

Current Trends in Fuel Cell Technology- A Review

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Abstract: Fuel cells are the devices converting chemical energy of various compounds to electrical energy, a more usable form of energy. Many researchers have been working on the fuel cells technology to get the energy efficiently with bulk quantity. The most important properties required for an ideal PEM can, therefore, be outlined as electrically insulating, good proton conductivity, and impermeable to gases and/or fuel to prevent gas/fuel crossover.

Methane Fuel cell, Graphite oxide fuel cells and the metal oxide fuel cells are the recent research fields, the advancements in these will be discussed in this paper.

Key Word: Fuel cells; Fuel cell performance; Methane fuel cell; Graphite oxide fuel cells.

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I. Introduction

A fuel cell is a device that converts chemical energy to electrical energy by a chemical reaction of positively charged Hydrogen ions and any Oxidizing agent. There are many types of fuel cells, but they all contain an arrangement of an Electrolyte, a Cathode and an Anode. At the anode a catalyst oxidizes the fuel, usually hydrogen, turning the fuel into a positively charged ion and a negatively charged electron. The electrolyte is a substance specifically designed so ions can pass through it, but the electrons cannot. The freed electrons travel through a wire creating the electric current. The ions travel through the electrolyte to the cathode. Once reaching the cathode, the ions are reunited with the electrons and the two react with a third chemical, usually oxygen, to create water or carbon dioxide.

There are many kinds of fuel cells, which operate on different fuels (acting as a source of hydrogen) and have varying operating temperatures, covering a whole range from 50°C up to 1000°C.

Fuel cells are commonly classified on the basis of their electrolyte¹ according to which they can be divided into five main groups:

1. Alkaline Fuel Cell (AFC),
2. Phosphoric Acid Fuel Cell (PAFC),
3. Polymer Electrolyte Fuel Cell (PEFC),
4. Molten Carbonate Fuel Cell (MAFC) and
5. Solid Oxide Fuel Cell (SOFC).

PEFC can be further sub-divided into three groups, the general PEFCs feed on hydrogen, Direct Methanol Fuel Cell (DMFC) and Direct Ethanol Fuel Cell (DEFC).

Direct alcohol fuel cells (DMFC and DEFC) are similar to the PEFC in operation but as the names suggest, they feed on methanol and ethanol, respectively. PEFC (including DMFC and DEFC) are preferentially used in small devices and in transport applications that require quick start-up and do not require very high power. Among the various components of a PEFC is the proton exchange membrane (PEM) that plays the most vital role, i.e. separating the anode from the cathode, which enables the current flow through an external circuit. The performance of a PEM strongly affects the durability and efficiency of the fuel cell system. The most important properties required for an ideal PEM can, therefore, be outlined as

- (1) electrically insulating,
- (2) good proton conductivity, and
- (3) impermeable to gases and/or fuel to prevent gas/fuel crossover.

Among these different types of fuel cells, SOFCs has big advantage on combination of environment-friendly power generation with fuel flexibility². As SOFCs are practicability at high temperature between 1073 K and 1273 K, several alternative fuels such as hydrogen, biogas, bio-ethanol and bio-methanol can be directly used in an SOFC³. Although hydrogen is considered as an ideal fuel, there exist problems associated with its production, storage and transportation⁴. Emission of greenhouse gases resulting from the use of natural gas and methane rules-out these as potential alternatives for energy production. In recent years, ammonia (NH₃) has emerged as a promising fuel for electricity generation in SOFCs because it is relatively cheap, carbon-free, easy

to store and transport, less flammable than other fuels and relatively safe because any leakage is easily detectable from the odour⁵.

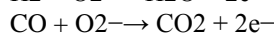
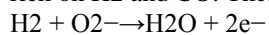
II. Literature Review

Bio-Methanol as Fuel

Methanol could be also easily reformed back to a mixture of CO and H₂ at relatively low temperatures compared with ethanol where decomposition reaction ($C_2H_5OH \leftrightarrow 3 H_2 + CO + C$) dictates the equilibrium products only above 850 °C giving rise of coking. It has been shown that although feasible the direct utilization of methanol can lead to cumulative carbon deposits which may modify the anode microstructure and reduce subsequently the SOFC performance. Many efforts have been made to investigate the effect of carbon deposition on SOFC performance. Saunders et al.⁶ found that carbon formed on Ni anodes when operated on methanol and the amount of carbon deposition was increased with decreasing temperature over the range of 700–850 °C. In contrast, Jiang and Virkar⁷ reported that there was no carbon deposition when SOFC operated on direct methanol at temperatures as low as 550 °C. Another investigation, reported by Bi et al.⁸, has also achieved good results when using Ni-GDC anodes without elucidating the carbon deposition problem that appears on the anode. Bio-methanol is much different from other hydrocarbon fuels. It carries sufficient oxygen to avoid entering into the thermodynamically unstable, carbon-forming regime. So, anode coking and deactivation and structure instability seems to be reduced using bio-methanol.

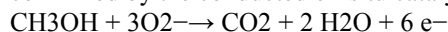
Recently, Li et al.⁹ reported an improvement of the SOFC performance using a reduced $La_2Ni_{0.9}Fe_{0.1}O_{4+\delta}$ (LNF) as anode under methanol feed. They proved that the carbon deposited shows a reasonable stability and has high degree of graphitization bringing a higher electrical conductivity which provides more conductive paths for electrons. Hence, methanol could be considered as a promising candidate fuel for SOFC. Therefore, the suitability to run SOFC on methanol makes it a possible choice for APUs used in several transportation applications. Indeed, an EU-funded METHAPU project started at 2006 and reached a significant milestone in 2010 when consortium proved for the first time the trials on a prototype of a methanol-based SOFC-APU. The prototype is tried and tested for performance and emissions under real-life conditions onboard a vessel (the MV Undine, a pure car truck carrier (PCTC)) involved in trading between Asia, Europe and the USA. The fuel cell unit, which has a nominal output of 20 kW, is based on planar SOFC technology fueled by methanol. Methanol is particularly suited for fuelling the Wärtsilä's WFC20 fuel cell unit since it can be easily reformed to a composition suitable for the unit. Methanol is available in all major harbours. This 20 kW SOFC-APU unit has been demonstrated, with a longer term target of 250 kW^{10,11}

In solid oxide fuel cells, methanol (CH₃OH) is directly fed to the anode chamber of the fuel cell, and air is supplied to the cathode chamber of the cell. When methanol reaches the anode/electrolyte functional layer, pyrolysis and catalytic decomposition reactions of methanol can take place leading to a gas-phase composition rich on H₂ and CO. These species will react electrochemically following Equations to produce current.

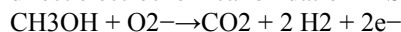


Oxygen in air available at the cathode/electrolyte interface reacts with electrons to form oxygen ions. The oxygen ions are transported through the ceramic electrolyte and electrons flow through the external electric circuit (load)

Lo Faro et al.¹² have also concluded that under OCV, part of methanol is not converted to syngas as also confirmed by the conducted ex-situ catalytic test giving rise to its direct oxidation reaction:



Other authors^{13,14} have reported also the possibility of total equation above and partial equation below methanol direct electrochemical oxidation in SOFC anode.



Thus, electrochemical oxidation of unconverted fraction of methanol could be considered through the mixed reaction dynamics of the anode fed directly by methanol.

Graphite-Oxide Incorporated CeP2O7 Electrolyte

Research on proton exchange membranes for mid-range temperatures (100–200 °C) has usually focused on polybenzimidazole (PBI)-based membranes, and PBI is typically loaded with phosphoric acid as a proton conductor. Phosphoric acid has specific properties, making it a desirable candidate for mid-range temperature electrolytes. However, the limitations of liquid phosphoric acid, such as its deactivation via phosphate anion adsorption at positive potentials, acid leaching, and its degradation to pyrophosphate at temperatures over 160 °C, restrict PBI use in higher temperature ranges^{15,16}. Therefore, the proton membranes that can endure temperatures over 200 °C for PEMFC are normally made of inorganic materials. Tetravalent metal ion pyrophosphates (MP₂O₇, M = Sn, Ti, Ce, or Zr) have been proposed in recent papers because they exhibit proton conductivities at temperatures between 200 °C and 400 °C^{17,18,19,20}. Hibino et al.^{21,22}

reported that the proton conductivities of SnP2O7-based materials increased via doping with trivalent or divalent elements. Park et al. researched CeP2O7, which exhibited a conductivity of $2.1 \times 10^{-4} \text{ S cm}^{-1}$ at 175 °C²³. However, the lower conductivity and weaker strengths of MP2O7 membranes limit their application to high-temperature PEMFCs; therefore, the modifiers that could improve the conductivity and form strong membranes have attracted more attention recently.

The structure of boron phosphate (BPO4) contains alternating PO4 and BO4 tetrahedral linked by shared oxygen atoms to form a three dimensional network, and BPO4 facilitates proton conduction in the absence of water to increase the conductivity. Furthermore, the strong water retention capability of the BPO4 powder could increase the strength of the MP2O7 particles to form stronger membranes. These properties provide the possibility of using solid BPO4 powders to replace liquid phosphoric acid as a proton conductor salt and binder for membranes. According to our previous work²⁴, GOs with different acidic functional groups, such as carboxylic acid and epoxy oxygen, offer more facile proton hopping, and a GO nanofiller extends the number of available ion exchange sites per cluster, resulting in increased proton mobility^{24,25,26,27,28}. Hence, GO should be a suitable filler for improving composite membrane proton conductivities at high temperatures.

The proton conductivity of the composite membrane was measured using a two-point probe method with an Autolab PGSTAT302. The impedance measurements were carried out between the frequencies of 1–106 Hz, and the applied voltage was 10 mV for the highest conductivities and reached up to 50 mV for the least-conductive materials. To ensure the membranes reached a steady state, the membranes were held at the desired conditions for 2 h before testing, and the measurements were taken at 30 min intervals. The crystal structures of different powders were analyzed using X-ray diffraction (XRD, X'Pert PROMPD), with a scan range of 5–80° over 2 h. The morphologies of the CeP2O7/BPO4 membranes were investigated via a SU-8020 Scanning Electron Microscope. Fourier transform infrared spectroscopy (FTIR) of the different samples was performed on a Nicolet 67FT-IR spectrometer system between 4000 and 300 cm⁻¹. The thermal behavior was analyzed by thermogravimetric analysis (TGA, STA449F3).

Inorganic composite CeP2O7/BPO4 and CeP2O7/BPO4/GO electrolytes without liquid phosphoric acid were prepared successfully, and the morphology and structure of the membranes were characterized. GO-incorporated CeP2O7/BPO4 composite membranes exhibited suitable conductivities ranging from 0.09 to 0.14 S cm⁻¹ at temperatures ranging from 200 to 250 °C, although the membrane conductivity subsequently decreased to 0.09 S cm⁻¹ as the temperature increased up to 300 °C. For the fuel cell tests, the CeP2O7/BPO4/GO composite membrane also provided superior performance results (with a peak power density: 240 mW cm⁻² at 250 °C) compared with the 215 mW cm⁻² result of the CeP2O7/BPO4 membrane. The composite with GO exhibited improved conductivity and fuel cell performance, especially under no-phosphoric conditions, indicating that CeP2O7/BPO4/GO is a suitable potential candidate as an electrolyte for high temperature PEMFCs.

Using Metal Oxide:

Solid oxide fuel cell (SOFC) technology has been developed successfully with viable commercialization. However, the wide implementation is not yet realized due to the high investment. One of the reasons is its complex technology, resulting chemical and thermal incompatibility issues amongst all cell components, i.e., anode, electrolyte, cathode and interconnectors^{29,30,31}. Single layer fuel cell (SLFC) was proposed due to the much reduced material requirement, only one material is used instead of three in conventional fuel cells components; anode, electrolyte and cathode. It is thus quite promising to avoid above mentioned issues. He et al. first introduced this concept based on Sr-doped LaInO3 (LSIO)³². However, a maximum power density of 3 mW cm⁻² at 800 °C was obtained for SLFC device even with Pt as anode and cathode current collectors. Very recently, Lan and Tao reported the performance of an analogous SLFC using the layered structure metal oxide, Lix-Co0.5Al0.5O2 (LCAO)³³ having a configuration of Ag/LCAO (0.79 mm)/Ag. This type of SLFC reached a much higher power output of 150 mW cm⁻² at 650 °C. The authors claimed that LCAO layered structure oxide displayed a high proton conductivity of 0.1 S cm⁻¹ at 500 °C in H2 containing atmosphere, beside its inherent electronic conductivity. Interestingly, such a mixed proton and electronic conductor did not cause the device short circuiting problem in fuel cell condition, which deserves further studies though the LCAO decomposition is an unavoidable challenge. In parallel, we successfully demonstrated that two-phase composite materials showed practical SLFCs application since 2010 because of mixed ionic and electronic or more precisely semi-ionic conductive property^{34,35,36,37,38,39}. We could intentionally adjust the two constituent materials composition and microstructure to reach a balanced electron and ion conductivities, which is one of the key principles for high performance target, close to even higher than 1000 mW cm⁻² at 550 °C^{40,41,42}.

Extensive efforts have been performed in our group on the fabrication and characterization of two-phase composites for SLFCs. In this regard, the single phase ionic conductors; doped ceria such as Ce0.8Sm0.2O2 (SDC) and Ce0.9Gd0.1O2 (GDC), or ceria-carbonate composite type, and the semiconductors: LiNiO2 and its doped analogues or composite with transition metal oxides, such as ZnO, CuO, SrO, FeO, along

with modified composite materials consisting of Mg_{0.6}Zn_{0.4}O/SDC have been developed^{39,42,43,44,45,46,47,48,49}. These semiconductor-ionic composite based SLFCs achieved excellent device performances to be technically useful for low temperature application (<600 °C): power outputs ranging from 400 to 1000 mW/cm² were delivered below 600 °C. Dong et al.,⁴¹ followed our approach and reported the performance of SLFC constructed by using two-phase composite material based on perovskite oxide, Sr₂Fe_{1.5}Mo_{0.5}O_{6-d} (SMF) and ionic Na₂CO₃-SDC (NSDC) materials. They found that a well matched electronic and ionic conductivity was achieved in 30% SMF/70%NSDC composite, which resulted in the highest power density of 360 mW/cm² at 750 °C.

III. Conclusion

Using Hydrogen Fuel Cell

Power density of about 350 mW·cm⁻² was achieved when IT-SOFC is fed with dry methanol at 750 °C. Carbon formation was observed on the Ni/SDC anode surface when fueled with pure methanol and then removed when a switch of H₂/methanol testing procedure was proposed. The occurrence of several chemical reactions mechanisms including the methanation, WGS and Boudouard reactions could be at the origin of the coking removing.

The GC measurements clearly confirmed that H₂/CO/ CH₃OH reactions may contribute to the electrochemistry the IT-SOFC anode fed directly by methanol at 700 °C. A series of probable reactions process taking into account the double (H⁺/O²⁻) conducting LN-SDC electrolyte is thus proposed. However, H₂ and CO electrochemical oxidation reactions seem to govern the anode process at 750 °C. This system has a great potential and several advantages to be used as an APU component in a specific eco-transportation sector. Indeed, tested cell is shown promising to be employed as a component in an APU-SOFC system in comparison to Gen 3 commercial cell developed by Delphi but still requires design optimization to limit ohmic losses and coking. Overall, the successful bio-methanol fed IT-SOFC has a great potential and impact on energy and environment whereas more research is required to achieve this technology up-scaling.

Using Graphite

Inorganic composite CeP₂O₇/BPO₄ and CeP₂O₇/BPO₄/GO electrolytes without liquid phosphoric acid were prepared successfully, and the morphology and structure of the membranes were characterized. GO-incorporated CeP₂O₇/BPO₄ composite membranes exhibited suitable conductivities ranging from 0.09 to 0.14 S/cm at temperatures ranging from 200 to 250 °C, although the membrane conductivity subsequently decreased to 0.09 S/cm as the temperature increased up to 300 °C. For the fuel cell tests, the CeP₂O₇/BPO₄/GO composite membrane also provided superior performance results (with a peak power density: 240mW/cm² at 250 °C) compared with the 215mW/cm² result of the CeP₂O₇/BPO₄ membrane. The composite with GO exhibited improved conductivity and fuel cell performance, especially under no-phosphoric conditions, indicating that CeP₂O₇/BPO₄/GO is a suitable potential candidate as an electrolyte for high temperature PEMFCs.

Using Metal Oxide

Based on the layered structure LiNi_{0.8}Fe_{0.2}-oxide, we have developed functional single layer composite for advanced SLFCs materials. The pure LNF has limited ionic conductivity in air while enables proton conductivity in H₂/air or hydrogen containing environment. By introducing the oxygen ion conducting phase e.g., SDC (samarium doped ceria) to form the two phase LNF-SDC composite, the ionic conductivity was significantly enhanced for high performance target. The comparable ionic and electronic conductivities at 0.1 S/cm level resulted a great increase of power density up to 760 mW/cm² of SLFC with LNF-SDC composite materials, much superior than 150 mW/cm² achieved using the pure LNF oxide. The mechanisms and scientific principles are very different from traditional electrolyte based fuel cell (anode/electrolyte/cathode). At this stage, probably the bulk p-n junction and new physical interfaces may explain the SLFC function and the excellent electrochemical performance. However, the investigation of the exact scientific nature of the underlying processes is still in progress.

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