

Full paper

A miniaturized, self-sustaining, and integrable bio-solar power system

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ABSTRACT

We demonstrate a practical and sustainable bacterial power source for low-power electronic applications. Self-sustainable electricity is produced from revolutionarily structured microliter-scale bio-solar cells using a co-culture of heterotrophic and autotrophic bacteria. Two bio-solar cells are integrated on a single chip and connected in series, continuously generating light-responsive electricity from heterotrophic bacterial respiration with the organic substrates produced by photosynthetic autotrophs. Agar-based anolyte, catholyte, and salt-bridge provided solid-state ionic environments for more efficient syntrophic interactions between co-cultures without bacterial competition, further enhancing the device performance and lifespan. The solid-phase platform allows versatile device configuration in a small footprint without a cumbersome fluidic feeding system and permits easy integration and operation with solid-state applications. The device is sealed with a gas-permeable polydimethylsiloxane (PDMS) membrane to facilitate gas exchange to the bacteria and cathodic reactions, even ideally allowing for replenishing bacterial gasses from environments for self-sustainable energy harvesting. A DC-DC booster circuit is integrated with the stacked bio-solar cells to increase the operational voltage (~ 500 mV) to a maximum output of >3 V for self-powering an on-chip, light-emitting diode (LED). This is the first demonstration of the microliter-scale bio-solar cell as a practical power source.

1. Introduction

Humanity is entering the new era of the “Internet of Things” or “IoT,” where billions of low-powered, miniaturized sensor nodes are connected to the Internet to monitor and surveil environmental conditions in real time [1,2]. In this context, many revolutionary paradigms have emerged for specific applications, such as the Internet of Disposable Things (IoDT) for connecting things inexpensively [3], the Agricultural Internet of Things (AIoT) for smart farming [4], or the Ocean of Things (OoT) for underwater exploration and disaster detection [5]. With advances in material science and device engineering, the size and energy consumption of the internet-enabled sensor nodes have considerably been reduced with improved performance, taking a major step toward ubiquitous deployment of intelligent devices in the environment [6,7]. However, fundamental breakthroughs that can supply power to these sensor nodes are needed to translate the promise of this technology as practical applications [8,9]. The development of power sources is in its pioneering stage. As the sensor nodes become more ubiquitous in remote, and even unreachable, environments, the power supply should provide long-term and even self-sustainable energy in an

environmentally friendly manner. In that sense, the conventional battery technologies become less practical because of their limited lifetime and environmentally hazardous components [1]. Furthermore, battery replacement will be very costly and infeasible for more remote areas. To enable relatively long-lasting operation without environmental issues, therefore, energy harvesting from the renewable sources will play a key role in future IoT technology [1,8,9].

Among various renewable energy sources such as airflow, vibration, or thermal energy [10], microorganisms are self-sustainable sources because they occupy essentially every environment and various microorganisms can continuously harvest electrical power from the ambient biomass or solar energy [11–14]. Particularly, microliter-scale bio-solar cells can be the most suitable power source for unattended environmental IoT sensors because the photosynthetic autotrophs that the technique typically uses as a biocatalyst can continuously generate electricity from microbial photosynthetic and respiratory activities through day-night cycles, offering a clean and renewable power source with self-sustaining and self-maintaining potential [15–18]. Many research groups generated a wealth of new scientific and technological results that clearly demonstrated the photosynthetic electrogenic

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activities of various cyanobacteria or algae [19–21].

Although macro-scale bio-solar cells have successfully operated under laboratory-controlled environmental conditions, micro-scale platforms have not realized their potential for practical applications because of their short life-times and relatively low power and output voltage. Even the latest micro-scale device was limited as a potential platform for long-term and practical use [22]. Normally, the electricity harvesting efficiency of the photosynthetic autotrophs is extremely low compared to the heterotrophic microorganisms used in microbial fuel cells or bioelectrochemical cells [21,23]. Our ability to harness the potential of the bio-solar cells lags because we lack an in-depth understanding of the mechanism for electron harvesting from photosynthetic microorganisms and the fundamental factors that maximize their power-generating capabilities. Furthermore, the enclosed microfluidic chamber of the micro-scale device does not allow replenishment of necessary gases (e.g. nitrogen) for microbial metabolism, preventing increases in performance and long-term operation [24]. Even worse, its initial operation requires a cumbersome fluidic control system to introduce microbial cultures and the need for handling of the liquid-state culture makes the bio-solar cell incompatible with solid-state sensor nodes [3]. Recently, we proposed the use of solid-state anolyte and catholyte to operate the microbial fuel cell without the need for external feeding of the device [3]. Even though, the all-solid-phase components of the device made it favorable in integration and operation with solid-state IoT devices, there was still the need for device breakthroughs to generate a relatively stable power to enable practical powering of an IoT system.

Here, we eliminate the aforementioned major technical hurdles by creating an indirect solar-driven power generating system that uses the syntrophic interaction between photosynthetic and heterotrophic electron-producing bacteria in solid-state microfluidic chambers (Fig. 1). Our microliter-sized bio-solar power system integrated two series-connected bio-solar cells into a well-designed low-power DC-DC booster circuit. The system demonstrated a simple, low-cost, but practical and long-lasting on-chip power supply. It produced a high power from well-studied heterotrophic microbes while their continuous and self-sustaining metabolism and growth were supported by solar-driven photosynthesis of the autotrophs. Nutrition and necessary gasses needed for heterotrophic microbial respiration and electricity harvesting were internally photosynthesized by the autotrophs whose reactants, such as carbon dioxide and water, were generated by the heterotrophic microbe, which consequently exhibited self-sustainable capabilities. The

photosynthetic microbes do not directly involve in electricity harvesting but indirectly support the heterotrophs' power production through photosynthesis. The device was sealed with a gas-permeable polydimethylsiloxane (PDMS) membrane to facilitate gas exchange to the bacteria and cathodic reactions, even ideally allowing for replenishing bacterial necessary gasses from environments for self-sustainable energy harvesting. Furthermore, the device used an agar-based salt bridge, and anodic and cathodic parts, providing solid-phase ionic pathways. This solid-phase paradigm can be readily integrated into a miniaturized system and operated with the solid-state IoT applications. Recently, our group created a microbial co-culture platform [25] and a solid-state microbial fuel cell [3] that establish the groundwork for advances in sustainable micro-energy systems. However, much of our previous work is in its nascent stages: the evolution of these methods will require additional exploration through a practical application of established techniques and comprehensive systematic integration. Therefore, this work is unique and innovative because we demonstrated, *for the first time*, a novel bio-solar cell architecture with co-culture integration and solid-phase device components, significantly contributing to the autonomy of the resulting system. The system is a practical self-sustainable power supply for actual applications.

2. Experimental

The developed bio-solar power system features (i) a dual micro-chambered bio-solar cell configuration with solid-state anodic and cathodic compartments and a salt bridge, (ii) a syntrophic co-culture of heterotrophic and autotrophic microorganisms, and (iii) a serial stack of bio-solar cells with a DC-DC booster converter.

2.1. Bacterial inoculums

Synechocystis sp. PCC 6803 (autotrophs) grew from -80°C glycerol stock cultures by inoculating 15 mL of BG-11 medium with gentle shaking under a 24 h light cycle (12 h light/dark). The BG-11 contained 1.5 g NaNO_3 , 40 mg K_2HPO_4 , 75 mg MgSO_4 , 36 mg CaCl_2 , 1 mg of EDTA, and 6 mg of citric acid and of ferric ammonium citrate per 1 L of distilled water. The culture system provided continuous aeration and illumination. *Shewanella oneidensis* MR-1 (heterotrophs) were grown from -80°C glycerol stock cultures by inoculating 20 mL of L-broth media with gentle shaking in air for 24 h at 35°C . The L-broth (LB) media consisted of 10.0 g tryptone, 5.0 g yeast extract and 5.0 g NaCl per liter. Both

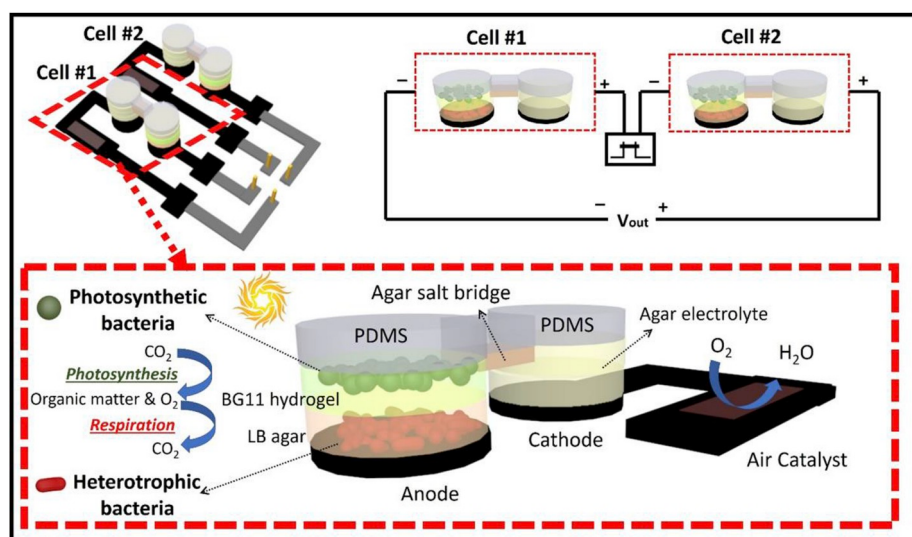


Fig. 1. A conceptual schematic diagram of the bio-solar power system. Each cell has a dual-chambered configuration with solid-state anolyte, catholyte and salt bridge. Two cells are integrated in a well-designed microsystem and will be connected in series for powering an application.

cultures were then centrifuged at 5000 rpm for 5 min to remove the supernatant. The bacterial cells were re-suspended in a new medium and used as an anolyte for the device. The bacterial growth was monitored by the optical density at 600 nm (OD_{600}). Both bacterial samples were cultured until their OD_{600} reached 2.5.

2.2. Device fabrication

Two dual-chambered bio-solar cells were integrated into a single device ($3.5\text{ cm} \times 2.4\text{ cm} \times 0.7\text{ cm}$) (Fig. 2). Each bio-solar cell contained 10 μL anodic and 10 μL cathodic chambers connected by a 7 μL salt-bridge chamber. Fig. 3 shows the fabrication steps of our novel bio-solar power device. The device was constructed by thermal bonding five patterned poly(methyl methacrylate) (PMMA) layers at 120°C for 20 min. By using laser micromachining (Universal Laser Systems VLS 3.5), the first three PMMA layers were micro-patterned to define chambers for bio-solar cells and windows for electrical connections. The anode and cathode were patterned on the fourth PMMA layer while nickel electrical paths were printed on the bottom PMMA layer and connected to the electrodes through thru-hole vias filled with silver paste. The electrical paths were linked to a 2-pin DIP connector that finally connected two bio-solar cells in series upon on-chip connection of the device to an external DC-DC booster circuit.

Graphite ink (Ercon Inc., USA) was screen-printed to define an anode (1.2 mm^2) and a cathode (1.2 mm^2). The cathodic catalyst was prepared by adding the mixture of Ag_2O (500 mg, 11407-14, Alfa Aesar, USA) in 10 mL poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS, Clevios PH1000, Heraeus, German) solution and was added on top of the cathode. Ag_2O is reduced to Ag with electrons and protons travelling from anode to maintain the electroneutrality of the system [26].

2.3. Solid-phase chambers

10 μL of *Shewanella oneidensis* MR-1 in LB-media was introduced in the anodic chamber to form a bacterial biofilm to improve power generation. With the addition of 1.5% (w/v) low temperature agarose, the anolyte was rapidly solidified at room temperature. Then, 20 μL of *Synechocystis* sp. PCC 6803 in BG-11-based hydrogel was introduced. The cathodic chamber was filled with 2.5 g agar in 100 mL distilled water, followed by introduction of the agar with an additional 5% (w/v) NaCl into the salt-bridge chamber to form ionic pathways between the anodic and cathodic parts. Finally, all chambers were sealed and covered with gas permeable PDMS to facilitate gas exchange to the

bacteria and further cathodic reactions.

2.4. Electrical measurement setup

The potentials between the anodes and the cathodes were measured with a data acquisition system (National Instruments, USB-6212), and recorded every 30 s via a customized LabView interface. The temperature was consistently maintained at 30°C and different external resistors were tested to connect the anode and the cathode while closing the circuit. The current through the resistors was calculated using Ohm's law.

3. Results and discussion

Our device innovated manufacturing, configuration, and operation of the solar-driven microbial energy harvesting technique and improved its power and device lifetime, translating it from laboratory novelties to practical use in IoT applications. This revolutionary achievement was realized by integrating two innovative techniques into an on-chip system. We exploited cross-feeding cooperation between heterotrophs and photosynthetic autotrophs, and we converted liquid compartments of the device into solid-phase counterparts. The heterotrophic microbes harvested electrons from the organic compounds produced by photosynthetic microbes. The by-products of the heterotrophs, such as carbon dioxide and water, were internally used for photosynthesis by the autotrophs, offering systemic self-sustainable capabilities. Furthermore, the agar-based device compartments are ionically conducting, which makes the device easily integrable and operable with solid-state IoT applications and provides flexible device designs in a miniaturized system. The agar-based anolyte, catholyte, and salt-bridge provided solid-state ionic environments for efficient syntrophic interactions between co-cultures without bacterial competition, further enhancing the device performance and lifespan. Typically, multi-species microbial communities form complex ecological interaction webs [27]. In most cases, acquiring substrates for metabolism sets the stage for interactions in bacterial communities. These interactions can be separated into four ecological niches: commensalistic, mutualistic, competitive, or parasitic [27]. In a commensalistic interaction, one bacterial species benefits from the interaction while the other is not affected. Mutualistic interactions benefit both species collectively, but not necessarily each individual. For competitive interactions, only one species benefits, while in a parasitic interaction, neither species benefits directly. The most frequent interaction for microorganisms is competition for nutrients. As shown in our previous work for co-cultured microbial power generation [25], the

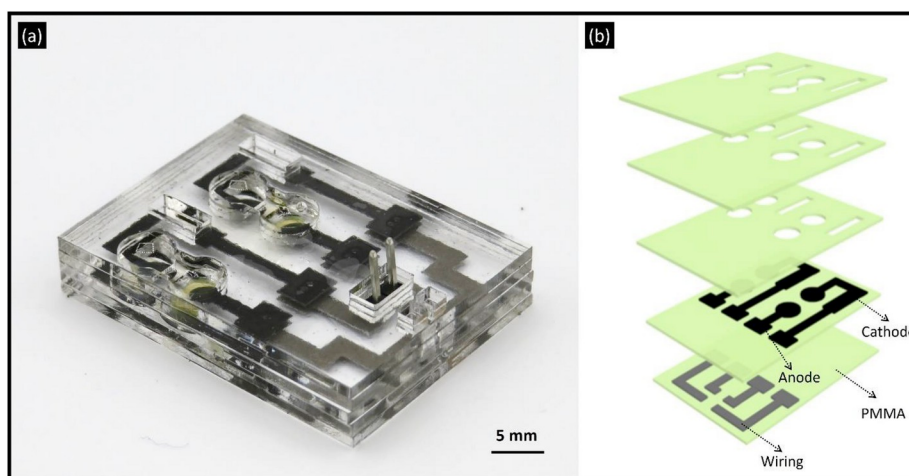


Fig. 2. (a) A photo-image of the device integrating two bio-solar cells and (b) a schematic diagram of the device individual layers. The device is constructed with five PMMA substrates. The PMMA layers are precisely cut by a laser cutting machine and patterned with conductive materials, and then aligned and assembled with high temperature and pressure.

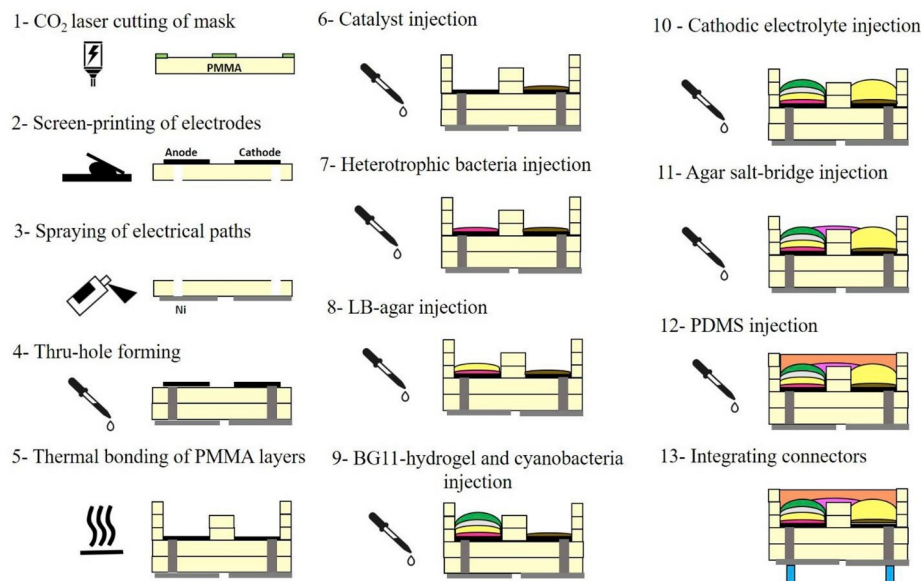


Fig. 3. Schematic illustrations of the fabrication process.

mixed culture with direct contact allowed 13 days of electricity generation, followed by a sudden drop in performance presumably because of the microbial competition. In this work, the agar separates two bacterial habitats but allows the interchange of their nutrients and gases (Fig. 1).

3.1. Current and power generation

To assess performance, two devices were prepared as controls; one with heterotrophs only and the other with autotrophs only. An external resistor of 100 k Ω was connected to one of the two bio-solar cells in the on-chip device and the generated current was evaluated. The second bio-solar cell was left unconnected and with an open circuit because a single unit needs to be tested before the series connection (Fig. 4a). All three

devices operated within a closed system and received no additional organic fuels. Under the operating condition of 12hr day and 12hr night consecutive cycles and consistent temperature of 30 °C, the devices required time to accumulate and acclimate bacteria on the anode but gradual increases in current were observed only from the co-culture and autotroph samples. The co-culture sample showed the current fluctuations that depended on the light availability despite the fact that their current generation is attributed to the heterotrophic bacteria (c.f. the heterotrophs only sample showed no light response). This indicates that two bacterial cells are syntrophically cooperating, where the heterotrophs' electricity generation relies on the by-products of the autotrophs, which are light-sensitive [25]. However, the current density from the co-culture sample was much higher than the autotroph-only

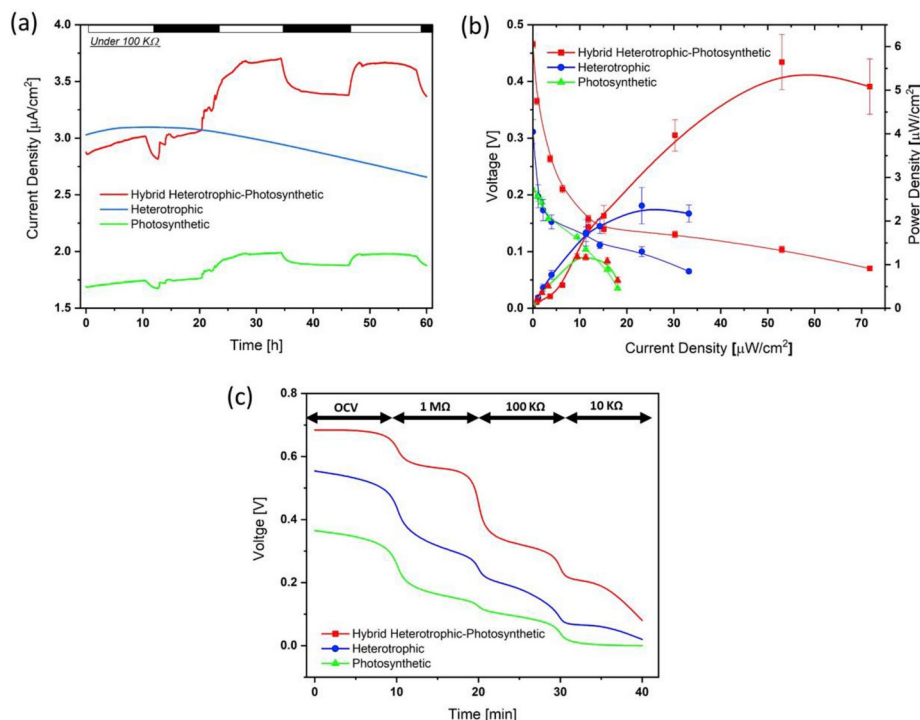


Fig. 4. (a) Current densities (at 100 k Ω resistor) as a function of time (the white and shadow bars indicate the illuminated and dark period, respectively). (b) Polarization curve and output power measured from heterotrophic bacteria (*Shewanella oneidensis* MR-1) only, photosynthetic bacteria (*Synechocystis* sp. PCC 6803) only, and a co-culture of the heterotrophic and the photosynthetic bacteria. (c) Output voltages in response to different loads when two units were connected in series.

control because the electron transfer efficiency of the heterotrophs is better than that of the autotrophs [20,21]. The current produced by the heterotrophs-only device decreased with time unlike the co-cultured- and autotroph-only devices. This indicates the autotrophs, which can produce their own food from light and necessary gasses, can be supported by the syntrophic interaction between bacteria and the gas-permeable PDMS membrane. In our previous device with a completely hermetic system, other necessary gases for bacterial metabolism, such as nitrogen, were rapidly depleted in the microchamber and could not be replenished even from the photosynthesis, hampering its long-term operation [24,25]. If the nitrogen concentration in the system is low, the photosynthetic activity of the autotrophs is limited and their metabolism slows down [28,29]. However, in our current device design, additional necessary gasses for microbial metabolism could be supported through the gas permeable PDMS layer from the air, significantly improving the device lifetime even in the extremely miniaturized chamber volume (Fig. S1), outperforming the gas-impermeable Kapton tape covered device. Electrochemical impedance spectroscopy (EIS) measurements evaluated ion transfer efficiency of devices (Fig. S2). The results were plotted as Nyquist curves and further fitted with an equivalent circuit. Each EIS curve was composed of a well-designed semicircle followed by a straight line. The intercept of the semicircle with the real impedance axis presented the total ohmic resistance of the device including the solution resistance (R_s) and the charge transfer resistance (R_{ct}) at the electrode/electrolyte interface. The ohmic resistance ($R_s + R_{ct}$) of the device with the salt-bridge membrane was estimated to be 1.1 k Ω , much lower than the device without the membrane (~ 1.9 k Ω). Fig. 4b shows the power and current outputs of the devices on Day 4 of operation. The curves were derived and calculated based on the stabilized current value at various external resistances. The maximum power density of 5.64 $\mu\text{W}/\text{cm}^2$ (15.2 $\mu\text{W}/\text{cm}^3$) and current density 53 $\mu\text{A}/\text{cm}^2$ (143 $\mu\text{A}/\text{cm}^3$) was obtained from the co-culture device. The single-cultured device only with heterotrophs or autotrophs produced much lower performances, generating maximum power densities of 2.35 $\mu\text{W}/\text{cm}^2$ (6.4 $\mu\text{W}/\text{cm}^3$) and 1.16 $\mu\text{W}/\text{cm}^2$ (3.1 $\mu\text{W}/\text{cm}^3$) and current densities of 23.2 $\mu\text{W}/\text{cm}^2$ (62.6 $\mu\text{W}/\text{cm}^3$) and 11.2 $\mu\text{W}/\text{cm}^2$ (30.2 $\mu\text{W}/\text{cm}^3$), respectively.

3.2. Bio-solar power system with DC-DC booster circuit

The produced sustainable voltage output from the single bio-solar cell is on the order of 0.4–0.6 V and load-dependent. To produce sufficient output voltage for practical low-power electronics, it is necessary

to use a DC-DC booster converter between the bio-solar cell and the application [30]. However, it is still challenging for a single bio-solar cell to generate a consistent and reliable minimum input voltage required by a typical commercially available low-power boost converter so that a complete self-powered system can be realized for practical applications. The catalysts of the bio-solar cells are living creatures, highly depending on environmental conditions such as temperature, pH, humidity, or gas composition in the air. Therefore, to continuously secure the minimum input voltage needed by the booster circuit, connecting multiple device units in series can be an excellent option. Here, we integrate two bio-solar cells in a single device, which were designed to be connected in series when the booster circuit was linked to the device through a 2-pin DIP connector. As shown in Fig. 4c, the highest performance was obtained from the device pack with the co-cultured configuration. For the low-power DC-DC booster converter, we used BQ255504 (Texas Instruments) and its evaluation board (BQ25504EVM-674) (Fig. 5). BQ255504 is well-suited to operate with the miniaturized low-power bio-solar cell because the booster converter requires only 0.6 V for an initial start. Once started, the converter can continuously release energy with only 0.13 V of input voltage (V_{in}) and ultra-low quiescent current of 0.33 μA . As shown in Figs. 5 and 6, the DC-DC booster circuit was capable of converting the relatively low DC voltage of the stacked cells ($V_{in} = 0.5$ V) to a more practical range for electronics of >3.0 V (V_{out}) which was enough to provide short

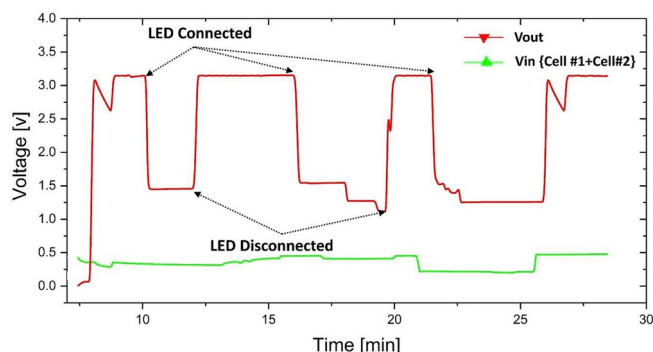


Fig. 6. Plots of output voltages as a function of time, measured from the serial stack of two cells (V_{in}), and the DC-DC booster circuit (V_{out}). The DC/DC booster circuit converts the relatively low DC voltage of the stacked cells (0.5 V) to a more practical range for electronics of >3.0 V.

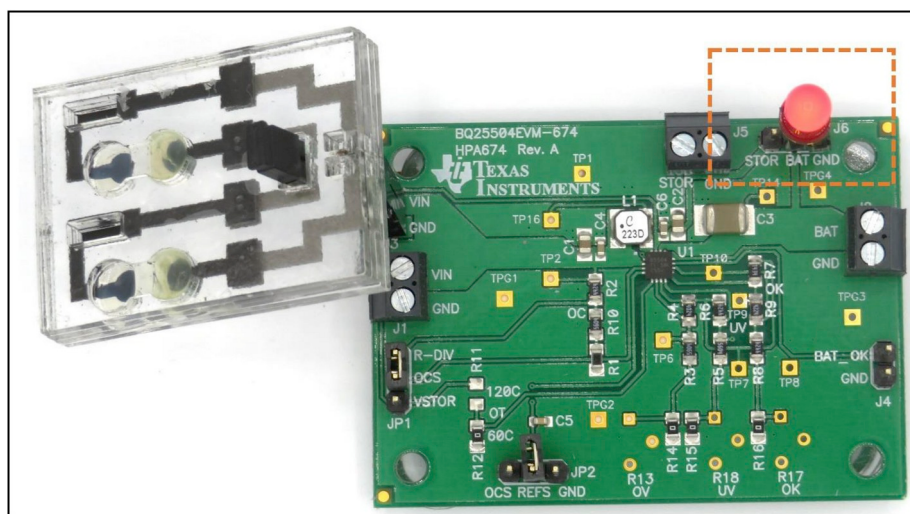


Fig. 5. A solar-driven microbial energy harvesting system integrating two bio-solar cells connected in series, a DC-DC booster circuit, and an on-chip, light-emitting diode (LED). This work ensures the practical efficacy of the biological fuel cells as a self-sustainable power supply for actual applications (See the Video 1).

operation of the connected LED (HLMP-4700-C0002, Digi-Key) with 1.6 V (Video 1). In this work, the DC-DC booster circuit and the LED were powered only with our bio-solar power device without any other power source or fluidic feeding system. Our device only needs light to generate power. Although further studies for longer-term operation are needed, our bio-solar power device demonstrated its potential use at least for short-term Internet of Disposable Things (IoDT) applications.

Supplementary video related to this article can be found at <https://doi.org/10.1016/j.nanoen.2020.104668>

4. Conclusion

This work created a simple and practical solid-state miniaturized bio-solar power system, delivering on-chip energy to the next generation of low-power IoT applications. We envision that the developed system can provide a novel strategy that can revolutionize the functionality of miniaturized bio-solar cells by syntrophic interactions between heterotrophic and autotrophic microorganisms in well-controlled solid-phase microchambers and can enable on-chip stacking of the cells in a small package for future on-board applications. The bio-solar power system could generate self-sufficient current and power density for more than 4 days. The obtained power performance represents a promising solution for self-sustainable remote wireless network applications. Furthermore, the optimization of our solid-phase integrated biosystem could be expected to provide opportunities for further studies on syntrophic reactions between many other microbial communities that could facilitate higher performance with a full self-sustainability.

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.

CRediT authorship contribution statement

Maedeh Mohammadifar: Investigation, Methodology, Data curation, Writing - original draft. **Mehdi Tahernia:** Investigation, Formal analysis, Writing - original draft. **Seokheun Choi:** Writing - review & editing, Funding acquisition, Project administration, Supervision, Conceptualization.

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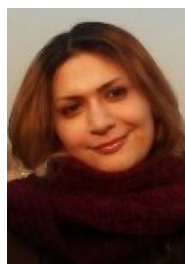
Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.nanoen.2020.104668>.

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