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Miniature microbial solar cells to power wireless sensor networks

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ABSTRACT

Conventional wireless sensor networks (WSNs) powered by traditional batteries or energy storage devices such as lithium-ion batteries and supercapacitors have challenges providing long-term and self-sustaining operation due to their limited energy budgets. Emerging energy harvesting technologies can achieve the longstanding vision of self-powered, long-lived sensors. In particular, miniature microbial solar cells (MSCs) can be the most feasible power source for small and low-power sensor nodes in unattended working environments because they continuously scavenge power from microbial photosynthesis by using the most abundant resources on Earth; solar energy and water. Even with low illumination, the MSC can harvest electricity from microbial respiration. Despite the vast potential and promise of miniature MSCs, their power and lifetime remain insufficient to power potential WSN applications. In this overview, we will introduce the field of miniature MSCs, from early breakthroughs to current achievements, with a focus on emerging techniques to improve their performance. Finally, challenges and perspectives for the future direction of miniature MSCs to self-sustainably power WSN applications will be given.

1. Introduction

Remarkable advances in portable and wearable electronics and wireless communication open up the new era of the internet of things (IoTs). Billions of wireless physical sensors on human bodies and in unattended working environments are linked to the internet, collecting and sharing real-time information for human safety and security (Dagdeviren et al., 2017; Fan et al., 2016). Complete power autonomy is the most critical requirement for stand-alone, always-on wireless sensor networks (WSNs) (Dahshan 2017; Hu et al., 2011; Wu et al., 2017). Conventional batteries are limited as power supplies because of their finite energy budgets and large size (Dagdeviren et al., 2017; Fan et al., 2016; Hu et al., 2011). Even emerging miniaturized energy storage devices such as supercapacitors and lithium-ion batteries cannot be sole platforms for independent and self-sustainable WSN applications because of their frequent recharging requirements (Pang et al., 2018). A promising solution is to infinitely harvest electricity from ambient sources such as kinetic, thermal, chemical, biological, and solar energy (Dewan et al., 2014). Miniaturizing these energy harvesting technologies can achieve the longstanding vision of self-powered, long-lived WSN deployments, allowing every corner of the world to be connected (Selvan and Ali, 2016). Among these technologies, miniature microbial solar cells (MSCs) can be the most widely distributed, especially for sensor nodes deployed in remote and resource-limited locations (Choi, 2015). MSCs are a bioelectrochemical system in which living microorganisms scavenge electricity continuously from microbial photosynthetic and respiratory activities during the day-night cycle (Rosenbaum et al., 2010; Strik et al., 2011). Because self-repairing and self-maintaining biological entities in the device work with abiotic components as a system that requires only sunlight, water, and carbon dioxide, MSC-powered WSNs can be fully self-sustaining autonomous systems. In particular, MSC miniaturization in a well-controlled microenvironment inherently produces favorable architectural designs to improve integration, performance, and operation of miniature sensing devices (Choi, 2015).

Here we highlight the latest progress in the development of the miniaturized MSCs for self-powered electronics and WSN applications. We will first discuss the MSC technology and limitations for scaling up MSCs to generate high-power output. Then, we will introduce a novel microscale MSC as a new avenue for powering future electronics and

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WSNs. Details of the frontier of research to improve the performance of the miniature MSCs will be discussed, followed by a critical perspective on strategic future directions.

2. MSCs and their limitations

A microbial fuel cell (MFC) is a widely used bioelectrochemical system, which uses microorganisms as the biocatalysts to produce electricity (Logan et al., 2019; Lovley, 2012). MSCs are a type of MFCs that use photosynthetic microorganisms, such as cyanobacteria or microalgae, to produce bioelectric power (Rosenbaum et al., 2010; Strik et al., 2011). Photosynthetic microorganisms use solar energy to split water and reduce carbon dioxide, generating oxygen and carbohydrates that are used for microbial respiratory reaction, which re-generates carbon dioxide and water (Lea-Smith et al., 2016; McCormick et al., 2015). Therefore, MSCs can offer self-sustainable capability without requiring an additional organic fuel. The photosynthetic and respiratory processes involve a cascade of electron transfer processes through redox-active molecules. Under illumination, an alternative electron pathway toward an anode of the MSC can be generated to partially deviate from the photosynthetic electron transport route while the microorganisms harvest electrons by respiring with the anode in oxygen-limited environments to obtain their biological energy (Beauzamy et al., 2020; Lovley, 2012). Moreover, the electrons harvested from microbial respiration can be boosted to higher energies through photosynthesis (Saper et al., 2018). Although an in-depth understanding of the key mechanisms underlying electron harvesting from photosynthesis remains unclear, some details appear in recent review articles (Fischer, 2018; Grattieri et al., 2020; Lea-Smith et al., 2016; McCormick et al., 2015; Tschortner et al., 2019). Then, the electrons move to the cathode through the external electrical circuit, where they re-form H₂O with O2. The photosynthetic and respiratory reactions in MSCs offer advantages over potentially competing sustainable power sources, such as MFCs or photovoltaic cells. MSCs do not require an energy-required feeding system that the MFCs typically need to continuously introduce an organic fuel and MSCs can produce power day and night while conventional photovoltaic cells suffer from interruption or considerably decreased power production at night and on cloudy days. Moreover, photosynthetic microorganisms can reduce atmospheric carbon dioxide (CO2) while generating electricity, which can be a realistic and accessible solution for CO₂ capturing technologies (Liu and Choi, 2020a).

MSCs can be a promising alternative technology that could alleviate energy crises and environmental pollution. However, no one has yet successfully ensured the practical efficacy of the MSCs as a green and clean energy technique suitable for real-world large-scale applications because they demonstrate persistent performance limits and scale-up bottlenecks. Much of this work is in its nascent stages and it may be difficulty to see how MSCs can meaningfully contribute to solving our societal energy and environmental issues in the short term. Even MFC techniques have not gone further than pilot-scale tests because of their low performance, and expensive core parts and materials (Gao et al., 2019).

3. Miniaturizing MSCs

Although macroscale MSCs for power generation and CO₂ reduction may not soon be practically realized, special applications to power battery-reliant small electronics that consume reasonably small amounts of power but require infinite energy, such as stand-alone, self-powered WSN applications, are within reach. Miniaturized MSCs can be a new avenue to energize small scale, low-power electronic devices that detours around the fundamental and practical roadblocks confronting large-scale MSC use. The emerging fields of Micro-Electro-Mechanical Systems (MEMS) and microfluidics can be attractive techniques for the MSC miniaturization as they offer microfabrication, economical mass production, and a controllable microenvironment (Choi, 2015; Choi

et al., 2011). In particular, as the size of the device decreases, surface area-to-volume ratio and mass transfer rate significantly increases, improving performance compared to macro-scale MSCs. However, the MSC miniaturization has many challenges, including microfabrication of an ion exchange membrane, improvement of sustaining capabilities, and enhancement of power performance.

The pioneer miniature MSC was reported by Lin's group in 2006 (Fig. 1a) (Chiao et al., 2006). Their MSC was constructed by stacking a transparent glass cover and a microfluidic Si substrate. The MSC used blue-green algae, *Phylum Cyanophyta*, in a 1 μL anodic chamber to produce light-driven electricity. Although the power density (40 pW/cm²) was too low to find practical applications (Table 1), it was the first demonstration of a miniature MSC that used MEMS and microfluidics, providing a potential platform for microfabrication of miniature MSCs

Howe's group has made significant contributions to the discovery, characterization, and understanding of fundamental electrogenic properties of various photosynthetic microorganisms by using miniature MSCs (Fig. 1b) (McCormick et al., 2015). They demonstrated that the photosynthetic microbial biofilms could generate electricity without exogenous mediators (Table 1) (McCormick et al., 2011). On a transparent, indium tin oxide anode in a miniature MSC, green algal or cyanobacterial biofilms were formed to generate self-sustained electricity for several weeks and run a digital clock. Recently, his group significantly enhanced the power generation (10 μ W/cm²) by integrating the MSC into a microfluidic device (Table 1) (Bombelli et al., 2015). A large surface area to volume ratio of the microfluidic MSC reduced the physical proximity of the microbial cells to the anode, leading to low internal resistance of the system and increasing its power.

For the last several years, our group has advanced techniques and applications of miniature MSCs (Fig. 1c & Table 1) (Choi, 2015). In 2014, we created our 1st generation miniature MSC which contains a typical two-chambered configuration integrating a thin gold film as a transparent electrode (Yoon et al., 2014). The power generation (7.09 nW/cm²) was much more powerful than the pioneer MEMS MSC. Our 2nd generation miniature MSC was based on a single-chambered configuration with a face-up anode to maximize the capture of solar energy (Lee and Choi, 2015). Furthermore, we provided a microfluidic space for the air-bubble trap to facilitate a gas exchange to the photosynthetic microorganisms. The device substantially increased power density up to 90 nW/cm² and 4-day sustainable operation. Our 3rd generation MSC revolutionized the performance by using an affinitive nanostructured anodic surface with superior electrocatalytic activity for greater electron transfer efficiency and integrating a microfluidic headspace to store gases necessary for bacterial metabolism (Wei et al., 2016). The device generated a maximum power density of 2.7 μ W/cm², which was a million times greater than the pioneer MEMS MSC and was sustained for 10 days. Recently, we revolutionized the performance of the miniature MSC (4th generation) and attained 43.8 µW/cm² of maximum electrical power and long-term operational capability (>20 days) with \sim 18.6 μ W/cm² during the day and \sim 11.4 μ W/cm² at night, which can provide a practical and sustainable power supply for some low-power lab-on-a-chip applications (Liu and Choi, 2017a). This remarkable performance was achieved by creating a 3-D conductive polymer-coated anode and a gas-permeable configuration in a well-controlled, tightly enclosed micro-chamber. The 3-D porous anode ensured a large surface area for the bacterial attachment and efficient mass transfer to and from the anode, which increased the power.

Other opportunities are to use miniature MSCs to study the behavior of individual photosynthetic microorganisms, providing excellent control of a smaller group of cells in a microenvironment. Several research groups used miniature MSCs for direct visualization of bacterial biofilm with simultaneous measurement of bacterial electrogenicity. The Fisher group developed a transparent miniature MSC that simultaneously enabled fluorescence imaging and current measurement (Inglesby et al., 2013). They showed that temperature and light intensity affected

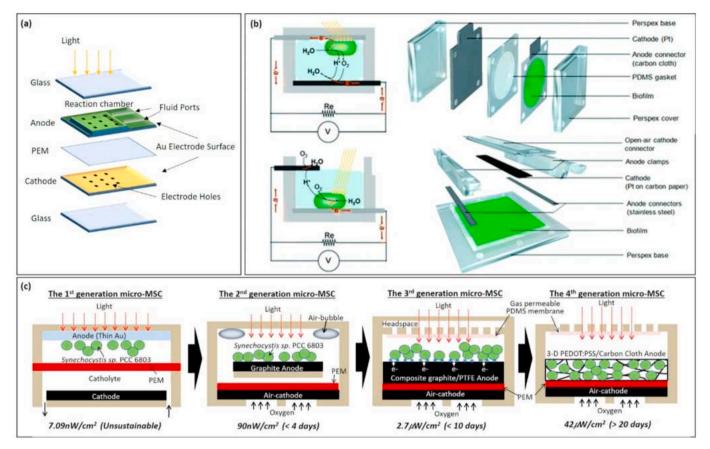


Fig. 1. (a) A MEMS-based MSC, (b) closed and open-air single-chamber MSCs, and (c) technological advancements in miniature MSCs. Reprinted with permission from (b) ref 25 and (c) ref 30, respectively. Copyright (b) 2011 and (c) 2017 The Royal Society of Chemistry, respectively.

electrical performance and uncovered a correlation between the fluorescence signal and electricity generation during the microbial photosynthesis. Our group developed a miniature MSC for simultaneous optical and electrochemical monitoring of photosynthetic microorganisms (Fraiwan and Choi, 2015; Liu and Choi, 2017b). A three-electrode configuration with a reference electrode provided reliable and accurate measurements of electrochemical activity of photosynthetic bacteria. At the same time, the transparent device allowed the direct microscopic visualization of bacteria at different stages of biofilm development. Multi-channel miniature MSC arrays were developed by research groups to compare quantitatively the performance of various microorganisms. Bombelli et al. created a five-channel MSC array that was sealed with polydimethylsiloxane (PDMS) (Bombelli et al., 2012). They showed how anodic materials can affect electrical performance, biofilm growth rate, and the kinetics of photo-bio-electrochemical activity. Our group developed a miniaturized nine-well parallel analyses platform for screening and characterization of the electrogenic capacity of eight microbial consortia and investigation of how light affects on their bioelectricity generation (Fraiwan et al., 2014).

4. Toward high-performance miniature MSCs

Despite these major improvements, the MSC performance is insufficient for applied use, and entirely self-sustainable power generation for WSNs is not possible. Given that the wireless sensors most commonly used for environmental monitoring require sustainable milliwatt-level power, it is not feasible to use the current format of miniature MSCs to continuously power remote WSN applications. This section reviews innovative approaches to revolutionize the performance of miniature MSCs and attain high power and long-term operational capability, which will provide a practical and sustainable power supply for WSN

applications.

4.1. Co-cultured MSCs

Normally, the electricity harvesting efficiency of photosynthetic microorganisms in MSCs is extremely low compared to the heterotrophic bacteria used in MFCs (Rosenbaum et al., 2010). Moreover, our ability to harness the potential of MSCs lags because we lack an in-depth understanding of the mechanism for electron harvesting from the photosynthetic microorganisms and the fundamental factors that maximize their power-generating capabilities. Recently, the "Plug and Play" co-cultured concept has been proposed to improve the power and life-time of the MSC (Jones, 2012). This is based on synergistic cooperation between heterotrophic and photosynthetic microorganisms where well-studied heterotrophic microorganisms generate power through their respiration while their metabolism and growth are self-sustainably supported by solar-driven photosynthesis of the photosynthetic microorganisms (Badalamenti et al., 2014; Nishio et al., 2013). Our group integrated, for the first time, these mixed microbial communities into a miniature MSC for sustainable high-power generation (Fig. 2a) (Liu and Choi, 2017b). The power generated from the mixed culture was about 70 times greater than that of the MSC with only photosynthetic microorganisms (Table 1). Moreover, the power generation in a microliter-sized MSC was self-sustainable for more than 13 days while the same size of the MFC with only heterotrophic culture lasted for a few hours. However, its power generation decreased after 13 days of operation because of imbalanced bacterial growth and reproduction originated from the microbial competition between physically contacted co-cultures. Recently, we developed a solid-phase MSC platform by separating the two bacterial habitats but allowing the interchange of their nutrients and gases (Fig. 2b) (Mohammadifar et al.,

Summary of the characteristics and performances of miniature MSCs. Output power densities were normalized by the anode area

Early Double-chambered Phylum Cyanophyta 1.5 cm x 1.5 cm x 2.5 0.04 nW/m2 At coated silkon On the chamber of mm 1.5 cm x 1.5 cm x 2.5 0.04 nW/m2 At coated silkon McCormid ce al. (% of met		MSC Configuration	Bacteria type	Dimension	Performance	Electrodes (anode/cathode)	References
Single-chambered Synechocystis sp. PCC 6803 O4 µL 10.5 mW/m2 ITO-PET/Pt-carbon paper Single-chambered Synechocystis sp. PCC 6803 0.4 µL 10.5 mW/m2 InSubil alloy/Pt Single-chambered Synechocystis sp. PCC 6803 90 µL 0.9 mW/m2 Gold/Gold Single-chambered Synechocystis sp. PCC 6803 90 µL 0.9 mW/m2 Gold/Gold Single-chambered Synechocystis sp. PCC 6803 90 µL 0.9 mW/m2 Gold/Gold Single-chambered Synechocystis sp. PCC 6803 90 µL 43.8 µW/cm2 Gold/Gold Single-chambered Synechocystis sp. PCC 6803 90 µL 43.8 µW/cm2 Gold/Gold Co-cultured Single Synechocystis sp. PCC 6803 90 µL 43.8 µW/cm2 Graphite (PTE) anotic Co-cultured Single Synechocystis sp. PCC 6803 90 µL 43.8 µW/cm2 Graphite ink/Ag2O-graphite ink Co-cultured Single Synechocystis sp. PCC 6803 90 µL 38. µW/cm2 Graphite ink/Ag2O-graphite ink Super-apacitive double Synechocystis sp. PCC 6803 90 µL 38. µW/cm2 Graphite ink/Ag2O-graphite ink	Early Breakthroughs	Double-chambered	Phylum Cyanophyta	$1.5~cm\times1.5~cm\times2.5~mm$	0.04 nW/cm2	Au coated silicon	Chiao et al. (2006)
Single-chambered Synechocystis sp. PCC 6803 0.4 µL 105 mW/m2 InSnBi alloy/Pt Double-chambered Synechocystis sp. PCC 6803 370 µL 6.05 µW/m2 7.09 mW/m2 Garbon cloth Air Cathode Single-chambered Synechocystis sp. PCC 6803 90 µL 6.05 µW/m2 Garbon cloth Air Cathode Single-chambered Synechocystis sp. PCC 6803 90 µL 8.05 µW/m2 Garbon cloth Air Cathode Single-chambered Synechocystis sp. PCC 6803 90 µL 8 mA/cm2 Garbon cloth Air Cathode Co-cultured Single Synechocystis sp. PCC 6803 90 µL 8 mA/cm2 Garbon cloth Air Cathode Co-cultured Single Synechocystis sp. PCC 6803 90 µL 8 mA/cm2 Garbon cloth Air Cathode Co-cultured Single Synechocystis sp. PCC 6803 90 µL 8 mA/cm2 Graphite ink/agO-graphite ink Co-cultured Single Synechocystis sp. PCC 6803 90 µL 38 µW/cm2 Graphite ink/agO-graphite ink Supercapacitive double Synechocystis sp. PCC 6803 0 µL 162 µW/cm2 Garbon Honorysis on carbon cloth/Air Cathode Superchased Synechocystis sp. PCC 6803	,	Single-chambered	Synechocystis sp. PCC 6803, Synechococcussp. WH 5701	12,560 mm3	0.3 mW/m2	ITO-PET/Pt-carbon paper	McCormick et al. (2011)
Double-chambered Synechocystis sp. PCC 6803 57 μL each chamber 7.09 nW/cm2 Gold/Gold Gold Gold Gold Gold Gold Gold Gold		Single-chambered	Synechocystis sp. PCC 6803	0.4 μL	105 mW/m2	InSnBi alloy/Pt	Bombelli et al. (2015)
Single-chambered Synechocystis sp. PCC 6803 300 µL 6.05 µW/mz graphite/polyterathoroethylene (PTFE) anodic single-chambered Synechocystis sp. PCC 6803 and Shewanella Single-chambered Synechocystis sp. PCC 6803 and Shewanella Synechocystis sp. PCC 6803 and Shewanella Super-capactitive double synechocystis sp. PCC 6803 and Shewanella Super-based Synechocystis sp. PCC 6803 and Shewanella Super-based Synechocystis sp. PCC 6803 and Shewanella Super-based Synechocystis sp. PCC 6803 and Shewanella Synechocystis sp. PCC 6803 and Shewanella Super-based Synechocystis sp. PCC 6803 and Shewanella Shewanella Synechocystis sp. PCC 6803 and Shewanella Shewanel		Double-chambered	Synechocystis sp. PCC 6803	57 μL each chamber	7.09 nW/cm2	Gold/Gold	Yoon et al. (2014)
Single-chambered Synechocystis sp. PCC 6803 and Shewanella Single-chambered Synechocystis sp. PCC 6803 and Shewanella Co-cultured single-chambered Synechocystis sp. PCC 6803 and Shewanella Optil. A3.8 µW/cm2 PEDOT:PSS Carbon Cloth/Air Cathode Co-cultured single-chambered Synechocystis sp. PCC 6803 and Shewanella Optil. A3.8 µW/cm2 Carbon cloth/Air Cathode Co-cultured single Synechocystis sp. PCC 6803 and Shewanella Shewanella Synechocystis sp. PCC 6803 and Shewanella Shewanella Synechocystis sp. PCC 6803 and Shewanella		Single-chambered	Synechocystis sp. PCC 6803	300 hT	0.9 mW/m2	Carbon cloth/Air Cathode	Lee and Choi (2015)
Single-chambered Synechocystis sp. PCC 6803 and Shewanella Co-cultured single- oneidensis MR-1 Co-cultured Single Synechocystis sp. PCC 6803 and Shewanella Co-cultured Single oneidensis MR-1 Co-cultured Single Synechocystis sp. PCC 6803 and Shewanella Synechocystis sp. PCC 6803 and Shewanella Co-cultured Single chambered Synechocystis sp. PCC 6803 Synechocystis		Single-chambered	Synechocystis sp. PCC 6803	7п 06	6.05 μW/cm 2	graphite/polytetrafluoroethylene (PTFE) anodic materials/Air Cathode	Wei et al. (2016)
Co-cultured single- Synechocystis sp. PCC 6803 and Shewanella 90 μL 8 mA/cm2 Carbon cloth/Air Cathode chambered chambered chambered chambered some step one idensis MR-1 Synechocystis sp. PCC 6803 and Shewanella one idensis MR-1 3.5 cm × 2.4 cm × 0.7 5.64 μW/cm2 Graphite ink/Ag2O-graphite ink Co-cultured Single chambered some chambered stambered supportive double- Synechocystis sp. PCC 6803 90 μL 162 μW/cm2 PEDOT:PSS on carbon cloth/Air Cathode Supercapacitive double- Synechocystis sp. PCC 6803 90 μL 162 μW/cm2 PEDOT:PSS on carbon cloth/Air Cathode Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS on carbon paper-Processed PEDOT:PSS on carbon paper-Processed Paper-based Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS paper/PEDOT:PSS + Ag2O paper Microfluidic channel Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS paper/PEDOT:PSS + Ag2O paper Microfluidic channel Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 11 nmm × 11 mm × 11 mm × 12 mm		Single-chambered	Synechocystis sp. PCC 6803	Ju 06	43.8 µW/cm2	PEDOT:PSS Carbon Cloth/Air Cathode	Liu and Choi (2017a)
Synechocystis sp. PCC 6803 and Shewanella oneidensis MR-1 3.5 cm × 2.4 cm × 0.7 5.64 μW/cm2 Graphite ink/Ag2O-graphite ink oneidensis mR-1 Synechocystis sp. PCC 6803 90 μL 38 μW/cm2 PEDOT:PSS on carbon cloth/Air Cathode Synechocystis sp. PCC 6803 90 μL 162 μW/cm2 PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Cathode Synechocystis sp. PCC 6803 2 cm × 2 cm 0.38 mW/m2 Carbon Manotubes/Carbon paper-Pt Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS paper/PEDOT:PSS + Ag2O paper Synechocystis sp. PCC 6803 250 μm × 6500 μm × 6500 μm × 6500 μm × 10.7 μW/cm2 0.5 W/m2 Indalloy 19/platinum wire Chlorella sp. (ATCC14854) N/A 150 nW/m2 ITO-PET/ITO-PET NA 11 mm × 11 mm × 5 6.15 mW/m2 Au and ZnO anode/Air-cathode	Latest Progresses	Co-cultured single-		7п 06	8 mA/cm2	Carbon cloth/Air Cathode	Liu and Choi (2017b)
one idensis MR-1 cm 38 μW/cm2 PEDOT:PSS on carbon cloth/Air Cathode Synechocystis sp. PCC 6803 90 μL 162 μW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 0.38 mW/m2 Carbon Monotubes/Carbon paper-Pt Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Synechocystis sp. PCC 6803 25 μm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Au and ZnO anode/Air-cathode 11 mm × 11 mm × 11 mm × 5 6.15 mW/m2 Au and ZnO anode/Air-cathode		Co-cultured Single		$3.5 \text{ cm} \times 2.4 \text{ cm} \times 0.7$	5.64 µW/cm2	Graphite ink/Ag2O-graphite ink	Mohammadifar et al.
Synechocystis sp. PCC 6803 90 μL 38 μW/cm2 PEDOT:PSS on carbon cloth/Air Cathode Synechocystis sp. PCC 6803 90 μL 162 μW/cm2 PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Synechocystis sp. PCC 7002 N/A 0.38 mW/m2 Carbon PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 μW/cm2 PEDOT:PSS/MnO2/CNTs Ternary Nanocomposite/Air Chlorella sp. (ATCC14854) N/A 0.5 W/m2 Indalloy 19/platinum wire Au and ZnO anode/Air-cathode 11 mm × 11 mm × 11 mm × 5 6.15 mW/m2 Au and ZnO anode/Air-cathode		chambered	oneidensis MR-1	cm			(2020)
tive double- Synechocystis sp. PCC 6803 90 µL 162 µW/cm2 PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Synechococcus sp. PCC 7002 N/A Carbon Carbon Nanotubes/Carbon paper-Pt Synechocystis sp. PCC 6803 2cm × 2 cm 10.7 µW/cm2 PEDOT: PSS paper/PEDOT:PSS + Ag20 paper channel Synechocystis sp. PCC 6803 250 µm × 6500 µm × 0.5 W/m2 Indalloy 19/platinum wire 25 µm LS µm 150 nW/m2 ITO-PET/ITO-PET site photoanode Synechocystis sp. PCC 6803 11 mm × 11 mm × 5 6.15 mW/m2 Au and ZnO anode/Air-cathode		Supercapacitive double- chambered	Synechocystis sp. PCC 6803	7п 06	38 µW/cm2	PEDOT:PSS on carbon cloth/Air Cathode	Liu and Choi (2019)
Synechococcus sp. PCC 7002 N/A 0.38 mW/m2 Carbon Nanotubes/Carbon paper-Pt Synechocystis sp. PCC 6803 2 cm × 2 cm 10.7 µW/cm2 PEDOT: PSS paper/PEDOT: PSS + Ag2O paper channel Synechocystis sp. PCC 6803 250 µm × 6500 µm × 0.5 W/m2 Indalloy 19/platinum wire 25 µm 25 µm 25 µm 150 mW/m2 ITO-PET/ITO-PET nim 11 mm × 11 mm × 11 mm × 5 6.15 mW/m2 Au and ZnO anode/Air-cathode		Supercapacitive double- chambered	Synechocystis sp. PCC 6803	7п 06	162 µW/cm2	PEDOT:PSS/MnO2/CNTs Temary Nanocomposite/Air Cathode	Liu and Choi (2020b)
Synechocystis sp. PCC 6803 $2 \text{ cm} \times 2 \text{ cm}$ $10.7 \mu\text{W/cm}2$ PEDOT: PSS paper/PEDOT: PSS + Ag2O paper channel Synechocystis sp. PCC 6803 $250 \mu\text{m} \times 6500 \mu\text{m} \times 0.5 \text{W/m}2$ Indalloy 19/platinum wire $25 \mu\text{m}$ $25 \mum$		Paper-based	Synechococcus sp. PCC 7002	N/A	0.38 mW/m2	Carbon Nanotubes/Carbon paper-Pt	Sawa et al. (2017)
channel Synechocystis sp. PCC6803 250 μ m × 6500 μ m × 0.5 W/m2 Indalloy 19/platinum wire 25 μ m Chlorella sp. (ATCC14854) N/A 150 μ m × 11 μ m × 11 μ m × 12 μ m Au and ZnO anode/Air-cathode mm		Paper-based	Synechocystis sp. PCC 6803	$2 \text{ cm} \times 2 \text{ cm}$	10.7 µW/cm2	PEDOT: PSS paper/PEDOT:PSS + Ag20 paper	Liu and Choi (2020c)
25 μm Chlorella sp. (ATCC14854) N/A 150 μm Site photoanode Synechocystis sp. PCC 6803 11 μm × 11 μm × 5 6.15 μm /m2 Au and ZnO anode/Air-cathode mm		Microfluidic channel	Synechocystis sp. PCC6803	250 μ m \times 6500 μ m \times	0.5 W/m2	Indalloy 19/platinum wire	Saar et al. (2018)
Chlorella sp. (ATCC14854) N/A 150 nW/m2 ITO-PET/ITO-PET site photoanode Synechocystis sp. PCC 6803 $11 \text{ mm} \times 11 \text{ mm} \times 5$ 6.15 mW/m2 Au and ZnO anode/Air-cathode mm				25 µm			
Synechocystis sp. PCC 6803 $11 \text{ mm} \times 11 \text{ mm} \times 5$ 6.15 mW/m2 Au and ZnO anode/Air-cathode mm		Microcavity	Chlorella sp. (ATCC14854)	N/A	150 nW/m2	ITO-PET/ITO-PET	Roxby et al. (2020)
mm		Nanocomposite photoanode	Synechocystis sp. PCC 6803	$11~\text{mm} \times 11~\text{mm} \times 5$	6.15 mW/m2	Au and ZnO anode/Air-cathode	Kim et al. (2020)
				mm			

2020). Agar-based analyte, catholyte, and salt-bridge provided solid-state ionic environments for more efficient syntrophic interactions between co-cultures without microbial competition, further enhancing the device's performance and lifespan (Mohammadifar et al., 2019, 2020). The device was sealed with a gas-permeable PDMS membrane. The permeability allowed a gas exchange to the bacteria, which facilitated cathodic reactions. The exchange ideally allows for replenishing bacterial necessary gasses from environments for self-sustainable energy harvesting. A DC-DC booster circuit and a light-emitting diode (LED) were powered only by our MSC without any other power source or fluidic feeding system (Table 1). Zhu et al. introduced a synthetic two-species microbial consortium into an MSC where engineered cyanobacteria generated p-lactate, which provided organic fuel and energy for engineered heterotrophic bacteria, thus improving the efficiency and longevity of the MSC (Fig. 2c) (Zhu et al., 2019). Although their MSC was not a miniatured platform, it is very meaningful because it provides a method to improve the electricity-producing activity of weak exoelectrogenic cyanobacteria.

4.2. Supercapacitive MSCs

Recently, extensive research has been made to improve the power density of bioelectrochemical systems without sacrificing their high energy capability (Dubal et al., 2017; Yan et al., 2014). This path toward a high power bioelectrochemical system is undergoing exciting development with an entirely new kind of electric power device. Researchers have built a supercapacitive bioelectrochemical system that generates energy from enzymatic fuel cells and MFCs, which is simultaneously stored within an integrated supercapacitor (Chen, 2017; Dubal et al., 2015; Pankratov et al., 2014). This innovative platform proves the possibility that even low-power MSCs can boost their performance to levels comparable with conventional batteries and other energy storage devices. Several research groups created a double-functional bioanode of the bioelectrochemical systems, which concurrently perform bioenergy harvesting and store the generated electric charges (Alsaoub et al., 2017; Chen, 2017; Dubal et al., 2015; Houghton et al., 2016; Pankratov et al., 2014a, 2014b; Xiao et al., 2017). This supercapacitive bioanode generates short but strong discharge pulses after it is charged by the bioelectrochemical system. This approach is gaining acceptance as a future energy technique, especially in the field of bioelectrochemical systems. Several hybrid energy devices have been developed for simultaneous energy conversion and storage (Alsaoub et al., 2017; Chen, 2017; Dubal et al., 2015; Houghton et al., 2016; Pankratov et al., 2014a, 2014b; Xiao et al., 2017). Even a couple of sub-cellular MSCs that use thylakoid or isolated photosystems have been integrated with the internal supercapacitors (Gonzalez-Arribas et al., 2017; Pankratova et al., 2017). However, the development of cellular MSCs has not been reported until recently. Our group demonstrated the first miniature supercapacitive MSC with whole photosynthetic microorganisms, producing stable high power and current density (38 μ W/cm² and 120 μ A/cm²) that can be a potential energy solution for the WSN applications (Fig. 3a and Table 1) (Liu and Choi, 2019). Moreover, the hybrid device could improve its performance by connecting multiple miniature units in series. Even more, considerable power enhancement of the supercapacitive MSC has been achieved by improving the supercapacitance of the dual-functional bioanode (Fig. 3b) (Liu and Choi, 2020b). A ternary nanocomposite of poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), manganese dioxide (MnO2), and carbon nanotubes (CNTs) with the cyanobacteria exhibited an excellent specific capacitance, leading to a maximum power density of 160 μ W/cm² and a maximum current density of 450 μ A/cm² at a pulse time of 0.1s, which was significantly greater than all existing miniature MSCs (Table 1). This hybrid device represents an important and novel technology breakthrough that offers a potentially viable long-term and high-performance resource for the WSN applications.

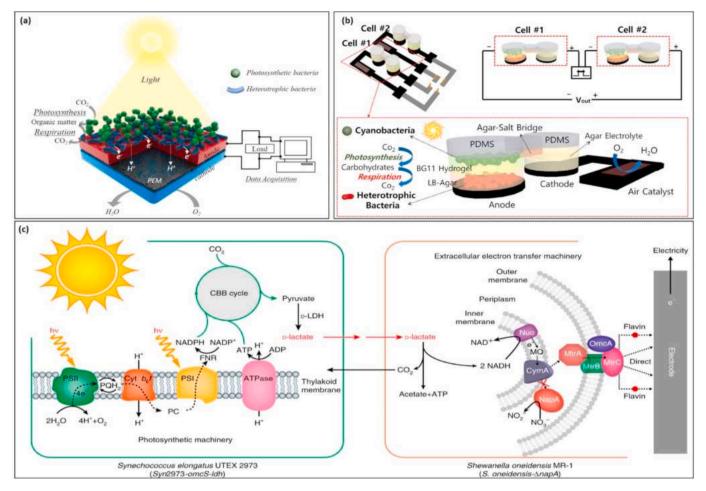


Fig. 2. (a) A miniature MSC with a mixed two-species microbial consortium in a single chamber, (b) a miniature MSC with two microbial strains separately placed in their agar layers, and (c) a mascroscale MSC with a two-species engineered microbial consortium. Reprinted with permission from (a) ref 32, (b) ref 39, and (c) ref 41. Copyright (a) 2017 Elsevier, (b) 2020 Elsevier, and (c) 2019 Nature publishing group.

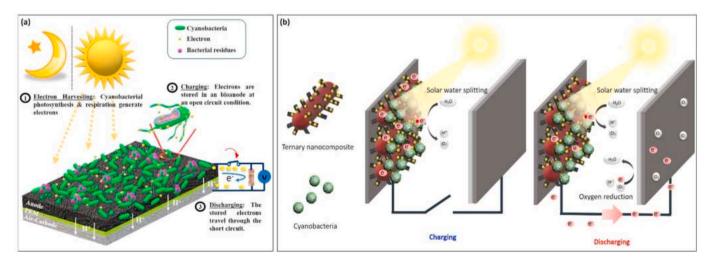


Fig. 3. (a) A supercapacitive MSC with a PEDOT:PSS anode, and (b) a supercapacitive MSC with a ternary nanocomposite anode of PEDOT:PSS, MnO₂, and carbon nanotube. Reprinted with permission from (a) ref 53, and (b) ref 54. Copyright (a) 2019 Elsevier, and (c) 2020 American Chemical Society.

4.3. Paper-based MSCs

Conventional miniature MSCs show a very long start-up time because they use gravity-induced cell deposition, which requires a long time to accumulate and acclimate bacterial cells on the anode. Moreover, many components demand manual assembly of the miniature MSCs, hampering batch fabrication. Recently, Sawa et al. demonstrated an innovative approach to address those issues (Fig. 4a) (Sawa et al., 2017). They used a simple inkjet printer to print bacterial cells onto paper, which has the great potential for device miniaturization, batch

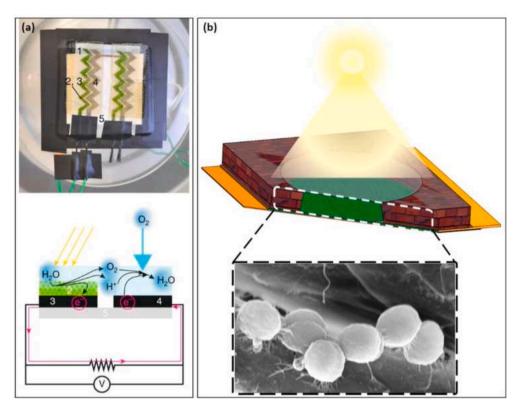


Fig. 4. (a) A MSC with digitally printed cyanobacteria on paper, and (b) a paper-based MSC. Reprinted with permission from (a) ref 55. Copyright (a) 2017 Nature publishing group.

production, and simple cell loading. The printed cells generated sustained electricity powering a small digital clock or low-power LED light (Table 1). Our group pioneered paper-based bioelectrochemical systems and initiated the field of paper bioenergy (Cho et al., 2020; Gao and Choi (2017), 2018, 2019; Mohammadifar and Choi., 2017, 2018; Tahernia et al., 2020a, 2020b). Instead of typical rigid materials (e.g. glass, plastic, or silicon), using paper as a device substrate allows rapid adsorption of bacterial cells through capillary force, reducing the start-up time (Fraiwan and Choi, 2014; Fraiwan et al., 2016). Furthermore, miniature devices can be easily batch-fabricated by using mature manufacturing processes such as screen-printing, dipping, coating, laminating and wax printing (Gao and Choi (2017)). A paper-based MSC was developed, in which porous, hydrophilic, and conductive paper reservoirs were formed by using water-dispersed conducting polymer mixture (Fig. 4b) (Liu and Choi, 2020c). The MSC generated a maximum current of 65 μ A/cm² and a power density of 10.7 μ W/cm² (Table 1).

4.4. Other MSCs

There are many other research efforts to improve the performance of miniature MSCs, which cannot be categorized into one of the subsections defined above. Saar et al. proposed an innovative miniature MSC that separates the energy harvesting process from power delivery in a two-chamber microfluidic device (Fig. 5a) (Saar et al., 2018). In one chamber, electrons harvested from cyanobacterial photosynthesis are stored in electron carrier molecules that flow to another chamber for electron transfer to the external anode. This decoupled operation allowed independent optimization of the energy conversion and the power delivery, improving the overall power output to 50 $\mu W/cm^2$ (Table 1). Roxby et al. proposed a new idea to increase the electricity of miniature MSCs by confining photosynthetic microorganisms in a Fabry-Perot microcavity (Fig. 5b) (Roxby et al., 2020). This technique could generate a strong energy coupling between the microcavity and photosynthetic resonance.

Kim et al. developed a miniature MSC that included a nanocomposite photoanode composed of Au nanoparticles and ZnO nanorods (Kim et al., 2020). The photoanode allowed multiplex energy harvesting of broadband regions of light, generating greater power than the control photoanode of indium tin oxide (ITO). Although reported work about MSCs using nanomaterials is quite limited, some lessons from other bioelectrochemical systems can apply to miniature MSCs (Mouhib et al., 2019). In particular, bacterial metallic nanoparticles synthesized from bacterial metabolism are garnering considerable attention because of their high potential for facilitating microbial extracellular electron transfer at the cell-electrode interface and their bottom-up manufacturing technique (Chen et al., 2018; Cheng et al., 2017; Deng et al., 2020; Dundas et al., 2018; Qian et al., 2015; Wu et al., 2011). Many nanoparticles can be biosynthesized directly on the surface of bacteria through their bioelectrochemical reduction of a chemical solution (Dahoumane et al., 2017; Kulkarni and Muddapur, 2014; Zhang et al., 2018a). Even many photosynthetic microorganisms can biosynthesize novel nanoparticles that improve light-harvesting efficiency and extracellular electron transfer (Hamida et al., 2020). These particles efficiently harvest sunlight and generate "hot" electrons via inter-band transition, and they extracellularly facilitate the transfer of all electrons produced from nanoparticle photosensitization and bacteria metabolism. Even non-photosynthetic bacteria could generate photocurrent through the Au nanoparticles biosynthesized in the bacterial cells (Fig. 5c) (Zhang et al., 2018b).

The low power performance of existing miniature MSCs may limit their ability to power useful WSN applications. The typical sustainable voltage and power outputs from a single miniature MSC are on the order of 0.1–0.5V and 10–100 μW , respectively. Most conventional WSN applications require voltage and power on the order of >1.5V and >1 mW for far-field wireless transmission of collected information or other energy-consuming functions. To produce sufficient voltage and power for residing within the operating range of electronics, arranging multiple miniature MSCs in an array is the most practical approach. Our group

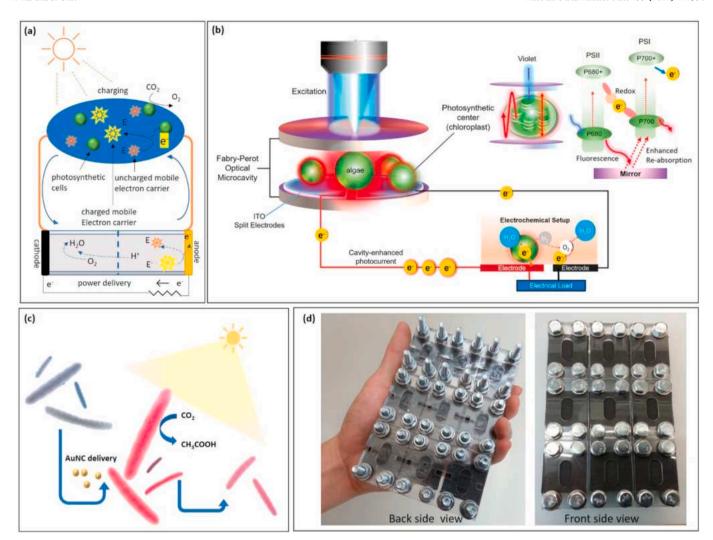


Fig. 5. (a) Decoupling storage and power delivery in a microfluidic MSC, (b) confinement of microalgae in an optical micro/nanocavity, (c) photosynthesis from non-photosynthetic bacterium by using intracellular Au nanoparticles, and (d) a miniature microbial solar panel. Reprinted with permission from (b) ref 64, and (d) ref 28. Copyright (b) 2020 Wiley, and (d) 2015 The Royal Society of Chemistry.

developed a 3×3 MSC panel that could be scalable (Fig. 5d) (Lee and Choi, 2015; Wei et al., 2016). Nine MSCs were integrated in a panel along with a common microfluidic channel for inoculum introduction. Although the power and output voltage were not sufficient for real-world applications, this work demonstrated the possibility of the microbial solar panel and established a standardized platform for its microfabrication.

5. Future directions

Various energy harvesting techniques have been well explored for stand-alone, always-on WSN applications over the past decade. Among possible technologies, miniature MSCs can offer the most suitable power source for WSNs deployed in nature environments because photosynthetic microorganisms can self-sustainably convert ambient light and water into electrical energy. Given that 75 billion sensing nodes are expected to be deployed in the next five years, conventional inorganic solar cells are sub-optimal as their power solution due to their high price and because they are subject to interruption or significantly reduced power production at night and on cloudy days. MSCs can generate electricity continuously from microbial photosynthetic and respiratory activities under day-night cycles in a cost effective and self-sustainable manner, which requires only sunlight, water, and carbon dioxide. Moreover, this technique resembles the earth's natural ecosystem,

where living organisms work with non-living components as a system to self-assemble, self-repair, and self-maintain operational capabilities. In particular, MSC miniaturization inherently produces favorable conditions for improving performance by effectively maximizing bacterial activities in a well-controlled, tightly enclosed micro-chamber. Despite the significant advances in miniature MSCs, when they are deployed in harsh environments, they can suffer from frequent functional failure caused by external damage that interferes with their requirement for a constant supply of water for microbial photosynthesis. Although recent all-solid-phase concepts using hydrogel made the MSC compatible with solid-state WSN devices (Mohammadifar et al., 2020), there is a need for device and material breakthroughs to generate a stable power with a longer lifetime, especially in wild environments. The hydrogel-based device platforms will be vulnerable to external mechanical damages over time, leading to deterioration, malfunction, and ultimately disappearance of their power-generating capability. Furthermore, in water-limited environments, evaporation will stiffen and dry the hydrogel. A self-healing material with hydroscopic properties could make the miniature MSC fully self-repairing and self-sustaining autonomous system in unattended harsh environments.

6. Conclusion

Miniature MSCs can be the most suitable power source for stand-

alone and always-on WSN applications because the technique resembles the earth's natural ecosystem where abiotic and biotic components work closely together to generate self-repairing and self-sustainable features. The miniature MSCs can continuously generate electrical power from bacterial photosynthesis and respiration through day-night (light-dark) cycles, offering a cost-effective, environmental-friendly, and clean power solution. However, the promise of this technology has not been realized as a real-world power source because of its low-performance and short lifetimes. These gaps have relegated conventional miniature MSCs to a laboratory test stage rather than being a practical real-world power source. To achieve the potential of miniature MSC technology as a superior substitute for batteries and other energy storage devices for WSN devices, many brilliant innovations have been proposed including synergistic cooperation between mixed bacterial communities, integration of supercapacitor for simultaneous energy storage, paper-based device platforms, and many other alternative ideas using nanomaterials and stacking. However, some additional work is required to avoid potential failure caused by external damage that interferes with their requirement for a constant supply of water for microbial photosynthesis. Once a functional mini-MSC becomes available, it could become a practical power source for supplying long-term power for small and low-power portable electronics as well as wireless sensors used at remote sites where frequent battery replacement is impractical. It is also expected that research into miniature MSC device platforms will enable crucial understanding of the extracellular electron transfer processes in a smaller group of photosynthetic microorganisms with excellent control over the microenvironment, thereby enabling a versatile platform for fundamental MSC studies.

Declaration of competing interest

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

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