



Performance enhancement of liquid antimony anode fuel cell by in-situ electrochemical assisted oxidation process



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ABSTRACT

Liquid Sb anode has been intensively studied for its capability to convert various solid carbon fuels into electricity. Performance of anode is one of the most important issues. In present research, performance of liquid Sb anode SOFC is promoted by an “assisted oxidation process”: fuel cell achieved larger current by electrochemically introducing extra amount of Sb_2O_3 into the anode region. By revealing the interfacial structure between anode and electrolyte, the mechanism of performance promotion is then explained by numerical model from a microstructural point of view: the oxidation process built ionic transporting pathways and extended the electrochemical reactive sites in liquid Sb anode. With the help of extended electrochemical reactive sites, the fuel cell reaches a better performance. The “assisted oxidation process” can be taken as an in-situ method to refine the structure of liquid Sb anode.

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1. Introduction

Direct carbon fuel cell is a promising power technology which converts chemical energy in solid carbon fuel directly into power. Due to its satisfactory energy efficiency and low level of emission, research of direct carbon fuel cell has been reported frequently [1–3]. Among various designs of anodes to achieve direct carbon conversion [4–6], liquid antimony (Sb) anode has drawn great attention from a number of researchers. Because of its stability of performance [7–9] and feasibility of fuel [9–11], liquid Sb anode has been systematically investigated. With metallic Sb itself being a good electronic conductor, current collection for anode can be largely simplified, solid oxide fuel cell with a liquid Sb anode is considered to be a promising technology for scaling up. Several batch scale demonstrations of tubular liquid Sb anode direct carbon SOFCs [10,12,13] have been reported. Another advantage of liquid Sb anode is that liquid Sb, apart from being carbon converting electrode, can also serve as an energy storage media. For the metallic liquid Sb would be electrochemically oxidized to maintain fuel cell performance when there is a scarce of fuel [14]. This operation condition without fuel adding is regarded as “battery

mode” of liquid Sb anode. The only reaction takes place during “battery mode” operation is electrochemical oxidation of metallic Sb to form its oxide (Sb_2O_3), so “battery mode” operation is a reliable way to investigate metal reaction kinetics and metal/metal oxide transport in the anode.

For liquid Sb anode associated with solid state electrolyte, the anode-electrolyte interface is of great importance because the oxygen ions transported through solid oxide electrolyte would oxidize metal in anode at this very place [15]. When carbon fuel is supplied to the interface, a high OCP (Open Circuit Potential) related with carbon oxidation can be observed [16]. Intensive research has been carried out to investigate metal oxidation behavior at the interface: Wang et al. [17] have revealed the influence of surface roughness on anode polarization; Zhou et al. [18] and Jayakumar et al. [19] have studied the interaction between metal oxide and solid state electrolyte during long term operation of the fuel cell. In order to investigate the electrochemical oxidation kinetics of metallic material in anode, during which long term issues of SOFC is no longer the dominating problem, bottom type fuel cells are often fabricated and tested [7,20] and regarded as a standardized procedure [8]. To focus on electrochemical oxidation of metal, as previously mentioned, electrochemical tests are often carried out under “battery mode” with anode chamber shielded in inert atmosphere without fuel fed. And during the test of liquid Sb

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anode, discharging fuel cell at a static working potential has become an approach to investigate the evolution of anode performance during fuel cell operation [18,21]. When a given potential is applied to the fuel cell, current of the fuel cell is recorded and various testing results have shown that the current density rises at beginning of the test, then the fuel cell performance reaches a steady state, and at last the current value decreases [21,22].

This phenomena was reported and explained in details in our previous work [22]. The profile of the constant potential discharge current curve is mainly attributed to the ionic conductivity of antimony oxide (Sb_2O_3) and microstructure of liquid Sb anode itself. The ionic conducting Sb_2O_3 [23] generated during fuel cell operation would form ionic conducting pathways in the anode. These pathways, starting from anode-electrolyte interface reaching through the liquid Sb anode, closely contacts with electronic conducting metallic Sb. Then electrochemical oxidation of metallic Sb can take place at the newly formed interface between metallic Sb and Sb_2O_3 . Therefore, generation and migration of Sb_2O_3 leads to an enhancement and extension of electrochemical reactive area in the anode. The formation of electrochemical reactive area in liquid Sb anode is significant enough to promote fuel cell performance.

Therefore, by introducing Sb_2O_3 properly into anode system, one can increase the density of electrochemical reactive sites in liquid Sb anode. In present study, extra amount of Sb_2O_3 is introduced in to the anode reaction system by an “assisted oxidation process” during fuel cell operation, and an increase in density of electrochemical reactive sites in the anode region has led to a more satisfactory fuel cell performance.

2. Experimental procedures

2.1. Fuel cell preparation

Fuel cell in this research was prepared base on a cylinder shaped 8YSZ crucible (Hengnai Ceramic Technology Co., Ltd., Shanghai, China). The crucible, with an outer diameter of 20 mm, inner diameter 16 mm and a height of 15 mm, was machined to serve as the container of liquid antimony anode. And with a flat bottom of 500 μm thick, strong enough to hold the anode, the crucible can also play the role of electrolyte. Platinum paste (MC-Pt100, Grikon Advanced Materials Limited Company, China) was screen printed onto outer surface of crucible bottom and sintered in air at 900 $^\circ\text{C}$ for 2 h. The well prepared reactive area of cathode was 16 mm in diameter and 15 μm in thickness.

The half-cell was then sealed onto one end of a vertical alumina tube with ceramic bond (Aremeco Ceramabond 552, Aremco Products Inc., NY, USA). An alumina plate, pulled by 3 springs, held a platinum mesh against the cathode for current collection. A rhenium (Re) wire (99.97%, 0.5 mm diameter, Alfa Aesar) was employed as current collector of anode. A K-type thermocouple was placed right next to the fuel cell as a temperature monitor. The details of test configuration is presented as Fig. 1.

The test setup was then heated up and held at 800 $^\circ\text{C}$ by a tubular furnace with anode chamber shielded in argon, and when the system reached steady state, 10 g of antimony powder (99.5 wt %, Sinopharm Chemical Reagent Co., Ltd., China) was introduced into anode chamber. The Sb powder then melt under the given temperature to form liquid Sb anode. Unlike feeding the anode under room temperature, this in-situ formation of antimony anode at elevated temperature avoid possible oxidation and volatilization of metallic antimony during heating process, and maintain the mass and purity of this metallic Sb anode.

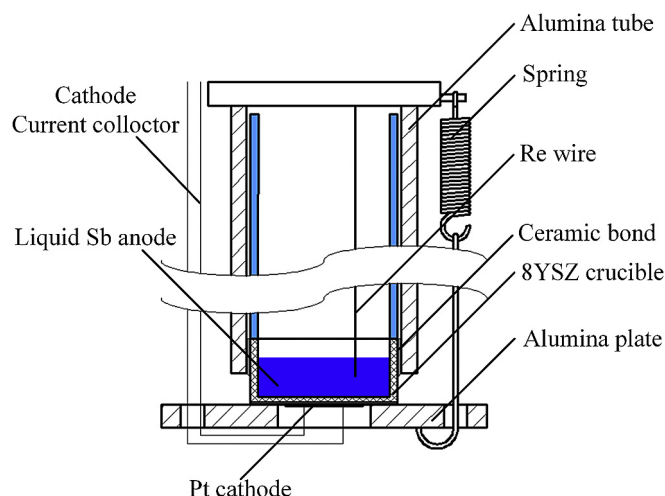


Fig. 1. Schematic of bottom cell test system.

2.2. Electrochemical test

Electrochemical measurements were taken by an electrochemical working station (Reference 3000™ Gamry Instruments Inc., USA) with the SOFC held at 800 $^\circ\text{C}$. And extra amount of Sb_2O_3 was introduced into anode system by “assisted oxidation process”. The working potential applied to the fuel cell consists of two periods: assisted oxidation period, which operates the fuel cell at a low potential (-0.1 V), and discharge period, which discharge the fuel cell at a constant potential of 0.3 V. Profile of working potential mentioned above is shown in Fig. 2. The signal was added to fuel cell by electrochemical working station.

Metal Sb in anode region is oxidized via electrochemical reaction shown in Equation (2.1).



During assisted oxidation period, potential as low as -0.1 V was applied to the fuel cell, this ultra-low potential led to large current drawn from the fuel cell and fast oxidation of metallic Sb, introduced extra amount of Sb_2O_3 into anode system. In this period, just as its name implies, the electrochemical working station assists the oxidation of metallic Sb. Then the working potential rose to 0.3 V to discharge the cell for 30min. Current values were recorded

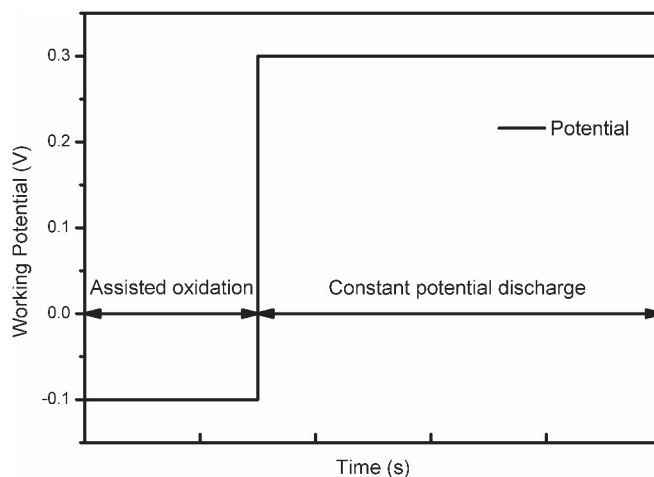


Fig. 2. Profile of step signal applied to the fuel cell.

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