Electronic Supplementary Information (ESI) for

# Mg–Air Oxygen Shuttle Battery Using a ZrO<sub>2</sub>-Based Oxide

# **Ion-Conducting Electrolyte**

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## **Experimental section**

### **Cell preparation**

A commercial 1.8-mm-thick Ca-stabilized  $ZrO_2$  (CSZ) tube (ZR-11, NIKKATO, CSZ, Volume; ca.14 ml) was used as the electrolyte. Pt paste (TR-7902, TANAKA KIKINZOKU KOGYO) painted on the inside and outside of the CSZ tube served as the electrodes. The system was calcinated at 1273 K for 30 min (surface area of electrodes: 13 cm<sup>2</sup>), and then metallic Mg (30 mg of WAKO) was inserted in a small alumina tube that was fixed inside the CSZ electrolyte tube. Next, the CSZ tube was closed except at the inlet and outlet for the stainless steel Ar gas lines to pass.

### **Electrochemical measurement**

After filling the tube with Ar in a Mg-set chamber, the cell was separated from the gas line using stop valves and then heated to 1073 K followed by electrochemical treatment at 3.5 V. Discharge measurement was performed at 1 mA (0.038 mAcm<sup>-2</sup>) in constant current mode, using the two probes method. Dry air was fed into cathodic side at a flow rate of 100 ml/min. An impedance analysis was performed under open circuit condition over the frequency range from 0.1 to 100000 Hz and an ac amplitude of 10 mV using a Sorlartron 1260/1287 impedance analyzer.

### Characterization of Mg powder after discharge

Changes in the morphology and elemental analysis of the Mg powder were measured using SEM-EDX (VE-7800, KEYENCE, EDAX type 7980). The current–voltage (I–V) and current–power curves (I–P) of the cell were measured using the two prove method. A constant current was applied using a galvanostat (HA-301, Hokuto Denko), and the potential was monitored using a digital multimeter (Advantest 6145, Advantest).

**Results of SEM-EDX** 



Fig.S1 (a) SEM image and (b) EDX results for the Mg powder after discharge.