



Review

Micro-fuel cells—Current development and applications

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Abstract

The importance of micro-fuel cell has been increased with the demand for uninterrupted power source in today’s power hungry portable electronics. Currently, there is aggressive research going on to commercialize the micro-fuel cell by many laboratories and companies. The three different fuels feeding systems, i.e. pure hydrogen, pure hydrocarbons (alcohol, i.e. methanol and ethanol; formic acid and ethylene glycol) and on-board hydrogen from reformed hydrocarbons like methanol or other compound like water can be used for operating the micro-fuel cells. The current status on the research and development of micro-fuel cell with all the above three types of fuels have been discussed. The different substrate materials used in micro-fuel cells for the suitability of the portable electronics have also been stated. The design aspects of micro-fuel cells and micro-reformers are discussed here. The current state of commercialization of micro-fuel cells for portable electronics has been reviewed based on the open literature. The hurdles to overcome in order to commercialize in full phase have been reported, whenever possible. Some very new technologies which can make the micro-fuel cell into a very promising system with a simple operation have also been focused.

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1. Introduction

The demand for power sources with superior performance has increased as a result of the rapid growth of the portable

electronics market. These power sources must yield adequate power output while at the same time maintaining criteria such as a very small volume and lightweight packaging. Micro-fuel cells are such distinctive, high energy density sources for next generation power hungry portable products like personal digital assistant (PDA), laptop, cellular phone and hybrid battery recharger-both in a separate way and integrated way with a maximum power of 1–50 W. The present manuscript is more

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Table 1
Energy density of various batteries and fuels [77]

Fuel	Energy density (Wh kg ⁻¹)	Comments
BB-2590	81	Secondary
BA-5590	150	Primary
BA-5390	235	Primary
BA-8180	345	Primary Zn–Air battery, large unit
Compressed hydrogen	500–1000	5000 psig, value includes container weight
Sodium borohydride	3600	[NaBH ₄ + 2H ₂ O] weight only
Methanol	5500	Based on lower heating value of fuel
Most liquid hydrocarbons	~12,400	Based on lower heating value of fuel
Hydrogen gas	33,200	Unpacked
Nuclear material	2,800,000	Raw power

focused on the type of micro-fuel cell having maximum power capability of <5 W. Batteries are a chemical process, but they do not last long enough. Fuel cells promise to provide more reliable, longer portable power than batteries. Since the energy is stored as a reservoir of fuel rather than as an integral part of the power source, fuel cells have advantages over batteries (Table 1). Micro-fuel cell products compete with power systems that utilize both direct and indirect energy conversion methods (Table 2). Direct conversion may involve fuels such as methanol, ethanol, formic acid, ethylene glycol and sodium borohydride that are converted into electrons through a direct fuel cell system. Indirect way is to first reform methanol, ethanol, sodium borohydride followed by feeding the reformat gas into fuel cell. Pure hydrogen feeding is relatively difficult for the application of electronic equipment due to storage problem (lack of efficient hydrogen storage material) in so small size. There is also concern in terms of safety while carrying hydrogen in wireless electronic equipment. The setup of a cost-effective, appropriate infrastructure for hydrogen canister is another major challenge for which most hurdles are unlikely to be overcome in the foreseeable future. In direct alcohol fuel cell system, the main studies center on methanol (direct methanol fuel cell, DMFC) though there is recent trend of studying with ethanol. DMFC has main drawback of high crossover rate and low rate of oxidation.

The micro-fuel cell market can be divided into three device categories namely, portable electronics, military portable and healthcare segments. In the portable electronics category, this represents handset, PDA, mobile phone, portable audio/video devices, digital imaging devices and in a larger version like personal computer (PC), notebook computer and digital segments.

Micro-fuel cell technologies face competition from advanced energy storages devices like secondary (chargeable) batteries such as lithium-ion batteries and nickel-based batteries. How-

Table 2
Types of μ -fuel cells in the literature

Based on fuels	Based on substrates
Hydrogen	Stainless steel
Reformed hydrogen	Aluminium
Methanol	PCB
Ethanol	Silicon wafer
Formic acid	LTCC
Ethylene glycol	PDMS

ever, micro-fuel cells, once commercialized, hold the promise for providing more back up ‘green power’ at lower cost than its competing battery technologies.

It is most unlikely that micro-fuel cells will compete with the primary battery. In order to compete with the primary battery, the cost component would not justify the use of micro-fuel cells. Theoretically competition to batteries from fuel cells impacts all sources of secondary batteries; however, practically it is only the high-cost, premium batteries (Li-ion and Li-polymer) that fuel cells really compete with. Those products which can realistically use Ni–Cd or Ni–MH are not likely to support the cost premium required for fuel cells, at least not initially. The added functions and features for more entertainment and sophisticated communications on modern handsets like third generation (3G) mobile phones, digital multimedia broadcasting (DMB) phones and a future portable device, iphone, to be launched by Apple suck up available power quickly. Therefore users are going to be faced with recharging their phones more often. In this case, micro-fuel cells have great potentiality to cope-up the situation. In addition, the price of 3G handsets is high enough to absorb the greater cost of fuel cells. Therefore micro-fuel cells will have to compete with the rechargeable battery not only in terms of cost but also the run time, life time and output power. Apart from mobile phones, another market opportunity of micro-fuel cell is in notebook computers. The potentially longer run time by micro-fuel cell may be competitive with the conventional battery for notebook computer. In addition to the individual portable consumer electronics products, the other potential market for micro-fuel cells is battery chargers. As the battery chargers are not integrated with the devices, these are much simpler to develop and market.

Secondly, there is great opportunity of micro-fuel cells in the military market where there is an aggressively requirement of continuously long time operation with light weight of portable batteries. Even micro-fuel cells can compete in this segment with high prices, if its requirements satisfy.

Thirdly, micro-bio-fuel cells can also be interesting product for implantable power sources (like Pacemaker, glucose sensor, prosthetic valve actuator power supply) and for the operation of robots [1,2]. The different types of bio-fuel cells and the current status on the development with the potential applications have also been nicely reviewed by Davis and Higson [1] and Bullen et al. [2].

2. Different types of micro-fuel cells in literature

2.1. Micro-reformed hydrogen fuel cell

Micro-reformed hydrogen fuel cell (μ -RHFC) refers to the small proton exchange membrane fuel cell (PEMFC) in addition to on-board hydrogen supplying system to the fuel cell either from hydrocarbon reforming or chemical hydride (like sodium borohydride) reforming. μ -RHFC can be divided into two categories based on the operating temperature of the reformer. Generally methanol and sodium borohydride are used as reformer feed at relatively high temperature and low temperature operation, respectively. There is growing consensus for RHFC with methanol feed that a combination of a micro-fuel cell system and a rechargeable battery will be the most applicable, in which the fuel cell continuously recharges the battery and the battery in turn provides electricity to the portable device. It should also be noted that incorporation of polybenzimidazole (PBI) based high temperature tolerating proton exchange membrane or other types of high temperature solid membrane [3] facilitates the design of the micro-fuel processor by direct feeding of reformed gas to the fuel cell without pretreatment. However there is a major disadvantage of this system. A fuel reformer constitutes a significant portion of the balance of plant (BOP) (which consists of a heating system, oxidant storage, pump, sensor and distribution components, control system, and dc–dc converter for power conditioning) for a fuel cell device, often constituting 30% or more of system cost and mass. Therefore it requires significant energy from the device, lowering total system efficiency [4]. It has also some time delay in the response at the starting point of hydrogen production.

To date, most micro-reformer and micro-PEMFCs were developed based on micro-electromechanical system (MEMS)

technology. Although this is an attractive approach given the maturity of silicon micro-fabrication, the processing techniques used to fabricate these micro-reactors are expensive, which eventually is directly reflected in the final cost of the fuel cell. Another problem emerging in micro-fuel cells based on Si wafer is that the Si substrate is quite fragile, making difficult to compress the fuel cell tightly for good seals and for lowering the contact resistance between membrane/electrode assembly (MEA) and Si-based bipolar plates. The other material such as low-temperature co-fired ceramic (LTCC) material is an alternative for the bipolar plate of micro-fuel cell systems. However, both silicon wafer and ceramic are nearly electrically insulated. Conductivity for current collection fully depends on the thickness of the conductive layer coated on these substrates, which will increase the cost significantly in order to minimize the resistance required in high power application. Printed circuit board technologies (PCB) offer an interesting platform to get the current collector on the body itself. It has also design flexibility, potentially higher power densities, ease of device integration, and improved packaging form factors. It is also possible to fabricate micro-fuel cell using thin stainless steel plate where there is also need of current collector layer.

Another type of substrate material for micro-reformer is polydimethylsiloxane (PDMS). PDMS has a number of material properties that make it attractive for micro-devices. PDMS micro-fuel cells can answer the need for compact and inexpensive fuel cells for developing fuel cell materials rapidly with little infrastructure.

There are many reported literatures on micro-reformers only. The exhaustive review on these reactors is given in Kundu et al. [5]. The two types of micro-reactors have been used namely micro-packed reactor and catalyst coated micro-channel reactor.

Pattekar and Kothare [6] presented a silicon based micro-packed reactor fabricated by deep reactive ion etch-

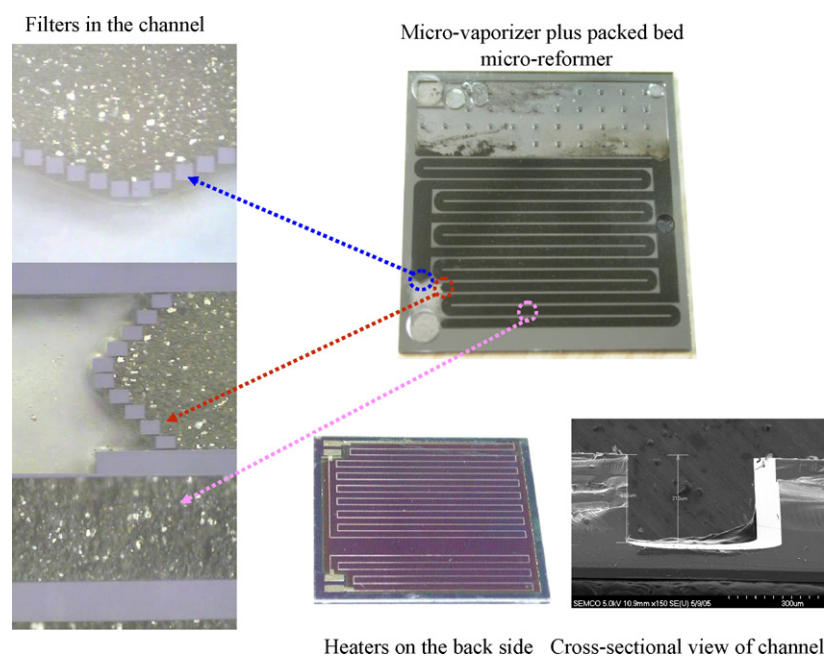


Fig. 1. Micro-reformer developed in Samsung Electro-mechanics [8].

ing (DRIE). Micro-channels were fabricated in 1000 μm thick silicon substrates using photolithography followed by DRIE. The catalyst used was commercial one (Sud Chemie, Cu/ZnO/Al₂O₃ catalyst). The temperature sensor is a platinum resistance–temperature device (RTD) with a linear temperature versus resistance characteristic. The heater is a platinum line meandering along the micro-channels (also on the backside of the substrate). They have obtained 88% methanol conversion at 1:1.5 molar ratio of methanol and water and the feed rate was 5 cm³ h⁻¹. Later, Pattekar and Kothare [7] developed a radial flow micro-packed-bed reactor which possesses less pressure drop compared to conventional one due to variable (increasing) cross section along the reaction path. More recently, Kundu et al. [8,9] presented the silicon based micro-channeled reactor with high performance of the catalyst with durability test (Fig. 1). The catalyst was Cu/ZnO/Al₂O₃ from Johnson Matthey. In their experiment, serpentine patterned micro-reformer showed superior activity than that of parallel patterned micro-reformer.

The catalyst coated micro-channel reactor and plate type reactor seems to be more attractive than micro-packed bed reactor due to low pressure drop and less channeling of gas, though it possess low catalyst loading which may affect on the performance and durability of the system. The methanol steam reforming reactions using catalyst coated micro-channeled stainless steel plate was carried out by Zapf et al. [10], Park et al. [11,12], Men et al. [13,14] and Kundu et al. [15,16]. The micro-channeled aluminium plate for the steam reforming of methanol was carried out by Pfeifer et al. [17,18]. De wild and Verhaak [19] and Fukuhara et al. [20] also used the wall coating catalyst on aluminium material. The catalyst coating on silicon-based micro-reformer was carried out by Kawamura et al. [21]. The volume of the micro-reformer is less than 1 cm³. The coated catalyst was their developed catalyst (Cu/ZnO/Al₂O₃). The catalyst showed 100% conversion of methanol at 250 °C. The key point for getting high performance and high durability is the uniform catalyst coating with large amount of catalyst loading.

Hydrogen production from sodium borohydride is advantageous with respect to operation at low temperature. The catalyst can be Pt, Ru or Pt/Ru on carbon support or other supports like LiCoO₂ and anion exchange resin. Though hydrogen from sodium borohydride in alkaline medium as a feed for micro-fuel cells seems to be easy and technologically feasible way, still there is a need to work on the deactivation of the catalyst, the treatment of the by-products, and the proper control of the reaction rate. The main drawback of the system is the production of boron oxide which is hazardous and also solid (which can affect the catalytic activity and clogging in the system may also occurred). Xia and Chan [22] tried to solve the latter problem by using low concentration of NaBH₄. They claimed the produced boron oxide could be dissolved in the resulting solution if 10 wt% NaBH₄ solution is taken. In this case, hydrogen generation can be controlled by inserting or removing the catalyst into/from the solution. Recently Prof. Gervasio of Arizona State University and his team claimed to use up to 15 wt% NaBH₄ solution with some external additive like ethylene glycol that dissolves boron oxide [23]. Other works on hydrogen production from sodium borohydride have been nicely reviewed by

Wee [24] and Wee et al. [25]. The reformed hydrogen fuel cell from sodium borohydride and direct borohydride fuel cell for the generation of 20 W power generations were also compared with respect to the rate of consumption of NaBH₄ and size and total cost of the fuel cell system by Wee [21]. The crossover problem makes the direct borohydride fuel cell inferior compared to that of RHFC from sodium borohydride. Richardson et al. [26] have demonstrated the flow type packed bed reactor for hydrogen production from sodium borohydride with the concentration of 20 and 25 wt%. They also claimed that the concentrations greater than 25 wt% only showed a greater propensity for clogging. The allowance of this higher concentration without clogging may be due to low reaction rate in the flow type reactor. Recently there is also an effort for hydrogen production from sodium borohydride by acid catalyzed reaction (e.g. with various edible acids such as ascorbic acid, malic acid, tartaric acid, etc.). Though the system is much simpler, hydrogen delivery may be problem. There is a need to look into this. Seiko Instruments [27] controlled the hydrogen flow rate in acid catalyzed sodium borohydride system by controlling pressure of the hydrogen reactor tank and acid solution tank.

2.2. Hydrogen fed micro-fuel cell

Hydrogen fed micro-fuel cell seems to be very simple and most effective with high efficiency, if the storage options become lightweight. There are efforts to get materials with high amount of hydrogen storage [28,29]. However, all the methods are not suitable for portable applications due to the low volumetric and gravimetric efficiency of hydrogen storage. Otherwise, there are many research activities on hydrogen fed micro-fuel cell (Table 3) hoping the availability of hydrogen storage materials in near future and also to help in the research of μ -reformed hydrogen fuel cell. It may also help to the micro-fuel cell system connected to the hydrogen producing system which produces pure hydrogen. One such system is hydrogen produced from metal (like Al) and water [30–32].

Lee et al. [33] developed a 2-cell and a 4-cell μ -fuel cell array with flip–flop interconnection with silicon as substrate material. The 2-cell array was supported on a borofloat glass with peak power density of 20 mW cm⁻², while the latter was on silicon substrate with peak power density of 42 mW cm⁻². Yu et al. [34] claimed that they have developed a miniature silicon wafer fuel cell with improved performance. Results showed that with the thickness of Cu/Au composite layers of 1.5/0.9 μm sputtered on the top of the silicon wafers as the current collectors, the peak power density has achieved 194.3 mW cm⁻² (at 450 mA cm⁻²) at room temperature. The superior performance of this μ -fuel cell was due to the use of oxygen instead of air and the high flow rates of hydrogen/oxygen flow (50 ml min⁻¹). The fuel utilization of this μ -fuel cell is around 6.8%. Another substrate material, PDMS was used commonly, though the current peak powder density is low with this material. Shah et al. [35] developed a hydrogen fed μ -fuel cell based on polymeric micromachining technology. PDMS was used as the cell support and housing. Its peak power density has achieved 35 mW cm⁻² when the cell was heated to 60 °C. Hsieh et al. [36] claimed to

Table 3
Experimental observations in the literature

Hydrogen fed micro-fuel cell	Authors	Substrate material and feed flow rate	Dimension of cell and MEA	Comments
	Lee et al. [33]	Glass and silicon; hydrogen and oxygen flow rate control by monitoring inlet pressure (35 kPa for two cell assembly and 100 kPa for four cell assembly)	Two cell assembly in wet etch glass: MEA area of each cell: 5 cm ² and split type channel with 150 μm deep. Four-cell assembly in dry-etched silicon: The flow chambers were etched 200 μm deep and the square distribution pillars were 100 μm × 100 μm in size, arranged in a uniform rectangular array 100 mm apart MEA area: 5 cm ²	Maximum power density: 20 mW cm ⁻² (glass). Maximum power density: 42 mW cm ⁻² (silicon wafer)
	Yu et al. [34]	Silicon wafer: H ₂ flow rate: 50 ml min ⁻¹ ; O ₂ flow rate: 50 ml min ⁻¹		Maximum power density: 107.3–194.3 mW cm ⁻²
	Yu et al. [78]	Silicon wafer: H ₂ flow rate: 40 ml min ⁻¹ ; O ₂ flow rate: 60 ml min ⁻¹	MEA area of each cell: 3 cm ² . Number of cells: 2. Pt loading in cathode and anode: 1 mg cm ⁻²	Maximum power density in twin cell: 190.4 mW cm ⁻²
	Shah et al. [35,79]	Silicon and PDMS; hydrogen flow rate: 1 sccm	MEA area: 1.4 cm × 1.2 cm; channel width of 5 μm and number of channels in each plate: 768 Size of micro-Fuel cell: <10 cm ³	Maximum power density: 0.35 mW cm ⁻²
	Hahn et al. [80]	Sandwiched metal–polymer foils; hydrogen flow: 0.5 sccm	Size of Planar micro-fuel cells 0.2 cm ³ ; MEA of each cell: 0.18 cm ² (total number of cells: 3); MEA of one cell: 0.54 cm ²	Maximum power density: 120 mW cm ⁻² ; it can be potentially applied in wireless sensor networks, chip cards or autonomous Microsystems; air-breathing
	Yamazaki [81]	Silicon wafer: hydrogen and oxygen	MEA area: 1 cm ²	Maximum power density: 37 mW cm ⁻² ; the porous silicon layer was there to support the MEA
	Hseih et al. [36]	Polymethyl methacrylate (PMMA); H ₂ flow rate: 10 cm ³ min ⁻¹ at 4 atm; air flow rate	Membrane: Nafion 117; MEA area: 5 cm ² ; flow field plate: parallel serpentine channel (width of 400 μm and depth of 200 μm); Pt loading in cathode and anode: 0.15 mg cm ⁻² ; size of micro-fuel cell: ~16 cm ³	Maximum power density: 31 mW cm ⁻²
	Hsieh et al. [82]	Cu and SU-8 photoresist H ₂ flow rate: 6, 7, 8, 9 and 10 sccm; air velocity: 25 cm s ⁻¹		Air breathing and forced air type; maximum power density: 22 mW cm ⁻² (air breathing) and 29 mW cm ⁻² (forced air)
	Chan et al. [83]	Polymethyl methacrylate (PMMA); hydrogen pressure: 10 psi; air flow rate: 50 ml min ⁻¹ ; oxygen flow rate: 20 ml min ⁻¹	MEA area: 3 cm ² ; flow field plate: spiral channel (width and depth of 220 μm); Pt loading in cathode and anode: 1 mg cm ⁻² ; size of micro-fuel cell: 3.5 cm ³	Maximum power density: 82 mW cm ⁻² (air in the cathode) maximum power density: 315 mW cm ⁻² (oxygen in the cathode)
Direct methanol fuel cell	Blum et al. [51]	Plastic	Membrane: Polyvinylidene Fluoride (PVDF), nano-powder SiO ₂ ; MEA area: 6 cm ² ; anode (Pt/Ru): 5–7 mg cm ⁻² ; cathode (Pt) 4–7 mg cm ⁻²	Air breathing type (1 M methanol); maximum power density: 12.5 mW cm ⁻²
	Yen et al. [47]	Silicon; liquid mixture flow rate (1 M methanol): 0.283 ml min ⁻¹ ; air flow rate: 88 ml min ⁻¹	Membrane: Nafion 112; MEA area: 1.625 cm ² ; flow field plate: serpentine channel (750 μm in width, 400 μm in depth, and 12.75 mm in length)	Maximum power density: 47.2 mW cm ⁻² (60 °C)

Table 3 (Continued)

Hydrogen fed micro-fuel cell	Authors	Substrate material and feed flow rate	Dimension of cell and MEA	Comments
	Lu et al. [48]	Silicon; liquid mixture flow rate (1–8 M methanol): $0.283 \text{ ml min}^{-1}$; air flow rate: 88 ml min^{-1}	Membrane: Nafion 112; MEA area: 1.625 cm^2 ; flow field plate: Serpentine channel ($750 \mu\text{m}$ in width, $400 \mu\text{m}$ in depth, and 12.75 mm in length). The loadings of Pt/Ru and Nafion in the anode catalyst layer were 4–6 and $1\text{--}1.5 \text{ mg cm}^{-2}$, respectively. The loading of Pt and Nafion in the cathode catalyst layer were 1.3 and 1 mg cm^{-2} , respectively	Maximum power density: 50 mW cm^{-2} (2 M methanol); uncompromising performance at room temperature for methanol feed concentration as high as 4 M, and only slightly degraded performance under a 8 M methanol solution
	Yao et al. [49]	Silicon	Size of micro-Fuel cell: 16.4 cm^3 . MEA area: 1 cm^2	Passive type fuel cell (driven by natural convection both at the anode and cathode; only water circulation pump from the cathode) maximum power density: 25 mW cm^{-2}
	Pan [84]	Stainless steel; liquid mixture flow rate (1–3 M methanol): 0.2 ml s^{-1}	MEA area: 5 cm^2 ; parallel style flow fields that have a 1 mm channel width and a 0.5 mm rib width	Air breathing; maximum power density: 65 mW cm^{-2} at 60C; development of new type MEA which lowers the methanol crossover
Direct formic acid fuel cell	Chu et al. [60]	Silicon wafer— $50 \mu\text{l}$ of 5 M formic acid + 0.5 M H_2SO_4 (batch)	Membrane: nano-porous silicon; MEA area: 0.0625 cm^2 ; catalyst: Pd/nafion solution(anode) and Pt/nafion solution (cathode)	Air breathing type; maximum power density: 30 mW cm^{-2}
	Yeom et al. [85]	Silicon wafer—anode: passive; cathode: passive/active	Size of MEA: $18 \text{ mm} \times 18 \text{ mm}$; membrane: Nafion-112; anode catalyst: Pt or Pd based catalyst; cathode catalyst: Pt	The maximum power density with Pd based anode catalyst using a 10 sccm stream of oxygen at the cathode: 30.7 mW cm^{-2}
Reformed hydrogen fuel cell				
Fuel: methanol	Yamazaki [81]	Silicon wafer: Size of reformer: $25 \text{ mm} \times 17 \text{ mm} \times 1.3 \text{ mm}$ (micro-channels with a width of $600 \mu\text{m}$ and a depth of $400 \mu\text{m}$)	95% and more of the conversion ratio at the reaction temperature of 280°C	
Fuel: methanol	Holladay et al. [86]	Two cells in series (area of each cell: 1 cm^2); the volume of fuel processor (a catalytic combustor, two vaporizers, a heat exchanger, and a catalytic methanol reformer): less than 0.25 cm^3 and a mass of less than 1 g	99% conversion of methanol at around $\sim 400^\circ\text{C}$	Maximum power density: $\sim 10 \text{ mW cm}^{-2}$

have a novel design and microfabrication on a μ -fuel cell, which consists of a polymethylmethacrylate (PMMA) flow field plate with narrow and deep channels cut by excimer laser. The peak power density achieved was 31 mW cm^{-2} at room temperature with an unusually high hydrogen pressure of about 4 atm.

Another factor that matters for the selection of the material is the contact resistance between MEA and bipolar plate. Generally the assembly of micro-fuel cell should be boltless in order to miniaturize the system. In that case, selection of the materials for bipolar plate is very important for getting lower contact resistance between MEA and flow field and getting uniform pressure distribution.

The one of the important factors for designing micro-PEMFC is flow field design especially with respect to transport phenomena. The flow phenomena in micro-PEMFC is quite different from that in large scale fuel cells. The channels are of micron cross-section and Reynolds numbers are typically very low (0.001–0.1) in the micro-fuel cells. Rawool et al. [37] studied theoretically the flow behavior in a simple serpentine channel attached to porous transport layer (PTL). The simulation for velocity profile and pressure drop using computational fluid dynamics (CFD) showed that the velocity magnitude along the interface between the microchannel and the PTL increases as the permeability is increased. That means that the flow through the PTL increases as the permeability of the porous layer is increased. Hsieh et al. [38,39] compared the performance of micro-PEMFC (with flow of air in cathode) with three different flow channels namely interdigitated, mesh and serpentine types of flow fields. The interdigitated type flow pattern produced superior power density to those of the mesh and serpentine channels. This is due to combined effects of convective transport mechanism (which refers to smallest diffusion resistance) for the reactant gas together with a better distribution. Cha et al. [40] investigated the performance of micro-fuel cell using flow field with three different width of channels (20, 100 and $500 \mu\text{m}$). Fuel cell performance was increased as the channel size decreases from 500 to 100 μm . By contrast, performance was decreased for the 20 μm channel. The initial increase of performance with the channel size is due to mass transport benefits by reducing dead zones and increasing gas flow velocity. In the case of 20 μm channel, the decrease in performance is due to water-blocking (or flooding) effects in the channels.

2.3. Direct methanol fuel cell (DMFC)

DMFC is a power generator that directly converts the chemical energy of methanol into electricity. Methanol fuel has several advantages including lightweight, small size, long duration, and easy fuel refilling. DMFC is one of the best candidates for micro-fuel cell applications, such as mobile phone, personal computer and electronic gadgets. Though there is considerable development on catalyst (especially to reduce the amount of catalyst with superior performance) of MEA and handling of high concentration of methanol without much crossover through the membrane, still it lacks the high performance based on power density.

There is extensive research on DMFC. Some of the reported information on DMFC is described here. The detailed research

and development on DMFC and anode catalyst in DMFC are given in Kamarudin et al. [41] and Liu et al. [42], respectively. In DMFC there are various types of attractive substrate materials used for bipolar plates in order to make it smaller. These are silicon [43], printed-circuit board (PCB) [44,45], photopolymer [40] and flexible polymer substrate [46].

Yen et al. [47] fabricated a direct methanol μ -fuel cell by silicon micromachining on a pair of $500 \pm 20 \mu\text{m}$ silicon wafers employed as the bipolar plates. The peak power density achieved was 47.2 mW cm^{-2} with 1 M methanol at 60°C . Lu et al. [48] developed a silicon based μ -fuel cell fed with 2 M methanol. The peak power density achieved was 16 mW cm^{-2} at 23°C and 50 mW cm^{-2} at 60°C . They also observed the unchanged performance up to the concentration of 4 M methanol solution and slight less performance with 8 M methanol solution.

Yao et al. [49] developed MEMS based μ -DMFC on silicon substrate with a power density of 25 mW cm^{-2} . The proposed DMFC contains several unique features: a silicon wafer with arrays of etched holes selectively coated with a nonwetting agent for collecting water at the cathode; a silicon membrane micro-pump for pumping the collected water back to the anode; and a passive liquid–gas separator for CO_2 removal. All of these silicon-based components are fabricated using micro-electromechanical systems (MEMS)-based processes on the same silicon wafer, so that interconnections are eliminated, and integration efforts as well as post-fabrication costs are both minimized.

Generally the preparation method of MEA for DMFC is “dry” hot-pressing method that relies solely on hot-pressing at a high pressure and temperature. Liang et al. [50] introduced the glue method for fabricating the MEA. One binding agent is used in this glue method in order to provide a better adhesion and stronger binding force between a membrane and an electrode, thereby facilitating a better interfacial contact between the electrode and the Nafion[®] membrane. This method may produce very stable MEA with respect to long term operation.

In DMFC, water management represents a critical issue to enhance the cell performance and sustained cell operation for an extended period of time. Blum et al. [51] discussed overall water balance between the cell’s net production rate and water loss through an air-breathing cathode. It was recognized that a water-neutral condition occurs when the water molecules lost per molecule of methanol consumed by the cell reaction approaches two, a stoichiometric value of the net water production per mole of methanol. The μ -fuel cell, with a plastic housing, showed a stable operation for 900 h and has achieved a peak power density of 12.5 mW cm^{-2} . Alyousef and Yao [52] introduced the concept of alternate strips of hydrophobic and hydrophilic channels in the cathode layer made of a silicon wafer for air and water management. In this design, water will be guided along the hydrophilic wetting channels and collected while air goes into the cathode from the hydrophobic dry areas.

Another important issue for designing DMFC is the flow field design especially the design of anode flow field through which, methanol solution is distributed to the diffusion layer and diffuses to the catalyst site. Generally carbon dioxide (CO_2) is

produced in the anode of DMFC under load and the mixture of gases of CO_2 and hydrogen evolves when DMFC is under open-circuit conditions and at low oxygen flow rates [53]. Therefore consideration of the flow behavior is not only important but also mass-transfer rates of diluted methanol solution from the flow channel to the reaction zone in the presence of produced CO_2 in the anode field design. Wong et al. [54,55] found the decrease in mass transfer area between methanol solution and the diffusion layer with a reduction in channel width while a narrower channel leads to a higher mass-transfer coefficient. Therefore the optimum channel size is required in order to get best performance. At high current density, the gas-void fraction in both the diffusion layer and the flow channel increases due to high amount of CO_2 evolved. This will increase the gas void fraction in the flow channel which increases the liquid-phase velocity, resulting in a higher hydrodynamic mass-transfer coefficient at the channel/diffusion layer interface under the operation with the same flow rate of methanol solution. However, increasing the gas void fraction in the DL also increases the mass-transfer resistance of methanol. Therefore, the current density is independent of the overall mass-transfer coefficient at low flow rates of methanol solution [56,57]. At high flow rate of methanol solution, though the mass-transfer resistance at the channel/diffusion layer interface remains more or less the same with increasing current density due to almost single phase behavior, the increased gas-void fraction in the diffusion layer as a result of the increased current density results in a lower effective mass transfer coefficient in the diffusion layer. Therefore the overall mass transfer coefficient decreased slightly with increasing current density in the case of high methanol flow rate.

Ilic et al. [58] compared direct methanol fuel cells (DMFC) and lithium-ion batteries. They estimated that there is a need of time of about 7–12 years when DMFC can compete with lithium-ion batteries in terms of size, cost and overall performance. Hotz et al. [59] compared the performance of μ -RHFC and DMFC using numerical models. The superiority of μ -RHFC in terms of exergetic efficiency has been reported at the same inlet flow rate of methanol, though there is complexity with μ -RHFC where three components at different levels have to be heated.

2.4. Direct formic acid fuel cell (DFAFC)

Chu et al. [60] performed micro-fuel cell with direct formic acid as fuel using nano-porous silicon membrane. The nano-porous silicon created by electro-chemical etching exhibited performance of maximum power density of 30 mW cm^{-2} with 5 M formic acid and 0.5 M H_2SO_4 at the anode and air breathing at the cathode.

Some groups [61] reported that higher power density (120 mW cm^{-2} at 0.5 V with 15 M formic acid) can be achieved with direct formic acid fuel cell (DFAFC) which can be fitted in cell phone to provide enough power to make calls without the need for a supplementary battery. The higher performance with DFAFC and more especially a significant low crossover of formic acid compared to that of methanol in DMFC was also claimed. Zhu et al. [62] also showed the high perfor-

mance DFAFC (maximum power density of 110 mW cm^{-2} at 30°C) with Nafion-112 as membrane and platinum and platinum–ruthenium black catalyst for the cathode and anode, respectively. Recently there is a much higher performance in DFAFC with Pd based anode catalyst [63,64]. Zhu et al [63] showed DFAFC with Pd catalyst can generate power densities of $255\text{--}230 \text{ mW cm}^{-2}$ at relatively high voltages of 0.40–0.50 V in a concentration range of formic acid from 3.0 to 15.0 M at a room temperature of 20°C with dry air and zero back-pressure.

3. Commercial developments of micro-fuel cells

3.1. μ -RHFC

Direct methanol fuel cells (DMFC) are being developed with an eye toward use in mobile phones, but they are poorly suited to applications demanding high output. There is a try to overcome it by using RHFC with methanol steam reformer. Currently there are four laboratories/companies involved to make this kind of product. These are Lawrence Livermore National Laboratory, Battelle, Casio, and Ultracell (Table 4). The compact methanol fuel processor developed by Casio Computer produced hydrogen from methanol heated to 280°C followed by preferential oxidation (PROX) reactor (Fig. 2) [65]. Initially a prototype of methanol reformer was announced in 2002, followed by an improved model in 2004 replacing the Si wafer substrate with a glass substrate. The problems of the older model, including poor insulation performance, a long start-up time and the fact that it generated high-concentration CO as a byproduct were resolved by the new design according to their claim. The target for sample-ship reformers is to use as power supplier in notebook personal computers (PC). Concerning thermal insulation,

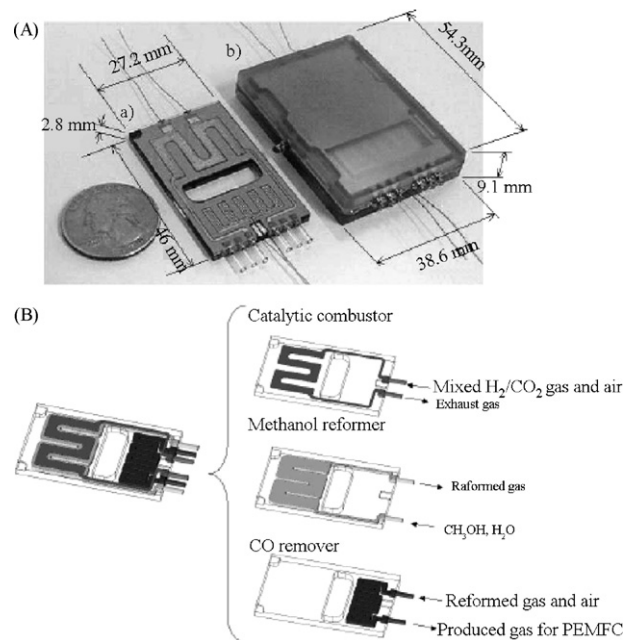


Fig. 2. (A) Photographs of newly developed (a) microreactor and (b) vacuum package enclosing microreactor. (B) Positions of three micro-channel reactors [65].

Table 4

Status of different companies and laboratories in the development of micro-fuel cell companies (RHFC, micro-formic acid fuel cell and hydrogen fed micro-fuel cell)

Type of micro-fuel cell	Company's name	Dimension and volume	Maximum power density and power	Potential applications
Micro-reformed hydrogen fuel cell	Ultracell [70]; fuel: methanol	150 mm × 230 mm × 43 mm (weight = 1 kg; with cartridge of 200 cm ³ and 550 cm ³)	Maximum power 25 W; cartridge duration: 9 h and 24 h at 20 W for 200 and 550 cm ³ volume of cartridge, respectively	Military purposes, portable electronics
	Casio [65]; fuel: methanol	Reformer + PROX system size: ~20 cm ³	2.5 W	Charger
	Seiko Instruments Inc. [27]; fuel: sodium borohydride	1 W system: 75 mm × 40 mm × 60 mm; 3 W system: 80 mm × 45 mm × 70 mm; 10 W system: 200 mm × 65 mm × 53 mm	Time of operation: 2–5 h	Maximum energy density of the system: 60 Wh l ⁻¹
Direct formic acid fuel cell	Korea Institute of Science and Technology (KIST) [87]	Stack: 88 mm × 70 mm × 50 mm; system: 205 mm × 85 mm × 82.5 mm; total weight: 1.8 kg (with fuel)	Maximum power density: 60 mW cm ⁻² ; delivered power: 13.7 W (maximum 30 W)	Laptop computer
Hydrogen fed micro-fuel cell	Angstrom Power Inc. [73]	25 mm × 66 mm × 100 mm	Maximum power: 2 W (through hydrogen storage); time of operation: 6 h; time of operation: 6 h	Charger for cell phones, PDAs, and digital cameras; needs infrastructure for refueling hydrogen into the system; energy density: 72.7 Wh l ⁻¹
	NTT DoCoMo and Aquafairy Co. [75]	24 mm × 24 mm × 70 mm; weight: 45 g	Maximum power: 2 W (800 mAh at 3.6 V) fuel cartridge (water plus hydrogen producing catalyst): 10 cm ³ ; time of operation: 5 h	Charger for lithium-ion battery in 3G handsets. Energy density of the system: 248 Wh l ⁻¹
	Hitachi Maxell Ltd. [66]	97 mm × 87 mm × 31 mm	Maximum power: 10 W; aluminium: 20 g; water: 40 cm ³ ; time of operation: 4–5 h	For laptop; energy density of the system: <191 Wh l ⁻¹
	Canon Inc. [76]	Fuel cell: 35 mm × 35 mm × 12 mm; metal hydride: 60 mm in length with 10 mm diameter and fuel cell: 55 mm × 77 mm × 3.5 mm; metal hydride: 55 mm × 66 mm × 5 mm	Stack power density: 0.2–0.3 W cm ³ ; system power density: 0.1–0.15 W cm ³	Maximum energy density of the system: ~160 Wh l ⁻¹

the main unit of the reformer consists of two glass substrates with vacuum insulation, while the inside faces of the substrates are covered with a thin film of primarily Au (gold) to minimize radiant heat from the interior. In the new design, the package surface temperature only rises to about 40 °C as a result of radiating insulator according to their report. The start-up time has been improved so that it now only takes 6 s to heat the reformer to 280 °C. The quick start was achieved by taking steps including improving insulation performance to minimize heat escape, directly heating the glass substrates with embedded heaters, and thinning out the glass substrates to reduce their thermal capacity [66]. Battelle has developed a prototype of methanol reformer for 15 W and 400 mW fuel cell [67,68]. The catalyst for methanol reforming is Pd/ZnO and the operating temperature is ~350 °C. The total weight of fuel processor/fuel cell assembly is around 1 kg for 15 W power in which total volume of the processor is 25 cm³ with a weight of 100 g for use in military purposes.

Total volume of the methanol reformer plus methanation reactor (as CO clean-up stage) for 400 mW is 0.3 cm³ with a weight of 1 g. The methanol reforming catalyst in this case is also Pd/ZnO. Lawrence Livermore National Laboratory has prepared silicon based micro-reformer (25 mm² area) for methanol steam reforming. The catalyst is copper and nickel oxide and deposited on the micro-channels by sputter coating. The complete conversion of methanol was obtained at 270 °C with this catalyst. The CO level in the reformat gas is decreased by PROX catalyst before feeding in the micro-fuel cell [69]. Recently Ultracell (exclusive license with Lawrence Livermore National Laboratory for micro-fuel cell technology) developed two products for the military purposes and portable electronics with the maximum power of 25 W [70]. The two models differ from the significant reduction of size of the system which facilitates to provide 2.75 times larger space for fuel cartridge with the same size and weight of the total system (including the cartridge). This results the increase of the operational time for about 2.67 times.

Table 5
Status of different companies and laboratories in the development of micro-DMFC

Type of micro-fuel cell	Company's name	Dimension and volume	Maximum power density and total power	Potential applications
DMFC	Samsung Advanced Institute of Technology [88]	100 cm ³	1.3 W at 3.6 V; 3000 mAh	PDA and Mobile phone; energy density of the system: 108 Wh l ⁻¹
	MTI (Mechanical Technology Inc.) Micro-Fuel Cells [89]	100% methanol	Average 800 mW (maximum 1 W), 35 Wh	RFID Reader and Mobile phone
	Motorola [90]	50 mm × 100 mm × 10 mm	Net power 100 mW	Mobile phone/recharger
	Fujitsu [91]	150 mm × 56 mm × 19 mm (160 ml), 190 g, cartridge 18 and 12 cm ³ (99% methanol)	Maximum 9 Wh	Mobile phone; energy density of the system: 56.3 Wh l ⁻¹
	Samsung SDI [88]	230 mm × 80 mm × 53 mm (1 l), cartridge 200c	Nominal 20 W, (maximum 50 W), 200 Wh l ⁻¹	Laptop computer
	SFC Smart Fuel Cell GmbH [92]	150 mm × 112 mm × 65 mm, 1.1 kg cartridge 125 ml	Average 25 W (maximum 50 W), 12 V	Laptop computer and portable application
	Toshiba [93]	50 mm × 98 mm × 40 mm, 160 g, cartridge 7 cm ³ (99.5% methanol)	30 mW cm ⁻² , 300 mW	Mobile phone
	NEC [94]	Methanol cartridge 300 cm ³ (10% below)	14 W (maximum 20 W), 12 V, 900 g	Laptop computer
	Antig [95]	110 mm × 7.6 mm × 55 mm; 480 mm × 345 mm × 315 mm	2 W; 100 W (maximum output)	Air-passive type; portable electronics; laptop computer

3.2. DMFC

Toshiba is widely recognized as the industry leader in DMFC development. The company has been working on a test DMFC prototype in two versions: one is geared for Flash-based MP3 players, the other is specifically designed for devices with a hard disk drive device. In Taiwan, Antig Technology is moving closer to delivering DMFC modules to its customers, which could potentially include some of the largest electronics contract manufacturers in the world. Antig is mainly focusing the development of DMFC prototype for notebook PCs. Antig foresees its first phase of DMFC deployment with solutions that offer a combination of conventional lithium-ion polymer and fuel cell hybrid. 3M is currently the dominant supplier of MEA [71]. The fuel-cell mobile phone co-developed by Toshiba/KDDI and Hitachi/KDDI is a hybrid type, with power supplied by a compact fuel cell and fuel tank plus an internal lithium-ion battery. Toshiba /KDDI uses high-concentration methanol to achieve a battery capacity 2.5 times the conventional value with a single refill [72]. Samsung SDI, Samsung Advanced Institute of Technology (SAIT), SFC, Fujitsu, motorola and MTI are the other prototype sample producer with DMFC for portable electronics (details are given in Table 5).

3.3. Hydrogen fed micro-fuel cell

As it is already mentioned that hydrogen fed micro-fuel cell shows the best performance provided that hydrogen can be stored safely in a small place or produced from water by convenient way. Angstrom Power Inc. has manufactured the prototype sample of 160 cm³ (350 g) micro-fuel cells for the

application of cell phones, PDAs, and digital cameras with 2 W power as recharger. The hydrogen storage material is metal hydride. According to their claim, it has the capability of running for 6 h by the hydrogen stored in the system [73]. They have also produced the prototype of recharging station for provision of the hydrogen refueling. Another interesting thing to get pure hydrogen is from water by some metal powders (like Al/Mg). Hydrogen is generated through the Al oxidation reaction, namely $\text{Al} + 3\text{H}_2\text{O} \rightarrow \text{Al}(\text{OH})_3 + 3/2\text{H}_2$. One gram of Al can produce 1.3 l hydrogen which is close to 95% of the theoretical maximum. It is claimed that sodium hydroxide acts as a catalyst as sodium hydroxide consumed in the hydrogen-generating reaction can be regenerated in the following reaction:



Hitachi Maxell has developed a device capable of generating hydrogen just by mixing Al powder into water [66]. The main aim was to improve the reactivity of aluminium by activating the aluminium fine particles obtained by milling aluminium alloy in water. The activation procedure comprised a thermal shock treatment wherein the aluminium fine particles were repeatedly heated and cooled down rapidly followed by storing in refrigerator [74]. It is intriguing because it only requires inexpensive Al and water, with no catalyst. From this article, it is obvious that the particle size is the determining factor for getting hydrogen. In another development, NTT DoCoMo and Aquafairy Co. co-developed the water based hydrogen generation system plus micro-fuel cell for charging the lithium-ion

battery (DoCoMo's FOMA 3G handset customers' device) [75]. The output of the system is 2 W and the electrical capacity is 10 Wh with 10 cm³ water plus hydrogen producing catalyst. The other properties like orientation-free property and safety and maintenance aspects are also in their consideration in order to improve the whole system. NTT DoCoMo is planned to develop an external recharger with higher performance than that of such rechargers as the company currently sells. Meanwhile, the company aims to develop a built-in fuel cell with a volume, current capacity and output of 10–20 ml, 1 Ah or more and 1 W or more, respectively. Canon Inc. has carried out the feasibility study for micro-fuel cell with metal hydride (LaNi₅) as hydrogen source. The total volume of the system is around 35 cm³ which can produce the maximum power of 2.1 W. This performance is increased with proper thermal exchange due to improvement of hydrogen supply [76].

4. Conclusions

There is a great potentiality of micro-fuel cells to deliver more energy per volume weight than conventional batteries. This is a motivating factor to consider micro-fuel cells for portable applications. The main challenge will be to identify the right technology and the configuration of fuel cells. Very small fuel cells for use in cell phones are a goal of many wireless carriers. However, engineers face a big challenge in getting fuel cells small enough to fit inside a cell phone. Therefore, miniaturization remains a design challenge despite much work is being done on this. Thus the initial focus is on using as recharger. The different types of micro-fuel cells namely μ -RHFC, hydrogen fed micro-fuel cell, DMFC and DFAFC are discussed and their commercialization status are also reported. Micro-RHFC is advantageous with respect to the high performance of PEMFC, though there is great concern issue of the system's complication and significant energy consumption for BOP. Most DMFC systems possess lower energy density which is mainly due to very low performance per cm² MEA area. In that aspect, PEMFC is better choice for micro-power services. The selection of PEMFC will further strengthen if it is possible to feed pure hydrogen with easy and control way on-board production. There is much work required on the further improvement of the performance of micro-PEMFC especially with respect to air breathing system. DFAFC system can also be an interesting alternative depending on the further progress on the improvement of the system's components based on the fact it has higher power density than DMFC. The theoretical energy density (Whr/lit) with respect to the overall system is still below 200 in the maximum cases of the prototype sample. The analysis on the commercialization status is based on the performance on prototype sample gathered from open literature, but when these devices will be available for series production is not clear yet.

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