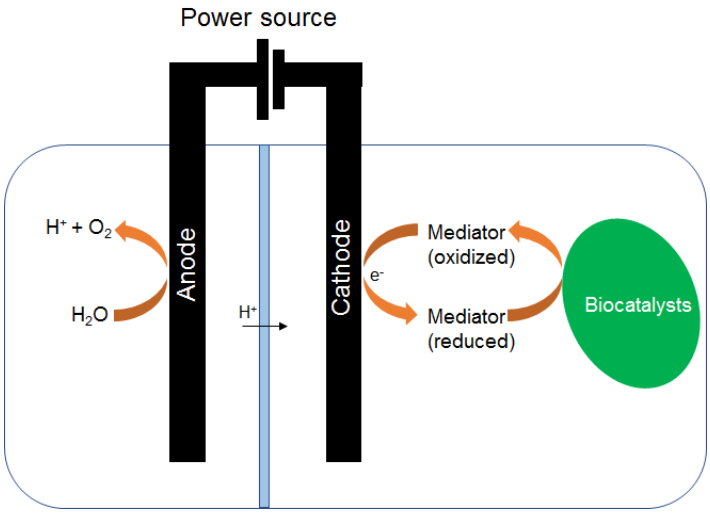

FIGURE 4. Electron transfer from electrode to biocatalysts through use of mediators in the microbial electrosynthesis cell.

In the indirect transfer of electrons mediators such as neutral red has been used, that accept electrons from cathode and then donate them to electron carriers in the membrane. Table 1 shows that in addition to neutral red (N^8,N^8,3-trimethylfenazine-2,8,-diamine IUPAC name; Figure 5A, used in half of the cases reviewed), anthraquinone-2,6-disulfonate (AQDS, Figure 5B) has been used as mediator in *MESynC* devoted to the production of lactate from glucose (Sasaki et al., 2014). These mediators have in common the presence of double bonds and/or nitrogen atoms that typically have a pair of electrons that could accept hydrogen/release proton plus electron.

In real world, use of mediators very often could have negative effects, such as increasing the cost of the process, possible toxicity to biocatalysts, chemical instability, and product target contamination that complicates the recovery and purification of organic products.

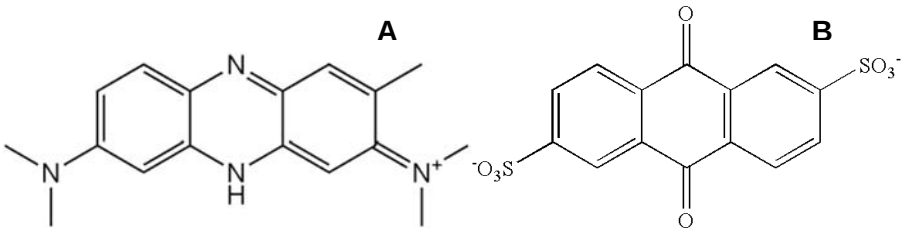

FIGURE 5. Neutral red and anthraquinone-2,6-disulfonate structures.

Direct Electron Transfer. Electrons can be directly provided to microorganisms (also known as *EAB* or electrotrophs) that couple direct electron consumption to the reduction of substrate or other compounds (Butler & Lovley 2016). The authors recommended this as the most straightforward mechanism and efficient strategy for producing biocommodities with the supply of electrical energy. Regarding these microorganisms able to take electrons directly from cathode, these microorganisms very often form a biofilm where the support is the electrode (Bond & Lovley, 2003; Nevin et al., 2010; Lovley, 2011). The common problems faced by biofilms are CO_2 and nutrient diffusion (whenever the *MESynC*) uses carbon dioxide as substrate, and likely high concentration of inhibitors at the external face of biofilm. Yet, advantages of biofilm structure are its great resistance and stability (Cheng et al., 2010; Rabaey et al., 2011).

It has been described for bacteria such as *Geobacter sulfurreducens* and *Shewanella oneidensis* that they have several protein complexes on their membranes and the periplasmic space that have participation in the electron transference. Moreover, this event is helped by pili-like structures or membrane appendices known as nanowires (Holmes et al., 2006; Reguera et al., 2005; Hartshorne et al., 2009; Rabaey & Rozendal, 2010; Pirbadian et al., 2014).

There is some evidence that poising the cathode of a cell could lead to morphological and likely physiological changes in the microbes. Interestingly, Choi et al. (2014) observed changes to extracellular structures and electronegativity of *C. pasteurianum* grown as biofilm attached to a cathode, when the latter was poised at +0.045 V *vs.* standard hydrogen electrode (*SHE*). For example, extracellular appendages were found in bacteria grown n the poised biocathodes but no in bacteria grown on graphite cathodes of control device (open circuit). Yet, the authors recognized that the role of the extracellular appendages observed in the bio-cathode cannot be elucidated based on only microscopic analysis applied in their research. Therefore, the possible interaction or selective pressure cathodic microbe-poised cathode is an area that deserves further research.

Electron Donor Compounds. Some chemical compounds can intervene in electron transfer based on their ability donate electrons. They act as intermediates in the electron transfer paths, but the oxidized form of the compound is not necessarily reduced (they are not true mediators). Examples of electron donor compounds are H_2, formate, ammonia, sulfide or Fe(II), that can be used for microbial growth and maintenance as well as *VAPs* production (Park et al., 1999; Hawkins et al., 2011; Khunjar et al., 2012; Li et al., 2012; Lovley & Nevin, 2013). Yet, it has been claimed that H_2 and formate have redox potentials sufficiently low to be used in the CO_2 reduction to produce either biofuels or more complex organic compounds (Lovley & Nevin, 2013). However, other potential electron donors (i.e., Fe(II), ammonia, sulfide) need electron acceptors, such as O_2, where $E_{O2} > E_{CO2}$ in such a way that cell growth is feasible. This feature would lead to inefficient design and performance of *MESynC*. For instance, physical separation between the site of electrochemical electron donor generation and microbial electron donor consumption would be required with added process cost and complexity.

VALUE-ADDED PRODUCTS AND OTHER COMMODITIES PRODUCED BY MICROBIAL ELECTROSYNTHESIS CELLS

We distinguish between value-added products (*VAP*s) and commodities. The differences are at least two, namely, the price per unit mass, and the scale of production/consumption worldwide. *VAP*s are chemicals (typically organic) with a high sales price and relatively low to moderate production/consumption, whereas commodities are chemicals and fuels with a low to moderate sales price and large mass production/consumption. Tentatively, a price limit could be traced at 1000 US/metric ton. If the price is below this value, it is a commodity whereas for prices higher than 1000 US/metric ton it would be a *VAP*.

For instance, acetic acid price is 400-500 USD/metric ton FOB (www.alibaba.com, accessed April 22, 2017), whereas the price of ethanol is *ca.* 560 USD/metric ton (www.trendingeconomic.com/commodity/ethanol, accessed April 23, 2017). On the other hand, the price of succinic acid in the range 7000 to 9000 USD/metric ton whereas lactate has an average price of 3000 USD/metric ton.

Examples of *VAP*s in *MESynC* and alike follow: succinic acid, butyrate, lactate, whereas examples of commodities are CH_4, H_2, acetic acid, and likely ethanol.

Intuitively, *MESynC* would compete more comfortably in the niche of *VAP*s production because in the area of commodities already existing, conventional production processes are well established for mass production and optimized. Yet, there is room for *MESynC* production of commodities as long as it can show specific advantages (use of wastes as substrates, less environmental impacts than conventional production, etc.).

Until nowadays, a wide diversity of *VAPs* and commodities have been production targets in *MESynC*, for instance hydrogen, caustic soda, hydrogen peroxide, methane, organic acids, alcohols (Rozendal et al., 2006; Lalaurette et al., 2009; Kato et al., 2012; Sharma et al., 2014; May et al., 2016). Commonly hydrogen has been produced with a bioelectrochemical device with an abiotic cathode. Choi et al. (2014) obtained butanol from glucose as well as 1,3-propanediol from glycerol using *Clostridium pasteurianum* DSM 525 in the cathode of a *MESynC* (Table 1).

Gildemyn et al. (2015) obtained acetic acid in a 3-chamber *MESynC*. This device allowed for extraction and concentration of the product at least 2 to 3 times the concentration of acetic acid obtained by conventional microbial processes. We included this reference just as an example of *MESynC* for acetic acid, since our focus was mostly *VAP*s. For more information on acetic acid production by this device, please consult May et al. (2016).

VAPs obtained through *MESynC* can be produced by microorganisms from low-cost carbon sources. For instance, Zhao et al. (2016) produced succinic acid from hydrolysate of corncob (Table 1). Some *MESynCs* use CO2 for producing the commodity acetic acid

(Table 1, Ganigué et al. 2015; Gildemyn et al., 2015). More works on CO_2 as substrate for *MESynC*s and alike can be found in the reviews by Lovley & Nevin (2013) and Rabaey et al. (2011) because in our review we highlight organic carbon sources. Regarding this, organic acids, wastewater, hydrolysate of lignocellulosic residues, among others have been used as carbon sources in *MESynC* (Moreno et al., 2016; Zhao et al., 2016, Table 1).

When the power and carbon sources are the sun and CO_2 respectively, the process is coined said artificial photosynthesis where the products are organic compounds and oxygen (Lovley & Nevin, 2013).

As it was briefly mentioned above, recently there has been a considerable interest to replace the expensive materials of construction of *MESynC* for more economic alternatives, for instance, one important cost is the membrane and electrode material.

Particularly for the electrode has been used as materials carbon, graphite and stainless steel (Soussan et al., 2013). Much work has been done in alternative membranes such as clay and agar (Hernández-Flores et al., 2015) but focused to MFC. Similar work and application to *MESynC* is still scarce. Most membrane materials used in *MESynC* are polymers such as Nafion (Sasaki et al., 2014; Zhao et al., 2016; Park et al., 1999) and Fumasep (Gildemyn et al., 2015).

Product secretion completely removes the need of cell lysis in product recovery, simplifying downstream processing and lowering production cost (Blankenship et al., 2011; Heeres et al., 2014). More efforts in the direction of secretion mechanisms of the biocatalysts.

TABLE 1. Microbial electrosynthesis cell production of value-added compounds and selected commodities.

Microorganism/substrate/ (initial conc. gL^{-1})	Product/ product concentration (gL^{-1}) ([a]target product)/ yield ($g_{product}/g_{sustrate}$)/ Product concentration in electrochemical vs non-electrochemical fermentation (%)	operation of reactor/ Redox potential (V vs. Ag/AgCl)/potential source/ Mechanism of electron transference (e-donor compound[b], shuttle[c] or cathode[d])	Reactor type/number of chambers/scale experiments-working volume (L)/mixing/temperature (oC/heating/pH control/Separator/Cathode materials	Integration to biorefinery/ system analysis/ energy and mass balances/Scale-up	Ref.
Corynebacterium glutamicum/ glucose (20)	lactate/15.5/0.76/120.9	Batch (24 h)/ -0.6/Potentiostat/ indirect/AQDS[c]	H-type/Dual/0.25/hotplate stirrer 330 rpm/30 oC/pH control No/Nafion 118 PEM/graphite plate C-porous graphite plate A	no/no/no/no	[1]
Actinobacillus succinogenes/ corncob hydrolysate (15)	Succinate/3.84/0.26/130.6	Batch (24 h)/ -1.8/power supply/ indirect/Neutral red[c]	NM/Dual/0.28/11.5/37/incubated/initial 7.2 pH control No/Nafion 117 PEM/carbon felt	no/no/yes incomplete/no	[2]
Actinobacillus succinogenes/ glucose (15)	Succinate/7.88/0.53/134.9	Batch (24 h)/ -1.8/power supply/ indirect/Neutral red[c]	NM/Dual/0.28/11.5/37/incubated/initial 7.2 pH control No/Nafion 117 PEM/carbon felt	no/no/yes incomplete/no	[2]
Actinobacillus succinogenes/ xylose (15)	Succinate/5.24/0.35/151.88	Batch (24 h)/ -1.8/power supply/ indirect/Neutral red[c]	NM/Dual/0.28/11.5/37/incubated/initial 7.2 pH control No/Nafion 117 PEM/carbon felt	no/no/yes incomplete	[2]
Actinobacillus succinogenes/ arabinose (15)	Succinate/4.7/0.31/209.82	Batch (24 h)/ -1.8/power supply/ indirect/Neutral red[c]	NM/Dual/0.28/11.5/37/incubated/initial 7.2/ Nafion 117 PEM/carbon felt	no/no/no/no	[2]
mixed culture dominated by species of genus *Clostridium*/ /CO_2	([a]butyrate-acetate-ethanol-butanol)/ (1.78-2.84-1.42-0.54)/ NR/ not control reported	Batch (34 d)/ -0.8/NM/ Donor compound/H_2[b]	H-type / /Dual/0.12/Stirred/33.9/wrapped with a coil of plastic tubing connected to a thermostatic bath/initial pH 6.5/ CMI 7000 CEM/ carbon cloth/titanium rod	no/no/no/no	[3]
acetate-reducing inoculum/ acetate (3)	([a]ethanol-methane-propionate-butyrate)/ (0.08-0.06-0.04-0.044)/ 0.075/ not control reported	Batch (10 d)/ -0.55/DC power supply/ Donor compound/hydrogen	flat plate/Dual/0.87/100rpm/30/regulated at 6 with a pH controller/ monovalente selective AEM/	no/no/yes incomplete/no	[4]

		[b]	graphite felt		
Escherichia coli/ glucose (20)	(lactate/Formate/ethanol/acetate/succinate)/ (3.72-1.06-0.86-1.08-0.35)/ 1.06/(115.43-101.95-115.42-120.58-125.42)	Batch (15.5 h)/ -0.65/custom-built potentiostat/ indirect/Neutral red[c]	Adams and Chittenden type/Dual/0.1/stir bars 150rpm/37/incubators/initial 6/ CMI 7000 CEM graphite felt/graphite rod	no/no/yes incomplete/no	[5]
Klebsiella pneumoniae/ glycerol (20)	(Propanediol-ethanol-acetate-lactate-succinate)/ (5.88-1.29-1.17-0.51-0.18)/ 0.74/	Batch (15.5 h)/ -0.65/ indirect/Neutral red[c]	Adams and Chittenden type/Dual/0.1/stir bars 150rpm/37/incubators/initial 6/ CMI 7000 CEM graphite felt/graphite rod	no/no/yes incomplete/no	[5]
Zymomonas mobilis/ glucose (20)	(Ethanol-acetate-succinate)/ (23.45-1.95-0.92) /0.55/(97.5-120.28-102.1)	Batch (21.5 h)/ -0.65/ indirect/Neutral red[c]	Adams and Chittenden type/Dual/0.1/stir bars 150rpm/37/incubators/initial 6/ CMI 7000 CEM graphite felt/graphite rod	no/no/yes incomplete/no	[5]
Actinobacillus succinogenes/ glucose (10.8)	([a]succinate-ethanol-formate-acetate)/ (9.79-0.97-0.41-0.23) /0.91/(161.7-405-24.35-58.6)	Batch (24 h)/ -2/ indirect/Neutral red[c]	NM/Dual/0.3/37/initial 7/ Nafion PEM/graphite felt	no/no/yes incomplete/no	[6]
Clostridium pasteurianum DSM 525/ glucose (18)	([a]butanol-butyrate-acetate) (*1-3.8-2) /0.38/(250-84.3-86.9)	Batch (40)/ -0.16/ Direct/cathode[d]	H-type/Dual/0.3/nm/37±1/heating tapes/initial 6.5/ Nafion 117 PEM/(graphite felt-Pt plate)	no/no/yes incomplete/no	[7]
Clostridium pasteurianum DSM 525/ glycerol (27.6)	([a]1,3-propandiol-butanol/butyrate/ethanol)/(7.14-4.35-1.1-0.57)/156.2-60.75-106.8-84.1)	Batch (50)/ -0.16/ Direct/cathode[d]	H-type/Dual/0.3/nm/37±1/heating tapes/initial 6.5/ Nafion 117 PEM/(graphite felt-Pt plate)	no/no/yes incomplete/no	[7]
Autotrophic acetate-producing community, dominated by *Clostridiales*/CO_2	1st cycle: Acetate/catholyte (1.8), extraction compartment (11.9), anolyte (7); 2nd cycle: Acetate/catholyte (2.3), extraction compartment (13.5), anolyte (8.5)	Batch (1st cycle day 10 to 43; 2nd cycle day 54 to 86) with recirculation (0.031L)/ -1.140/ Direct/cathode[d]	three compartments 0.2/21/regulated at 8.4/AEM & CEM/ carbon felt/titanium-coated TiO2/IrO2 (35/65 %) mesh	no/no/no/no	[8]

Notes: [a]target product/ yield ($g_{product}/g_{sustrate}$); Mechanism of electron transference: [b] e^- donor compound, [c] shuttle or [d] cathode

References: 1. Sasaki et al., 2014; 2. Zhao et al., 2016; 3. Ganigué et al., 2015; 4. Steinbusch et al., 2010; 5. Harrington et al. 2015; 6. Park et al., 1999; 7. Choi et al., 2014; 8. Gildemyn et al., 2015.

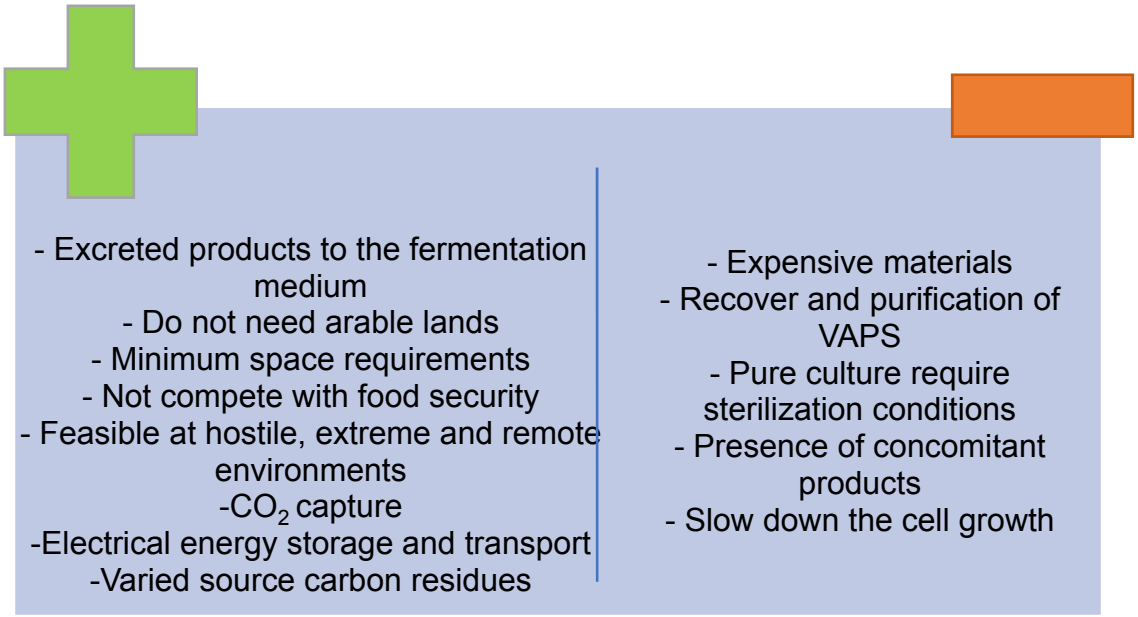

FIGURE 6. Pros and cons of the application of microbial electrosynthesis cells for the *VAPs* production (Lovley, 2011; Hawkins et al., 2011; Rabaey et al., 2011).

Figure 6 shows a counterpoint of benefits and drawbacks of the application of *MESynC* technology for production of *VAP*s. In the benefits side, it can be cited the following (Cheng et al., 2009; Lovley, 2011; Hawkins et al., 2011; Rabaey et al., 2011):

- so far, the *VAPs* are extracellular compounds, thus avoiding extra costs due to disruption and separation of the targeted product;
- need of arable land is minimum, particularly for *MESynC* fed with CO_2 and hydrolyzates of residues;
- relatively small footprint;
- no competition between *MESynC* technology and food supply/security;
- mitigation of environmental impacts, see the arguments in arable land;
- some target products can be used as stored energy and transported to the point of use, for example ethanol, butanol
- easy integration with renewable energy sources (sunlight, eolic, *MFC*s)
- possibility of minimizing concomitant products thus abating costs of target product separation/purification. For instance, in succinic acid production by *A. succinogenes* other organic acids such as acetic and formic acids are generated (Zhao et al., 2016).

There are perceived disadvantages as well. One of the most significant is the lagging on scaling up of the technology. So far, most experiments *MESynC* have been implemented at lab scale, batch operation.

ENERGY EFFICIENCY, MASS AND ENERGY BALANCES

If *MESynC* has to become a commercial technology, it is required to maximize product yield to reach competitive production costs. Also, to establish this, it is important to

determine the values of parameters related to electrochemical and fermentation performance, such as coulombic efficiency, conversion efficiency, and comparison electrofermentation vs. conventional processes efficiencies. Energy efficiency reflects the energy input (electric, thermal, and mechanical energies) *vs.* how of this energy is recovered in the product. Energy efficiency depends of the voltage and coulombic efficiencies, the reaction between cathodic and anodic electrodes (Hamelers et al., 2009). Coulombic efficiency indicates the quantity of electrons supply to system through electric energy in the product as it will be discussed below. Voltage efficiency (η_E) in a *MESynC* is defined as:

$$\eta_E = \frac{-E_{emf}}{-E_{app}} \quad [1]$$

where E_{app} is the applied voltage and E_{emf} is the thermodynamic voltage (reversible) determined by the reactions at the electrodes at equal pH. Both potentials are typically negative as power is supplied to the system for the *VAPs* production (Sleutels et al., 2012). By providing external voltage to the *MESynC*, this must be enough to overcome the thermodynamic barriers, for this reason it is desirable minimize the internal resistance of the *MESynC* (Sleutels et al., 2012).

Steinbusch et al. (2010) and Marshall et al. (2012) used the coulombic efficiency as a performance indicator of *MESynC*. Steinbusch et al. (2010) also advocated the use of a conversion efficiency and traced its conception to McCarty (1972).

The coulombic efficiency was defined as the percentage of supplied electrons that was converted to product (s) (*Pr)* in the cathode of the *MESynC*. Assuming a batch process with initial and final times t_1 and t_2, respectively, we have:

$$\eta_{Coul}\,(\%) = [([Pr]_{t2} - [Pr]_{t1})VbF / \int_{t1}^{t2} I.\mathrm{d}t\,] \times 100\% \quad [2]$$

where
$[Pr]_t$ is the product concentration at time *t* (mol L-1); *V* is the working volume of the device (L); *b* is number of electrons involved in the reduction, *F* the Faraday constant (96,485 Coul (mol electrons)$^{-1}$); *I* the current intensity (A). Steinbusch et al. (2010) proposed to neglect the electron equivalents that might derive from the inoculum.

According to Steinbusch et al. (2008, 2010) conversion efficiency ($\eta_{conversion}$) was defined as the efficiency of electron flow from reactants to products, where electron flows were expressed in electron equivalents (mol of e^-) based on concentrations, carbon atoms, and degree of reduction of each individual compound. In their work, the conversion efficiency (referring to their mediator experiments) was defined as the ratio of consumed electron equivalents of acetate and hydrogen that were converted to electron equivalents of ethanol or other products as *n*-butyrate. The authors proposed to correct the expression for the electron equivalents contributed by sludge decay, and that enter the liquid phase. Unfortunately, they neither provided an explicit equation of the conversion efficiency nor a worked example.

These efficiencies allow to compare performances between or among several *MESynC* processes. Yet, both the η_{Coul} and the $\eta_{conversion}$ do not seem to be adequate parameters to make comparisons between *MESynC* and conventional fermentations or *MESynC* and non bioelectrochemical processes.

Other indices that can be useful to evaluate and compare *MESynC* performance (that are related to mass and energy balances) are the following: the yield and pseudoyield of the produced *VAP*, borrowed from Microbiology (*Y* and *Y'*, respectively, Madigan et al.,

2012), the electrofermentation index η_{EF}, the conventional fermentation index η_{cf}, and the efficiency that compares performance of electrofermentation vs. conventional fermentation of the same *VAP*, $\eta_{EF/cf}$. This group of indices (Eqs. 3 to 10) has been proposed by Poggi-Varaldo (2017) (private communication, 2017). Their definitions follow:

$$Y = m_{VAP,gen}/m_{US} \quad [3]$$

$$Y' = m_{VAP,gen}/m_{IS} \quad [4]$$

$$\eta_{EF} = m_{VAP,EF}/(E_{electr} + E_{heating} + E_{mixing}) \quad [5]$$

$$\eta_{cf} = m_{VAP,cf}/(E_{heating} + E_{mixing}) \quad [6]$$

$$\eta_{EF/cf} = \eta_{EF}/\eta_{cf} \quad [7]$$

$$E_{electr} = \int P.\mathrm{d}t = \int E_{applied} * I.\mathrm{d}t \quad [8]$$

$$E_{heating} = m_{electrolytes} * c_{p\ electrolytes} * (T_{oper} - T_{amb}) + m_{equip} * c_{pequip} * (T_{oper} - T_{amb}) + \\ + U * A_{equip} * (T_{oper} - T_{amb}) * \Delta t \quad [9]$$

$$E_{mixing} = \pi * V * \Delta t \quad [10]$$

where $m_{VAP,gen}$ is the mass of *VAP* produced, either in kg or mole; m_{US} is mass of uptaken (consumed) substrate, either in kg or mole; m_{IS} is the initial mass of substrate, either in kg or mole; $m_{VAP,EF}$ is the mass of *VA*P produced in the *MESynC*, in kg or mol; $m_{VAP,cf}$ is the mass of *VAP* produced in the conventional fermentation; E_{electr} means the electrical energy spent in the electrofermentation, in J; $E_{heating}$ is the thermal energy required to heat the electrolytes and equipment, as well as the heat losses, in J; E_{mixing} stands for the energy of mixing requirements, if any, in J; *P* is the power supplied to *MESynC*; *I* is the current intensity in the *MESynC*; $m_{electrolytes}$ is the mass of the electrolytes in the cell; $c_{p\ electrolytes}$ is the specific heat (constant pressure) of the electrolytes; T_{oper} is the operation temperature, either ^{o}C or K, typically in the mesophilic range or room temperature; T_{amb} is the ambient temperature, typically 15 ^{o}C in an indoor facility, either ^{o}C or K; m_{equip} is the mass of equipment, $c_{p\ equip}$ is the specific heat of the equipment; *U* is the overall heat transfer equipment for heat loss of the equipment; A_{equip} represents the external surface area of the equipment; Δt is the period of operation; π is the unit volumetric power of mixing for fermenters/reactors that is an index that depends on the mixing intensity sought and the scale size of the reactor); *V* is the liquid volume in the device that is being mixed.

Regarding the comparative efficiency $\eta_{ef/cf}$ (Eq. 7) please note that when

$$\eta_{EF/cf} > 1 \quad [11]$$

Then the electrofermentation of a given *VAP* outperforms the conventional fermentation.

By inspecting Eqs. 5 to 7, it is apparent that in order the electrofermentation process to outperform the conventional one, the influence of an eventual increase of the mass of *VAP* generated in the *MESynC* should offset the increase in the denominator due to E_{electr} expenses.

From Table 1, it can be seen that no work out of 14 cases have reported complete mass and energy balances for *MESynC* experiments. Most references have performed mass balances, whereas nearly half have carried out electron-equivalent balances (electrochemical). None has included energy balances that take into account not only the electrochemical energy but heating and mixing energies as well. From our perspective, energy balances are incomplete and constitutes a pending matter. As we mentioned above, indices and efficiencies that allow us to evaluate *MESynC* performance are necessarily based on mass and energy balances (Fast & Papoutsakis, 2012). Therefore, we claim that any article on *MESynC* should be accompanied by complete mass and energy balances, as well as performance indices and efficiencies. This is the only way to establish feasibility of the reported process and its results and eventual advantages over competitor processes. If the numbers are poor, it is a clear indication that either the research approach likely should be abandoned or a critical appraisal of issues to improve the process should be discussed in the article. In case that calculations of indices and efficiencies were long and fastidious, they can be submitted as *Supplementary Information*. One or another approach should be followed because it would be very debatable, from our point of view, to omit such an information on *MESynC* research results.

SYSTEM ANALYSIS OF MICROBIAL ELECTROSYNTHESIS CELL PROCESS

It is worth noting that the feasibility and sustainablility of *MESynC* (as well as that of other *BES*) should be evaluated following a system approach, or “cradle-to-grave” method. If not, it may occur that some less perceived drawbacks of *MESynC* or *BES* were not taken into account when performing conventional energy or economic assessment thus resulting in a ‘false positive’ conclusion, i.e. to integrate *BES* or *MESynC* in a biorefinery on the grounds of mere increases of yield of a desired *VAP* or biofuel, when actually this decision decresases the sustainability of the project. Very often other factors such as environmental impact, energy consumption, etc., not considered in a superficial analysis could offset the perceived “gain” of *BES*/*MESynC* integration to a biorefinery scheme.

There are system analysys techniques that allow the unearthing of more or less hidden factors and effects. Pioneering efforts of von Bertalanffy and others in the previous century lead to recognition of systems complexity as well as the need for system analysis and assessment (von Bertalanffy, 1968). Among a menu of analytical tools, life cycle assessment (*LCA*) can be considered a contemporary, widely used, and powerful tool of system analysis. *LCA* consists of a set of methods, techniques, and protocols that perform system (and systematic) assessment of project(s) or process(es), either individually or in the context of critical comparisons (ISO 14040, 2006 a; ISO14044, 2006b). *LCA* evaluates environmental impacts as well as consumption of resources that occur during every stage of services/facilities and products manufacturing, use, and final disposal. *LCA* takes into account and analyzes all the inputs of resources and energy needed to perform a process or product fabrication, the wastes generated, and the health and environmental burdens associated to that process/product (Menon & Rao, 2012).

According to Romero-Cedillo et al. (2016), *LCA* can prevent/minimize bias and missing of hidden costs and environmental subsidies in the analysis. *LCA* becomes a solid, scientific replacement of the commonly used (and debatable) return on investment evaluation of process and product manufacturing (Poggi-Varaldo et al., 2014).

The *LCA* allows to carry out detailed analysis of energy and mass exchanges on regional and global scales; it naturally leads to the quantitative estimation of the overall advantages and benefits as well as unearthing drawbacks or hidden disadvantages of the given process/ project. Another important feature is that *LCA* also helps to identify opportunities for process improvement (Kemppainen & Shonnard, 2005).

The *LCA* has gained widespread acceptance as an imprescindible instrument for systematic and system analysis of environmental and industrial processes.191–194 At present, bodies of the scientific and engineering community such as the European Union Research Commission considers that *LCA* is an objective evaluation tool of processes and projects that leads to fair comparisons and conclusions and should be a mandatory part of large R&D projects. One such application of *LCA* is for evaluation of biorefineries innovations (Cherubini et al., 2010).

The *LCA* methodology has been established and standardized in the ISO 14044 standards (ISO, 2006a; ISO, 2006b) There is a variety of software devoted to *LCA*, that includes examples from private companies (*i.e.*, SimaPro, Pré Consultants 2013) and academic groups (Easetech and Easewaste, DTU, Lynbgy, Denmark). Technical databases that feed data to *LCA* are ancillary although key tools for *LCA*, such as EcoInvent Ecoinvent (R) v2.1 database (Poeschl et al. 2012; Buratti et al., 2015).

A few years ago, Pant et al. (2011) in a significant paper reviewed the application of *LCA* to bioelectrochemical systems (*BES*s). They emphasized that *BES* are novel technologies based on the ability of several microbes to catalyze oxidation or reduction reactions at solid surfaces, typically the electrode (anode and cathode) surfaces. They recognized that *BES* could replace more energy intensive wastewater treatment technologies, and either generate sustainable energy from the organic pollutants present effluents or produce value-added chemical products. In spite of certain lags on scaling up *BES* technology, Pant et al. (2011) pointed out the need to scrutinize it by using *LCA*. Yet, they recognized that studies on *LCA* of these systems with the current state of the art are very scarce. In addition, the focus of the analysis has been centered on energy-producing *BES*, i.e., *MFC*. Less works, if any, are available for *MEC*s and *MESynC*. The authors also proposed a methodology to implement *LCA* of the *MFC*s and give a series of valuable recommendations. Regarding *MFC*s, the authors anticipated that direct benefits in terms of energy saved in aeration (considering activated sludge as a typical conventional wastewater technology), as well as earnings due to bioelectricity generated should be factored in *LCA* of such devices and help establishing *MFC*'s advantages. They identified anaerobic digestion as a conventional technology that is a direct competitor of *MFC*s, since the first also abate the pollutant load of effluents while simultaneously producing energy (biofuel methane) (Poggi-Varaldo & Rinderknecht-Seijas, 1996; Robles-González et al., 2012), similarly to *MFC*s. With optimism, Pant et al. (2011) concluded that *LCA* of *MFC*s would show how well these devices compare with more conventional, already existing treatment technologies, and particularly with anaerobic digestion.

We advocate that *LCA* implementation to evaluate *MESynC* technology will provide a clearer idea of the benefits and environmental/resource burdens related to *VAPs* production. Six years after the prophetic article by Pant et al. (2011), works on *LCA* of *MESynC* are still scarce and the current *status* is unsatisfactory. According to Table 1, no work on *MESynC* out of a total of 14 cases have implemented some sort of *LCA*, that is, 0%. This is clearly insufficient to gain knowledge on the true feasibility and sustainability of *MESynC*. Therefore, the *BES* community along with the experts on system analysis should devote more efforts to gain knowledge of *MESynC* by *LCA*. Second, and related to *LCA* probable results, we foresee that the negative environmental impacts would be related to the use of electric energy, substrate transport, shuttles addition to the culture medium and its sterilization, as well as *MESynC* construction. Among positive environmental impacts would be the CO_2 capture or the use of the lignocellulosic residues as substrates for *VAP* production in *MESynC*.

MICROBIAL ELECTROSYNTHESIS CELLS AND BIOREFINERIES

Pant et al. (2011) pointed out that microbial biorefinery could be the next logical step in the already existing green biorefinery, forest and lignocellulosic biorefinery, aquatic or algal biorefinery and integrated biorefinery concepts. Moreover, we foresee that integration of *BES* and particularly *MESynC* to biorefineries likely will enhance effectiveness and sustainability of waste-based biorefineries.

Recently, Poggi-Varaldo et al. (2014) discussed the growing interest on in-series coupled processes to obtain other products besides bioH_2, such as *DF* followed by another stage like microbial fuel cells (*MFC*) or other systems and their implementation in waste-based biorefineries. This approach aims at reclaiming increased amounts of bioenergy from organic wastes (Escamilla-Alvarado et al., 2012; Robledo-Narvaez et al., 2008). Considering the bioH_2 dark fermentation as a pivotal stage of such biorefineries, and the value of CH_4 as source of energy a methanogenic stage has been successfully coupled to bioH_2, which increased energetic potentials (Escamilla-Alvarado et al., 2012; 2013). Interestingly CH_4 production also could be coupled to profitable and environmentally friendly bioremediation processes (Estrada-Vazquez et al., 2001; Garibay- Orijel et al., 2005).

According to Poggi-Varaldo et al. (2014), another approach to increase energy gains in waste-based biorefineries is to couple bioH_2-*DF* with microbial electrolysis cells (*MEC*). Microbial electrolysis cells (*MEC*s) can be considered a subytpe of *MESynC*, where the VAP is the biofuel H_2. They have been proposed as an alternative technology for production of bioH_2 from wastes. It has been reported that simple organic substrates that are metabolites already present in effluents of bioH_2-*DF* can be profitably employed in *MEC*s to increase bioH_2 production (Wang et al., 2011).

As it was mentioned above in this review, a typical *MEC* consists of an anode and a cathode in their respective chambers, commonly separated by a selective membrane. *MEC*s are generally operated with poised potential of 0.3 V or higher. The *MEC* is typically fed with an effluent that contains organic substrate(s) and biocatalysts (Borole and Mielenz, 2011; Manish and Banerjee, 2008).

Wang et al. (2011) reported results for an integrated system for hydrogen production from cellulose that consisted of *DF*, *MFC*s and *MEC*s. The H_2 yield from the single *DF* process was significantly increase by *ca.* 40%, from 10.1 to up to 14.3 mmol H_2 $(g_{substrate})^{-1}$. Borole & Mielenz (2011) achieved in a *MEC* an energy efficiency of 72% for the conversion of acetate to bioH_2. According to the authors, the net energy gained in this way could meet nearly 60% of the distillation energy demands in a lignocellulosic biorefinery.

Importantly, poising the *MEC* with applied potential implies energy expenses of high quality energy (i.e., electricity). Poggi-Varaldo et al. (2014) advocated that there is a need to determine the net energy gain of bioH_2 from *MEC*s and to evaluate whether this increase can offset the electrical energy spent. On the one hand, there will be a positive amount of energy associated to the H_2 produced in the *MEC*; however, this energy should be compared with, or discounted from, the electric energy expended in poising the *MEC*. To the best of our knowledge, such studies are scarce or not yet available in the open literature. Therefore, there is an urgent need to perform research on this issue.

Unfortunately, Poggi-Varaldo et al. (2014) did not provide information on integration on other types of *MESynC* to waste-based biorefineries.

Since biorefineries are facilities with an integrated, efficient, and flexible conversion of the renewable biomass to *VAPs* and commodities, through a combination of biochemical/biological, physical, chemical, and sometimes thermochemical processes (Sadhukhan et al., 2014; Romero-Cedillo et al., 2016) it is very likely that *MESynC* could be integrated/coupled since several waste streams in the biorefinery can become the substrate for *VAPs* and commodities production in such devices (Sadhukhan et al., 2016). This would translate into improved and more sustainable performance of the biorefinery

facility by optimizing the Principle of Cascading (Poggi-Varaldo et al., 2014). Table 1 shows that no work reviewed in this article out of a total 13 cases have dealt with experiments of *MESynC* coupled to waste-based biorefineries, that is, 0%. Clearly, there is a need to conduct further research on this fertile area. We anticipate a blossoming body of research in the not so far future dealing with coupling of *MESynC* to waste-based biorefineries.

PERSPECTIVES AND CONCLUSION

*MESynC*s have a great potential, and this technology could become soon more important in a primary process for the *VAPs* and commodities production. However, *MESynC*s need to overcome large barriers as economic costs, electrode materials, membranes, microorganisms, scale-up, etc. There are still few published works on producing chemical products from microorganisms powered with electricity. Also, considerable further work will be necessary to reach a complete understanding of bacterial-electrode electron flow at a molecular level that will, in turn, be applied for improving development of *MESynC*. On the other hand, there is more information on the bioelectrochemical production of methane and hydrogen, although these compounds are more commodities than typical *VAPs*.

There are few studies regarding the system evaluation of the *MESynC* performance that determine energy efficiency as well as environmental impacts. According to the present review, *LCA* is an accepted method for determining these aspects. Though in the last years have been published some studies, definitive incorporation of *LCA* (as well as mass and energy balances to *MESynC* R&D) is still weak. Past research shows that no work reviewed in this article, out of 14 cases have included LCA or some kind of system analysis of *MESynC*, *i.e.*, a deafening 0%. This gap, along with that of scarce experience with scale up of the processes, should be filled up in the near future in order to demonstrate the sustainability and sound commercialization of *MESynC*.

Since biorefineries are facilities with an integrated, efficient, and flexible conversion of the renewable biomass to *VAPs* and commodities, through a combination of biochemical/biological, physical, chemical, and sometimes thermochemical processes it is very likely that *MESynC* could be integrated/coupled since several waste streams in the biorefinery can become the substrate for *VAPs* and commodities production in such devices. This would translate into improved and more sustainable performance of the biorefinery facility by optimizing the Principle of Cascading (Poggi-Varaldo et al., 2014). Previous research shows that 0% of the total references reviewed in our article have dealt with experiments of *MESynC* coupled to waste-based biorefineries. Clearly, there is a need to conduct further research on this fertile area. We anticipate a blossoming body of research in the not so far future dealing with coupling of *MESynC* to waste-based biorefineries. This integration may lead to accelerated development of sustainable environmental technologies as well as new paradigms in modern societies.

NOTATION

AQDS	anthraquinone-2,6-disulfonate
A_{equip}	external surface area of the equipment, in m^2
BECSR	bioelectrochemical soil-slurry reactors
BES	bioelectrochemical systems
$c_{p\ electrolytes}$	specific heat (constant pressure) of the electrolytes, in J/(kg ^oC)
$c_{p\ equip}$	specific heat of the equipment, in J/(kg ^oC)
EAB	electrochemically-active bacteria
E_{app}	applied voltage to the *MESynC,* in V

EET	extracellular electron transfer
E_{electr}	electrical energy spent in the electrofermentation, in J
E_{emf}	thermodynamic voltage (reversible) determined by the reactions at the electrodes, V
$E_{heating}$	thermal energy required to heat the electrolytes and equipment, as well as the heat losses, in J
E_{mixing}	energy of mixing requirements, if any, in J
$E^{o'}$	standard redox potential at pH = 7.0
I	current intensity in the *MESynC*
LCA	life cycle analysis
MDC	microbial desalination cell
MEC	microbial electrolysis cell
$m_{electrolytes}$	mass of the electrolytes in the cell
m_{equip}	mass of equipment
m_{IS}	mass of uptaken (consumed) substrate
MESynC	microbial electrosynthesis cell
MFC	microbial fuel cell
MRC	microbial remediation cell
m_{US}	mass of uptaken (consumed) substrate, either in kg or mole
$m_{VAP,cf}$	mass of *VAP* produced in the conventional fermentation
$m_{VAP,EF}$	mass of *VAP* produced in the *MESynC*
$m_{VAP,gen}$	mass of *VAP* produced in a fermentation, either in kg or mole
P	power
Pr	product
R&D	research & development
SHE	standard hydrogen electrode
T_{amb}	ambient temperature, either in ^{o}C or K
T_{oper}	operation temperature, either in ^{o}C or K
U	overall heat transfer equipment for heat loss to the surroundings, W/(m^2 K)
VAPs	value-added products
Y	product yield
Y'	product pseudo-yield

Greek characters

η_{cf}	conventional fermentation index
$\eta_{conversion}$	conversion efficiency
η_{Coul}	coulombic efficiency
η_{E}	voltage efficiency
η_{EF}	electrofermentation index
$\eta_{EF/cf}$	efficiency that compares performance of electrofermentation *vs.* conventional fermentation of the same *VAP*
π	volumetric power intensity in mixed vessels
Δt	period of time

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