

Operando observation of electrode reactions in a solid oxide fuel cell by an environmental high-voltage electron microscope

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Electrode reactions play important roles in a solid oxide fuel cell (SOFC) which transforms the chemical energy to the electrical energy through electro-chemical reactions. The reactions in electrode/solid-electrolyte interfaces, however, are complicated due to combinations of various phenomena such as a redox reaction and ion conduction. In order to understand electrode reactions directly, we have developed a dedicated specimen holder for *operando* observation of SOFC in an environmental high-voltage electron microscope (E-HVEM) [1]. In this study, we will report oxygen distribution around an electrode/solid-electrolyte interface in a SOFC during operation by using electron energy loss spectroscopy (EELS).

The E-HVEM used in this experiment is a JEM-1000K RS (JEOL) equipped with an equivalent Gatan Image Filter Quantum [2]. The E-HVEM was operated in two modes of transmission electron microscopy (TEM) and scanning transmission electron microscopy (STEM) at the acceleration voltage of 1000 kV. The dedicated specimen holder can heat and apply the external voltage to the specimen [1]. The cross-sectional specimen has a SOFC's structure, is prepared by a focused ion beam technique. We made the SOFC's structure which has platinum electrodes and a solid electrolyte (Yttria-Stabilized Zirconia: YSZ), using a pulsed laser deposition method. Oxygen distribution images were reconstructed from the data cube including O K-edge EEL spectrum obtained by STEM-EELS. Observation conditions were as follows. A specimen temperature, an applying voltage for the specimen and a pressure in the specimen chamber were 673 K, 0-1 V and 0.5 Pa (O₂ atmosphere), respectively.

Figure 1(a) shows a low-magnification TEM image of the SOFC specimen. We applied the external voltage to the platinum electrode of the upper side in Fig. 1(a). Figure 1(b)(c) show the oxygen distribution around the Pt/YSZ interface. The interior of YSZ has high intensity for the oxygen concentration due to that being an oxide crystal, whereas the oxygen concentration of the Pt/YSZ interface decreases from YSZ to Pt. In order to evaluate the electrode reaction from oxygen distribution, we measured the length L which was defined as shown in the lower left inset of Fig. 1(b). We found that the length L are extended about 6 nm by the positive extra-voltage to Pt electrode. This indicated that Pt oxide are formed during the anodic reaction at the interface. This result is in agreement with a previous study using electrochemical impedance spectroscopy [3].

Reference

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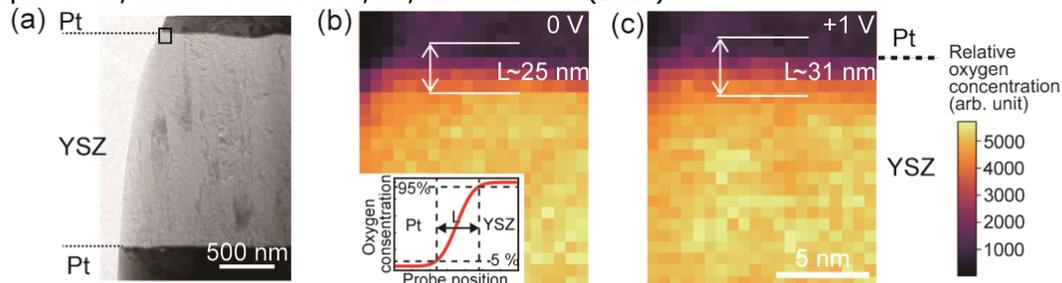


Fig. 1 (a) Low-magnification TEM image of the SOFC specimen, (b)(c) oxygen distribution around the Pt/YSZ interface. The black box as shown in (a) is the acquisition area of STEM-EELS. The Pt electrode have bias potentials, where the case of (b) and (c) have 0 V and +1 V, respectively. The lower left inset in (b) shows a schematic drawing of a line-profile around the Pt/YSZ interface.