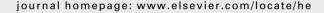
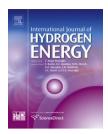


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The possible failure mode and effect analysis of membrane electrode assemblies and their potential solutions in direct methanol fuel cell systems for portable applications

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ABSTRACT

One of the major challenges in direct methanol fuel cells (DMFCs) is to design reliable and stable FC systems that satisfy the very high dynamic demand in various environmental conditions for portable devices. This paper provides an overview of several failure modes and effect analyses (FMEAs) which can have significant consequences on the durability and stability of DMFCs, including high and sub-zero temperature storage, dry and high humidification atmospheres, and fuel/oxidation starvation by breakdown of fuel/air supply components. Firstly, some characterization methods are discussed to investigate changes of membrane electrode assemblies (MEAs) in terms of their physiochemical and electrochemical properties after testing in various simulated failure modes. Secondly, possible mitigating solutions to minimize the hazards associated with them are suggested through a fundamental understanding and scientific approach. The relationship between the causes and symptoms in DMFC systems is determined by examining a variety of failure sources.

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1. Introduction

Recently, advanced power sources have become increasingly important due to their applications in portable electronic devices such as digital multimedia broadcasting phones, laptops, notebooks, and GPS car navigation systems that are now indispensable in our daily life [1–3]. However, as computing technologies have become integrated with wireless digital communication, today's leading-edge battery technologies for

multi-convergence electronic devices are theoretically reaching their critical point in terms of energy density, operational time, and recharge time [4,5]. Even more significantly, safety has become one of the key considerations in battery manufacture due to the danger of sporadic battery explosion caused by the misuse or malfunction of high capacity battery packs. In addition, hazardous chemicals (e.g., lead, mercury, and cadmium) found in some types of battery have a high environmental impact and raise the necessity of recycling or proper disposal.

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In this regard, the top-tier consumer electronics manufacturers such as Samsung, Hitachi, and Toshiba have devoted significant R&D resources to micro fuel cells (FCs). Compared to other FCs, polymer electrolyte membrane FCs (PEMFCs) and direct methanol FCs (DMFCs) have been envisaged as promising electrical power sources for portable applications due to their superior fuel density compared to conventional batteries. The theoretical specific energies of hydrogen and methanol are 33 Whg⁻¹ and 6.1 Whg⁻¹, respectively, in PEMFC and DMFC, while rechargeable Libatteries have a specific energy of around 0.2 Whg^{-1} [6]. DMFC technology which directly uses methanol as a fuel are the most viable for converged communication devices because of smaller system volume, lower weight and easier handling of fuel without any complex reforming process. In contrast, PEMFCs remain relatively unexploited since hydrogen production, storage, and distribution are complicated, even though hydrogen is the best fuel in terms of high specific energies and low environmental impact.

Over the last decade, the increasing research activity on DMFCs has given rise to considerable development in the performance of membrane electrode assemblies (MEAs) through the use of highly active electrode materials and optimization of gas diffusion layers (GDLs) with reduced methanol crossover [7–9]. Moreover, comprehensive and systematic research based on the degradation of MEAs and using various electrochemical and physiochemical analysis techniques has increased our understanding of the major sources and mechanisms of performance deterioration in the DMFC operation [10–12].

Despite the advances in the design and components of MEA, several technical issues in DMFC MEAs still need to be overcome before commercial micro FC power packs can be produced for consumer electronic devices. One of the challenges is to design reliable and stable FC systems that satisfy the very high dynamic demand in various environmental conditions for portable devices. As an example, MEAs for this system should withstand extreme environmental conditions like the torrid desert, tropical rainforests, and the cold polar regions. That is, FC systems must be able to provide stable average power even after storage and during operation at subzero temperatures and above 60 °C. However, the operation of portable FC systems at sub-zero and high environmental temperatures has not been greatly investigated and much effort should be devoted to mitigate this stability issues in these temperatures. Additionally, DMFC operation under high humidification conditions is yet another source of stability challenges due to the possibility of cathode flooding in the MEAs. On the contrary, performance degradation due to membrane dehydration may be more severe due to the low proton conductivity of the MEA with dry inlet gases when operating under high-temperature dry conditions compared to wet conditions.

An understanding of the endurance and stability of DMFC MEAs in extreme conditions is also important for portable applications to be accepted as a viable product. For instance, in the case of possible failure modes of DMFC systems such as malfunction of fuel/air supply components as the key ingredient of balance of plant (BOP), the performance and robustness of MEAs deserve special attention in the design of secure

FC systems. Failure mode and effects analysis (FMEA) of MEA can predict the power loss and determine the sources of catastrophic failure, thereby demonstrating the approaching commercial viability of FC systems. However, the failure causes and mechanisms in various failure modes have not been well investigated and remain to be elucidated.

In this research, the FMEA of MEAs on DMFC systems is conducted for application of portable power sources in various simulated environmental conditions such as sub-zero, high temperature, completely dry and very humid. In addition, changes of MEA in the view of physiochemical and electrochemical properties are investigated during the breakdown of the fuel/air supply components in the FC BOP. This research may contribute to developing the necessary technology to eliminate or mitigate the probable design and process failures by increasing our understanding of a variety of failure sources occurring in the DMFC operation and by providing their potential solutions to thereby minimize the hazard associated with them.

2. Experimental

2.1. MEA preparations and performance measurements

The MEAs used in this work were prepared with Nafion 1135 (Dupont, USA). The catalyst slurries were prepared by Pt/Ru black and Pt black (both from HiSpec 1000, Johnson Matthey, UK) for the anode and cathode, respectively. The catalyst slurries were prepared by mixing the catalyst with Nafion ionomer solutions (Dupont, USA) and water. The catalyst inks were sprayed directly onto the GDL of each electrode. For the GDLs, commercial SGL carbon papers (21BC brand, Germany) were used as received without any treatment for both electrodes. The catalyst loading for both electrodes was 6 mg cm⁻² and the active area of the MEAs was 25 cm². To fabricate the MEAs, the catalyst-coated GDLs were hot-pressed onto a membrane.

All electrochemical experiments on the DMFCs were controlled by an FC testing system equipped with potentio-static/galvanostatic operational options (Won-A Tech, Korea). To measure the polarization curves, the anode and cathode were fed with 1.0 M methanol solution and dry air, respectively, at a constant flow rate of three times stoichiometry at 0.1 A cm⁻². The power density was determined from the current—voltage polarization curves at 70 °C. The design of the bipolar plates was triple-pass, serpentine flow-channels (POCO, USA) on each side of the electrodes.

2.2. Failure mode and effect analysis (FMEA) in DMFCs

The high-temperature storage test was performed to determine the effect on DMFCs at elevated temperatures (60 °C) without the application of any electrical stress. Three MEAs were tested to determine the temporal effect of long-term storage at elevated temperatures. The environmental stress chamber (AR series, ESPEC, Japan) was heated to 60 °C and kept at 60 °C for 10 h, 24 h, and 48 h. Before and after high-temperature storage, the MEA performance was evaluated by current—voltage polarization curves at 70 °C.

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