Prospects in bioelectrochemical technologies for wastewater treatment
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ABSTRACT
Bioelectrochemical technologies are emerging as innovative solutions for waste treatment, offering flexible platforms for both oxidation and reduction reaction processes. A great variety of applications have been developed by utilizing the energy produced in bioelectrochemical systems, such as direct electric power generation, chemical production or water desalination. This manuscript provides a literature review on the prospects in bioelectrochemical technologies for wastewater treatment, including organic, nutrients and metals removal, production of chemical compounds and desalination. The challenges and perspectives for scale-up were discussed. A technological strategy to improve the process monitoring and control based on big data platforms is also presented. To translate the viability of wastewater treatment based on bioelectrochemical technologies into commercial application, it is necessary to exploit interdisciplinary areas by combining the water/wastewater sector, energy and data analytics technologies.

Key words | bioelectrochemical systems, bioenergy, data analytics, water resources wastewater treatment

INTRODUCTION
The management of water resources has been strongly influenced by the water shortage. According to UNESCO, the water demands will increase by 55% by 2050 and, in the industrial field, the water requirements will be 400% higher in the next 50 years (UNESCO 2015). Linked to it, the high costs associated with wastewater treatment call for a consideration of water use rationalization, adequacy of treatment methods and the viability of reuse techniques (Mirre et al. 2011).

Conventional wastewater treatment technologies are focused on purification rather than resource recovery. In the field of biological treatment, the activated sludge process is the most common (Shariati et al. 2011), with alternatives like sequencing batch reactors and membrane bioreactors (Viero et al. 2008). However, these technologies consume large amounts of energy due to the necessity for aeration, in order to provide the amount of oxygen required for microorganisms involved in the process (Ren et al. 2013). Nevertheless, this power consumption can be avoided through the adoption of innovative engineering processes, promoting resources recovery and providing a reusable water resource (Chen et al. 2015). In this context emerges the technologies based on bioelectrochemical systems (BES).

BES have been intensively studied and developed only recently, opening a new interdisciplinary field for research and development through integration of microbiology, electrochemistry, materials science and engineering (Wang & Ren 2015). BES have a range of potential configurations and applications, including wastewater treatment, biofuels production, water desalination, biosensors and as a source of energy power for remote areas (Logan et al. 2015). However, there are several challenges to be overcome in order to translate these technologies into commercial applications. Given these prospects, this paper reviews the potential applications of BES to treat wastewater, focusing on the challenges and opportunities to improve their performance. Also, a technological strategy to improve the process monitoring and control based on big data platforms is presented.

INTEGRATION OF BES INTO WASTEWATER TREATMENT
The basic BES principle is the reaction of microbial oxidation. However, the way electrons are used on the cathode shows how attractive this technology is: any
reduction reaction can be performed in the cathode chamber, creating numerous application options (Figure 1). Thus, BES have been specified into different designations, such as MxC, where M stands for microbial, C stands for cell and x stands for different applications, for example, fuel (microbial fuel cells (MFCs)) or desalination (MDC) (Torres et al. 2010; Luo et al. 2016). Figure 1 shows a representation of these technologies and their main configurations are summarized in Supplementary Information Table S1 (available with the online version of this paper).

Studies on BES have been focused on wastewater treatment, energy recovery, desalination, and synthesis of high-value products (ElMekawy et al. 2014; Majumder et al. 2014; Pandey et al. 2016). There are several effluents with potential application, such as sewage (Kim et al. 2015) and industrial sources (i.e. food and beverages (Dong et al. 2015), paper and cellulose (Cheng et al. 2011), textile (Pushkar & Mungray 2016), agribusiness (Vilajeliu-Pons et al. 2016; Molgononi et al. 2018), pharmaceutical (Zhang et al. 2015), petrochemical (Ashwani & Perumalsamy 2017; Sevda & Abu-Reesh 2017), landfill leachates (Iskander et al. 2016) and, most recently, water desalination (Carmalin Sophia et al. 2016)).

Organic wastes treatment

Organic wastes contain more internal energy than the amount required to treat them. Given a scenario with a daily chemical oxygen demand (COD) per capita contribution from 60 to 120 gCOD inhabitant⁻¹ day⁻¹ and a wastewater energy value of 14.7 kJ gCOD⁻¹ (Heidrich et al. 2011), the expected energy available in this source is from 2.45 to 4.90 × 10¹⁵ kJ year⁻¹ (considering the current world population is 7.6 billion according to the most recent United Nations estimates (https://esa.un.org/unpd/wpp/)). However, this energetic potential is not exploited yet, mainly when traditional technologies are applied to treat wastewater.

Under aerobic conditions, for example, the energetic consumption for aeration systems is around 0.5 kWh m⁻³, which give a per capita consumption of 30 kWh inhabitant⁻¹ year⁻¹ (Wei et al. 2003). Also, these systems produce large quantities of sludge (~0.4 kg of sludge per kg of oxidized COD) (Rabaey & Verstraete 2005). On the other hand, BES consume small amounts of energy and can simultaneously generate useful products, such as electricity, hydrogen, and chemicals, such as carboxylic acids, caustic compounds, acetate, hydrogen peroxide, methane, ethanol, and hydrogen (Xu et al. 2015; Srikanth et al. 2018).

There are numerous possibilities to integrate BES into wastewater treatment plants (WWTPs) (Liu et al. 2016). MFCs can complement biological treatment units, such as activated sludge reactors, generating from 10 to 20% more energy. The low biomass yield is another positive factor, saving costs with sludge disposal. As an MFC is based in a biofilm system, and the cell yield of exoelectrogenic bacteria (0.07–0.16 gVSS gCOD⁻¹ (VSS, volatile suspended solids)) is lower than that from activated sludge (0.35–0.45 gVSS gCOD⁻¹), the sludge production can be reduced by 50 to 70% and, consequently, the WWTP operational costs can be reduced by 20 to 30% (Pant et al. 2010). The integration of an anaerobic fluidized bed membrane bioreactor and MFCs can achieve high levels of COD (92.5%) and total suspended solids (>99.0%) removal (Ren et al. 2014).

Nutrients removal

The removal of nutrients in BES has also been studied (Park et al. 2017; Roustazadeh Sheikhyousefi et al. 2017). Sotres

Figure 1 | Schematic representation of MxC technologies.
et al. (2016) evaluated nitrogen dynamics in an MFC treating swine wastewater. It was observed that nitrification of the diffused ammonia (50.4%) was the main process occurring at the aerated cathode, and denitrification processes reach a nitrate removal efficiency of 41.2% when alternating aeration cycles and acetate were applied. The performance of a microbial electrolysis cell (MEC) for autotrophic nitrate removal from groundwater was assessed by Cecconet et al. (2018). According to the authors, the MEC system achieved a nitrate removal rate of 62 gNO₃-N m⁻³ d⁻¹.

A promising single-chamber MFC was proposed by Wang et al. (2015a, 2015b) to remove organic nitrogen compounds (pyridine and methyl orange). Degradation efficiencies of 82.9% for pyridine (initial concentration of 200 mg L⁻¹) and 90.4% for methyl orange (initial concentration of 50 mg L⁻¹) were achieved. Cusick et al. (2014) investigated the performance of an MEC with a fluidized bed cathode chamber (FBR-MEC) developed to improve phosphorus removal. Results indicated that energy consumption (0.2–0.3 W L⁻¹) was lower than observed in conventional systems (~1.0 W L⁻¹) used to recover struvite (Battistoni et al. 2001).

Metals removal

Metal contamination is an environmental concern, as these compounds are not biodegraded and can be transferred across trophic levels, accumulating in the biota (Nancharaiah et al. 2015). In BES, metals can be spontaneously reduced on the cathode due to favorable half-cell redox relative to organic matter (Cheng et al. 2013; Huang et al. 2015; Nancharaiah et al. 2016a, 2016b). There are several mechanisms related to the cathodic metal recovery, involving the direct metal recovery using biotic or abiotic cathodes, supplemented or not by an external power source. These mechanisms are discussed in detail in the review of Wang & Ren (2014).

Chemical compounds recovery

When BES are designed to produce chemicals, the main advantage is the raw material sources which can be from renewable and/or waste materials (Liu et al. 2010). Carbon dioxide has been captured and used to produce organic compounds like methane (for fuel) and bioplastics (e.g., poly-β-hydroxybutyrate). Organic compounds commonly found in industrial wastewaters have been recovered in the form of ethanol (used, for example, as a biofuel) from acetate and butanol (used, for example, as a biofuel) from butyrate (Rabaey & Rozendal 2010).

Watson et al. (2015) produced hydrogen from a microbial reverse-electrodialysis electrolysis cell treating dark fermentation wastewater utilizing synthetic cellulose as the carbon source. An increase in the current production was observed when the number of cell pairs was duplicated (from 5 to 10 cell pairs). With 10 cell pairs, hydrogen yield was 1.1 L gCOD⁻¹ and the coulombic efficiency was between 69 and 79%. By using acetate as the electron donor, Rabaey et al. (2010) could generate up to 1.05 A in a MEC system and, allowed the caustic production of 3.4 wt%.

Desalination

The MDC appears as an innovative BES option to desalination technologies, as they can convert the energy stored in wastewater directly into electricity and utilize it in situ to drive desalination (Perazzoli et al. 2018). MDCs can be used as either a stand-alone for simultaneous organic and salt removal with energy production or a pretreatment for conventional desalination processes such as reverse osmosis, reducing salt concentration, and minimizing energy consumption and membrane fouling (Li et al. 2017; Ebrahimi et al. 2018). The development of MDC-based systems to treat wastewater integrating both nitrogen removal, electricity generation and desalination is also possible. In this case, the cathodic chamber can be fed with nitrogen ions (e.g. nitrate, nitrite) as electron acceptor. However, studies exploiting this possibility are scarce (Meng et al. 2014; Kokabian et al. 2018).

Energy recovery and costs

BES applications are attractive as a complement to traditional wastewater treatment technologies, reducing energetic requirements as well as recovering resources and synthesizing new products by using wastes as raw material. Several studies have demonstrated the ability of BES to treat wastewater with simultaneous electricity production (Liu et al. 2017; Molognoni et al. 2018). However, the small amount of energy generated would be sufficient only for low-power applications. Thus, the simple production of electricity is not yet economically feasible when comparing well-established processes, such as anaerobic digestion.

Alternatively, it would be an advantage to utilize the electricity to conduct desalination (Al-Mamun et al. 2018;
Ebrahimi et al. 2018) or even to synthesize new products (Watson et al. 2015; Xu et al. 2015). Studies have suggested the integration of BES into the anaerobic processes, improving acidogenesis, and hydrogen or methane production (Liu et al. 2016). An economic comparison of organic removal efficiencies and energy recovered from MFC and MEC treating winery and/or domestic wastewaters was performed by Cusick et al. (2010). At a produced cost of $4.51 kgH₂⁻¹ (all costs are US dollars) for winery wastewater and $3.01 kgH₂⁻¹ for domestic wastewater, hydrogen costs less than its estimated commercial value ($6.0 kgH₂⁻¹). These results show that energy recovery and organic removal from wastewater can be more effective with MFCs than MECS, but hydrogen production from MECS using wastewater as a carbon source can also be cost-effective, based on electrical energy requirements.

CHALLENGES AND PERSPECTIVES INVOLVING SCALE-UP

As a novel technology, there are several challenges to be overcome before the successful BES scale-up and commercialization (Hernández-Fernández et al. 2015; Pandey et al. 2016), requiring a solid integration among academia, research institutions and industry (Nunes et al. 2012). Substantial advances have occurred in the last years; however, the major bottlenecks to scale-up BES include the low power densities, and high capital and operational expenses (Seelam et al. 2018). In fact, to overcome these gaps it is necessary to improve the electron transfer and electrode materials, reduce the costs involving membranes and separators, improve reactor design and also the technologies for process monitoring and control.

Electron transfer and electrodes

The successful BES applications and research efforts require a better understanding of the exoelectrogenic bacteria and their biochemical pathways used to release electrons to the acceptors outside the cell (Logan 2010). There are numerous reviews on this subject of the bacteria in BES (Lamberg & Bren 2016; Qiao et al. 2017; Zhang et al. 2017; Kondaveeti et al. 2018) and, thus, this will not be addressed here.

In general, electrode materials should ideally be biocompatible, conductive, porous, easily made at low cost, recyclable, and scalable. In addition, they should possess high specific surface area, corrosion resistance, and high mechanical strength (Kalathil et al. 2017; Han et al. 2018). When the main goal is to treat wastewater, for example, electrode material costs need to be lower than $110.00 m⁻² to make them economically viable (Sleutels et al. 2012) for this application. Numerous materials have been used as the electrode in BES, including metal, carbon-based and activated carbon (Figure 2).

With recent advances in materials science and nanotechnology, the development of three-dimensional electrodes is promising, for example, graphene-based materials (Tang et al. 2015; Cai et al. 2016), carbon nanotubes (Cui et al. 2015; He et al. 2015), carbon nanofibers (Peng et al. 2016) and composites (Dai et al. 2016; Yasi & Nakhla 2017).

Carbon paste electrodes also can be useful for this application. These electrodes are made from a paste of finely granulated carbon mixed with mineral oil (like Nujol), paraffin oil or silicon grease; where the paste is packed into the electrode body cavity. However, they have the disadvantage of being prone to mechanical damage during use (Lamberg & Bren 2016). For more details, a review of recent developments of anode materials for MFCs is presented by Sonawane et al. (2017).

Membranes and separators

The membrane, as well as structural materials, play a critical role in continuing the development of BES, because the architecture, materials and overall geometry, significantly affect performance levels and unit cost (Santoro et al. 2017). According to Daud et al. (2015), those materials should be inexpensive, low oxygen and fuel crossover, and enable higher proton transfer and long-term stability. In the case of membrane-based technologies, the main challenge comes from membrane fouling (biofilm growth) and scaling (hardness-causing cation deposition) (Song et al. 2017; Rudra et al. 2018).

Porous materials, such as ceramic, terracotta, earthenware, mullite and so many other clay materials, are promising materials. However, the porosity, proton conductivity, and brittleness need to be improved (Winfield et al. 2016). Khallili et al. (2017) evaluated the influence of unglazed ceramics as separators on the performance of dual-chamber MFCs, achieving a maximum power density of 321 mW m⁻². According to the authors, due to the low production cost, high mechanical strength and increased output power density of the MFC, these separators proved to be a suitable alternative to replace costly polymeric membranes.

Chitosan-based membranes have been developed for biofuel cell applications (Ikram et al. 2017). Salar-García et al. (2017) evaluated the performance of silicone, polyvinyl
chloride, colloidal silica and chitosan as the binder for carbon-based cathodes in ceramic MFCs. Colloidal silica and chitosan were the most sustainable options. Cathodes prepared with 2.5 wt% of chitosan reached a maximum power of 510 μW, representing 60.3% of the power output from MFCs with traditional polytetrafluoroethylene (PTFE)-based cathodes. Also, chitosan-based MFCs reached COD removal rates of up to 26%, which was slightly higher than the COD removal rate measured for MFCs using PTFE cathodes (23.5%). As can be observed, these studies report the potential application of alternative materials for advancing BES, as they are not only comparable with conventional ion exchange membranes in terms of performance, but also substantially less expensive.

Reactor design

BES commercialization could be limited by the reduction of the power produced at larger scales. In this case, the major difficulty is to maintain the relationship between reactor geometry and electrode. In experiments assessing different MFC configurations treating brewery and piggery wastewaters, a higher power density of 4.1 W m⁻³ was produced using a 5.7 L reactor with an electrode specific surface area of 62 m² m⁻³ (Zhuang et al. 2022a, 2022b), compared to 1.1 W m⁻³ using a 5.0 L MFC with an electrode specific surface area of 5 m² m⁻³ (Zhao & Song 2014) and 1 W m⁻³ in a 90 L reactor with an electrode specific surface area of 6 m² m⁻³ (Dong et al. 2015). These findings show that electrode surface areas need to be maintained for larger reactor sizes (Logan et al. 2015), thus enabling the system operating performance.

Continuous research is needed to better understand and find appropriate solutions for those gaps addressed here. It is fundamental to develop low-cost and effective materials associated with pilot-scale studies to assess their performance and maintenance at a larger scale, including parameters such as longevity and behavior with variations in wastewater composition and temperature (for example to control fouling on electrodes) (Hatzell et al. 2014).

Process monitoring and control

The successful application of new technologies is highly dependent on the development of reliable strategies for process monitoring, control, and optimization. Nowadays,
smart meters, sensors and real-time control solutions are essential to improve the overall performance of water/wastewater treatment systems (Li 2018). The global market for control and monitoring systems in the water and wastewater sector was $21.3 billion in 2016 and it is estimated to be $30.1 billion in 2021. In the next years, the investments in advanced solutions for data management and analytics are expected to grow by 11.9% a year (Global Water Intelligence 2016).

Due to the dynamic nature of the BES operations, there are numerous variables influencing process performance which need to be closely monitored, such as periods with fluctuations in the volume of influent to be treated, climatic conditions and electricity production variations. Therefore, improved sensors and advanced analytics tools bring possibilities to utilize this information to predict, identify and correct eventual failures that affect the process performance, thus reducing losses and energy consumption (Helmbrecht et al. 2017; Bibri 2018).

Concerning the monitoring processes, it is necessary to collect, store and analyze the data generated during the operational process. Also, these data need to be processed and transformed into high-value information. Due to the low power produced in BES, an alternative to scaling the process is to design blocks containing sets of stacked reactors, in order to reach the power requirements for a given application (Jiang et al. 2013). Thus, the power produced is a function of the total number of electrodes throughout the system as presented in Equation (1):

\[
P_{\text{max}} = \frac{V^2}{R_{\text{in}}} * n
\]

where \(P_{\text{max}}\) (watts) is the maximum power required, \(V\) (volts) is the system voltage, \(R_{\text{in}}\) (ohms) is the internal resistance and \(n\) is the number of electrodes interconnected throughout the system.

For each cell there is a need for at least one sensor (i.e. to monitor the power output), and the amount of data generated can be determined according to the following equation:

\[
D = \frac{P_{\text{max}} R_{\text{in}}}{V^2} * f_s * 8
\]

where \(D\) is the data volume (bytes s\(^{-1}\)), \(f_s\) is the sampling rate (s) and 8 is the volume (bytes) occupied for each sampling point.

Considering a machine with a random-access memory (RAM memory) processing capacity of 8 GB, we assume the following cases:

- Case 1: For systems generating a data volume until 8 GB d\(^{-1}\), data can be processed by conventional platforms;
- Case 2: For systems generating data volumes higher than 8 GB d\(^{-1}\), there is a need to adopt alternative strategies (i.e. distributed systems) to enable the processing of large datasets. In this context, the ‘big data’ platforms emerge as suitable alternatives (Figure 3).

It should be noted that in systems using smart sensors there is a need for high sampling rates with measurement intervals lower than 1 second (Nielsen et al. 2017). Thus, the relationship between the volume of data generated and the power produced in a bioelectrochemical reactor can be graphically represented in Figure 3 (where \(P_{\text{max}} = 3.47\) W; \(R_{\text{in}} = 30\) Ω; \(V = 0.3\) V; \(f_s = 10\) samples s\(^{-1}\)).

Big data platforms are attractive as they offer support to scale a large number of metrics. Also, they are strongly consistent (data will not be lost), fault tolerant (if any component fails, the system will not stop) and can distribute the system in several infrastructures (Oussous et al. 2017). Figure 4 presents a big data-based framework for data management during BES operation.

Data acquisition occurs during the steps of monitoring and control. According to Boe et al. (2010), to develop a suitable strategy for monitoring parameters that reflect the real behavior of the reactor, ensuring precision, stability and easy maintenance is still a big challenge. This is mainly due to the complexity of the organic residues and fluctuations (composition, pH, temperature, conductivity) that occur during the operating cycles (Recio-Garrido et al. 2016). These
fluctuations can result in the system performance drop –
electrical properties (Gonzalez del Campo et al. 2013) and
microbial communities (Ren et al. 2013).

After the acquisition, data are processed, allowing
extraction of knowledge and value. To process big data
with scalability and high performance there are two main
tools: Hadoop MapReduce and Spark. While MapReduce
stores the data on hard disk, Spark does the same operation
in RAM memory. MapReduce is, therefore, suitable for cases
where data operations are static, especially when processing
is done in batches. However, when there is a need for con-
tinuous data analysis (i.e. sensors), Spark is the most
appropriate tool (Günter et al. 2011; Bibri 2018).

Once processed, data are stored in distributed file sys-
tems such as Hadoop distributed file systems. In this step,
files are divided into blocks that can be accessed simulta-
neously and replicated to be fault tolerant (Garcia et al.
2017). All this information is extracted and managed by web
services in the cloud computing (Oussous et al. 2017). Web
servers can operate in remote and customized infrastructure
named cloud services. For example, Amazon Web Services
offer a wide range of computing, storage, database, analysis
and deployment tools, allowing monitoring and analysis of
data in real time, reducing operational costs and developing
high-scalable processes (Amazon Web Services 2018).

Finally, the user interface involves a web navigator or
even tablet or smartphone applications. It is the space
where occur the interactions between user and machine,
allowing effective operation and control, whilst the machine
simultaneously gives feedback of information to help in the
user decision-making process. Here, there are two possibili-
ties. The first involves the development of an exclusive
application to a particular case. The second opens an oppor-
tunity for the use of already available platforms, which may
be customized in accordance with the user needs. The
Grafana platform, for example, enables the analysis and
visualization of metrics. Grafana is used in a wide variety
of areas, such as monitoring of climate change, industrial
sensors, automation and process control for real-time data
visualization (Betke & Kunkel 2017).

The design of smart infrastructures linked with the big
data technologies has the potential to improve the manage-
ment of wastewater treatment systems. Moreover, the small
investments in hardware and software required for cloud-
based computing make possible the implementation of
these technological tools in small- and medium-sized
utilities.

CONCLUSIONS

Bioelectrochemical technologies have potential for waste-
water treatment. Herein, BES applications were
considered as a complement to traditional technologies,
thus reducing the energetic requirements by using wastewater as raw material.

However, there are several challenges to be overcome before BES commercialization. As these technologies have generated small amounts of energy, which would be sufficient only for low-power applications, the simple production of electricity is not economically feasible if compared to well-established processes. Alternatively, it would be an advantage to utilize the electricity generated in situ to drive desalination or to synthesize new products.

On the other hand, the adoption of technological approaches has the potential to improve BES performance. Thus, by exploiting the combination of the water/wastewater sector, energy, information, and data analytics strategies, the wastewater treatment based on BES can be translated into a commercial technology.

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