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Fully printed and integrated electrolyzer cells with additive manufacturing for high-efficiency water splitting

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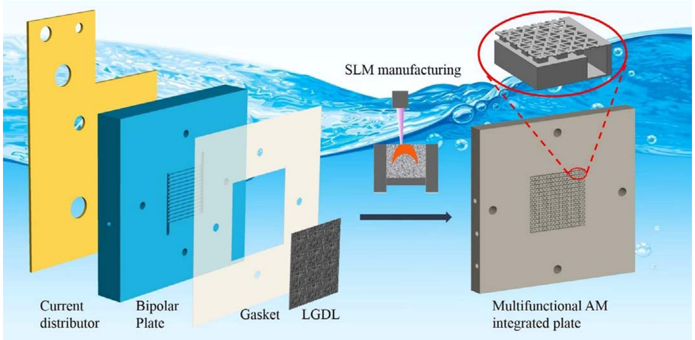
HIGHLIGHTS

- Integrated electrolyzer cells with complex inner structure were fully printed with additive manufacturing.
- Four components in PEMECs were integrated into one part.
- An ultralow electrical resistance of electrodes was achieved.
- AM PEMECs provide an energy efficiency of up to 86.48% at 2 A/cm^2 and 80 °C.
- AM PEMECs open a door for developing other energy conversion devices.

GRAPHICAL ABSTRACT



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ABSTRACT

Using additive manufacturing (AM) technology, a fundamental material and structure innovation was proposed to significantly increase the energy efficiency, and to reduce the weight, volume and component quantity of proton exchange membrane electrolyzer cells (PEMECs). Four conventional parts (liquid/gas diffusion layer, bipolar plate, gasket, and current distributor) in a PEMEC were integrated into one multifunctional AM plate without committing to tools or molds for the first time. In addition, since the interfacial contact resistances between those parts were eliminated, the comprehensive *in-situ* characterizations of AM cells showed that an excellent energy efficiency of up to 86.48% was achieved at 2 A/cm^2 and 80 °C, and the hydrogen generation rate was increased by 61.81% compared to the conventional cell. More importantly, the highly complex inner structures of the AM integrated multifunctional plates also exhibit the potential to break limitations of conventional manufacture methods for hydrogen generation and to open a door for the development of other energy conversion devices, including fuel cells, solar cells and batteries.

1. Introduction

Hydrogen is considered as one of the best vectors/carriers for storing renewable and intermittent energy from sources including wind and solar, or off-peak energy from the grid [1–9]. Water electrolysers, especially low-temperature proton exchange membrane electrolyzer cells (PEMECs), provide a sustainable method for producing hydrogen from water splitting with rapid response to the power input, higher

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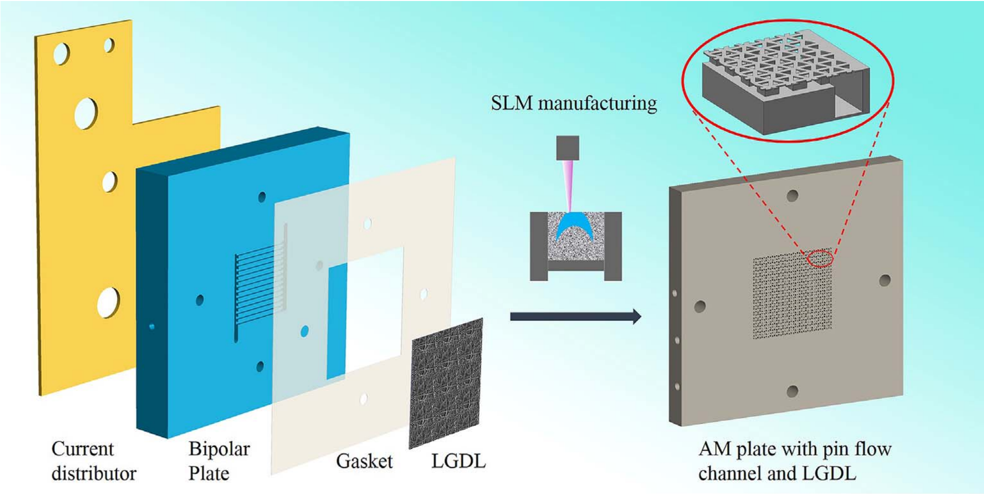

Fig. 1. Schematic illustration of the integration of current distributor, bipolar plate, gasket, and LGDL into one AM plate via additive manufacturing.

purity product (up to 99.995%), high operation current density (above 2 A/cm^2), and high operation pressure (up to 350 bar) [4,10–16]. A PEMEC typically consists of a catalyst-coated membrane (CCM), sandwiched by two liquid/gas diffusion layers (LGDLs), two bipolar plates (BPs), two gaskets, and two current distributors at anode and cathode sides. Nevertheless, the relatively low efficiency and high cost/complexity have retarded its wider commercialization [3,12,17,18].

To reduce the cost and promote performance of PEMECs, some efforts are focusing on reducing the use of the precious catalysts (e.g., Pt and Ir), or exploring the alternatives, such as Fe, Co, Cu, Ni, Mo, and W-based nonprecious catalysts, while another approach is to optimize other components and the whole system [11,19–21]. BPs and LGDLs significantly affect the performance of PEMECs especially at the high current density, where the ohmic overpotential and concentration overpotential become the dominant factors for total cell voltage [22,23]. BPs usually account for 30% - 48% of the cost and 60% of the weight of PEMECs [3,24]. The cost of labor, tooling, and machining for large-volume BP manufacturing is almost the same as that of the materials, and the cost even become much higher for small batches [25]. Furthermore, the LGDLs are expected to simultaneously transport electrons, reactants and products at minimum voltage, fluidic, and heat losses in a severe electrochemical environment, and they mainly made of titanium and carbon by using weaving, sintering or etching processes. But the woven and sintered felt LGDLs will result in nonuniform interfacial contacts and random pore morphologies. And the etching method involves several complicated processes, and the chemical and disposal costs will limit its application [3,26,27].

Researchers have recently reported to improve the PEMEC performance by reducing or eliminating contact resistances between mating components such as the BPs, LGDLs, CLs, and current distributors [28,29]. Several strategies have been proposed to solve this problem, by coating precious metals (Pt, Au, etc.) and abundant metals (Ti, Ni, etc.) on the surface of BPs and LGDLs [30,31], or by optimizing the components properties and structures [32,33]. Coatings increase the cost of PEMECs because of the complex chemical processes and expensive materials. Structures and morphologies of BPs and LGDLs are optimized by changing the channel shape and the pore size [3,32,34,35]. But the interfacial contact resistance has not been eliminated, and the optimization of PEMEC materials and structures for better performance reaches a bottleneck. Furthermore, when cells are scaled up to stacks, assembling the large number of components is very time intensive, and requires exacting control. Therefore, simplifying the components design and configuration with low cost and high efficiency remains an important challenge for the commercial application of PEM energy devices.

Additive manufacturing (AM) technology, also called rapid prototyping, freeform fabrication, or 3D printing, allows material to "print" three-dimensionally from three-dimensional (3D) digital models instead of cutting materials from an initial bulk [36,37]. Due to its potential to offer cost and time-efficient parts with complex 3D structure, AM has received significant attention in electrochemical sciences. Cronin et al. tried to print a plastic BP, which was coated with Ag and Au, but the voltage in the PEMEC reached 2.5 V at 1 A/cm^2 [38]. Mo et al. fabricated LGDLs from Ti using electron beam melting (EBM), however the performance was not good compared with conventional PEMECs [26]. Several researchers also tried to fabricated BPs for energy devices from carbon and plastic materials with AM, but dimensional shrinkage and structure inaccuracy as a result of curling and high voltage at certain current density remain unresolved issues [18,38,39]. Nevertheless, none of those efforts has been devoted to integrate components in the energy devices into one part, and the devices' performances in those publications are not comparable to that of conventional one. Therefore, the integration of different parts for low-cost and high-efficiency water electrolyzers is unexplored and highly desired.

Herein, one of the AM technologies, selective laser melting (SLM), is used to meet the aforementioned challenges, and to develop novel multifunctional AM integrated plates that serve as BPs, LGDLs, gaskets and current distributors simultaneously, shown in Fig. 1. Both ex-situ and in-situ characterizations of the AM integrated plates are comprehensively conducted, and the equivalent electrochemical circuits (EECs) are introduced to quantify the AM plates for PEMEC performance. The integration of several conventional components into one multifunctional AM part not only significantly improves the performance of PEMECs with lower resistances, but also greatly simplify the configuration and reduce the weight and volume. The study also indicates AM can accomplish radical structure changes for electrolyzer and other energy conversion devices.

2. Experimental

2.1. Fabrication of AM plates

Using AM technology, the conventional BP, LGDL, gasket, and current distributor were integrated into one part, as shown in Fig. 1. The patterns of AM SS plates were designed using SOLIDWORKS 2016, then solid models were sliced and converted for building using Materialise Magics 20. A Renishaw AM250 laser powder bed printer was employed to build the plates. Parameters during manufacturing included laser power of 200 W, focus offset of 0 mm, layer thickness of 50 μm, point spacing of 60 μm, exposure time of 80 μs, hatch spacing of