

# Review on the Unitized Regenerative Fuel Cells Materials

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## Abstract

Unitized regenerative fuel cell is a unique fuel cell can work in both electrolyser and fuel cell in one cell at different mode, its useful in some application in which the weight is an important issue. This article reviews current literatures with the objective of identifying the latest development in membrane and bipolar plate materials for the Unitized Regenerative Fuel Cell (URFC). The result shows that the choice of both the bipolar plates and the catalyst for URFC electrodes is a delicate task especially for oxygen side because a very good candidate for fuel cell mode will not function well in the electrolyser mode and therefore it is suggested that a compromise should be considered. It is recommended that coated aluminum, titanium or titanium with gold-coated layer to be very suitable candidates as the bipolar plate material and  $\text{IrO}_x$ , Ru or both with Pt is suitable for oxygen side catalyst in the URFC. for hydrogen side conventional Pt even with carbon will function well for both electrolyzer and fuel cell mode.

*Keywords:* Fuel cell; bipolar plate; Unitized; titanium; membrane, catalyst

## 1. Introduction

Fuel cell based power plants offer one of the most lucrative possibilities for future power generation and the fuel cell is also found to be potentially more efficient than the conventional plants since the fuel reacts electrochemically instead of combustion. In this way there is far less air, thermal, and noise pollution issues to be considered. A conventional fuel cell is an electrochemical device that produces electricity by separating the fuel (generally hydrogen gas) via a catalyst. The protons flow through a membrane and combine with oxygen to regenerate water with the help of the catalyst used while the electrons flow from the anode to the cathode to create electricity.

An electrolyzer operates reversely to the fuel cell which splits water into hydrogen and oxygen by the power supplied. To make a hydrogen system self dependant energy source, an electrolyzer

should exist to generate gases which then be consume to generate power through the fuel cell. For some applications a specific weight (power per unit weight) is an important issue to be considered, thus researches succeed in getting one cell to function as electrolyzer and fuel cell in one cell at different modes. This cell is known as a unitized regenerative fuel cell (URFC). The URFC works logically in the electrolyzer mode first to store hydrogen and then use it in the fuel cell mode, thus the URFC behaves like a battery as a self independent power source but in contrast to the battery system, the URFC is unaffected by the depth of the discharge or length of the cycle duration. In the URFC, energy and power are not linked i.e. the cell is sized for power but the storage tank is sized for energy [1]. A diagram of the electrochemical reaction that occurs in the proton exchange Membrane (PEM) of the URFC is shown in Figure 1.

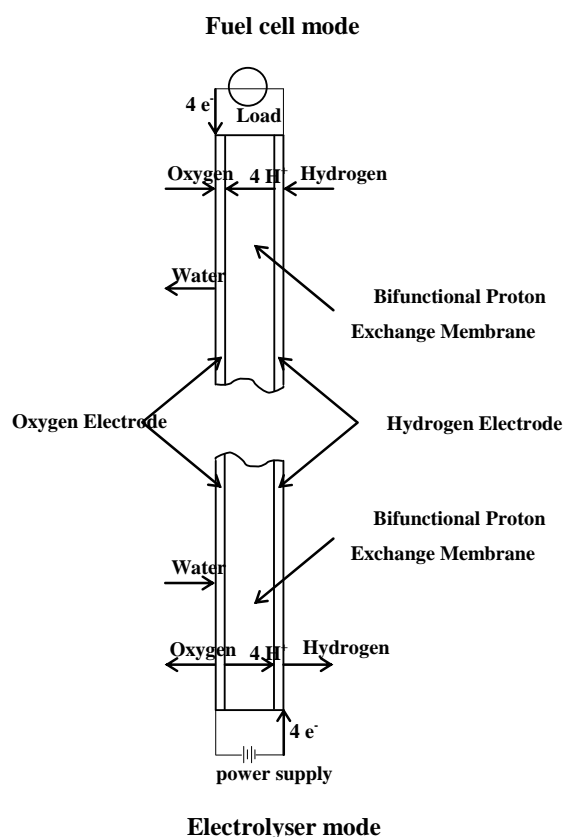


Fig.1. Electrochemical reaction in the URFC

The objective of this paper is to study the materials used for bipolar plate and the membrane in the URFC. For that purpose a review of the latest development in the materials used for the bipolar plate and the development in the Membrane Electrode Assembly (MEA) for URFC was carried

## 2. Bipolar plate

Flow channels are conveniently machined into a bipolar plate or field plate to allow a high electronic, good thermal conductivity and stability in a chemical environment inside a fuel cell [2]. In a fuel cell stack, each bipolar plate supports two adjacent cells and the bipolar plate is known to have five typical functions: (1) to distribute the fuel and its oxidants within the cell (2) to facilitate water management within the cell (3) to separate individual

cells in the stack (4) to transport currents away from the cell and (5) to facilitate heat management [3]. The sketch of a bipolar plate is shown in figure 2.

A cell potential of 1.6-2 V in an electrolysis mode with  $O_2$  is highly reactive and leads to a corrosive atmosphere. Thus, the bipolar plate material for URFC has to be corrosion resistant. Watstd [4] choose titanium as a bipolar plate and end plate material for his URFC. Hodgson [5] used coated titanium as a lightweight bipolar plate and it is found from this study that titanium can be employed to produce fuel cell with a very high volumetric and gravimetric power densities. Wind [6] investigated the effect of coating on a metallic bipolar plate for PEM to prevent the formation of oxide layers with high receptivity. Hung [7] compared the fuel cell performance of aluminum coated bipolar plate with a conventional fuel cell bipolar plate made of graphite. The results indicate that coated metal bipolar plates show a 22% savings in hydrogen consumption, higher efficiency and durability and without any sign of power degradation.

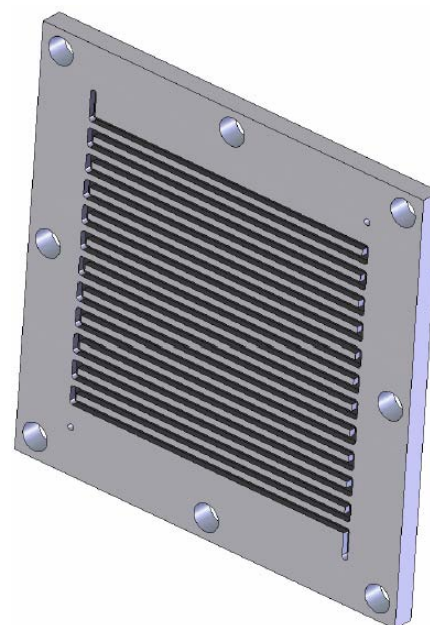


Fig.2: Bipolar Plate Used in PEM Fuel Cell Stack.

The electric conductivity for titanium (refer to Table: 1) is not as high as some other materials, but its high resistivity to corrosion makes it a very suitable candidate for URFC. For that reason an attempt to increase the electric conductivity was done by coating the surface of the titanium with a thin layer of gold. Wang [8] developed a light weight and corrosion resistant bipolar plate for the PEMFC. The bipolar plate was made from titanium and coated with gold. Wang [9] investigated the effect of surface modification on the titanium bipolar plate. Two different surface modification materials, iridium oxide (IrO<sub>2</sub>) and platinum were studied and the performance of these two plates is found to be very similar to the graphite plate.

Table 1  
Electrical conductivity for selected materials

Material	Electrical Conductivity (S/m)
Gold	45.2x10 <sup>6</sup>
Aluminum	37.7x10 <sup>6</sup>
Ruthenium	3.19x10 <sup>6</sup>
Titanium	2.34x10 <sup>6</sup>
SS 316	1.33x10 <sup>6</sup>
Graphite	1.27x10 <sup>5</sup>

### 3. (MEA) for URFC

Two problems are encountered when designing electrode structures for URFC systems. For PEM fuel cells, highly

hydrophobitised carbon paper or carbon cloth is usually adopted as the gas diffusion layer or electrode materials but however, they cannot be used as the gas diffusion layer (GDL) of a URFC for the following two reasons: firstly, the carbon materials tend to corrode at high potential on the oxygen electrode side during the water electrolysis operation; secondly, GDLs have to achieve an appropriate balance between the hydrophobic and hydrophilic properties for both the fuel cell and water electrolysis operations. The fuel cell operation requires that the oxygen GDL have a hydrophobic property to prevent water flooding, on the other hand, water electrolysis requires that the GDL has a hydrophilic property to supply water to the oxygen electrode.[10].a schematic sketch of a MEA for a URFC at different modes is shown in figure 3.

Some researchers have demonstrated to overcome or reduce the effect of those two problems. Swette [11] used Na<sub>x</sub>Pt<sub>3</sub>O<sub>4</sub> as a bifunctional oxygen electrode catalyst in the URFC. The results show a highly efficient bifunctional oxygen electrocatalysis. This study also used another approach by integrating two catalysts into dual-character electrodes like Pt/IrO<sub>2</sub>, Pt/RhO<sub>2</sub> or Rh/RhO<sub>2</sub>.The same approach can be applied to a single bifunctional catalyst as such electrodes also show efficiency as electrodes. Ahn [12] developed a new concept of the bifunctional electrode. In this new concept, oxidation and reduction reactions are assigned to the electrodes.

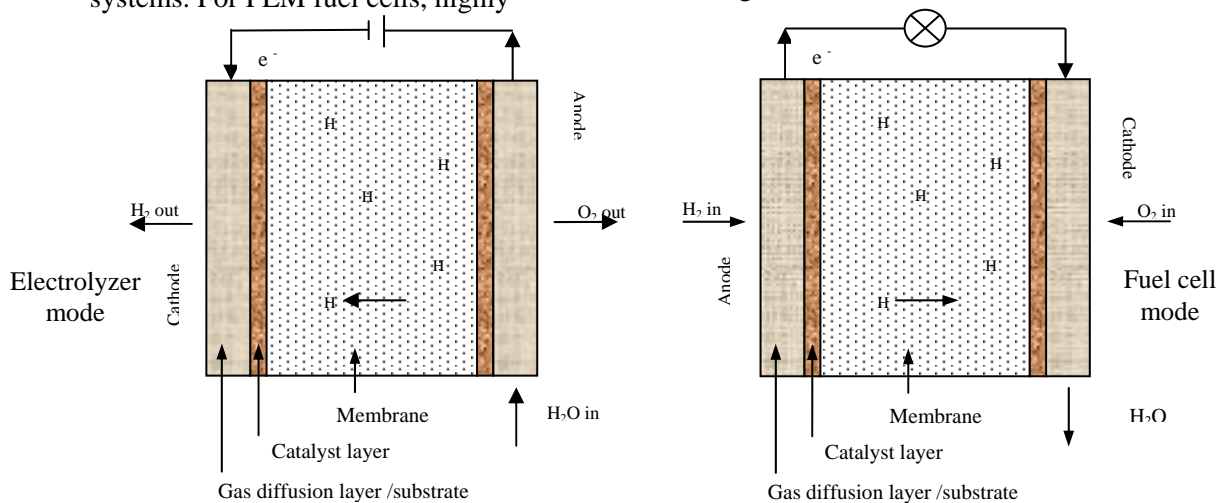


Fig.3. A Schematic of Membrane Electrode Assembly

Thus hydrogen oxidation (fuel cell mode) and oxygen evolution (electrolysis mode) proceed alternatively on one electrode, namely the oxidation electrode, whereas the two remaining reactions proceed at the reduction electrode. The suggested cell operating with these types of electrodes is known as an 'integrated water electrolysis and hydrogen /oxygen fuel' as opposed to the 'reversible' fuel cell. By using this concept, it is found to be less difficult to identify suitable electrode material for both electrodes.

Hari [13] tested a URFC with Pt: C ratio 20:80 at a loading of 1 mg/cm<sup>2</sup> as an electrode for the hydrogen side and Pt/Ir-on-C with a 20% iridium content and impregnated with 5% Nafion at approximately 0.5 mg/cm<sup>2</sup> as an electrode for the oxygen side. The data collection for the regenerative is first from the fuel cell mode and then the electrolyzer shows better electrolysis performance and finally as it slightly lowers for Pt/Ir-on-C than those for Pt-on-C electrodes. Swette [14] tested different oxygen electrode catalyst materials and different decompositions and compared the results with the baseline electrode with Pt at both the hydrogen and oxygen side. The results show that the regenerative PEM fuel cells display an efficient bifunctional performance that can be fabricated with the traditional Pt for the hydrogen side and Pt-Ir catalyst if the electrode structures are properly designed. This investigation has also identified RuO<sub>x</sub> as a catalyst with a good potential for improving the O<sub>2</sub>-evolution component of a bifunctional positive electrode. Ledjeff [15] shows the manufacturing procedure of coating Nafion117 membrane with a variety of different noble catalysts such as Ru and Ir. The resulting electrodes are very thin and show a high performance.

Zhigang [16] used platinum as a bifunctional catalyst for a hydrogen electrode and 50 wt. % Pt + 50 wt. % IrO<sub>2</sub> for a bifunctional catalyst of the oxygen electrode. The study also shows that using a thin catalyst layer can reduce the loading of the catalyst to 0.4 mg /cm<sup>2</sup> and minimizes mass transport and ohmic overvoltage at the same time giving the

cell a promising stability under any number cycle operations. Prakash [17] investigated the effect of using the ruthenium as a catalyst for both oxygen reduction and generation in URFC. The results show that ruthenium is quite an effective catalyst for the oxygen reduction and generation reaction. However; it suffers stability problems in an anodic mode. The study proposes the use of anion-conducting polymers both as membrane over layers and as integral components of gas-diffusion electrode as it promises to alleviate this problem.

Ioroi [18] showed that the preparation procedure of the IrO<sub>2</sub> /Pt catalyst can affect the performance of the URFC, comparing the result of catalyst prepared by mixing IrO<sub>2</sub> /Pt and the one with deposited IrO<sub>2</sub> /Pt. The deposited method shows 2-3 % improvement of the roundtrip efficiency and this enhancement is due likely to the microstructure of the electrocatalyst. Chen[19]prepared electrode arrays containing 715 unique combinations of five elements (Pt, Ru, Os, Ir, and Rh) by the Borohydride reaction of aqueous metal salts. Catalysts that showed high activity for both reactions and good resistance to anodic corrosion have been identified in the Pt-Ru rich region of the Pt-Ru-Ir ternary. The ternary catalyst Pt<sub>4.5</sub>Ru<sub>4</sub>Ir<sub>0.5</sub> (subscripts indicate atomic ratios) is significantly more active than the previously described Pt<sub>1</sub>Ir<sub>1</sub> bifunctional catalyst for both reactions.

Ioroi [20] showed that the PTFE content in the electrocatalyst layer affected only the fuel cell performance. The electrode containing 5-7 wt. % PTFE was appropriate for URFC and also the results show that the Nafion content in the electrode affected both the fuel cell and water electrolysis performance; the electrode containing 7-9 wt. % Nafion showed good performance, and the addition of a small amount of iridium catalyst (about 10 at. %) to the oxygen electrode layer significantly improved the URFC performance. Ioroi [21] studied the relation between the Polytetrafluoroethylene (PTFE) coating amount on the Gas Diffusion Backing (GDB) and the URFC

performance. The PTFE coating on the GDB of the H<sub>2</sub> electrode does not affect the cell performance. But however, the URFC performance depends significantly on the coating of O<sub>2</sub> electrode side. The study also concludes that the cell with the smaller PTFE loading amount on the O<sub>2</sub> GDB exhibited a higher fuel cell and lower water electrolysis voltage. The results of the alternate operations between fuel cell and water electrolysis cycle tests reveal that a stable operation of the URFC cycle is achieved using the appropriate amount of PTFE loading on the GDB (16 mg cm<sup>-2</sup>).

Irooi [22] also reported the testing of the URFC system for the fuel cell technology program at the Glenn research Center (GRC) and plans to minimize the system weight, volume, and parasitic power as its goal. The design concept currently being developed uses no pumps to circulate coolants or reactants, and minimizes the ancillary components to only the oxygen and hydrogen gas storage tanks, a water storage tank, a loop heat pipe to control the temperature and two pressure control devices that control the cell stack pressure during operation.

Yim [23] employed various oxygen electrode catalysts for URFC. The results show that Pt+Ir was the best in URFC's performance and cyclic stability, revealing the least activity loss in fuel cell mode and significant activity improvement in water electrolysis mode by adding Ir to Pt black. Burke [24] developed a unique lightweight URFC and the cell is unique in that it uses regenerative gas dryers/humidifiers that are mounted on the surface of the gas storage tanks that act as the radiators for thermal control of the URFC system. The system's function is to dry the hydrogen and oxygen gases produced by electrolysis when the gas storage tanks cool down during the charging mode and to humidify the hydrogen and oxygen gases used by the fuel cell.

Lee [25] proposed a novel method of direct deposition of Pt catalysts on Nafion membrane modified with polypyrrole for the URFC. The study demonstrated that the polypyrrole could play a role as an electronic sink material for Pt catalyst

deposition and the subsequent chemical reduction. Yim [26] tested experimentally a single cell URFC system. The results show that the fuel cell mode performance in the order of Pt black >PtIr>PtRuO<sub>x</sub>>PtRu~ PtRuIr>PtIrO<sub>x</sub> and water electrolysis performance PtIr ~ PtIrO<sub>x</sub>> PtRu> PtRuIr>~ Pt black. For both modes PtIr shows the best URFC performance and with increasing Ir or IrO<sub>x</sub> composition from 0 to 50 % in PtIrO<sub>x</sub> catalyst the fuel cell performance decreases while the water electrolysis significantly improves. Wittstadt [4] sputtered platinum on the oxygen electrode for URFC after hot pressing. The main enhancement is caused by the higher fuel cell mode efficiency, the round-trip efficiency is 4% higher than the one of the standard MEA. Song [27] developed a novel bifunctional oxygen electrode with a thin-film electrocatalyst layer and a corrosion-resistive gas diffusion layer (GDL) prepared by the carbon paper backing and a protective micro-porous layer (MPL). The protective MPL is made of the IrO<sub>2</sub> and deposits fine Ti powders. This electrode shows slight performance loss during 20 cycles.

Yao [28] prepared a bifunctional electrocatalyst for oxygen electrodes of URFC by chemical deposition of platinum on IrO<sub>2</sub> powder in aqueous solutions. The study also explains the differences between the deposition and mixing methods of preparing Pt/IrO<sub>2</sub> on the electrode surface. The results show that the oxygen electrode with IrO<sub>2</sub>-supported Pt prepared by the deposition method exhibits slightly lower oxygen reduction reaction (ORR) activity but markedly a higher oxygen evolution reaction (OER) activity than a mixture of Pt and IrO<sub>2</sub>. Zhang [29] tested the performance of fuel cell/water electrolysis of URFC. The electrocatalyst for the oxygen side of the URFC is bifunctional RuO<sub>2</sub>-IrO<sub>2</sub>/Pt. The results show that there are differences in morphology and structure between deposited RuO<sub>2</sub>-IrO<sub>2</sub>/Pt and mixed RuO<sub>2</sub>-IrO<sub>2</sub>/Pt. and that the performance of URFC using deposited RuO<sub>2</sub>-IrO<sub>2</sub>/Pt electrocatalyst with a high Dispersion is better than that of URFC using mixed RuO<sub>2</sub>-IrO<sub>2</sub>/Pt electrocatalyst.

Table 2  
Summary of the studies catalyst

Author	Year	Catalysts
Swette	1991	Pt/Ir(O <sub>2</sub> side) Pt/RhO <sub>2</sub> (O <sub>2</sub> side) Rh/RhO <sub>2</sub> (O <sub>2</sub> side)
Hari	1993	Pt/Ir (O <sub>2</sub> side)
Swette	1994	Pt (H <sub>2</sub> side) Pt/Ir (O <sub>2</sub> side) RuO (O <sub>2</sub> side)
Ledjeff	1995	Ir (O <sub>2</sub> side) Ru (O <sub>2</sub> side)
Zhigang	1999	Pt (H <sub>2</sub> side) Pt+IrO <sub>2</sub> (O <sub>2</sub> side)
Prakash	1999	Ru (H <sub>2</sub> side) Ru (O <sub>2</sub> side)
Ioroi	2001	IrO <sub>2</sub> /Pt (O <sub>2</sub> side)
Chen	2001	Pt <sub>4.5</sub> Ru <sub>4</sub> Ir <sub>0.5</sub> (O <sub>2</sub> side)
Ioroi	2002	Ir (O <sub>2</sub> side)
Yim	2004	Pt+Ir (O <sub>2</sub> side)
Yim	2005	Pt+Ir(O <sub>2</sub> side) Pt+Ir(H <sub>2</sub> side)
Song	2006	IrO <sub>2</sub> (O <sub>2</sub> side)
Yao	2007	P+IrO <sub>2</sub> (O <sub>2</sub> side)
Zhang	2007	RuO <sub>2</sub> –IrO <sub>2</sub> /Pt (O <sub>2</sub> side)

The listed of the materials used for URFC membrane electrode assembly are listed in table 2.

#### 4. Hydrogen storage

As mentioned before a URFC have double functions ability over the conventional fuel cell in which it can be work as electrolyzer as well as fuel cell, for that to make the system self dependant source of energy ,the generated hydrogen should be stored in some media to use it during the fuel cell mode .generally hydrogen storage is a key enabling technology for the advancement of hydrogen and fuel cell power technologies in transportation, stationary, and portable power applications [30].

Hydrogen has the second lowest boiling point and melting points of all substances, second only to helium. Hydrogen is a liquid below its boiling point of 20 K ( $-253\text{ }^{\circ}\text{C}$ ) and a solid below its melting point of 14 K ( $-259\text{ }^{\circ}\text{C}$ ) and atmospheric pressure. The density of hydrogen also very low ( $0.08245\text{ kg/m}^3$ ) therefore the hydrogen only exist as a gas at the ambient conditions[31].

Basically hydrogen can be stored mechanically and /or chemically; four basic hydrogen storage options are [32]

- **Liquid hydrogen storage**

The liquefaction of hydrogen can only achieved cryogenically by mechanical compression and cooling [32]. Lowering the temperature of hydrogen to its boiling point at ( $-253\text{ }^{\circ}\text{C}$ ) at atmospheric pressure requires approximately 79 kJ/mol.To put this value into perspective ,this energy amounts to a third of the lower heating value (LHV 242 kJ/mol) and over a quarter of higher heating value (HHV 286 kJ/mol )of hydrogen. In other words, the overall energy efficiency has already significantly dropped by the time the cryogenic tank is filled[2]

- **Adsorber storage**

Hydrogen can also be adsorbed (as molecules in the gaseous state) in adsorbing material such as an active carbon. However, in order to obtain a specific energy comparable to  $\text{LH}_2$  it is

necessary to cool the adsorber to very low temperatures (about  $-200\text{ }^{\circ}\text{C}$ )[32].Another ,interesting adsorber method is the graphitic nanofibers (GNFs) this fiber consists of graphene sheets aligned in a set direction (dictated by the choice of catalyst )and this material have the ability to adsorb hydrogen.Reversible storage capacities for this technique has been found to be 4-7 % ,but these values need further verification to provide a reliable benchmark for nanofibers [33].

- **Metal hydride storage**

Metal hydride storage is another promising method to store hydrogen in which the hydrogen stored in solid state. Solid materials such as lithium nitride ( $\text{Li}_3\text{N}$ ) and ammonia boron ( $\text{NH}_3\text{BH}_3$ ) have high potential storage capacities that exceed the 2015 US DOE target of 9 wt %hydrogen Woolf [34]used a ternary nitrides lithium boron nitride (  $\text{Li}_3\text{BN}_2$ ) and magnesium boron nitride (  $\text{Mg}_3\text{BN}_3$ ) as a light metal hydride.

- **Pressurized hydrogen ( $\text{PH}_2$ ) gas storage**

The traditional method to store hydrogen is to compress and store it in small vessels or large tanks[32].Conventional carbon wrapped aluminum cylinders can store hydrogen at pressure up to 550 bar ,although national legislation and codes of practice may limit the allowable pressure to a value well below this. In most countries, gas cylinder are typically filled up to a maximum 248 bar [2] .

The compression of hydrogen is achieved by using of conventional compressors which is still an economically and simple way for high volume production .for several applications, like small electrolyzers, it is necessary to compress the hydrogen in low quantities to high pressure .for this low power performance region, mechanical hydrogen compressors are not very efficient .An alternative way to compress hydrogen is the use of an electrochemical hydrogen pump. New material developments for the polymer electrolyte membrane (PEM) fuel cells make it possible to realize PEM

hydrogen compression with more than 80% efficiency [35].conventional gas cylinders used to store hydrogen up to about 250 bar are shown in figure 4.



Fig. 4 hydrogen cylinders

## 6. Conclusions

This paper reports the development on bipolar and membrane material for conventional and unitized regenerative fuel cells and the following conclusions from this study can be summarized as:

1. Selecting titanium for URFC is a good choice in terms of its high resistivity to corrosion but since its electric conductivity isn't high so it's recommended to coat the titanium bipolar plate with a gold layer to enhance the conductance ability.
2. Choosing URFC oxygen side catalyst is a delicate task because it's a very good candidate for fuel cell mode and will be not suitable for the electrolyser mode, thus a compromise should be made to get an acceptable dual cell function by adopting  $\text{IrO}_x$ , Ru or both with platinum. This will be a very good candidate for the oxygen side URFC
3. For the hydrogen side the matter is less difficult because the corrosion environment does exist anymore in the hydrogen side and thus a Pt/C will be an acceptable candidate

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