

## Anodic reaction of dense Pd film in SOFC using Y-doped BaCeO<sub>3</sub>— Effect of mixed ionic conduction of electrolyte

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### ABSTRACT

The anodic reaction of a dense Pd film in a solid oxide fuel cell (SOFC) was investigated by using BaCe<sub>0.8</sub>Y<sub>0.2</sub>O<sub>3-α</sub> (BCY) as an electrolyte. The anode performance of the film was better than the anode performances of porous Pd and Pt at 873 K. Although protons were predominant conduction in the porous Pt anode, oxide ion conduction and hole conduction slightly occurred in it at 873 K. In contrast, in the dense Pd film anode, small oxide ion conduction was compensated by hole conduction at 873 K. Hence, proton conduction occurred predominantly and hole conduction occurred to a small extent in it. However, at 1073 K, its performance decreased considerably because partial oxide ion conduction could not be inhibited by the dense Pd film. At the same temperature, conduction of oxide ions of BCY occurred predominantly in the porous Pt anode.

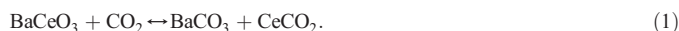
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### 1. Introduction

Mugikura and Asano introduced a performance equation that can be used to analyze basic performance factors and calculate the operating characteristics of various types of fuel cells (PEFCs, PAFCs, MCFCs, and SOFCs) [1]. They also speculated that it might be possible to fabricate proton-conducting fuel cells that would have similar performance to high-temperature fuel cells in an intermediate-temperature range of 523–873 K.

It is known that proton conduction occurs in BaCeO<sub>3</sub> in a H<sub>2</sub>-containing atmosphere [2–4]. Since the activation energy of proton conduction in this oxide is small, it exhibits high proton conductivity at low temperatures. Therefore, it may be used as an electrolyte in an intermediate-temperature SOFC (IT-SOFC).

However, in a CO<sub>2</sub>-containing atmosphere, it does not always possess sufficient chemical stability [5–7] and loses its proton conductivity. Thermodynamically, it easily decomposes into CeO<sub>2</sub> and BaCO<sub>3</sub>, according to the following reaction:



The brittleness of BCY induced by CO<sub>2</sub> can be reduced by depositing a dense Pd film on a BCY substrate that is used as an anode in an

SOFC, under fuel cell conditions [8]; the Pd film functions as an anode, despite its high density. However, BCY partly functions as an oxide ion conductor as the temperature increases. Therefore, it is assumed that oxide ion conduction in BCY mainly affects the anodic reaction of the dense Pd film that exhibits hydrogen permeability. In this study, we evaluated the influence of the mixed

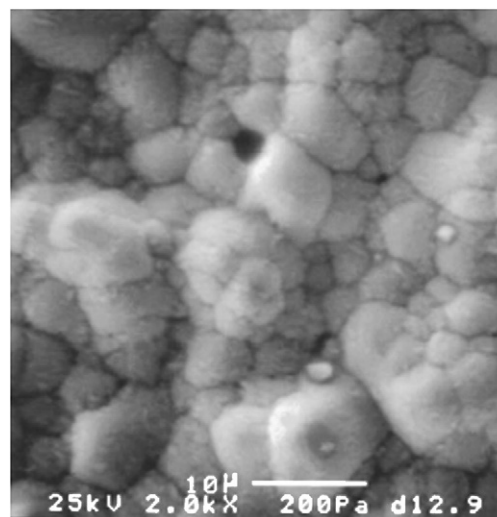


Fig. 1. SEM image of surface of dense Pd film after electroless plating and heat treatment.

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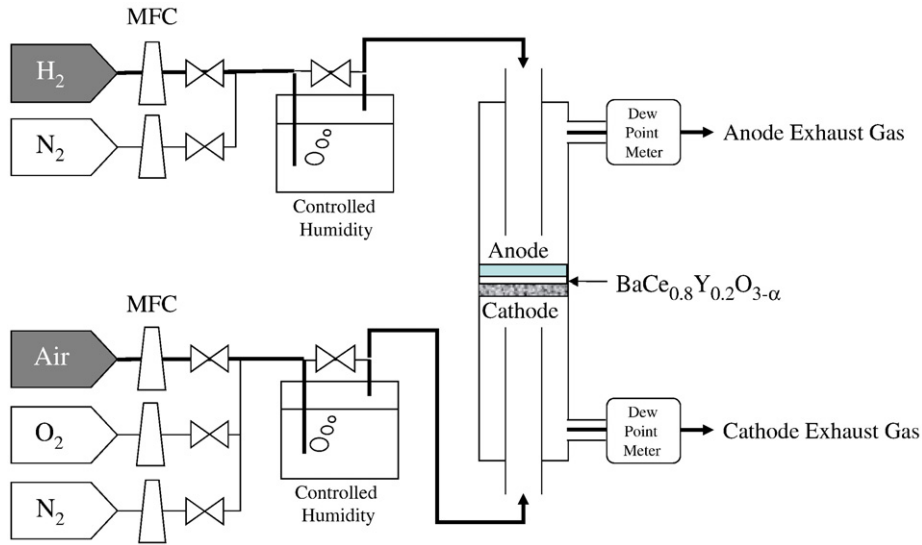


Fig. 2. Schematic illustration of test cell.

ionic behavior of BCY during the anodic reaction of a dense Pd film in an SOFC.

2. Experimental

A dense Pd film anode with a diameter of 13 mm was deposited onto a BCY substrate (TYK, Japan, diameter: 17 mm, thickness: 0.6 mm) by electroless plating and subsequently subjected to heat treatment at 1123 K. The dense Pd film is shown in Fig. 1; its thickness was approximately 5 μ using Ag paste, a porous cathode was attached to the substrate at a position facing the anode.

Fig. 2 shows the experimental apparatus. To eliminate the ohmic drop of the Pt wires from the cell, we attached two Pt-lead wires to each electrode; one of them was used to measure current, and the other was used to measure voltage. Each electrode compartment was sealed by a Pyrex glass ring gasket. A Pt wire was wound around the side of the BCY substrate as a reference electrode. The cells were discharged under fuel cell conditions, and their performances were

examined by measuring the terminal voltage for any discharge current.

The anodic polarization of a dense Pd film has been examined by the current interruption method [9]. A current pulse generator (Nikko Keisoku, NCPG-101, Japan) was used as an electric load and a pulse source. An electrometer (Hokuto Denko, HC-104) with a high input resistance ( $10^{11} \Omega$ ) was connected to the voltage terminals. The potential pulse measured by the electrometer was observed by using an oscilloscope (Agilent Technologies, DSO3152A, USA), and the ohmic drop was also observed by the current interruption method.

The proton and oxide ion currents in the electrolyte during the discharge of the cell can be determined from the rates at which water vapor was formed at the cathode and anode, respectively. These rates were determined by measuring the humidity and flow rate of the exhaust gas from each electrode compartment. A dew point meter (Shinyei Co., HygroFlex, Japan) was used for the measurement of humidity. From the formation rates of water vapor, the ratio of each ionic current to the total current, namely, the ionic transport number,

$$t_{H^+} = i_{H^+} / i_{total} \quad t_{O^{2-}} = i_{O^{2-}} / i_{total} \quad (2)$$

in the specimen was calculated. Here,  $i_{H^+}$  and  $i_{O^{2-}}$  are the ionic currents determined from the formation rates of water vapor at the

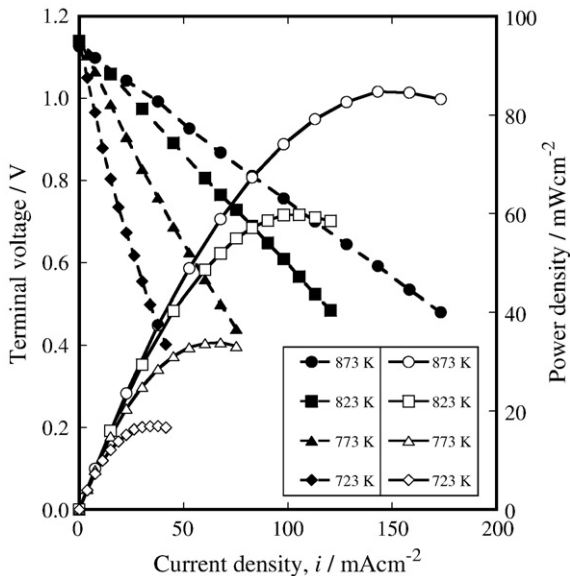


Fig. 3. Typical discharge characteristics of IT-SOFC; H<sub>2</sub>, dense Pd film |BCY| porous Ag, air. Closed symbols: IV properties, open symbols: power densities.

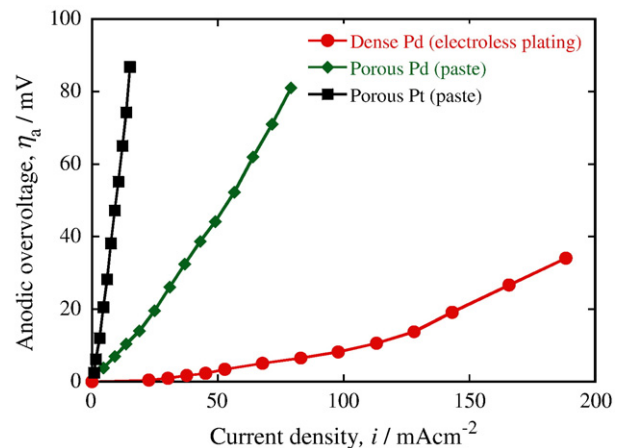


Fig. 4. Anodic overvoltages of dense Pd film, porous Pd, and porous Pt at 873 K. The cathode is made of porous Ag.

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