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Effects of gas composition just before fuel pulses on power generation characteristics at different fuel supply frequency in Pulse Jet-Rechargeable Direct Carbon Fuel Cells

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INTRODUCTION

Rechargeable Direct Carbon Fuel Cells (RDCFCs) in which the solid carbon deposited on the anode by thermal decomposition of hydrocarbon is used as fuel, have been proposed and researched by our group [1-4]. One application of RDCFCs is a Pulse Jet-Rechargeable Direct Carbon Fuel Cell (PJ-RDCFC) in which small amounts of a liquid hydrocarbon, typically isooctane, are supplied via pulse jet [5]. Because power density can be maintained by repeated charging and power generation at intervals of several seconds, continuous operation is realized, thus making PJ-RDCFCs suitable as mobile power sources such as for vehicles. PJ-RDCFCs are expected to have high energy conversion efficiency and fuel utilization because not only solid carbon but also hydrogen, methane and other hydrocarbons generated by isooctane decomposition can be used as fuel. Furthermore, because solid carbon can be removed frequently in PJ-RDCFCs via the electrochemical reaction with oxide ions, degradation of the anode is expected to be prevented. Operating conditions of PJ-RDCFCs can be controlled by adjusting the interval time of the pulse jet (T_{int}), amount of a single pulse (PJ_{vol}) and the current density (j). Our group previously suggested that the chemical species contributing to an electrochemical reaction can be controlled, suggesting that the gas composition just before the onset of each jet pulse depends on the operating conditions [5]. However, neither the anode reactions nor its relationship with the power generation characteristics has been evaluated.

In this study, power generation characteristics of PJ-RDCFCs were measured over a wide range of fuel supply frequency, and the relationship between gas composition just before fuel supplying and anode reactions were investigated to determine the optimum operating conditions.

EXPERIMENTAL

In the PJ-RDCFC used here, the electrolyte was an YSZ disk (8 mol% Y_2O_3 –stabilized 1 mol% ZrO_2, 0.25 mm thickness, 20 mm diameter), the anode was a Ni / $Gd_{0.10}Ce_{0.90}O_{2-\delta}$ composite, and the cathode was a $La_{0.8}Sr_{0.2}MnO_3$ / ScSZ composite. The operating temperature was 900ºC and oxygen was introduced to the cathode at STP 60 ccm. After 3 jet-pulses of isooctane followed by a 1-min wait, power generation at constant j was initiated. Charging was again carried out by pulse jetting at fixed T_{int}. After two or more generation/charging cycles, pulse jetting was stopped, and power generation was considered done when terminal voltage V reached 0 V. First, a power generation test at extremely high fuel supply frequency f_{fs} (1 pulse/sec) was conducted at j = 800 mA/cm^2, PJ_{vol} = 0.72 μl. Then, power generation tests at different T_{int} (136, 102, 68, 34 sec) were performed at j = 100 mA/cm^2 and PJ_{vol} = 1.20 μl. Finally, a current interruption test was carried out to separate V into electromotive force (EMF), overvoltage, and IR loss.

RESULTS AND DISCUSSION

Figure 1 shows V as a function of time at j = 800 mA / cm^2, and at f_{fs} = 1 pulse/sec. During quasi continuous fuel supplying (0-124 sec), V was stable. The average power density reached 445 mW / cm^2, which is the highest reported performance of a PJ-RDCFC.

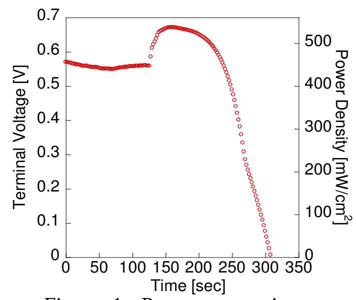

Figure 1. Power generation characteristics of a PJ-RDCFC (j = 800 mA/cm^2, PJ_{vol} = 0.72 μl, T_{int} = 1 sec)

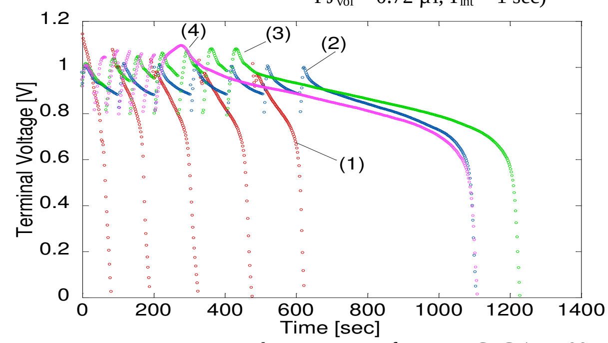

Figure 2. Power generation characteristics of a PJ-RDCFC (j = 100 mA/cm^2, PJ_{vol} = 1.20 μl, T_{int} = (1)136 sec, (2)102 sec, (3)68 sec, (4)34 sec)

Figure 2 shows V as a function of time at different T_{int}, and Figure 3 shows the last cycle in Fig. 2. The maximum V increased with increasing f_{fs}. In general, V is determined by EMF, overvoltage, and IR loss. The current interruption test separating V revealed that EMF is dominant at low current density, namely, 100 mA/cm^2. Because fuel utilization is low at high f_{fs}, the partial pressure of O_2 just before the fuel supplying was small. Therefore, EMF became large and thus affected the maximum V.

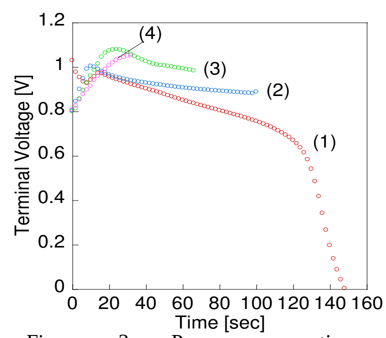

Figure 3. Power generation characteristics for a single cycle of a PJ-RDCFC (j = 100 mA/cm^2, PJ_{vol} = 1.20 μl, T_{int} = (1)136 sec, (2)102 sec, (3)68 sec, (4)34 sec)

Fig. 2 also shows that the increase rate of V depends on T_{int}. This increase in V might be caused by the generation of H_2 or CO, which in turn causes a decrease in overvoltage. Because fuel utilization is high at low f_{fs}, the partial pressure of H_2O and CO_2 is high near the anode just before fuel supplying and these two chemical species promote reforming reactions that generate H_2 or CO after fuel is supplied.

In conclusion, our study achieved the highest power density of a PJ-RDCFC, 445 mW / cm^2. Furthermore, because both the maximum V and the increase rate of V depend on the fuel utilization, the fuel utilization determines the partial pressure of H_2O just before fuel pulses.

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