



The recovery mechanism of proton exchange membrane fuel cell in micro-current operation



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HIGHLIGHTS

- A new recovery mechanism in micro-current operation is proposed and validated.
- Fuel cell performance recovers by 40% after 20 hours' micro-current operation.
- Changes of membrane electrode assembly status shows recovery mechanism.
- Water content and proton conductivity show better characters in micro-current.
- A new method is provided to on-line performance recovery of fuel cell vehicles.

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ABSTRACT

In the rapid development of new energy technologies, fuel cells have exhibited their great potential for green, low-carbon applications. However, the deterioration of their performance after a certain time of use is considered as a serious handicap for their wide application. In this study, a micro-current working condition was found to be effective in recovering the performance of proton exchange membrane fuel cell. To investigate the mechanism behind, a series of experiments were conducted to study the fuel cell's performance recovery and its membrane electrode assembly parameters, and membrane water content and proton conductivity were simulated by a fuel cell model. The results showed that fuel cell saw a performance recovery by 40% after 20 hours' micro-current operation. The reduced hydrogen crossover and ohmic resistance, and the increased electrochemical active surface area should explain the recovered performance of the fuel cell. In addition, the higher, more uniform membrane water content and proton conductivity found in simulation results may also contribute to fuel cell's performance recovery. This study proposes a new recovery mechanism for proton exchange membrane fuel cell and offers plausible explanations, which is a new addition to fuel cell theory and provides a theoretical basis for on-line performance recovery of fuel cells.

1. Introduction

Hydrogen is the most promising renewable energy for future power generation systems. Fuel cell technology, which can efficiently generate electricity using hydrogen as fuel, has attracted widespread attention in recent years. The proton exchange membrane fuel cells (PEMFCs), can greatly reduce the burning of fossil fuels and emission of carbon dioxide. The pollution-free production of energy [1], high power density [2] and high energy conversion efficiency [3] of PEMFC make fuel cells a most promising candidate for future energy industry. Staffell et al. [4] reported that fuel cell power conversion efficiency was much higher (up to 60%) compared with traditional internal combustion engines.

Despite PEMFC vehicles' great potential for development, there are still some major obstacles, such as durability and cost, which must be resolved [5]. Many researches have been conducted to investigate the mechanism of deterioration for fuel cells. Panha et al. [6] and Pei et al. [7] announced that frequent changes of vehicle load, starting, stopping and idling all contribute to accelerating fuel cell aging. Pei et al. [8] reported the inconsistent parameters of single cells in cell stacks also seriously affected their durability. In addition, membrane electrode assembly (MEA) conditions could also lead to fuel cell aging, for example, dissolution [9,10] and contamination [11] of MEA, agglomeration or the specific types of catalysts used [12]. Among these influencing factors, the MEA humidification condition is an important

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Nomenclature

List of abbreviations

PEMFC	proton exchange membrane fuel cell
MEA	membrane electrode assembly
OCV	open circuit voltage
ECSA	electrochemical active surface area
F	fluorine
S	sulfur
SLPM	standard liter per minute
T	temperature
P	pressure
RH	relative humidity
A	anode
C	cathode
EOD	electro-osmotic drag

Variables

Φ_w^{EOD}	water flux caused by EOD
n_d	EOD coefficient
I_{ion}	ionic current density
λ_{nf}	non-frozen water content
Φ_w^{diff}	water diffusion non-frozen water flux
ρ_f	dry membrane density
M_m	equivalent weight
D_w	diffusion coefficient
Φ_w	water transport flux
a	water activity
P_{sat}	saturated vapor pressure of water vapor
S_{lq}	volume fraction of liquid water
w	MEA water content
X_{vp}	mole fraction of water vapor in the pore regions
P_g	pressure of the gas mixture in pore regions

Parameters of PEMFC model

L_c	cathode channel length
$C_{w,c}$	cathode channel width
$C_{d,c}$	cathode channel depth
L_a	anode channel length

$C_{w,a}$	anode channel width
$C_{d,a}$	anode channel depth
$C_{l,c}$	cathode ridge width
$C_{l,a}$	anode ridge width
$\delta_{pl,c}$	cathode collector plate thickness
$\delta_{pl,a}$	anode collector plate thickness
$\delta_{\text{diff},c}$	cathode diffusion layer thickness
$\delta_{\text{diff},a}$	anode diffusion layer thickness
$\delta_{\text{cata},c}$	cathode catalytic layer thickness
$\delta_{\text{cata},a}$	anode catalytic layer thickness
δ_{mem}	membrane thickness
A	active area
$\varepsilon_{\text{diff},c}$	cathodic diffusion layer porosity
$\varepsilon_{\text{diff},a}$	anodic diffusion layer porosity
$\varepsilon_{\text{cata},c}$	cathodic catalytic layer porosity
$\varepsilon_{\text{cata},a}$	anode catalytic layer porosity
ε_{mem}	membrane porosity
$K_{\text{diff},c}$	cathodic diffusion layer permeability
$K_{\text{diff},a}$	anodic diffusion layer permeability
$K_{\text{cata},c}$	cathodic catalytic layer permeability
$K_{\text{cata},a}$	anodic catalytic layer permeability
K_{mem}	membrane permeability
D_{diff}	diffusion layer conductivity
D_{cata}	catalytic layer conductivity
D_{pl}	collector plate conductivity
ζ	membrane conductivity
$i_{c,\text{ref}}$	cathode exchange current density
$i_{a,\text{ref}}$	anode exchange current density
α_c	cathode exchange coefficient
α_a	anode exchange coefficient
γ_c	oxygen concentration index
γ_a	hydrogen concentration index
V_{oc}	open circuit voltage
T_{in}	cooling water temperature
$P_{in,c}$	air inlet pressure
$P_{in,a}$	hydrogen inlet pressure
λ_c	air stoichiometric ratios
λ_a	hydrogen stoichiometric ratios
RH_c	relative humidity of air inlet
RH_a	relative humidity of hydrogen inlet

factor. Jeon et al. [13] investigated the humidification shortage of reaction gases would accelerate the membrane electrolyte degradation, leading to membrane damage and gas permeation. Knights et al. [14] proposed that excessive humidification could make catalyst agglomerates to occur more easily, reducing active area.

There have been many studies in the literature which were conducted to recover PEMFC performance. Shi et al. [15] proposed a recovery method to treat air contaminated PEMFCs. Air purges are proposed by Mohtadi et al. [16] to reactivate fuel cell catalysts and a 70 hours' air purge could recover 20% of fuel cell's initial activity. Colbow et al. [17] reported periodic reactant starvation could lead to potential changes in the starved electrode, which helps remove electrocatalyst poisons and recover fuel cell performance. Urdampilleta et al. [18] took 3 hours to recover fuel cell performance partially by exposing cell to open circuit voltage (OCV). A method to improve fuel cell electrical performance on catalyst-containing cathode was proposed by Gould et al. [19]. Moreover, many PEMFC activation methods have also been proposed and studied, such as load control or temperature control and hydrogen evolution [20].

In addition to the recovery methods mentioned above, we found that micro-current conditions in fuel cell system when it switched from

standby state to idle speed could also help recover PEMFC performance. A standby state is a working condition in which a fuel cell stack can be started up at any time with OCV but no current output; Running at idle speed means that a fuel cell stack outputs a small current to maintain power requirements of auxiliary system, and the fuel cell system has no output power. PEMFC performance recovery by micro-current condition is applicable to both aged fuel cells and normal ones. Before a new fuel cell can be used, it also needs initial activation to improve performance. Therefore, MEA is wetted by water vapor in reaction gas and water produced by reactions, resulting in better proton conductivity, smaller resistance [21] and larger platinum electrochemical active surface area (ECSA) [22]. Xu et al. [23] reported that increasing the number of transport channels of reactive gases, electrons, protons and water could help improve the performance of fuel cells.

In this paper, the recovery mechanism of PEMFC in micro-current operation was probed. Tests on fuel cells were carried out to investigate micro-current's influence on polarization curves and MEA parameters. In addition, membrane water content and proton conductivity were also studied in simulations using validated model to investigate micro-current recovery mechanism.

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