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PEM FUEL CELLS FOR TRANSPORTATION AND STATIONARY POWER GENERATION APPLICATIONS

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Abstract

We describe recent activities at Los Alamos National Laboratory devoted to polymer electrolyte fuel cells in the contexts of stationary power generation and transportation applications. A low cost/high performance hydrogen or reformat/air stack technology is being developed based on ultra-low platinum loadings and on non-machined, inexpensive elements for flow-fields and bipolar plates. On board methanol reforming is compared to the option of direct methanol fuel cells, because of recent significant power density increases demonstrated in the latter.

1. INTRODUCTION

The fuel cell is presently the most efficient device for the conversion of hydrogen fuel to electric power. As such, the fuel cell represents a key element in efforts to demonstrate and implement hydrogen fuel utilization for electric power generation.

For the last thirty years, the benefits provided by fuel cell technologies, including high efficiency, low environmental emissions and modular construction, have fostered continued interest in their development for transportation and customer-side-of-the-meter utility applications. Among the family of fuel cell technologies, polymer electrolyte membrane (PEM) fuel cells have some uniquely attractive features. They provide high power density, high chemical-to-electric energy conversion efficiency, and fast and easy start-up. Additionally, they are very reliable and are constructed from durable and benign materials. Until recently, however, the large-scale, terrestrial application of PEM fuel cell technology for transportation and for utility or stand-alone power generation was considered to be too expensive and too sensitive to the low levels of carbon monoxide present in reformed carbonaceous fuel feed streams. These barriers to the practical implementation of PEM fuel cells have been significantly lowered in the last few years, in part as a result of work done at Los Alamos National Laboratory. This paper describes some of our efforts to lower the cost of PEM fuel cell stacks and to demonstrate some effective approaches for the mitigation of anode catalyst deactivation effects resulting from low levels of CO or other organic impurities. These recent technical developments increase the attractiveness of PEM fuel cell technology for utility and stand-alone power generation applications as well as for transportation, where it has previously held the strongest attraction.

Even though these and other recent developments have been very effective in raising the profile of PEM fuel cell technology as a viable technology for transportation, the commercial implementation of hydrogen-fueled PEM fuel cell stacks is likely to occur first for stationary power generation applications, because of the somewhat less stringent requirements on overall cost, space, and the nature/source of the hydrogen fuel.

Some recent achievements at Los Alamos that have advanced our PEM fuel cell technology to the point of readiness for the demonstration of a low-cost/high performance hydrogen-fueled stack are summarized below.

- Low cost membrane-electrode assemblies (MEAs) with ultra-low platinum loadings of 0.2 g Pt/kW have been demonstrated in 5000 hour life tests, generating areal power densities of 6 kW/m² on pressurized hydrogen/air [1-4]. These MEAs have been recently implemented successfully in stacks by some developers.

- Effective water management principles have been identified, based to a large extent on advanced membrane electrolytes evaluated jointly with membrane manufacturers [5,6].

- Detailed evaluation of losses in the PEM fuel cell has been based on long-term investigation of fundamental processes in the cell, including all electrocatalytic and transport phenomena [6].

- Based on the above evaluation, an effective computer code and a set of experimental diagnostics for the PEM fuel cell have been established [7].

- The tolerance to carbon monoxide levels expected from steam-reforming of carbonaceous fuels has been significantly improved. In previous work we have shown that air bleeding into the anode corrects for CO levels in the anode feed stream as high as 50-100 ppm under normal operation conditions [8,9]. However, we have recently identified ways to increase the CO level tolerance to well above 100 ppm.

These achievements place the PEM fuel cell technology developed at Los Alamos in a strong position to demonstrate that high performance may be achieved at low cost in hydrogen-fueled PEM fuel cell stacks. Activities based on these technologies are taking place at some PEM stack development sites in North America.

Most recently, the Fuel Cell Core Technology Program at Los Alamos addressed PEM fuel cell issues related to both transportation and stationary power generation applications. In the transportation arena, providing hydrogen/air and reformate/air PEM fuel cell stacks at low cost and coupling the PEM fuel cell stack to reformers of carbonaceous fuels remain important challenges to overcome. Fuels being considered range from methanol to petroleum mid-distillates. In a system designed for transportation, based on the reforming of liquid hydrocarbons, the challenge of operating PEM fuel cell stacks on hydrogen that is diluted by CO₂ (and nitrogen) and contains significant levels of CO, is similar to that for stationary power applications where the stack is fed by reformed natural gas. The situation in the latter case is significantly less severe, however, due to stationary operation conditions, which require less stringent cost and power density targets.

This paper describes some of our research efforts devoted to low-cost/high performance PEFCs for stationary power generation. It also briefly mentions some recent achievements with direct methanol PEM fuel cells at Los Alamos.

2. STATIONARY POWER APPLICATIONS [10-12]

2.1 Background

While large utility applications are envisioned as belonging to the domain of molten carbonate fuel cells (MCFCs) and solid oxide fuel cells (SOFCs), there are many applications such as utility peak power generation, demand-side-management, and dispersed power generation for which the phosphoric acid fuel cell (PAFC) and the polymer electrolyte fuel cell (PEFC) could be more attractive. Natural gas-fueled PAFC

technology for such applications has become commercialized, with a 200 kW PAFC unit (from ONSI) costing approximately US\$2,500/kW. The unit operates at 40% electrical efficiency and 85% overall thermal efficiency (including heated water). Ballard, originally in conjunction with Dow, is developing a 250 kW natural gas-fueled PEFC power plant after demonstration of a 10 kW plant [12].

The PEFC appears to have some important advantages over PAFC technology in terms of energy conversion efficiency (higher voltage at same current density) and power density. In the long term and in light of recent advancements, the life-cycle cost of PEFC technology could be more favorable. A stable, inert polymer electrolyte and an easy start-up/shut down characteristic may make the reliability and longevity of the PEFC superior. The PEFC could compete for the near-term markets in which the PAFC is expected to be successful. Note the following background discussions, culled from studies that were oriented towards the PAFC [13-16].

Many applications in the U.S. utility sector require power capacities in the range of 1 to 10 MW. This is particularly true for peak demand in some of the small public utility systems that are required to pay high charges for peak demand, usually to a private utility grid. These modest utility applications could be more suitable for the PAFC and PEFC. Indications are that at US\$1,100/kW, fuel cell technologies could be attractive for peak power requirements. A conservative estimate of this market in the US is 14,000 to 17,000 MW for fuel cells. The efficiency, compact design, low maintenance and potentially lower cost of the PEFC could allow it to capture a substantial portion of this market. Ideally, the PEFC system could be in a cogeneration unit, with the waste heat utilized for its thermal value. Increasing the PEFC operating temperature would reduce the effect of CO on the anode catalyst and would allow the low grade waste heat to be used more effectively. Some increase in operating temperature is possible even in present day PEFCs.

It is becoming more difficult to obtain regulatory approval for large-scale power stations or to secure rights-of-way for high-tension power lines. Thus in the relatively near future it is reasonable to expect that a substantial amount of additional power capacity may be from neighborhood or apartment building power stations that tap into the massive natural gas distribution grid. Efficient, low emission, and quiet fuel cells are ideal for this type of application. Though gas turbine systems by comparison are less expensive, it may be more cost effective to pay more for an environmentally benign fuel cell system and in effect to bypass regulatory processes and the associated costs. Commercial and industrial facilities will probably take the lead in implementing fuel cell systems. According to GRI, approximately 18,000 MW of the latter power market in the US should be economical for fuel cell cogeneration systems, provided the installed cost is US\$1,000/kW [15]. The industrial sector is similar to the commercial sector in size and in required fuel cell characteristics. Approximately 10% of this potential market is for small (less than 1 MW) cogeneration systems. Waste heat is at a premium in these applications, thus the PEFC may need to operate as a cogeneration system to be competitive.

2.2 Low Cost Stack Components

With a target of only US\$1,000/kW, a PEFC stationary power plant, with significant power conditioning and natural gas reforming costs, will require relatively low cost fuel cell stacks. At Los Alamos, we are experimenting with the scale-up and automated fabrication of the low platinum loading catalyzed membranes that we developed based on "thin-film" catalyst layers [1-4]. A recently developed scheme for the

lab-scale (areas up to 200 cm²), automated preparation of membrane electrode assemblies by applying thin film catalysts directly to the membrane, is shown in Fig. 1. This mode of preparation is computer- controlled and provides high repeatability [17].

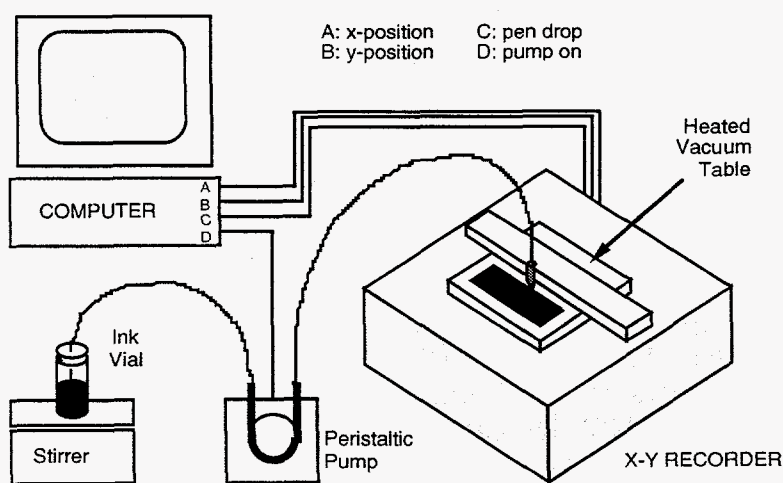


Fig. 1. MEA fabrication using a computer controlled process for applying a uniform layer of catalyst ink to either a membrane or a decal blank positioned on a heated vacuum table.

At present some of our work concentrates on the development of low-cost gas diffusion backing, flow-field and bipolar plate materials. We have investigated metal-based hardware components as part of our PEFC stack development for stationary power applications, where the weight density of the stack is not as critical as for transportation applications. One promising stack configuration is based on the use of stainless steel screens as flow-fields. Researchers elsewhere [18] have reported the use of stainless steel flow-fields with no apparent effects of corrosion after 1000 hours of testing. Though the stainless steel element is not in direct contact with the membrane electrolyte, the surface of the metal is exposed to the corresponding humidified gas streams. Therefore some form of surface treatment may eventually be required to achieve extended long-term stability. Initial short-term tests of PEFCs based on ultra-low Pt catalyst loadings [1-4] and stainless steel screen flow-fields show performance equal or exceeding that obtained under similar conditions with machined metal flow-fields [17]. The ability to eliminate machining costs of flow-fields by using off-the-shelf metal screens would be advantageous to a stack manufacturer attempting to lower the cost of PEFC stacks. Figure 2 depicts a polarization curve for a 100 cm² active area PEFC, employing a Los Alamos MEA with 0.14 mgPt/cm²/electrode and stainless steel screen flow-fields. The screens seem to be effective flow-fields under ordinary PEFC conditions of mixed gas/liquid(water) feed streams, enabling effective gas distribution across relatively large electrode surface areas.

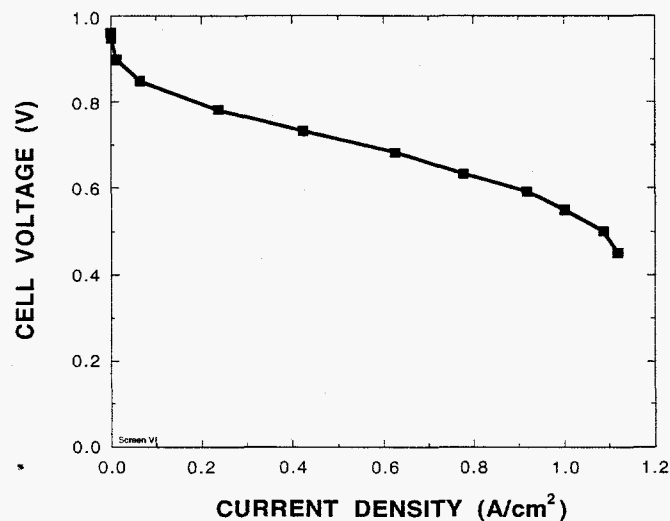


Fig. 2. Internally manifolded 100 cm² cell with 0.14 mg Pt/cm² thin-film electrodes on Nafion 112 membrane and stainless steel screen flow-fields. H₂/Air = 3/3 atm, 50% air utilization at 1 A/cm², T_{cell} = 80°C.

2.3 Coupling of a Natural Gas Reformer and a PEFC Stack

Stable PEFC anode operation on a feed stream of natural gas reformat is a challenging requirement for utilization of PEFCs for stationary power generation. Some of the problems associated with the operation of PEFCs on realistic fuel feed streams have been addressed recently by operation of PEFCs on methanol reformat. The roughly 1% CO in a methanol reformat can be decreased to about 10 ppm using shift reactors followed by a preferential oxidation reactor (PROX) utilizing a Pt/alumina catalyst [19]. The sensitivity of the PEFC to such residual levels of CO (10 ppm) is still very significant at the normal operation temperature of 80°C. We have demonstrated the ability to remediate effects of residual CO in the fuel feed stream by bleeding small levels of O₂ or air into the anode compartment of the fuel cell [8,9]. There have been recent indications of success by others in implementing this approach with PEFC stacks. We also have shown [20] that a Pt-Ru anode catalyst does not catalyze the reverse shift process that generates CO from CO₂ within the fuel cell anode, but the high level of CO₂ in the reformat can create dilution effects. A combination of i) shift reactors to lower the roughly 10% CO level of the methane reformat down to about 1%, ii) a PROX reactor upstream of the fuel cell to lower the CO to the 1-10 ppm level, and iii) limited air bleeding into the anode to eliminate the residual CO effect, could conceivably provide fuel cell performance very nearly equal to that of a neat hydrogen cell. The general system is depicted in Fig. 3a. An overall loss of fuel caused by the oxidative treatments in such a system should not exceed 4%.

Several recent efforts with Pt-Ru/C catalysts [20-22] have indicated that some of the problems caused by residual CO within the PEFC anode can be solved by the use of appropriate Pt-Ru anode catalysts that seem to exhibit sufficient CO electrooxidative activity. The identification of a CO tolerant or rather, CO consuming, anode catalyst would naturally have a significant impact on the design and efficiency of the system.

2.4 Purifying Natural Gas Reformat with a Hydrogen Separator

As discussed above, a major issue impacting the coupling of the PEFC to a natural gas (methane) reformer is the effect of contaminants, particularly CO, on the fuel cell anode. A possible approach to circumvent this difficulty is to utilize some form of gas separator, which would supply pure hydrogen to the fuel cell. In this section, we describe a recent effort at Los Alamos to develop an effective hydrogen separator and use it to provide a pure hydrogen feed stream from simulated reformates to a polymer electrolyte fuel cell. This approach, depicted in Fig. 3b, not only eliminates the anode contamination difficulties, but decouples to some extent the operation of the fuel processor and the fuel cell. The hydrogen feed to the fuel cell could be effectively dead-ended, as is done in a number of fuel cell systems that use a neat hydrogen feed. This simplifies control of the complete system and allows stockpiling of hydrogen during low power demand. Thus, the fuel cell stack could then be brought up instantaneously with a "cold" reformer.

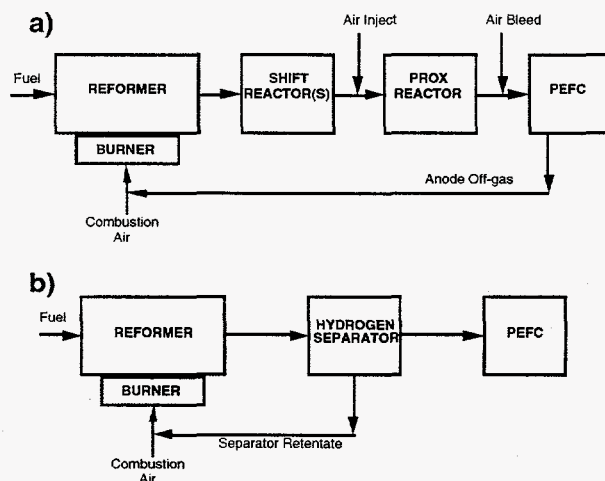


Fig. 3. Reformer/fuel processors for PEFCs: a) a more conventional system with shift and PROX reactors and air injection at two points, and an b) alternative concept incorporating a hydrogen permselective separator.

The use of a membrane separator module usually has been associated with high cost. Commercial hydrogen purifiers traditionally use Pd/Ag films, either free-standing or supported, which have significant cost and integrity drawbacks. Several groups are studying ways to increase the stability and lower the Pd content of hydrogen separator membranes. A fellow group at Los Alamos is developing Pd clad Ta films [23]. Tantalum supplies phase and structural stability, while providing high hydrogen permeability. The Pd is applied to either face of the Ta film to facilitate the interfacial process of hydrogen dissociation. At this point, the total Pd loading is only about 1 mg Pd/cm² of separator membrane. We have operated a 5 cm² single cell PEFC on the permeate of a 4 cm² Pd/Ta membrane separator operating at 315°C with a pseudo-methanol reformat feed (1% CO, 24% CO₂, 74% H₂). Figure 4 indicates that about 600 mA/cm² is obtained at 0.6 V. In contrast, the presence of only 100 ppm of CO (in H₂) fed directly to the anode suppressed the current density at this voltage to about 60 mA/cm², one-tenth the cell performance on the permeate, as is also shown in Fig. 4. It is anticipated that the separator will be equally effective with the much higher CO levels expected from a natural gas reformer. On-going development objectives for this

membrane separator include improving its throughput and further lowering the Pd loadings. Currently, the Pd film used can be thought of as adding the need of 1mg of Pd per cm^2 of fuel cell membrane area.

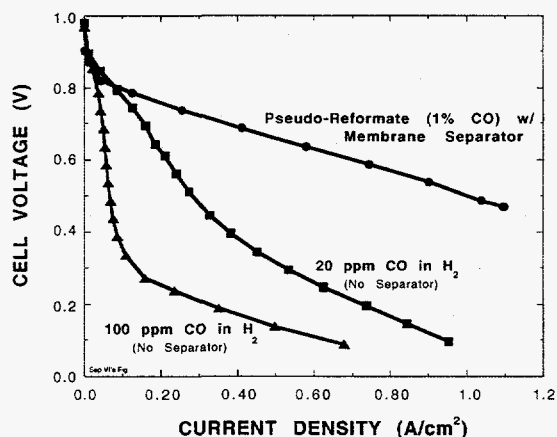


Fig. 4. Polarization curves depicting a single cell PEFC operating at 80°C on i) pseudo-reformate (1% CO) fed via a high throughput hydrogen permselective separator, ii) 20 ppm CO in hydrogen, and iii) 100 ppm CO in hydrogen.

Preliminary process simulations for the reformer/separator/fuel cell scheme depicted in Fig. 3b are quite promising, particularly for a methanol-fueled system. If a perm-selective separator is paired with a methanol reformer in the most effective way, the need for high and low temperature shift and partial oxidation reactors would be eliminated because the roughly 1% CO reformat product is useful as burner fuel. The natural gas-fueled system is more problematic when using a separator because of the lower hydrogen concentration in the reformat, however, a number of approaches to increase the separator throughput are possible.

3. DIRECT METHANOL PEM FUEL CELLS

Until very recently, the PEFC systems considered for transportation applications have been based on either hydrogen carried on board the vehicle, or on the steam-reforming of methanol to generate a mixture of hydrogen and CO_2 , as the fuel feed stream for the fuel cell stack. A central reason for resorting to methanol reforming on-board the vehicle has been the much more limited performance of the direct methanol fuel cell (DMFC). The demonstrated power density for the latter has been very low indeed and until recently its application for transportation has not been a serious option at all.

However, recent advancements in DMFC R&D have been quite dramatic, achieving power densities which are significant fractions of that provided by the reformat/air cell (RAFC) [24-28]. The more applied DMFC R&D work, aimed primarily at the demonstration of enhanced stable performance of DMFC single cells or stacks, has been supported in the U.S. by the Department of Energy (Office of Transportation Technology), the Advanced Research Projects Agency (ARPA) and the U.S. Army. Similar efforts in Europe resulted in a demonstration at Siemens of a high-performance polymer electrolyte DMFC operating at temperatures approaching 130°C [24].

As in the previous case of the hydrogen/air PEFC or the RAFC, the strong recent advances in DMFC performance have been achieved without any dramatic breakthroughs in electrocatalysis. The use of established Pt-Ru anode electrocatalysts and Pt cathode electrocatalysts in conjunction with polymer electrolyte DMFCs has resulted in very significant enhancements in DMFC performance when such cells are operated at temperatures as high as 110-130°C and particularly, as shown here, when catalyst layer composition and structure have been optimized. Operation at elevated temperatures is facilitated in such polymer electrolyte fuel cells by the anode being continuously in contact with liquid methanol-water mixtures.

Recent experimental work at Los Alamos National Laboratory resulted in record high performances of polymer electrolyte DMFCs [28], demonstrated by the final two figures, which show a polarization curve for a methanol/air cell and power density curves for DMFCs with oxygen and with air cathodes, achieved in cells with Nafion™ membranes operated at elevated temperatures. These recent results have been subsequently used in a comparative evaluation [29] for transportation applications of (i) a polymer electrolyte DMFC stack, and (ii) a system comprising a methanol reformer and PEM reformat/air fuel cell stack.

Based on that evaluation, it seems that the DMFC could provide comparable power densities and energy conversion efficiencies, thus becoming a serious candidate for transportation applications, provided the following requirements are also met:

- (1) Catalyst loadings are further reduced (or alternative anode catalysts developed).
- (2) Long term stable performance (1000 hours time scale) is demonstrated.
- (3) Strategies to minimize methanol cross-over to the cathode [30] increase the DMFC fuel efficiency to the 90% level.

4. CONCLUSION

We have investigated the use of ultra-low Pt loading MEAs (0.14 mg Pt/cm²) and stainless steel screen flow-fields in 100 cm² single cells as part of our efforts to develop low cost/high performance hydrogen or reformat/air PEFC stacks for stationary power applications such as utility peak power generation, demand-side-management and dispersed power generation. In general, these low-cost component cells provide power densities (approximately 0.6 A/cm² at 0.7 V) approaching those obtained with commercial cells, which typically utilize much higher Pt loadings (4 mg/cm²) and more expensive flow-field materials (i.e. machined graphite plates). On-going research includes further development of inexpensive flow-field, bipolar plate and gas-diffusion backing materials. Additional research at Los Alamos is focused on reducing the Pd content of hydrogen membrane separators for RAFC applications and improving overall DMFC performance, in which greatly increased power densities have been recently obtained (at high temperature with Pt-Ru anode electrocatalysts and Nafion membranes).

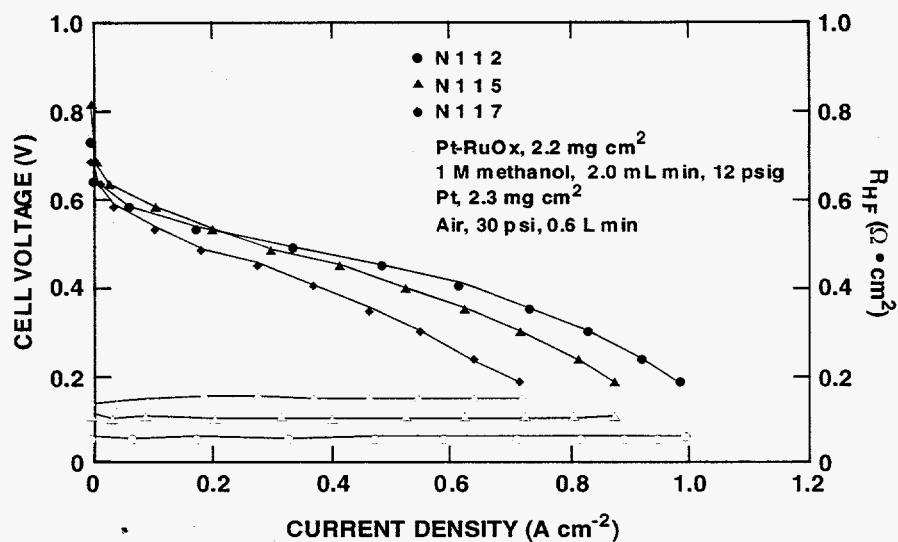


Figure 5. Polarization curves for 110°C, air cathode DMFC based on thin-film catalyzed Nafion 112, 115 and 117 membranes. Anodes: 2.2 mg/cm² Pt-RuO_x, 1 M methanol at 2 ml/min and 1.8 atm. Cathodes: 2.3 mg/cm² Pt-black, 3 atm air at 0.6 l/min.

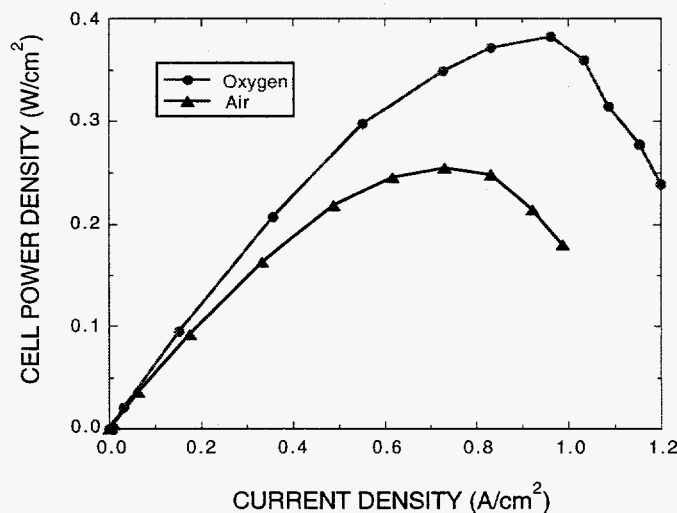


Figure 6. Power density curves for a thin-film catalyzed Nafion 112 assembly operating at 130°C on oxygen and at 110°C on air.

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