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Liquid antimony-silver alloys as anodes for direct carbon solid oxide fuel cells

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HIGHLIGHTS

- The Sb-Ag anode produced comparative performance to the pure Sb anode.
- The Sb-Ag anode produced higher power densities due to an activation process.
- A quasi layered-structure was formed in anode at working mode.
- Ag appeared a concentration gradient from low to high along gravity.

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ABSTRACT

Sb_{1-x}-Ag_x (x = 0.1, 0.2, 0.3) alloys, with a lower melting point and a higher density than Sb, are used as anode for tubular 8 mol% yttria stabilized zirconia supported direct carbon solid oxide fuel cells which are prepared through a slurry-casting and co-sintering method. The initial performance of alloy anode cells is lower than that of Sb anode cell, indicating that the addition of Ag has negative effects on the electrochemical reaction between Sb and oxygen ions. After running at different current density from small value until 0.5 A cm⁻² for several hours at 750–800 °C, the final performance is very close to that of Sb anode cell. The energy dispersive spectroscopy results and scanning electron microscopy examination of tested cells reveal that a quasi-layered structure instead of a uniform distribution of Sb and Ag is formed in the anode. The Sb and Ag inter-diffusion between carbon-reduced Sb phase and Sb-Ag solution phase is not fast enough to form a uniform phase, thus the original uniformly distributed Sb-Ag solution is gradually replaced by a gradient distribution in which the Ag concentration is from low to high along the gravity direction.

1. Introduction

Electricity has been the most popular energy source for human society because it is clean, quiet and easy to be transmitted and stored. Unfortunately, most of the electricity is generated by thermal power plants, where fossil fuels are burned, producing significant amount of pollutants into the atmosphere, such as particulates, CO₂, CO, NO_x, and SO_x, which cause global warming and damages in human health, agriculture and creature system. In order to ease the present conflict between energy demands and environmental degradation, global effort on reducing extensive consumption of fossil energy must be increased by several orders of magnitude through development of high-efficiency energy conversion technologies. Solid oxide fuel cell (SOFC) is one of

such technologies for directly conversion of the chemical energy stored in fuels into electricity [1,2]. The fuels can be hydrogen, natural gas, biomass, fossil fuel derivatives and any other hydrocarbons. Direct-carbon SOFC (DC-SOFC) directly utilizes solid carbonaceous fuels for electricity generation and has received increasing attention due to its potential high efficiency [3,4].

Conventional SOFC with a porous cermet anode is not suitable for directly utilizing solid carbonaceous fuels, because the solid fuel particles have only limited and impermanent contact with the electrolyte that transports oxygen ions [5,6], so that the cell performance can't be maintained at a reasonable level for long time. In order to improve cell performance and durability, Gür et al. suggested two approaches to ensure the contact between the fuel and electrolyte [7]. One way is to

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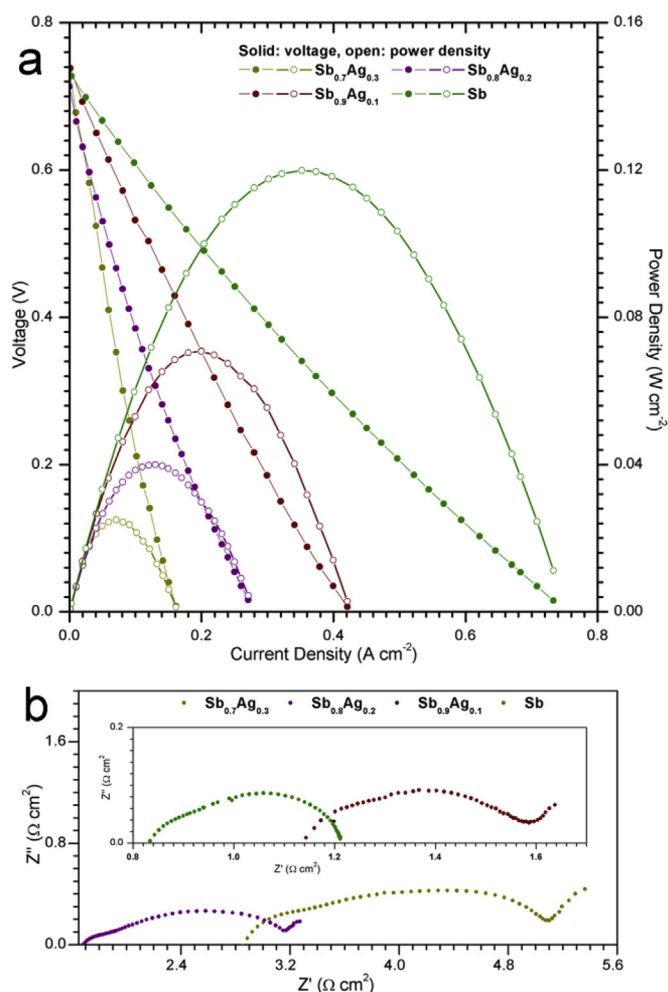


Fig. 1. Initial performance of cells with $\text{Sb}_{1-x}\text{Ag}_x$ ($x = 0.3, 0.2, 0.1, 0$) anodes at 750 °C: (a) I-V and I-P curves and (b) EIS.

gasify the fuel (fluid-bed SOFC) by oxidation gases, such as CO_2 [8,9] and H_2O [10,11]; another way is to mix solid fuels with molten carbonate (hybrid SOFC) [12,13]. Different from the above approaches, the newly developed DC-SOFC uses liquid metal as the anode. The liquid metal is electrochemically oxidized to metal oxide by the oxygen ions transported from the electrolyte and the metal oxide is then chemically reduced by the solid carbonaceous fuel [14–16]. Tin (Sn) was the first low-melting-point metal used as anode for DC-SOFC [17,18]. In fact, by using liquid Sn as anode, other types of fuels, such as JP-8 kerosene, methane, coal, biomass and plastics, also can be directly utilized for electricity generation [19,20], indicating liquid tin anode has more extensive fuel applicability than cermet anode. The Nernst potential corresponding to reaction between Sn and O_2 (air atmosphere) at 700 °C is 0.93 V, so if the working voltage is below this threshold value, the electrochemical reaction between Sn and oxygen ions will happen. What's more, the energy stored in metal Sn endows an extra battery mode for liquid tin anode SOFC. One critical issue for tin anode is the high melting point of electrochemically generated SnO_2 . According to previous reports, after several hours' test, a layer of SnO_2 formed at the electrolyte/Sn interface and led to an increasing resistance and performance degradation [21]. Indium (In), plumbum (Pb), and bismuth (Bi) were another three low-melting-point metals that had been used as the anode for SOFCs [22]. At 700 °C, the open circuit potential (OCP), which is also called threshold voltage, established between these metals and air atmosphere was 1.03 V, 0.65 V, and 0.45 V, respectively. Besides, Fe and Ag also had been used as electrode

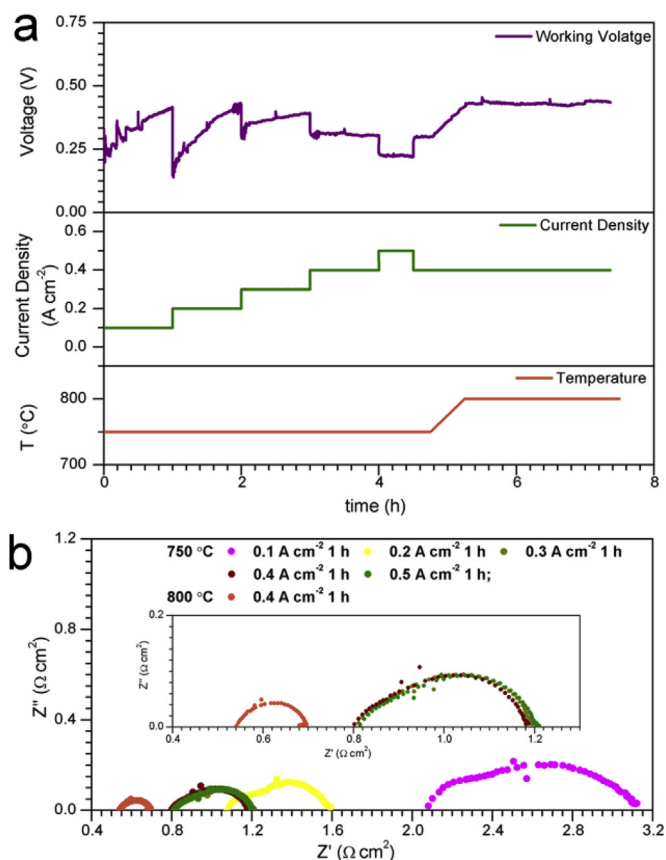


Fig. 2. Performance of cell with $\text{Sb}_{0.7}\text{Ag}_{0.3}$ alloy anode at 750–800 °C: (a) cell voltage, current and temperature; (b) EIS.

materials for solid oxide cells [23,24]. Javaddekar et al. used molten Ag as electrode for direct-carbon SOFC, they suggested that the diffusion of oxygen ions within the electrode is likely to be the limiting factor in controlling the performance [25]. Worth of attention, carbon was not used as reductant but directly reacted with oxygen ions in their proposal.

The melting point of antimony oxide (Sb_2O_3) is 656 °C, which is much lower than that of SnO_2 . At working temperature above 656 °C, liquid instead of solid Sb_2O_3 is generated in contacted with the electrolyte, and a hierarchical structure of Sb and Sb_2O_3 is formed in the anode, which was driven by the density difference between them, as reported in our previous works [26]. Liquid Sb_2O_3 layer between electrolyte and liquid Sb is moveable instead of forming a thickness-increasing solid layer, so Sb anode DC-SOFC has drawn more and more attention. Furthermore, preliminary research about antimony-based alloy anode was reported by Jayakumar et al. [27]. They suggested that the characteristics of alloy anode are similar to that of the easier oxidized metal except for two activity-approached metals. Different atoms are competing to participate in the electrochemical reaction with oxygen ions in order of the chemical potential from low to high. The metal oxidation reactions of alloy anode might also relate with threshold voltage and working voltage. The reaction between a metal and oxygen ions will happen only if the working voltage is smaller than its threshold voltage.

Alloying Sb with another high density metal can increase the density of metal phase and thus promote the mobility of Sb_2O_3 . Adding Ag into Sb will increase the density of metal phase and decrease the melting point of anode. Since silver oxide decompose to silver at DC-SOFC working temperature, Ag would not participate in the reaction between oxygen ions and metal atoms and keep staying in metal phase.

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