Progress in the Proton Exchange Membrane Development and Application of Fuel Cells

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Abstract:- *Since the development of chemically stable per-fluorinated membrane (Nafion) by Dupont, membranes are extensively used in fuel cells. But technical limitation e.g. thermal stability, proton conductivity and membrane dehydration at higher temperature and higher cost of polymeric membrane are the major hindrance in the commercialization of this technology. In last 10 years, emphasis was to find membranes for use in fuel cell with high proton conductivity and chemical stability in aggressive fuel cell environment. The intention of this paper is to provide an overview of progress made in the development of proton-conducting membranes for use in fuel cell.*

Keywords: Fuel cell, Proton conductivity, Thermal stability.

1. INTRODUCTION

Environmental problems and energy security are the biggest challenges of twenty first century. Enormous efforts are being made to overcome the demand of energy. Automobiles are the major candidates who contribute to environmental problems and are the bulk consumers of the fossil fuel available. This scenario leads to intensive research to solve these two problems. In this age of ecofriendly energy systems, PEM fuel cells may be the building blocks, since these can be used as a replacement of fossil fuel in transport applications (buses, trucks, cars) as well as in stationary power applications(energy supply for public building), and in mobile applications(computers, cell phones). Due to their enormous applications fuel cell research and development efforts have increased extensively in last decade. But the cost and technical limitations are the main hurdle in the commercialization of this technology. In this paper we would summarize the developments in the area of membrane systems.

2. DEFINITION OF PROTON EXCHANGE MEMBRANE

A thin sheet or film which may be used to separate ions by allowing the preferential transport of either cations (Protons) (in the case of a Cation- Exchange Membrane) or anions (in the case of an Anion-Exchange Membrane) is characterized as ion exchange membrane. If the membrane material is made from only ion-exchanging material, it is called a Homogeneous Ion-Exchange Membrane. If the ion- exchange material is embedded in an inert binder, it is called a Heterogeneous Ion-Exchange Membrane. Ion exchange membranes are classified based upon the type of charged (positive or negative) group attached to the membrane backbone, therefore cation exchange membranes contains negatively charged groups, such as $-SO_3-$, $-COO-$, $-PO_3^2-$, $-PO_3H-$, $-C_6H_4O-$, etc., fixed to the membrane backbone whereas anion exchange membranes contains positively charged groups, such as $-NH_3+$, $-NRH_2+$, $-NR_2H+$, $-NR_3+$, $-PR_3+$, SR_2+ etc., fixed to membrane backbone. In 1970s, a chemically stableproton exchange membrane based on sulfonated polytetra-fluorethylene was first developed by Dupont as Nafion, leading to a large scale use of this membrane in the chlor-alkali production industry and energy storage or conversion system (fuel cell) simultaneously.

3. DEVELOPMENT IN MEMBRANE OF FUEL CELLS

As membrane is the heart of the different types of fuel cells, therefore extensive research is going on the different types of membranes used in fuel cells.

3.1. Alkaline fuel cells

While there remains some interest in AFCs for commercial applications, notably by ZeTek as a range extender for battery powered vehicles, this is still at a low level compared with that for SPFC systems for transport applications [1]. A circulating KOH electrolyte is preferred to an immobilized system for transport applications as in addition to providing the means for cooling the stack, it enables the electrolyte to be replaced in the event of substantial CO_2 absorption, without the need to

disassemble the stack. Modified anode and cathode catalysts have now been developed that supersede the high concentrations of Pt and Pt/Au used in the space Orbiter vehicle. However, there is no consensus on whether precious or base metals are the preferred system. The choice depends on the balance between the required performance and acceptable cost. The choices range from low loaded all Pt systems [2] through Ni anodes and silver cathodes to all base metals now being developed by ZETEC. With increasing interest in the use of hydrogen for fuel cell powered vehicles, interest in the AFC system may increase. On the other hand, it is reported that the new fuel cells for the space shuttle Orbitor vehicles will be SPFCs in preference to the established AFC[3].

3.2. Solid polymer fuel cells

By 1989 and the first Grove Fuel Cell Symposium, the early work by General Electric which established the SPFC system for use on the Gemini space missions[4] had been re-evaluated and developed by Ballard Power Sources [5] to become one of the most significant fuel cell developments of the 1980s. It is suggested that General Electric chose not to pursue commercial applications for the SPFC as, in its then existing form, it required high Pt metal loaded electrodes and was more sensitive to CO poisoning than the established PAFC system. In addition, it was potentially more expensive as a result of the cost of the proton conducting polymer electrolyte and the high Pt metal loaded electrodes. While the sensitivity to CO poisoning remains, but less so with anode catalysts such as Pt/Ru, rapid developments were achieved in improving the power density while at the same time reducing Pt metal electrode loadings. For example, Nafion had been identified as a suitable proton conducting membrane electrolyte. By substituting other sulphonated fluorocarbon polymer materials, a four- fold increase in current density could be achieved at the same operating cell voltage. Further, by optimising catalyst and electrode structure, Pt metal loadings were reduced from a high of 28 to 0.4 mg/cm2 and recently to 0.2 mg/cm2 while maintaining current densities in excess of 1 A/cm2 at 0.7 V[6]. The details of what, at the time, were unexpected but exciting developments are described in a paper given by Pratter at the first Grove Fuel Cell Symposium [7].

3.3. SPFC fuel cells for transport applications

These developments attracted the interests of the major car companies who were seeking ways to eliminate CO, HC and NOx emissions from vehicles, while at the same time reducing fuel consumption and the associated CO2 emissions [8, 9]. In 1994, California introduced the 'zero emissions mandate' which required that 2% of all new vehicles sold in 1998 should produce no emissions, rising to 5% by 2001 and 10% by 2003. These time scales have now been relaxed, largely as a result of the failure of battery powered vehicles to meet the required performance and range targets. However, the ZEV standards, together with the US Government's PNGV (Partnership for a New Generation of Vehicles) initiative to develop light duty vehicles with a fuel economy of 80 miles/gal by 2004 remain.

Together, they have provided the stimulus for major investments by the car companies in fuel cell technology for both cars and buses. Developments in the technology of SPFC systems and their application to vehicles have been rapid in the last 10 years. In 1993, Ballard demonstrated an SPFC powered bus. Following the announcement of the first fuel cell stack with a power density of 1 kW/l [23], a Ballard phase 2 bus was demonstrated powered entirely by a 200 kW unit. This bus had no reduction in passenger seating or on road performance compared with the standard bus. Today, XCELLSIS, the Daimler Chrysler/Ballard/Ford consortium has now developed and is demonstrating buses in Canada, America and Europe with the aim of making them commercially available by 2005. Progress in developing fuel cell powered light duty vehicles has been equally impressive [24]. But thereremain significant technical challenges to be addressed. These include the fuel such vehicles will use and the cost of the fuel cell system and drive train.

Pure hydrogen is the preferred fuel for SPFC fuel cells but it is debatable whether this is a realistic choice for widescale consumer use in the short to medium term [10]. Hydrogen storage on vehicles has been demonstrated and includes high pressure light weight cylinders, cryogenic liquid systems and solid state metal hydride stores. Although substantial progress [11]. has been made with these systems to a level that, for example, compressed hydrogen may be acceptable for use on buses, they are generally considered not to meet volume and weight criteria for light duty vehicles. Following reports on high levels of hydrogen adsorption in carbon nanotubes [12]. this, together with improvements to metal hydride systems are being actively sought by the research community [13]. A liquid-fuel-based system with on-board reforming would be the preferred choice as the existing production and distribution system could be used or easily adapted with minimal additional investment. However, with today's onboard reformer technology, methanol is the preferred choice and has been demonstrated by Daimler Chrysler in their prototype NECAR3 vehicle [14]. Although gasoline or other petroleum derived fuel is significantly more difficult to reform for on-board fuel cell applications, the challenge to solve the problems is being addressed with the aim of bringing fuel cell vehicles into widespread use prior to the dawn of the 'hydrogen economy' [15]. Steam reforming of natural gas or liquid hydrocarbon fuels is the established technology for producing hydrogen rich gas for stationary fuel cells.

Such systems are not suited to the requirements of vehicle applications where space, weight and in particular fast start up and response are essential. Auto thermal reforming, while intrinsically less efficient than steam reforming, does provide the basis for a mobile system. A successful development of auto thermal technology is the Hot Spot reactor demonstrated by Johnson Matthey [16, 17]. Starting from cold, with vaporized methanol, 100% output is achieved in 50 s; subsequent changes in through put result in instantaneous changes in output.

3.4. SPFC fuel cells for stationary applications

In the early 1990s, as interest in SPFC systems for transport applications gathered pace, it became apparent that the performance and cost objectives set by the car manufacturers would provide an attractive and competitive fuel cell system for stationary applications, albeit with a limited temperature heat output. A 250 Kw unit has been developed by Ballard Generation Systems operating on natural gas. Other versions are planned which will operate on propane, hydrogen or anaerobic digester gas. Demonstration units in America, Japan and Europe are now in place with some having achieved over 1 year of operation. Cell voltage decay rates are reported to be <0.3% per 1000 h [18]. The applications for 250 kW units in distributed power systems are well established. An exciting and potentially new development with major implications for reducing energy consumption in the housing sector is the adaption of the SPFC units for mobile applications to smaller systems in the 2–10 kW range. Units in this capacity range were considered to be impractical until the advent of the SPFC fuel cell. Up to 100 units have now been built and demonstrated by companies such as Plug Power, H Power and Sanyo [19, 20].

3.5. Further developments in SPFC technology

SPFC fuel cell developments and technology have been particularly rapid during the 1990s. However, there is scope for further improvements notably concerning the proton conducting membranes presently available. In addition to the cost, existing materials such as Nafion need to be hydrated to be effective proton conductors. This limits their operating temperature to 90°C and adds further complications to the design of the fuel cell stack and system. For these reasons, a material with an operating range up to 200°C would be beneficial. In addition, the higher temperature operation would result in an increase in the CO tolerance of the anode catalysts and extend the range of applications where high grade heat is a requirement. Proton conducting membranes with a 200°C operating capability and no hydration requirement have recently beenannounced by Celanese [21]. An extensive review of SPFC technology and its applications has been published by Hoogers [22].

3.6. Direct methanol fuel cells

Fuel cell technology, particularly for transport applications, would take a leap forward if a viable system were to be developed that could use a liquid fuel without the need for reformation. The prospects for anode catalysts being developed having the activity to operate on petroleum derived hydrocarbon fuels are poor. However, Shell and others in the 1960s established that Methanol, with anode catalysts such as Pt/Ru, had some potential. The early work utilized sulphuric acid as the electrolyte. With the introduction of proton conducting membranes, interest in DMFC systems in the 1990s has been renewed with projects in America, Japan and Europe. Of particular significance has been the work of Los Alamos National Laboratory. If the power density required for vehicle applications are to be achieved, further improvements to anode catalyst performance are necessary. In addition, existing membrane materials are subject to what is known as 'methanol crossover', which in turn contributes to poor cell performance. In this context, it is interesting to speculate on how high temperature membranes such as that developed by Celanese would perform in a DMFC fuel cell [23].

In addition to transport applications for DMFC fuel cells, there are also defense requirements and with appropriate technology, consumer applications such as laptop computers, video cameras and mobile phones. In other words, any small scale applications dependent upon rechargeable batteries.

3.7. Phosphoric acid fuel cells

Following the successful development of AFCs for space exploration, attention turned to commercial applications, initially in American in 1967 with the TARGET programme for small scale units, followed in 1971 with the FCG1 programme for large scale multi- MW units. The phosphoric acid system using Pt metal containing electrodes was chosen as the most viable technology at the time for use with hydrocarbon fuels such as natural gas. Similarly, established steam reforming technology was adapted to provide hydrogen rich gas for the fuel cell. The decision, by Pratt and Whitney to use phosphoric acid as the electrolyte and adapt steam reforming technology to produce hydrogen were ground breaking at the time. For these developments, Mr. William Podolni, head of fuel cell development at Pratt and Whitney, later to become United Technologies, was awarded the Grove Medal in 1995. Much of the technology relating to SPFC systems originates from the development of the PAFC units and their demonstration in a variety of on-site locations. Notable in this, is the use

of highly dispersed Pt metal catalysts, Teflon bonded electrode structures, graphite bipolar plates and last but by no means least, reformer technology and the associated balance of plant. PAFC systems such as the ONSI Corporation PC25 200 kW CHP unit are still the only units that are commercially available, albeit at a price that is still not competitive with established CHP systems. They have established the viability and reliability for on-site electricity and heat generation with the added feature, where appropriate, of absorption chilling for air conditioning applications. There are now some 65 MW of PAFC systems worldwide.

Most of the plants are in the 50–200 kW capacity range but large plants of 1 and 5 MW have been built including an 11 MW plant for Tokyo Electric Power. Details on performance, reliability and cost reduction initiatives are contained in several reviews [24-26]. Despite extensive fuel cell and systems development and wide spread demonstration in a wide range of potential applications, the PAFC system has not yet achieved the commercial goals to fulfil projections of 2000 MW of installed capacity by the year 2000 [27].

3.8. Molten carbonate and solid oxide fuel cells

These high temperature fuel cells operating at 650 and 1000°C, respectively, were developed largely with the intention of overcoming the limitations of the low temperature SPFC and PAFC systems. Their two main advantages are that their performance is not affected by carbon monoxide and the residual heat, which is available at temperatures in excess of 600°C, makes them applicable to industrial as well as commercial uses.

Although neither system with existing anode catalyst technology is capable of truly operating in a direct fuel mode and hence hydrocarbon reforming is necessary, the stack operating temperatures are such that internal reforming is possible. In fact, the systems which today have been developed and demonstrated all employ internal reforming either directly within the cell [28] or in a separate reformer contained within the stack module [29]. This basically simplifies the system with benefits in terms of cost and power efficiency.

Despite the benefits of these high temperature systems, their development and demonstration is still at a comparatively early stage compared with that of PAFC.

3.9. Molten carbonate fuel cells

The largest demonstration of a MCFC system was a proof of concept natural gas fuelled 2 MW unit which was operated from 1996 to 1997 at Santa Clara, CA. The plant was built by Energy Research Corporation (now Fuel Cell Energy Inc.) and incorporated internal reforming in what is known as the Direct Carbonate Fuel Cell TM (DFC). The plant achieved a maximum output of 1.93 MW AC at an electrical efficiency of 43.6% and operated grid connected for 4100 h. One of the problems during the extended run was attributed for the decomposition of thermal insulation materials which caused deposition of carbon and short circuits in electrical components including the stacks [30]. Based upon the technology and experience gained from the 2 MW demonstrations, 250 kW units have been designed and demonstrated by FCE and their partner, MTU, in Germany. Several MCFC developers are also active in Japan and Europe including BCN in The Netherlands, Ansaldo (Italy), Hitachi, IHI, Mitsubishi Electric and Toshiba in Japan. Interestingly, recalling early attempts to use coal in fuel cells, a 250 kW DFC unit will be installed at the Harrison Mining Corporation coal mine in Cadiz, OH, USA to demonstrate the feasibility of fuelling an MCFC with coal mine methane emissions. The recent progress in demonstrating MCFC systems is being supported by an extension to the existing FCE production plant to increase its annual production capacity from 50 MW in 2001 to 400 MW in 2004.

3.10. Solid oxide fuel cells

Last but by no means is least of the six fuel cell systems that make up today's range the SOFC. Less developed than the rest of the fuel cell range, but possibly having more and wider potential. Until recently, the SOFC was seen as only having application in large scale multi-MW stationary plants. Now, SOFC systems have been developed and demonstrated by Sulzer Hexis Ltd. at the 1 kW level for residential CHP applications. These are primarily intended to be fuelled with natural gas but have been demonstrated using low sulphur home heating oil [31]. The maximum electrical efficiency of these demonstration units that has been achieved so far is 36% (LHV) [23]. Although an SOFC system operating at 1000°C is not an initial first choice for transport applications, in fact, small scale systems are now being developed in Europe and America for use as auxiliary power units (APU) in cars. A duel fuel hydrogen/gasoline vehicle with a SOFC auxiliary power unit is being demonstrated by BMW [33]. The unit provides electric power for on road use as well as for accessories, heating and cooling when the car is stationary. Fuel cell APUs with larger capacities is also being developed to replace engine driven generators and refrigeration units on trucks. The first demonstration of a SOFC system for cogeneration was carried out at Westervoot in The Netherlands. The 100 kW unit built by Siemens Westinghouse and operated by Elsam and EDB (a consortium of Dutch utility companies) began operation in 1998. At the end of the demonstration project in

December 2000, the plant had accumulated a total of 16,612 h. The fuel cell stack used the Siemens Westinghouse tubular cell system and was shown to be reliable under plant operating conditions which included several thermal cycles and sulphur contamination. The plant had an electrical efficiency of 46% (net) and a total energy utilization of 75% [34].

3.11. Hybrid SOFC–gas turbine (GT) developments

The electrical efficiency of fuel cell plants are often compared with that of combined cycle gas turbine units (CCGT) with electrical efficiencies of up to 50%. While high temperature fuel cells such as the MCFC and SOFC, in the early stages of their development, are capable of a similar performance, the question is frequently raised as to what benefits fuel cells offer over and above the established CCGT systems. It is now widely believed that a hybrid SOFC–GT system could achieve electrical efficiencies of up to 70% in a system where the GT combustor is replaced with a SOFC fuel cell. A proof of concept 220 kW demonstration has been carried out at the National Fuel Cell Research Centre, Irvine, USA [35].

Further demonstrations of SOFC–GT hybrids are planned in America and Europe including a 1 kW unit. In a further development of the SOFC hybrid concept, a study has been made of combining SOFC and SPFC fuel cells in a single system. In this system, the SOFC is used to produce electricity and carry out reforming simultaneously. The exhaust stream from the SOFC is passed through shift converters and is supplied to a low temperature SPFC unit. It is predicted that the overall efficiency of the hybrid system will be significantly higher than the separate fuel cell units with additional benefits to capital and running costs [36].

3.12. Future technical developments

Although SOFCs have the potential for a wider range of applications than any of the other fuel cell systems, there remain significant materials as well as cell and stack assembly challenges to be met before confidence in the technology is justified. Experience from more advanced systems such as PAFC and MCFC, has shown that extensive demonstrations in real situations are required before operating deficiencies become apparent. This is particularly true with balance of plant components and materials. As far as materials are concerned, there remain three principle areas of research and development. They are:

- The manner in which cells are constructed and stacks assembled;

- The development of interconnect or bipolar plate materials for 1000°C operation;

- The development of electrolyte and electrode materials compatible with a reduced operating temperature of 600–800°C.

Three types of cell/stack assembly have been identified, namely, the tubular, planar and monolithic [37]. The tubular system, developed by Siemens Westinghouse, is to date the most advanced and demonstrated system [38] but it remains to be seen how successful further scale up and cost reduction programmes prove to be. Others, notably Sulzer Hexis Ltd., Global Thermal electric Inc., Rolls Royce Plc. and Ceramic Fuel Cell Ltd., are actively developing innovative versions of the planar system for use in single or hybrid systems [39]. Although the monolithic cell structure offers significant advantages, little progress has been reported, possibly as a result of difficult materials and application technology. An area that has received considerable research interest in the last 10 years is the development of what is generally described as the 'low temperature' SOFC operating in the range of 600–800°C. At these temperatures, cheaper materials may be used for stack components as well as heat exchangers used in the balance of plant. Success depends upon using thinner electrolyte layers of established materials such as yttria-stabilised zirconia or mixed oxides such as ceria, gadolinia. This and other materials including electrodes with low temperature activity have been presented at several Grove Fuel Cell Symposia [40].

4. Conclusions

Since the first Grove Fuel Cell Symposium was held in September 1989, progress in developing fuel cell technology for both stationary and mobile applications has been steady if not spectacular. The most significant has been the rapid development of the SPFC system for transport applications, together with the realisation that success in this field would provide the basis of a low temperature stationary unit for combined heat and power applications (CHP).

Following the successful demonstration of fuel cell powered buses and light duty vehicles by all of the major vehicle manufacturers, attention has now turned to choice of fuel and fuel infrastructure. Technical solutions to two challenging areas will, I predict, go a long way to resolving the question. First, the development of a hydrogen storage system. It should provide a vehicle range of at least 300 miles with no significant increase in volume or weight compared with that of the equivalent gasoline tank.

Second, a reformer and gas clean up system for use with gasoline with a start up and response time giving a fuel cell powered vehicle similar performance to that of the conventional ICE vehicle. In the 1980s, projections for stationary fuel cells anticipated there being at least 2000 MW of capacity in use by 2000. Not only have these projections been over optimistic, but also opinions on the size of the units have changed. For example, much of the market was seen to require large stationary multi- MW units. The smallest size that was thought to be commercially viable was 200 kW. While this may still be the case in some circumstances, both low-temperature and high-temperature micro-CHP fuel cell units are seen to have wide scale application in domestic applications.

Why these changes of direction? What are the driving forces? In part, interest in micro CHP fuel cell units results from technical developments in stack and reformer systems.

However, deregulation of the energy utilities and environmental issues, including Kyoto climate change aims, have stimulated a rethink of the relative benefits of large base load power stations versus distributed power, including heat, power and cooling for individual buildings.

In all, the drive to develop the zero emission vehicles with improved fuel economy, together with the contribution fuel cells have long been predicted to make to stationary applications should this time truly see a significant start to their commercialization in the recent few years.

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