Recent Perspectives and Crucial Challenges on Unitized Regenerative Fuel Cell (URFC)

(Perspektif Terkini dan Cabaran Kritikal mengenai Sel Fuel Regeneratif Terunit (URFC))

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ABSTRACT

Renewable sources of energy are becoming increasingly popular in recent years. The idea of using alternative energy that utilizes renewable sources is to slowly replace our dependence on fossil fuels. Conventional fossil fuels are not viable due to the fact that they are predominantly unsustainable over the long run. Furthermore, pollution will be reduced through the use of cleaner and more environmentally friendly renewable energy. Polymer electrolyte membrane fuel cell (PEMFC), which utilizes hydrogen as fuel, has a high potential as an alternative power generator. The unitized regenerative fuel cell (URFC) is a type of PEMFC that can perform both in charge mode (as a fuel cell) and discharge mode (as an electrolyzer). This review looks into the recent researches on the structure and different components of the URFC. In particular, emphasis is placed on bifunctional electrodes. Recent development in URFC research has produced a more stable bifunctional electrode with improved energy efficiency and overall stability and durability. Various works have been carried out to replace Pt as the electrocatalyst, including the use of graphene as a low cost non-metal graphene-based electrocatalyst. Electrocatalyst support also plays an important role in increasing conductivity while reducing the catalyst resistance to corrosion. The technological challenges and limitations of the URFC system are also discussed in this review.

Keywords: Regenerative Fuel Cell; Water Electrolysis; Gas Diffusion Layer; Bifunctional Electrode

INTRODUCTION

Nowadays, there is a major challenge in finding a feasible renewable energy source to replace the widely used fossil fuels. Alternative energy source is desperately required by the increasing demand for energy due to the explosive development of industrialization in many countries. Climate change and global warming due to the excessive emission of pollutants have also been a warning trigger to migrate towards an emission free and green source of renewable energy. A polymer electrolyte membrane fuel cell (PEMFC) is a fuel cell type that can be an alternative power generator as PEMFC is able to continuously operate at high current densities, low temperature. PEMFC is also tolerant to shock and vibration as well as having a compact and durable form that makes it the currently preferred commercially viable fuel cell type (Guerrero et al. 2015). The performance of PEMFC is bounded by polarization, temperature, pressure and gas stream composition.
Therefore, researches on the effects of designs and operating conditions of the cell potential is important to reduce these setbacks (Kamarudin et al. 2007). Membranes used for PEMFC are mostly related to ionomers, i.e. perfluorosulfonic acid ionomers (PFSA) such as Nafion® or Aquivion®, due to their excellent chemical and electrochemical stability (Assumma et al. 2015). Among other commercially available PFSA ionomers that exhibit high proton conductivity and chemical stability are Flemion, DAM-3G, Fumion, Gore Select, Dow, 3M and Aciplex (Campagne et al. 2013).

The reactions occurring in the polymer electrolyte membrane electrolyzer (PEMEL) are as shown in Equations 1 – 3. The reactant (water) is oxidized into oxygen, protons and electrons at the anodic reactive site by the catalyst. Meanwhile, the hydrogen ions conducted through the polymer electrolyte membrane (PEM) and the electrons that travel through the external circuit are combined at the cathodic reactive site to form gaseous hydrogen (Rahim et al. 2016). The operating principle of PEMEL is the opposite of PEMFC, as illustrated in Figure 1.

The combination of PEMEL and PEMFC into URFC is an efficient method of storing energy and reducing the sophistication of the two separate systems. A typical structure of a regenerative fuel cell (RFC) consists of an electrolyzer (EL) that converts water into hydrogen and oxygen using electricity, a fuel cell (FC) that utilizes hydrogen as fuel to generate electricity, and a tank for storing the oxygen and hydrogen gases used in the process (Guarnieri et al. 2015). Figure 2 illustrates the concept of a RFC.

A unitized regenerative fuel cell (URFC) is a type of fuel cell that can produce clean energy efficiently as it is an electrochemical energy conversion and storage system that can effectively generate electricity in FC mode as well as split the water into hydrogen and oxygen in EL mode. The reactions occurring in the URFC in EL and FC modes are as shown in Equations 4 – 9 and illustrated in Figure 3.

On the anode side (the oxidation electrode), water is supplied and the molecules are dissociated into oxygen, protons, and electrons through oxygen evolution reaction (OER) during EL mode, whereas hydrogen molecules experience hydrogen oxidation reaction (HOR), in which they are dissociated into protons and electrons, during FC mode. On the cathode side (the reduction electrode), incoming protons and electrons from the oxygen electrode experience hydrogen evolution reaction (HER) to produce hydrogen during EL mode, whereas oxygen molecules experience oxygen reduction reaction (ORR) to produce water during FC mode. Therefore, a bifunctional electrode is required for the URFC operation (Swider-Lyons and Campbell 2013).

Other types of water electrolysis technology are also currently being researched. The silicon oxide electrolyzer cells (SOEC) are capable of operating at a very high temperature range of 700-1000°C., whereas an alkaline electrolyzer (AEL) that uses sodium hydroxide (NaOH) and potassium hydroxide (KOH) as electrolytes is inefficient in terms of energy production but applicably cost effective (Lee et al. 2016).

**TECHNOLOGICAL CHALLENGES ON URFC**

**WATER MANAGEMENT**

The electrode is designed to be purposely flooded during EL mode. Conversely, the electrode for the FC mode is designed to repel water in order to increase the mobility of H₂ and O₂ to reach the catalyst surface. Development of flow channels
for the bipolar plates (BPPs) is crucial for the management of oxygen, fuel and water in the fuel cell system. Hwang et al. (2013) concluded that the parallel and serpentine-dual flow designs for BPPs can improve fuel cell efficiency and electrolysis performance. They used PTFE-treated Ti-felt GDL in the oxygen electrode and this also contributed to the improvement of water discharge during FC mode.

Apart from that, Lele et al. (2014) introduced a prototype design of UV-catalyzed porous polymer wicks, which improved the water flooding by diverting the water produced towards the water storage chamber. This passive water management was able to reduce the roundtrip efficiency loss and was found to be a better strategy compared to the active method that resulted in parasitic loss and a decrease in system efficiency.

COST

One of the most crucial disadvantages of the URFC system is the cost of the materials used. The high cost is associated with the use of rare platinum (Pt) metal on the surface of bifunctional electrocatalyst and the cost of the polymer electrolyte in the cell region (Fuentes et al. 2014; García et al. 2013; Vesborg and Jaramillo 2012).

Figure 4 describes the analysis of the cost status of fuel cell stack at 500,000 systems per year as compared to the cost in the 2020 targets. This shows that the development of technology for the URFC system can potentially lower the cost of the overall system by using different materials and overcome the durability issue currently faced by the system.

STRUCTURAL SETUP OF URFC STACK

Nafion, a widely used PEM for hydrogen fuel cells, provides high proton conductivity and low gas crossover (Antolini 2014). The electrolyte is essentially a conductive polymer membrane that acts as a barrier for the fuel and electrons but a conductor for protons (Boutsika et al. 2016). Nafion is expensive and does not work well in high temperature and low humidity. The use of hydrocarbon-based PEM increases the proton conductivity but also increases the chance of water flooding. Therefore, it is generally required to reinforce the composite PEM with excellent mechanical properties. The performance of different PEM composites is as summarized in Table 1.
For the gas diffusion layer (GDL), its functions consist of transporting the reactant gas from the flow channel to the catalyst layer, draining the water from the catalyst layer to the flow channel, and maintaining a constant moisture condition of the membrane at low humidity (Park et al. 2015). The BPPs deliver the reactants to the active catalyst layer through the GDL and the subsequent porous electrode while maintaining a minimum pressure drop (Kahraman and Orhan 2017). The advantages and limitations of using different types of GDL and BPPs are summarized in Table 2 and Table 3, respectively.

**TABLE 2. Performance of URFC with different GDL**

| Author               | GDL                  | Advantage                                                                 | Limitation                                  |
|----------------------|----------------------|---------------------------------------------------------------------------|---------------------------------------------|
| Hwang et al. (2012)  | Titanium (Ti)-felt   | The Ti-powder loading increase the effectiveness of water flooding prevention in wet condition (RH = 100%) | Not effective under dry condition (RH = 66%) |
| Ito et al. (2015)    | Through-plane Polytetrafluoroethylene distribution in Ti-felt      | Current density increased and improve liquid water discharge in wet condition (RH = 100%) | –                                           |
| Huang et al. (2012)  | Conventional carbon substrate as GDL & Iridium–titanium nitride (Ir–TiN) as MPL | Reduced carbon corrosion during water electrolysis mode | –                                           |
| Lee and Kim (2014)   | IrO₂/Pt/IrO₂ layered-electrode structure: Pt sandwiched between two IrO₂ layers in contact with a membrane carbon-based GDL | The electrochemical carbon corrosion is decreased and cyclic performance is significantly increased | –                                           |
| Hwang et al. (2013)  | Titanium (Ti)-felt   | Performance in FC mode increased in the dry condition as PTFE contents increase in FC mode | Flooding increased as PTFE contents increased |
| Alvar et al. (2014)  | Wet coating of carbon paper with Nb-doped TiO₂                      | Increased stability towards oxidation at the oxygen electrode               | –                                           |

**TABLE 3. Performance of URFC with different BPPs**

| Author               | Bipolar Plates                  | Advantage                                                                 | Limitation                                  |
|----------------------|---------------------------------|---------------------------------------------------------------------------|---------------------------------------------|
| Zhang et al. (2012)  | Honeycomb-like nanocomposite Ti-Ag-N Films | Showed excellent conductivity by the Ag nanoparticles and increased corrosion resistant. | –                                           |
| St. John et al. (2010)| Au-Nanoparticle Polyaniline Hybrid Coating | AuNPs-PANi showed excellent corrosion resistance at 0.61 V SHE and stability in cathodic PEFC environment | –                                           |
| Lin et al. (2014)    | SS304 Stainless Steel and Titanium Coated with Alternate Layers of TiN and ZrN | Improved corrosion resistance by 243 times                                | –                                           |
| Hwang et al. (2013)  | Serpentine-single flow field design | Improves the performance of FC mode electrolysis mode.                    | Deteriorate during the FC mode              |
| Lin et al. (2013)    | SS304 substrate coated with (Ti,Zr)N thin films | Improved corrosion resistance by 215 times                                | –                                           |

The results show that outstanding performance and increased corrosion resistance contribute to the improved efficiency of the URFC system. Studies into light weight and low cost bipolar plate materials are also actively carried out, such as the optimization work on conductive polymer composites by Irmayani and Suherman 2017. Regardless, further research is needed to find alternative BPP materials for a more favorable URFC system cost and performance.

The catalyst layer in URFC is crucial as it essentially exists to provide continuous pathways, i.e., an efficient path for the transport of protons, steady path of the pore network for the reactants/products and water withdrawal, and also a constant path for the conduction of electrons between the catalyst layer and the current collector (Zamel 2016). Some of the main setbacks of using PEMFC are the low durability and high cost of the electrocatalysts used, especially Pt (Lobato et al. 2016). Pt has been widely used as the primary catalyst, which also contributes to the excessive cost of PEMFC (Ebrahimi et al. 2016).
URFC CATALYST

URFC BIFUNCTIONAL HYDROGEN ELECTRODE (BHE)

Pt has been widely used as the main component in BHE. However, the use of non-Pt-based metal is preferred despite its lesser performance as a substitute for the electrocatalysts due to the scarcity and exorbitant cost of Pt.

Wang et al. (2016) suggested the idea of using an electrochemical catalyst of Phosphorus (P) and Nitrogen (N) dual-doped cobalt-based carbon nanofibers (Co-N-P-CNFs) using an electrospinning method. The doping of N/P atoms in the Co-based CNFs increases the electrocatalytic performance, showing low onset potential of ~0.216 V and a stable current density of 10 mA cm\(^{-2}\) at the potential of ~0.248 V in the HER.

Ding et al. (2015) employed the use of nickel-decorated carbon nanofibers via an electrospinning method. A Ni/C nanofiber hybrid with 8% Nickel (II) nitrate hydrate (Ni(NO\(_3\))\(_2\)-6H\(_2\)O) was prepared. The membrane exhibited low overpotential of ~0.17 V and high current density of 3.05 mA cm\(^{-2}\) at overpotential \(\eta = 200\) mV.

Huan et al. (2016) utilized nickel bis(diphosphine) complex [Ni(P\(_6\)N\(_5\))\(_2\)]\(^{2+}\) in 0.5 M H\(_2\)SO\(_4\) Aq. electrolyte attached on a multi-wall carbon nanotubes (MWCNTs) for bidirectional and reversible hydrogen evolution. They found that Ni-based electrode was ~35% less active for the hydrogen oxidation at 85°C and hydrogen production by ~20%. This cost-effective, molecular-engineered nanomaterial is able to transcend the conventional Pt electrodes but further research is needed to improve the long-term stability for the hydrogen oxidation.

URFC BIFUNCTIONAL OXYGEN ELECTRODE (BOE)

Currently, Pt in its reduced form is the best catalyst for oxidation-reduction reaction. However, it is not suitable for OER and therefore a different mixture of Pt, Ir, Ru, IrO\(_2\), or RuO\(_2\), and other compounds have been utilized as a bifunctional oxygen electrode (Yim et al. 2005). The oxygen electrode is responsible for both the oxygen reduction and evolution reaction. The oxygen electrode of choice should possess high catalytic activity, long-term durability, high electronic conductivity as well as being cost efficient. Currently, the oxygen electrode is only limited to using noble metal catalyst, which is Pt in its reduced form (Wang et al. 2016). The performance of various bifunctional electrodes is summarized in Table 4.

Various researches have been carried out to replace Pt as the electrocatalyst, including the use of graphene. Mao et al. (2014) identified N-doped crumpled graphene-CoO (Cobalt oxide) as the OER bifunctional electrocatalyst. The overpotential of the OER study showed ~0.34 V, which is the smallest reported for a Co-based OER catalyst.

Ye et al. (2018) employed PtRuIr nanoclusters as the bifunctional electrocatalyst for the OER and ORR. The PtRuIr electrocatalyst showed superior active catalytic activity, specifically for the 10 mol% of Ir that exhibited the highest electrocatalytic activity.

Iorioi et al. (2001) studied the use of iridium oxide/platinum electrocatalyst as a substitute for the conventional Pt catalyst. It was found that the fuel cell performance was marginally degraded but showed an increase in water electrolysis performance.

Tian et al. (2014) investigated the use of Chemical Vapor Deposition (CVD) method to fabricate a novel N-doped graphene (NG) with a single-walled carbon nanotube hybrid (NGSH). It was found that there is an increase in the OER and ORR of oxygen activity comparable to a Pt/C catalyst. This in turns demonstrates high ORR activity and also better durability and resistance to crossover effect.

Lin et al. 2013 used pyrolysis to fabricate a NG as an alternative cost-effective and facile method. The study discussed how an immense anodic current exists when electric potential is applied between ~0.6V and 0.8V, revealing the OER at the NG surface. Further testing showed that the current increased when the potential was extended to 1.0V.

Jahan et al. (2013) suggested that the graphene oxide (GO) is incorporated with copper-centered metal organic framework composite (Cu-MOF) as a tri-functional catalyst for ORR, OER, and HER. It was found that in PEMFC testing, the GO-incorporated Cu-MOF composite delivered 76% higher current density as compared to the commercial Pt. Based on the power density curve, they obtained a maximum power density of 145 mW cm\(^{-2}\) for the Pt catalyst and 110.5 mW cm\(^{-2}\) for the GO-incorporated Cu-MOF.

Unni et al. (2015) investigated the use of graphene nanotube as an electrocatalyst derived from single-walled carbon nanohorns (SWCNH) containing thin layers of graphene nanotube with iron oxide nanoparticles (FeGNT). It was found that FeGNT showed a 750 m\(^2\) g\(^{-1}\) surface area and assisted the distribution of Fe-Nx and quaternary N-based activation centers, providing stability towards ORR in acidic and alkaline media. The PEMFC performance at the cathode with Nafion as PEM was 200 mW cm\(^{-2}\) at 60°C.

ELECTROCATALYST SUPPORT

A suitable electrocatalyst support increases the surface area for catalyst to be dispersed on, thus reducing the loading of metal catalyst. Additionally, the support controls wettability and increases conductivity while reducing its resistance to corrosion. Carbon black is not suitable in the application of URFC as a support material since the material is easily corroded at high voltages during EL mode. Numerous researches have been carried out to find a suitable electrocatalyst support. Table 5 summarizes the compilation of electrocatalyst supports previously studied. Titanium carbide and nitride are shown to be superior to the conventional Pt-based catalyst (Fuentes et al. 2014; García et al. 2013; Roca-Ayats et al. 2014). Iridium oxide compounds were fabricated in different methods and revealed higher OER and ORR activity compared to Pt/C catalyst (Kim et al. 2015; Kong et al. 2012a). Additionally, antimony-doped tin dioxide shows promising stability and increased conductivity (Cruz et al. 2012; Gurrola et al. 2013).
TABLE 4. The performance of bifunctional electrodes in URFC

| References          | BOE                  | T (°C) | Onset Potential (V) | OER Activity | ORR Activity | Electrolyte          | Stability                       |
|---------------------|----------------------|--------|----------------------|--------------|--------------|----------------------|---------------------------------|
| Ye et al. (2018)    | PtRuIr nanoclusters  | 25     | –                    | 63.48 mA cm² | 53.68 mW cm² | Nafion (5 wt%, 45 mg cm⁻³) | ~13% activity loss after 7000 Seconds (RDE) |
| Mao et al. (2014)   | N-doped crumpled graphene-CoO (cobalt oxide) | 25 | ~0.90 | ~0.34 @ 10 mA cm⁻² Current Density | – | 5% Nafion in ethanol | ~8% (7.9) Relative current loss Chronoamperometric response after 16000s @ 1600 rpm |
| Tian et al. (2014)  | N-doped graphene w/single-walled carbon nanotube hybrid (NGSH) | 25 | 0.88 | 1.63 @ 10 mA m⁻² Current Density | 2.6 mA µg⁻¹ Current Density | Nafion (% wt% water solution) | |
| Lin et al. (2013)   | NG-1000 (N doping 2.4 at.%) | 25 | – | – | – | Nafion (0.05 wt%) | – |
| Jahan et al. (2013) | GO-incorporated Cu-MOF | 25 | -0.202 | 1.54 | -0.21 | 5 wt% Nafion | – |
| Ioroi et al. (2001) | Pt-IrO₂ | 80 | – | 51% at 500 mA cm⁻² | – | Nafion 115 | – |

TABLE 5. Electrocatalyst support performance

| Author            | Electrocatalyst support | Performance                                                                 |
|-------------------|-------------------------|-----------------------------------------------------------------------------|
| Won et al. 2018   | Ti₄O₇                  | Ti₄O₇ showed increased OER and ORR; Better roundtrip efficiency than Pt/C & Pt/Ti₄O₇ support. |
| Roca-Ayats et al. (2014) | TiC, TiCN & TiN | TiCN support showed highest activity towards OER & ORR. Stability of the MEA and roundtrip efficiency increased compared to Pt/C catalyst. |
| Roh et al. (2016)  | SiO₂-SO₂H              | ATO support showed promising stability with 0.6 V in PEMFC mode & 1.55 V in PEMWE mode at 80°C. |
| Cruz et al. (2012) | Sb-doped SnO₂ (ATO)    | ATO support showed increased stability compared to conventional carbon black. |
| Garcia et al. (2013) | TiC & TiCN | PtIr/TiCN catalyst showed enhanced ORR activity & stability rather than PtIr/TiC. |
| Pai and Tseng (2012) | Graphitized carbon | Graphitized carbon showed an increase in stability & roundtrip efficiency compared to conventional carbon black. |
| Gurrola et al. (2013) | Sb-doped SnO₂ (ATO)    | ATO support showed enhanced stability & conductivity compared to carbon black Vulcan XC-72. |
| Kim et al. (2017)  | Crumpled rGO           | Heat-treated Pt-Ir/rGÖs at 600°C showed increased ORR activity and nearly identical OER polarization trend compared to Pt/C catalyst. |
| Kong et al. 2015   | SBA-15 template (s-IrO₂) | s-IrO₂ catalyst revealed to have enhanced OER catalytic behavior and ORR activity rather than Pt/commercial IrO₂. |
| Kim et al. (2015)  | IrO₂, fabricated by flashlignt irradiation | Pt-IrO₂ showed higher OER activity compared to Pt/C catalyst. |
| Kong et al. (2012b) | Porous IrO₂            | Pt/porous- IrO₂ showed high OER & ORR activity compared to Pt/ commercial- IrO₂. |
| Fuentes et al. (2014) | TiC | Supported Pt-Ir/TiC showed better kinetic current per mass than Ir-black & Pt-based catalyst; enhanced roundtrip efficiency |
There are a variety of fabrication methods that have been developed essentially aimed at reducing the fabrication cost and efficiently producing the components of the URFC system. Kong et al. (2012c) incorporated Adams fusion method to prepare Irx(IrO_x)10-x composites catalyst support. This fast and efficient method provides oxides with a high specific area especially for the preparation of platinum oxide, ruthenium oxide, and other similar groups. Pai and Tseng (2012) developed an ultrasonic mixed technique process to fabricate a bifunctional graphitized carbon-supported Pt oxygen, producing fine Pt/graphite electrocatalyst in order to make it highly corrosion-resistant and stable.

Studies have also looked into the works of the catalyst layer application process. Ng et al. (2013) incorporated the use of the commercial anion exchange membrane (AEM) (Fumapem FAA-3, Fumatech) to fabricate their membrane electrode assembly (MEA). Lee and Kim (2014) suggested the use of catalyst-coated membrane (CCM) method to fabricate their MEA. Meanwhile, Ng et al. (2014) used the conventional catalyst-coated substrate (CCS) technique to synthesize their MEA. However, according to Ito et al. (2016), both CCS and CCM methods are suitable for MEA fabrication.

CONCLUSION

This review has carried out a comprehensive study on the development of the unitized regenerative fuel cell (URFC) system. The URFC system has a limitation that causes severe backunders towards the overall efficiency and this mainly concerns the degradation of the catalyst support, gas diffusion layer, and BPs due to carbon corrosion during the water electrolysis mode. Therefore, this issue is critically important to be identified in future studies to improve the URFC performance. It was found that the low cost non-metal graphene-based electrocatalyst shows promising stability and performance in fuel cell applications. Moreover, TiC is potentially a good electrocatalyst support to reduce the amount of Pt loading in the fuel cell. However, further research is needed for the non-metal electrocatalyst and electrocatalyst support to ultimately become viable and efficient for its application in URFC.

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