ELSEVIER

Feeding PEM fuel cells

A.A. Kulikovsky[a,1], A. Kucernak[b], A.A. Kornyshev[b,*,1]

[a] *Institute for Materials and Processes in Energy Systems, Research Center Juelich, D-52425 Juelich, Germany*
[b] *Department of Chemistry, Faculty of Physical Sciences, Imperial College London, SW7 2AZ London, UK*

Abstract

This paper discusses the laws of the fuel consumption in a polymer electrolyte fuel cell and its related performance. Based on simplifying but realistic approximations these laws are presented in analytical form. We verify these laws with space resolved current–voltage measurements in a *single channel fuel cell*. The results give a clear picture of the inside-the-cell distribution of performance associated with reactant consumption. This picture helps to rationalize the modes of optimal fuelling and is lessening for design of the flow-fields.
© 2004 Elsevier Ltd. All rights reserved.

Keywords: Polymer electrolyte fuel cells; Fuelling; Oxygen utilisation; Flow-field

1. Introduction

Modern polymer electrolyte fuel cells (PEFC) and direct methanol fuel cells (DMFC), based on polymer electrolyte membranes [1] are seen as promising, highly efficient renewable electrical power generators for emission free vehicles, portable power sources for laptops or cellular phones, and power sources for small scale or remote residential applications [2,3].

As progress in production of efficient catalyst layers and membrane materials proceeds, more emphasis is placed on the issue of fuel distribution and consumption. Earlier work on fuel cells paid little attention to the utilisation of reactants, and fuel cells were run at high stoichiometry of feed gases. Under these conditions, the concentration of reactants varies little with position. Such a situation is unreasonable in any realistic fuel cell within which we would try to utilise as much of the reactants (hydrogen and oxygen) as possible in order to improve cell efficiency.

In PEFCs, the protons generated at the anode by hydrogen oxidation migrate through a film of a polymer electrolyte membrane towards the cathode. There, pairs of protons are consumed in recombination with two electrons and one oxygen atom to produce a water molecule. Both sets of reactants are supplied through flow-fields which are the macroscopic channels running along the current collector plates (Fig. 1). These plates are separated from the catalyst layers on the two sides of the membrane by backing layers made typically of electron conducting carbon cloth or paper that function as an electronic current collector whilst remaining porous so gas can diffuse through it. This is the most popular, standard geometry, although some other geometries have also been suggested [4,5]. The topology of the feed channels can vary from 'through'-channels, to dead-end channels where all the gas has to be consumed within the cell. Channels may have a single or parallel meander geometry [6], interdigitated geometry [7] or a structure of parallel meanders with differential gas pressure [8].

In the simplest case at constant inlet flux, ideal humidification of membrane, and negligible losses on hydrogen side, the lower the stoichiometric ratio on the cathode side, the more oxygen will be consumed close to the inlet and the

---

* Corresponding author. Tel.: +44 20 759 45786; fax: +44 20 759 45801.
  E-mail address: a.kornyshev@imperial.ac.uk (A.A. Kornyshev).
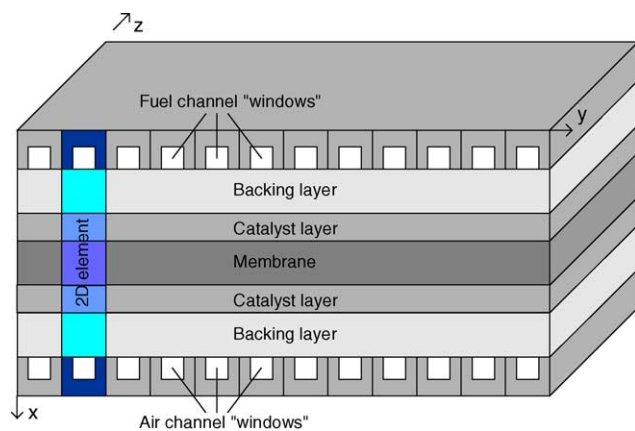[1] ISE member.

Fig. 1. Sketch of the PEFC cross-section.

less left to consume close to the outlet. This will result in a highly non-uniform current distribution—an aspect which is overlooked in most fuel cell literature in which *average* current densities are reported. If the downstream part of the channel is thus *starving*, then this later region of the cell will under-perform or not perform at all. Moving to a higher stoichiometry will lead to a more uniform reaction distribution, but this has the consequence of reducing the overall efficiency of the system. Thus there is a tension between two major design parameters—the overall reactant utilisation, and the current distribution—increasing the reactant utilisation seems to require greater non-uniformity of current distribution, and hence greater energetic losses within the fuel cell. Minimisation of energetic losses within the fuel cell requires higher stoichiometries which in turn lowers the overall fuel utilisation.

The above description of how reactant utilisation affects the profile of current distribution represents the simplest possible case. Of course, in a real fuel cell we have added complications. The main aspect, which complicates the situation is water management. The conductivity of the electrolyte is a growing function of its water content. High flow rates and currents, especially when the flow of reactant contains little water may dehydrate the membrane. This will result in a change in membrane conductivity with position along the channel. Of course, the counter case may also occur—an air flow may become so saturated that it is no longer able to abstract product water. This may then lead to flooding in the catalyst and backing layers and a dramatic reduction in local performance.

Whereas the starvation effect was well known, less known were the laws of fuel distribution along the gas supply channels, which have only recently became clearer due to a systematic set of experimental [9,10], theoretical [11,12] and computer modelling [13,11,14] investigations. Understanding these laws may help to establish the rationale behind a proper 'diet' for a fuel cell which optimises performance without sacrificing efficiency. In this paper we present experimental and theoretical study of the effects due to non-uniform

oxygen and local current distribution along the channel. We discuss how the obtained knowledge may help to rationalize the mode of fuelling or improve the flow-field design.

2. The laws of feed gas consumption

Stationary patterns of fuel distribution along the gas supplying channels and across the backing layers have different length scales. This allows the separate development of transport models for these two scales which may then be combined into one picture. Initially, we consider the most transparent case of negligible transport limitations in the backing layers, and then we consider more subtle effects due to such limitations.

2.1. The case of negligible gas diffusion limitations in the backing layer

2.1.1. Constant inlet flux
The simplest imaginable mass balance equation for feed gas concentration, c, in a linear channel is given by

$$\frac{d(vc)}{dz} = -\frac{S}{nFh} j(z) \tag{1}$$

Here z is the coordinate along the channel, h the channel depth, v the gas flow velocity, F the Faraday's constant, j the local current density, generated in the electrochemical reaction in which n electrons take part, S the number of molecules which consume or produce the n electrons in the reaction. Particular numbers are: (i) in hydrogen fuel cell, $n = 4$ and $S = 2$ for the hydrogen electrode (anode) and $S = 1$ for the oxygen electrode (cathode); (ii) in DMFC $n = 6$, $S = 1$ for the anode and $S = 3/2$ for the cathode. In this section we assume ideal transport of feed gas in the porous layers.

The reaction current itself depends on the oxygen concentration c and overpotential η

$$j = j_T(c, \eta) \tag{2}$$

The simplest approximation for this dependence is

$$j(c, \eta) = \left[\frac{c}{c_{ref}}\right]^\gamma f(\eta) \tag{3}$$

Here c_{ref} is a reference concentration to which the concentration of the feed gas scales, γ an empirical *effective order of reaction*. In the Tafel approximation $f(\eta)$ depends exponentially on η [15]

$$f(\eta) = i_* l \exp\left(\frac{\eta}{b}\right) \tag{4}$$

Here b is the Tafel slope, i_* the product of exchange current density and the volume density of the reaction surface, which has a dimensionality of current per unit volume; l the thickness of the catalyst layer (or a thickness of the 'slab' in the catalyst layer, if the latter is thick, where the reaction takes place [16]). Both the effective order of reaction and the 'Tafel