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Review Article

Fuel Cell Technology for Vehicles

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Abstract

The development and usage of fuel cells in a variety of applications have come a long way. Fuel cells hold great promise for fueling alternative fuel vehicles. Fuel cells offer a promising alternative to conventional fossil fuel systems, due to their high efficiency, low environmental impact and flexible application. The direct methanol fuel cell (DMFC) has been discussed recently as an interesting option for a fuel-cell-based mobile power supply system in the power range from a few watts to several hundred kilowatts, which has a proton conducting polymer membrane as electrolyte. While the fuel used in most fuel cells is hydrogen (e.g. in the polymer electrolyte membrane fuel cell: PEMFC), the DMFC uses methanol as fuel. Fuel cells are attractive for several applications; however, there are several barriers, which must be overcome before they can become an alternative to internal combustion engines.

Keywords*: fuel cell, alternative fuel, direct methanol fuel cell, membrane.*

Introduction

A fuel cell is a device in which the energy of a fuel is converted directly into electricity direct current (DC) by an electrochemical reaction without resorting to a burning process, rather than to heat by a combustion reaction. The chemical energy of the fuel is released in the form of an electrical energy instead of heat when the fuel is oxidized in an ideal electrochemical cell. Energy conversion by a fuel cell depends largely upon catalytic electrodes, which accomplishes the electrochemical reaction to convert fuel into electric energy without involving the burning process. Efficiencies of fuel cells (40–85%) are considerable high compared to heat engines (A.Demirbas et al ,2007)

The first fuel cell was invented in 1839 by Sir William Robert Grove (Aravindhababu, P et al,1999). He is known as father of the fuel cells. At the London Institution, where he was professor of physics (1840–1847), he used his platinum-zinc batteries to produce electric light for one of his lectures. The energy chemically stored in the fuels is converted into electric current by means of an electrochemical process in the fuel cell.

A fuel cell produces electricity directly from the electrochemical reaction of hydrogen, from a hydrogen-containing fuel, and oxygen from the air. H_2 is the ideal fuel for a fuel cell, the infrastructure for producing and storing. Hydrogen is industrially produced by steam reformation of naphtha oil, methane, and methanol. High purity hydrogen has been mainly used as a fuel for low temperature fuel cells such as polymer or alkaline electrolyte fuel cells (Lin, Y.-M et al 2000)

Fuel cells offer a promising alternative to conventional fossil fuel systems, due to their high efficiency, low environmental impact and flexible application. One of the suggested systems for residential, automotive and portable applications is the direct methanol fuel cell (DMFC), which has a proton conducting polymer membrane as electrolyte. While the fuel used most fuel cells is hydrogen (e.g. in the polymer electrolyte membrane fuel cell: PEMFC), the DMFC uses methanol as fuel. The advantage of methanol is that the existing distribution infrastructure could be used for fuel supply, unlike for hydrogen (M.A. Smit et al ,2003)

The possible applications of fuel cells range from stationary power production in megawatt dimensions down to portable systems to supply mobile computers with a few watts. In between these two extremes lies their application as vehicle power sources, with almost all major car manufacturers now having their own research programs. Recently, after many years of research and development on fuel cells, the initial euphoria has somewhat vanished, as many problems are yet unsolved. Especially for mobile applications, most material components in fuel cell systems are still too expensive, the systems are more complex than initially anticipated, sometimes difficult to control and still the discussion is far from an end which will be the best fuel for them (Hˆhlein, B et al; Thomas, C et al ,2000).

While hydrogen is the best fuel in terms of operating the fuel cell itself, its production, storage and distribution is complicated. Alternatively liquid fuels are discussed,

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like conventional gasoline or methanol. These are easy to store and to distribute, but their conversion in a fuel cell system is difficult. Either one produces hydrogen from them on-board the vehicle to feed a standard Polymer Electrolyte Fuel Cell (PEMFC), or one uses a fuel cell which can convert a liquid fuel directly, like the Direct Methanol

History of the Development of Fuel Cells

- William Grove invented the fuel cell in 1839.
- General Electric invented proton exchange membrane fuel cells in the 1950s
- Francis Bacon demonstrated a 5kW alkaline fuel cell in 1959.
- NASA's use of fuel cells during the Apollo space missions in the 1960s was the first commercial use of fuel cells.
- Alkaline fuel cells have flown over 100 missions and operated for more than 80,000 hours in spacecrafts operated by NASA.
- The US Navy has been using fuel cells in submarines since the 1980s
- Fuel cell buses are running in several cities around the world, the largest being the European Union backed CUTE project (Clean Urban Transport for Europe).
- All major automakers have prototypes of alternative fuel vehicles using fuel cells on the road-some have already been leased to customers.
- Iceland has plans to convert its fishing fleet from diesel engines to hydrogen fuel cells as part of a national project to create a fossil fuel free economy
- Several car manufacturers are hoping to produce their first semi-commercial models of fuel cell cars by 2005, yet they will most probably not be mass produced until 2010.
- Numerous fuel cell products will be coming to market-portable direct methanol fuel cells will power mobile phones, laptops and cameras in the near future
- A fuel cell is around 60% efficient at converting fuel to power, double the efficiency of an internal combustion gas engine-which makes it perfect for alternative fuel vehicles.

Advantages

- Fuel cells reduce pollution that is caused by the burning of fossil fuels-their only by-product is water
- If the hydrogen used in the fuel cell comes from the electrolysis of water, then using fuel cells will eliminate greenhouse gases
- Because fuel cells don't need conventional fuels like oil or gas, they eliminate economic dependence on politically unstable countries
- Since hydrogen can be manufactured anywhere there is water and electricity, production of potential fuel can be allocated in various areas
- Fuel cells operate at a higher efficiency than diesel or gas engines which makes them an ideal source of efficient power for alternative fuel vehicles
- Most fuel cells operate silently, while internal combustion engines do not
- Fuel cells can operate for longer times than batteries, therefore to double the operating time, only the fuel needs to be doubled and not the capacity of the unit itself
- The maintenance of fuel cells is relatively straightforward since there are few moving parts in the system

Disadvantages

- Energizing fuel cells continues to be a major problem while production, transportation, distribution and storage of hydrogen remains difficult
- Reforming hydrocarbons via a reformer to produce hydrogen is technically challenging and not actually environmentally friendly
- The refueling and the starting time of fuel cell vehicles are longer, while the driving range is shorter than in a conventional vehicle
- Fuel cells are normally somewhat larger than comparable batteries or engines, however, the size of the units continues to decrease with research and testing
- Fuel cells are currently expensive to produce, since most units are hand-made and some use expensive materials
- The technology is not yet fully developed, therefore few products are readily available

Although hydrogen fuel cells appear to be the most promising source of alternative fuel, other sources are being researched and tested. Alternative transportation fuels provide economic advantages while also offering significant environmental benefits. They offer air quality advantages through reduced emissions and some fuels produce less greenhouse gas emissions than gasoline. There's significant research being conducted worldwide. Canada, for example, is recognized as a world leader in the development and use of alternative transportation fuels with more than 170,000 alternative fuel vehicles in use across Canada. (avtorentacar.com/en/2008-10-09/)

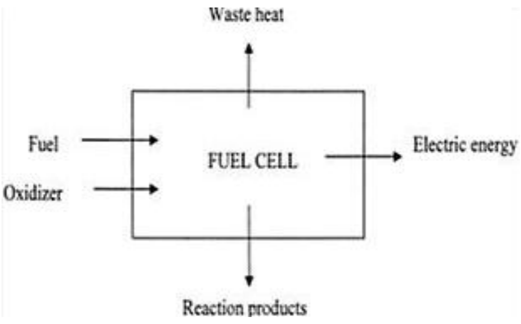


Fig. 1 General block diagram of fuel cell

Table 1 Characteristics of the Main Types of Fuel Cells

Type of fuel cell	**Operating Temperature, oC**	**Efficiency, %**	**Output, kW**	**Electrolyte**	**Ion**	**Fuel**
AFC	60-120	60–70	0.3–5	35-50% KOH	OH^-	H_2
MCFC	620-660	60–80	0.10	Molten Carbonate Melts (Li2CO3/Na2CO3)	$CO3^{2-}$	Hydrocarbons, CO
PAFC	160-220	40–80	50–200	Con. H_3PO_4	H^+	H_2
PEMFC	50-80	40–50	50–200	Polymer Membrane	H^+	H_2, CH_3OH
SOFC	800-1000	50–60	50–100	ZrO_2	O^{2-}	Hydrocarbons, CO

Different types of Fuel Cells

Various types of the fuel cells have been developed to generate power according to the applications and load requirements. Several types of the electrolyte play a key role in types of fuel cells. It must permit only the appropriate ions to pass between the anode and cathode.

Fuel Cells can be classified based on their temperature of operation: high, medium and low temperature fuel cells or based on the type of electrolyte used. Primarily, the latter method of classification is used for easier understanding and practical reasons and the six common types of fuel cells are Proton Exchange Membrane Fuel Cells, (PEMFC), Direct Methanol Fuel Cell (DMFC), Alkaline Fuel Cell (AFC), Phosphoric Acid Fuel Cells (PAFC), Molten Carbonate Fuel Cells (MCFC) and Solid Oxide Fuel Cells (SOFC). The classification determines the chemical reactions that take place, type of catalysts required, operating temperature and fuel used. These factors in turn determine the most suitable applications for each type of fuel cell. The essential characteristics of the major types of fuel cells and their applications are given in following table 1.

The waste heat from the fuel cell, depending on the temperature of operation can be utilized for water and space heating and owing to their cogeneration applications; the overall efficiency of the fuel cell system is increased (Y. Matsumoto et al,1994, M. Echigo et al,2003).

Table 2 Thermodynamic Enthalpy, Free Energy and Efficiency of some Fuel Cell Reactions

Reaction	$-\Delta H^o$	$-\Delta G^o$	$-\Delta H^o/-\Delta G^o$
	KJ/KMol	KJ/KMol	KJ/Kmol Efficiency
$H_2+1/2O_2=H_2O$	286	237.3	83
$CH_4+2O_2= CO_2+ H_2O$	890.8	818.4	91.9
$CH_3OH+3/2O_2= CO_2+ H_2O$	726.6	702.5	96.7
$CO+1/2O_2=CO_2$	283.1	257.2	90.9

Proton exchange membrane fuel cells (PEMFCs) work with a polymer electrolyte in the form of a thin, permeable sheet. The PEMFCs, otherwise known as polymer electrolyte fuel cells (PEFC), are of particular importance for the use in mobile and small/medium-sized stationary applications (Pehnt, M.et al,2001)

The PEM type fuel cells are considered to be the most promising fuel cell for power generation (11). Efficiency of PEMFCs are about 40 to 50%, and operating temperature is about 255 K. The PEMFCs and direct methanol fuel cells (DMFCs) are considered to be promising power sources, especially for transportation applications. The PEMFCs with potentially much higher efficiencies and almost zero emissions offer an attractive alternative to the internal combustion engines for automotive applications. This fuel cell has many important attributes such as high efficiency, clean, quiet, low temperature operation, capable of quick start-up, no liquid electrolyte and simple cell design (Hu, G et al, 2003)

Direct Methanol Fuel Cell ((DMFC)

Direct-methanol fuel cells or DMFCs are a subcategory of proton-exchange fuel cells where the methanol (CH_3OH) fuel is not reformed as in the indirect methanol fuel cell, but fed directly to the fuel cell operating at a temperature of 90 – 120 °C . Because the methanol and water is fed directly into the fuel cell, steam reforming is not required. Storage of methanol is much easier than for hydrogen as it does not need high pressures or low temperatures, because methanol is a liquid from -97.0 °C to 64.7 °C (-142.6 °F to 148.5 °F). The energy density of methanol (the amount of energy contained in a given volume) is an order of magnitude greater than even highly compressed hydrogen. The waste products with these types of fuel cells are carbon dioxide and water

The complexity of a system combining a hydrogen production unit and a hydrogen-consuming fuel cell have led many people to the conclusion, that the DMFC is the most favorable option for certain mobile and portable applications (Höhlein, B et al,2000).

The overall reaction of the DMFC is shown below and the basic operating principle is shown in fig.2

Anode:

$$CH_3OH(l) + H_2O(l) \xrightarrow{Pt/Ru} CO_2(g) + 6\ H^+ + 6\ e^-$$

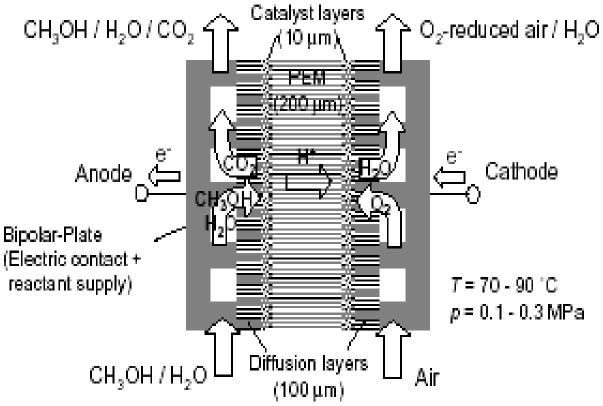
Cathode:

$$3/2\ O_2(g) + 6\ H^+ + 6\ e^- \xrightarrow{Pt} 3\ H_2O(l)$$

Overall:

$$CH_3OH(l) + 3/2\ O_2(g) \longrightarrow CO_2(g) + 2\ H_2O(l)$$

The DMFC relies upon the oxidation of methanol on a catalyst layer to form carbon dioxide. Water is consumed at the anode and is produced at the cathode. Positive ions (H^+) are transported across the proton exchange membrane (often made from Nafion) to the cathode where they react with oxygen to produce water. Electrons are transported through an external circuit from anode to cathode, providing power to connected devices.



At the anode, methanol and water are converted to carbon dioxide, protons and electrons. The protons are transported to the cathode side through a polymer electrolyte membrane (PEM). The electrons are transported through the external circuit, where they can be used to perform work. At the cathode, the protons and electrons reduce oxygen (from air) to form water. (Thorsten Schultz et al, 2001)

Methanol Crossover

The proton conducting membrane generally used in the PEMFC and DMFC is the Nafion (Dupont) membrane, a perfluorosulfonic acid. It has excellent chemical, mechanical and thermal stability and high protonic conductivity in its hydrated state. While it has shown very good performance in the PEMFC, Nafionperfluorosulfonic acid polymers are the most commonly used fuel cell membranes. Although it would be desirable methanol could be spontaneously oxidized at the cathode, however, a methanol transport across the membrane has been observed. It causes depolarization losses at the cathode and conversion losses in terms of lost fuel. In order to improve the performance of the DMFC, it is necessary to eliminate or, at least, to reduce the loss of fuel across the cell, usually termed ‘‘methanol crossover’’. In this sense, the membrane technology is one of the alternatives for trying to solve this problem. (A. Heinzel et al,1990).

In the DMFC there exists the problem of cross-over of methanol from the anode to the cathode side, leading to secondary reactions, mixed potentials, decreasing energy and power densities, and hence a reduced performance. This cross-over is caused by permeation of methanol due to a concentration gradient, indirectly dependent on the operation current, and by molecular transport due to electro-osmotic drag, directly related to proton migration through the membrane which increases with increasing current density (R. Jiang et al ,2002 K. Sundmacher et al,2001)

Direct methanol fuel cells (DMFCs) are promising candidates for applications in portable power sources, electric vehicles and transport applications because they do not require any fuel processor and can be operated at room temperature (Hampson NA et al ,1979, Heinzel A et al,1999, Ravikumar MK et al,1996)

However, the DMFC is hindered by methanol crossover through the electrolyte membrane as a result of diffusion and electro-osmotic drag. This results in a mixed cathode potential, as well as a reduction in power output and fuel utilization. Therefore, the suppression of methanol crossover has been a major research focus, and various methods for reducing methanol crossover have been examined including the development of a new electrolyte (Wainright JS et al,1995), the surface modification of a Nafion membrane (Pu C et al,1995) and the incorporation of Pt and hygroscopic oxides into the Nafion membrane (Gummaraju RV et al,1996).

Methanol crossover arises from electro-osmotic drag and an ion clustering with the membrane. Methanol diffusion or crossover from the anode to the cathode lowers fuel utilization, increases cathode polarization and causes excess thermal load in the cell and consequently lowers the cell performance (M.W. Verbrugge et al,1989).

At the cathode, the reduction of oxygen to water takes place on (usually supported) platinum catalysts. This reaction has been broadly examined in the last twenty years accompanying the development of hydrogen-consuming low-temperature fuel cells (PEMFC), (Eikerling, M. et al,1989;, Broka, Ket al,1987). The reaction is much slower than hydrogen oxidation, therefore it plays a major role in optimizing the performance of these cells. In the DMFC, though, the anode reaction is even slower, so the cathodic oxygen reduction cannot be assumed to be the rate determining step under most operating conditions. In the DMFC, a second reaction also takes place at the cathode platinum catalyst: The direct oxidation of methanol permeating through the PEM. This undesired side reaction leads to a mixed potential formation at the cathode, which results in a severely reduced electrode potential, and therefore also a severely reduced overall cell voltage. Thorsten Schultz et al (14.) Compared the open circuit cell voltage of a DMFC with standard NAFION to the thermodynamic cell voltage according to the Nernst equation. The dramatic voltage difference between thermodynamic and experimental data is to a large extent due to the oxidation of crossover-methanol.

Possible Solutions

Two different pathways exist to solve this problem of methanol cross-over, the first being the development of ion-conductive membranes based on alternative polymers or polymer composites, the second being the modification of the existing Nafion membrane, in order to prevent cross-over.

Therefore, to establish a better performance, a significant reduction in the methanol permeation through the PEM is necessary. This can either be achieved by PEM materials less permeable for methanol, byoptimized (dynamic) methanol feeding strategies, by simply using low methanol feed concentrations (at the moment values around 1 mol/dm^3 show best performance (Broka, K et al ,2000) or by realizing high methanol conversion at the anode (i.e. high fuel utilization.). Another important aspect is a possible water flooding of the cathode pore structure due to the water transport through the membrane and the water production at the cathode. Basically, this aspect plays a similar role like for the PEMFC, and can therefore be treated likewise, (Baschuk, J.J et al,2000)

An alternative to new membrane technology, in order to minimize the effect of the methanol crossover, is to improve the activity of methanol electrooxidation catalysts.

The studies about the influence of the different operating parameters on the methanol crossover show that high temperatures and high cathode pressures make methanol crossover decrease and improve the cell performance. In relation to the influence of the concentration of methanol, in spite of increasing the methanol concentration the crossover increases, it is necessary to take into account the polarization phenomena at the lowest concentration and so, to find the optimal concentration under the operating conditions of the fuel cell. The experimental results found in the literature show that methanol crossover decreases when the thickness and the equivalent weight of the membrane increase. However, due to the increase of the membrane resistance with the increasing of these parameters, the combined effects of ionic conductivity and fuel crossover determine the performance of fuel cell with membranes or various thickness and equivalent weights (A. Heinzel et al,1990).

Lots of researchers made efforts to reduce methanol crossover by modifying the Nafion membranes via hybridizing Nafion with inorganic nano-particles, such as silicone oxide, tetraethoxysilane, diphenyl silicate, zirconium phosphate (ZrP), and phosphotungstic acid, etc. Methanol might crossover the Nafion membranes either via diffusion or via electro-osmosis through the ionic clusters of Nafion membranes. Mixing inorganic nano-particles into Nafion membranes and leading nanoinorganic particles to locate inside the ionic clusters of Nafion membranes could reduce methanol crossover the membranes (29 D.H. Jung et al ,1998, Z.X. Linag et al, 2006; V.S. Silva et al,2006).

Sharp and the Massachusetts Institute of Technology, detailed technology advances in making liquid fuel methanol a source for fuel cells. Sharp claimed to have achieved the highest density ever with its prototype direct methanol to fuel cell (DMFC). The fuel cell has a longer continuous-use life span than a same-size lithium ion battery. Also MIT announced that researchers have managed to improve the power output of a methanol fuel cell by 50 percent. They developed a new technique for creating the membrane material that sits between the anode and cathode ends of a fuel cell. The material is less expensive than Nafion, which is typically used, and will not absorb as much methanol, making it more efficient. (news.cnet.com/)

Conclusion

The fuel cell is a very promising power generation system with several possible applications. The increasing research activity of the past ten years has resulted in a considerably improved cell performance. Nonetheless the most important problems (methanol crossover, anode kinetics, carbon dioxide evolution) are as yet unsolved. Possible solutions are being investigated though these are still far from final application; their stable long-term operation has yet to be proved.Current efforts in PEMFC research are focused on (1) reducing membrane cost via the use of non-fluorinated polymer electrolytes and (2) reducing system complexity via the development of ‘water-free’ electrolytes.

Methanol crossover from the anode to the cathode appears to be a major limitation at present for DMFCs to become a commercially viable alternative. Although Nafion membranes are the most usually used as solid polymer electrolyte in DMFCs, the nvestigations found in literature shows that methanol readily transports across perfluorosulfonic acid membranes and, in order to minimise the effects of methanol crossover, alternative membrane materials have been sought.

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