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A review on unitized regenerative fuel cell technologies, part B: Unitized regenerative alkaline fuel cell, solid oxide fuel cell, and microfluidic fuel cell

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ABSTRACT

In part A of this review, we have introduced the progress of the research and the application status of unitized regenerative proton exchange membrane fuel cells. In addition to this Proton Exchange Membrane (PEM)-based Unitized Regenerative Fuel Cell (URFC), other URFC technologies with different electrolytes have also been reported in the literature, which form the basis for emphasis in this part of the review. Unitized Regenerative Alkaline Fuel Cells (UR-AFC) have long been utilized for aerospace applications, while the recent development of Anion Exchange Membrane (AEM) has stimulated their further development, especially on the AEM-based UR-AFCs. Vast research works have been reported on the bifunctional oxygen catalyst development, while the latest UR-AFC prototypes are also being briefly introduced. Despite their potential cost-efficiency and better reactivity, cell performance and round-trip efficiency of the current UR-AFCs are still lower than their PEM-based counterparts. Unitized regenerative solid oxide fuel cell, which is more commonly cited as Reversible Solid Oxide Fuel Cell (RSOFC), is a high-temperature URFC technology with superior performance and reversibility. Review works conducted on this type of URFC are separated into two categories, that is, RSOFC with oxygen ion conducting electrolyte and RSOFC with proton ion conducting electrolyte. Despite the highest efficiency among various URFC technologies, the application of RSOFCs, however, is restricted by their limited long-term stability and poor cycle ability. Unitized regenerative microfluidic fuel cell, also referred to as the reversible microfluidic fuel cell, is a newly-emerging URFC research trend which benefits a lot from its membraneless configuration. However, limited research works have been conducted on this new technology.

1. Introduction

In part-A of this review, we have presented a detailed introduction to the Unitized Regenerative Proton Exchange Membrane Fuel Cell (UR-PEMFC), which is currently the most represented and advanced

Unitized Regenerative Fuel Cell (URFC) technology recorded to date [1–5]. The introduction follows a sequence from the inner cell components, that is, the bifunctional catalysts, to the outer cell components, that is, the Gas Diffusion Layer (GDL) and Bipolar Plates (BPPs). Till date, Pt catalyst is predominantly utilized as a

Abbreviations: AEM, Anion exchange membrane; AFC, Alkaline fuel cell; ASR, Area specific resistance; BCY, Yttrium-doped barium cerate; BCZY, Yttrium-doped barium cerate zirconate; BCZYbCo, BaCe_{0.48}Zr_{0.40}Yb_{0.10}Co_{0.02}O_{3–δ}; BHC, Bifunctional Hydrogen Catalyst; BHE, Bifunctional hydrogen electrode; BOC, Bifunctional Oxygen Catalyst; BOE, Bifunctional oxygen electrode; BOP, Balance of plant; BPPs, Bipolar plates; BSCF, Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3–δ}; BZC, BaZr_{1–x}Co_xO_{3–δ}; EC mode, Electrolysis cell mode; FC mode, Fuel cell mode; GC, Glassy carbon; GDC, Gadolinium-doped ceria; GDL, Gas diffusion layer; gRGO, Gently reduced graphene oxide; HER, Hydrogen evolution reaction; HOR, Hydrogen oxidation reaction; LC, Lanthanum cobaltite; LCC, Lanthanum calcium chromite; LCFr, La_{0.3}Ca_{0.7}Fe_{0.7}Cr_{0.3}O_{3–δ}; LNO, Lanthanum nickelate; LSC, Lanthanum strontium chromite; LSCF, Lanthanum strontium cobalt ferrite; LSCM, (La_{0.75}Sr_{0.25})Cr_{0.5}Mn_{0.5}O₃; LSCN, La_{2–x}Sr_xCo_{0.5}Ni_{0.5}O_{4±δ}; LSCuF, Lanthanum strontium copper ferrite; LSF, Lanthanum strontium ferrite; LSGM, Lanthanum strontium gallate magnesite; LSM, Lanthanum strontium manganite; MEMS, Micro-electro-mechanical systems; MFC, Microfluidic fuel cell; MH, Metal hydride; MOF, Metal-organic framework; MWCNTs, Multi-walled carbon nanotubes; NCNTs, Nitrogen-doped carbon nanotubes; N-rGO, Nitrogen-doped reduced graphene oxide; N-rmGO, Nitrogen-doped reduced graphene oxide; N/S-rGO, Nitrogen and sulfur co-doped reduced graphene oxide; OER, Oxygen evolution reaction; ORR, Oxygen reduction reaction; PEM, Proton exchange membrane; PEMFC, Proton exchange membrane fuel cell; PPD, Peak power density; PTFE, Poly Tetra Fluoro Ethylene; qPDTB-OH⁺, Quaternary ammonium Poly(DMAEMA-co-TFEMA-co-BMA) ionomer; rGO, Reduced graphene oxide; RMFC, Reversible microfluidic fuel cell; RP, Ruddlesden-Popper; RSOFC, Reversible solid oxide fuel cell; RSOFC-H, RSOFCs with proton-conducting electrolyte; RSOFC-O, RSOFCs with oxygen ion-conducting electrolyte; RT-efficiency, Round trip efficiency; ScSZ, Scandia-stabilized zirconia; ScCeSZ, Scandia and ceria stabilized zirconia; SDC, Samaria-doped ceria; SFM, Sr₂Fe_{1.5}Mo_{0.5}O_{6–δ}; SOFC, Solid oxide fuel cell; SOEC, Solid oxide electrolysis cell; SSC, Samarium strontium cobaltite; URFC, Unitized regenerative fuel cell; UR-PEMFC, Unitized regenerative proton exchange membrane fuel cell; UR-AFC, Unitized regenerative alkaline fuel cell; UR-SOFC, Unitized regenerative solid oxide fuel cell; UR-MFC, Unitized regenerative microfluidic fuel cell; UR-PAFC, Unitized regenerative phosphoric acid fuel cell; UR-MCFC, Unitized regenerative molten carbonate fuel cell; WGSr, Water gas shift reaction; YDC, Ytria-doped ceria; YSZ, Ytria-stabilized zirconia

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Bifunctional Hydrogen Catalyst (BHC) which shows excellent performance in both Hydrogen Oxidation Reaction (HOR), and Hydrogen Evolution Reaction (HER). As for the oxygen electrode, a combination of the best Oxygen Reduction Reaction (ORR) catalyst (that is, Pt), and the Oxygen Evolution Reaction (OER) catalyst (that is, Ir, Ru and their oxides), are used to prepare Bifunctional Oxygen Catalysts (BOC). Due to the sluggish reactions of oxygen [6], cell performance in both Fuel Cell (FC) and Electrolysis Cell (EC) mode, and the Round-Trip (RT) efficiency of the present UR-PEMFCs, are primarily hindered by the electrode performance of oxygen. Therefore, numerous research works have been conducted to optimize BOCs including their composition, ratio, preparation method, support, binder, and layer structure. As for the GDL and BPPs, research works focus on the improvement of their stability mainly, especially in EC mode operation. Furthermore, for practical applications, several systematic issues have been carefully investigated to ensure adequate and steady power output, such as heat and water management, electrode configurations, fuel cell stacking, and so on. Till date, UR-PEMFCs have been applied in many areas already, including aerospace and aviation, renewable energies, grid supplement, transportation, and so on. However, due to their relatively high-cost, most applications are still limited to aerospace and military field. As for civil applications, tentative trials have been conducted to couple this technology with wind and solar energy, power grid, electric vehicles, or employ them as backup powers and uninterrupted power supplies.

Apart from UR-PEMFC, other URFC technologies are also getting more Research and Development (R&D) interests considering their specific advantages, including the low-temperature Unitized Regenerative Alkaline Fuel Cell (UR-AFC), the high-temperature Unitized Regenerative Solid Oxide Fuel Cell (UR-SOFC), and the recently-emerged Unitized Regenerative Microfluidic Fuel Cell (UR-MFC). To the best of our knowledge, no research work has been reported on either the Unitized Regenerative Phosphoric Acid Fuel Cell (UR-PAFC), or the Unitized Regenerative Molten Carbonate Fuel Cell (UR-MCFC), so far.

Compared to the Proton Exchange Membrane (PEM) electrolyte in UR-PEMFC, UR-AFC utilizes either aqueous alkaline solution or Anion Exchange Membrane (AEM) as its electrolyte. This alkaline reactive environment provides various benefits such as the improved kinetics reaction, and the utilization of non-noble catalysts. However, so far, the development of UR-AFC is still hindered either by the CO₂ poisoning problem, or the imperfect AEM [7]. Moreover, the performance and efficiency of present UR-AFCs are also not satisfactory. Different from UR-PEMFC and UR-AFC, UR-SOFC generally works at high temperatures (500–1000 °C). Benefiting from this high temperature effect, outstanding performance and low over-potentials are achievable, while all the cell components can be made from cost-efficient ceramic materials. In general, UR-SOFC is very promising for the purpose of energy storage and conversion in high-temperature environments. However, their stability during the regenerative mode of operation still needs further improvement. UR-MFC is a URFC technology that emerged recently, and is based on microfluidic reactors. This kind of device utilizes the interface between two laminar flows as a virtual membrane instead of polymer membranes. Therefore, the fabrication cost is reduced greatly, and the pH environment of both electrodes can be independently tailored to optimize the performance and efficiency [8,9]. However, UR-MFC generally suffers from poor energy density and difficulties in scalability, due to the complex fluid management.

In this part of the review, research works related to the three URFC technologies mentioned above are introduced, respectively. For UR-AFC, emphasis will be placed on both catalyst and fuel cell development. For UR-SOFC, two main types, that is, the oxygen ion-conducting UR-SOFC and the proton-conducting UR-SOFC, are discussed separately, while the related modelling works are also summarized. As UR-MFC is only a newly-emerged technology, only a brief report is given at the present stage.

2. Unitized regenerative alkaline fuel cell (UR-AFC)

Alkaline Fuel Cell (AFC) is one of the most developed fuel cell technologies which has been put into practical services, since the early 20th century [7]. During its working process, the conducting ion in the electrolyte is hydroxyl (OH⁻) moving from cathode to anode, which is similar to proton (H⁺) moving from anode to cathode, in the proton exchange membrane fuel cell (PEMFC). As for the electrolyte, either a static porous matrix saturated with aqueous alkaline solution or a flowing aqueous alkaline solution can be adopted. Moreover, recently, a new form of electrolyte for AFCs has also been developed, named the AEM, which eliminates the problems that are aqueous solution related, such as CO₂ poisoning and potential electrolyte leakage. However, the ionic conductivity and chemical stability of the present AEM is still not as good as that of its acidic counterpart (that is, PEM) [7].

UR-AFC, also known as the rechargeable alkaline fuel cell, is developed by combining an alkaline fuel cell with an alkaline electrolyzer into a single unit. One early UR-AFC prototype was proposed in 1960 by Ludwig, for the Apollo program [10]. Afterwards, more prototypes were developed by NASA and Giner, Inc. in the 1980 s [3]. In the late 1980 s, Swette et al. developed a series of electrocatalysts and catalyst supports for the oxygen electrode of UR-AFCs with good electrochemical activity and stability [11–13]. Recently, research interests have placed emphasis on the AEM-based UR-AFCs, due to their simpler structure and the elimination of possible electrolyte leakage.

Belonging to the low-temperature URFCs (20–120 °C), one of the most significant advantages of the UR-AFC against the UR-PEMFC is its ability to use non-noble catalysts instead of the conventional noble metal catalysts, which makes it much more cost-efficient for large-scale manufacture and application. Such non-noble catalysts include nickel or metal hydride for the hydrogen electrode, and transition metal oxides or heteroatom-doped carbon for the oxygen electrode. Another merit of UR-AFC is the improved reaction kinetics in alkaline medium, which helps to improve the sluggish oxygen reaction activity, due to the multi-electron transfer processes and strong reaction irreversibility of oxygen [6].

In this section, a brief summary of the latest bifunctional catalysts of both hydrogen and oxygen electrode is given first. Furthermore, recent UR-AFC cell development, including performance optimization and stability improvement, will also be introduced.

2.1. Bifunctional Hydrogen Catalyst (BHC)

Pt is commonly used as BHC in UR-PEMFCs, because of its high activity towards both HOR and HER. As for UR-AFCs, even though the HOR/HER kinetics on Pt are several orders of magnitude slower in alkaline than in acidic medium [14], Pt catalyst with a relatively small loading (for example 0.1–1 mg cm⁻²) is still competent, considering both catalytic activity and cost efficiency [15–18]. Nevertheless, future perspectives of UR-AFCs will still expect non-noble metal based catalysts instead of Pt.

Metal Hydrides (MH) can be utilized in fabricating bifunctional hydrogen electrode in UR-AFCs. Such an integrated electrode can not only participate in catalyzing the HOR/HER reaction, but also function as a storage medium for hydrogen. Hu et al. [19] investigated the possibility of an AB₅-type MH (MmNi_{3.88}Co_{0.85}Mn_{0.39}Al_{0.4}), and a Zr-based AB₂-type MH (ZrNi_{1.2}Mn_{0.48}Cr_{0.28}V_{0.13}) for hydrogen oxidation and generation, and found that the AB₅-type MH exhibited higher HOR and HER activity than the Zr-based AB₂-type MH, almost comparable to 10 wt% Pt/C. In addition to MH, nickel and silver alloy are also considered to be promising when used as BHC in alkaline medium. Tang et al. [20], synthesized Ni-Ag alloys with various ratios, found that Ni_{0.75}Ag_{0.25} can exhibit two times HER activity than pure Ni but with comparable HOR activity.

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