Electronic Supplementary Material (ESI) for ChemComm.

Copper cobalt spinel as a high performance cathodes for Intermediate temperature solid oxide fuel cells

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Experimental

Preparation of $CuCo_2O_4$

$CuCo_2O_4$ phases were synthesized through standard the citric-ethylene diamine tetraacetic acid (EDTA) method. Stoichiometric amounts of copper nitrate ($Cu(NO_3)_2{\cdot}3H_2O$) and cobalt nitrate ($Co(NO_3)_2{\cdot}6H_2O$) were weighted and dissolved in deionized water. Subsequently, the equal moles of citric acid and EDTA acid to the total metal ions were added. The pH of the resulting solution was adjusted to about 6-7 using ammonium hydroxide and heated on a hot plate. Water was evaporated and the resultant gels were combusted to remove organic compounds, which then was ignited to form fine powder. The obtained ash after combustion was calcined at 450°C for 4 h in air to obtain the final powders.

10 mol% Sc2O3 stabilized ZrO2 (SSZ, Tero ChemicalCo., China) was chosen as the electrolyte material. SSZ electrolyte disk and anode-supported SSZ electrolyte half cells were fabricated by co-tape casting. The NiO and SSZ mixed 55:45 by weight for anode slurry, and followed by sintering at 1450 °C. The anode supported Ni-SSE|SSZ half cell discs were about 12 mm in diameter and 850 μm in thickness. The thickness of the SSZ electrolyte film was approximately 10 μm. The thickness of the SSZ symmetric disk is 110 μm. To achieve the single cell and the symmetric cell, the $CuCo_2O_4$ cathodes were fabricated by screen-printing the cathode slurry onto surfaces of SSZ electrolyte and calcining the painted cell at 1000 °C for 2 h in air. The final thickness of cathode was 25 μm and its effective area about 0.196 cm^2.

Characterizations

The crystal structures of the samples were identified by X-ray diffraction (XRD) using Cu Kα radiation (λ = 1.5405 Å) over 2θ range of 10° to 90° with a step size of 0.02° at room temperature. The electrochemical impedance spectra (EIS) were typically investigated on symmetric cells under open-circuit conditions using an electrochemical workstation (PARSTAT 2273) at 650-800 °C in air. The applied frequency range was from 0.1–10^5 Hz with the signal amplitude of 10 mV. The current-voltage characteristics of an anode-supported single cell were monitored by BT2000 instrument (Arbin). The electrical conductivity measurements were performed from 350 to 850 °C in air with a four probing DC technique on sintered $CuCo_2O_4$ using a Keithley 2400 source meter. The bar-shaped samples used for conductivity measurements were prepared by dry pressing and sintered at 1100 °C for 6 h, sizes of 2 mm × 5 mm ×10 mm. Linear thermal expansion was measured by a dilatometer (Netsch DIL 402PC) between room temperature and 900 °C with a heating rate of 5 K min^{-1}. The surface and cross-sectional microstructures of the cathode coated onto the SSZ electrolyte were characterized by a scanning electron microscope (SEM, SU8000).

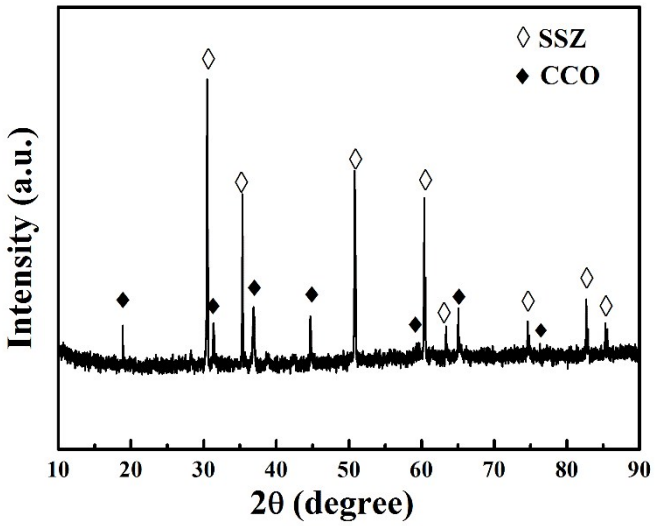

Fig.S1 XRD patterns of the $CuCo_2O_4$ and SSZ mixtures calcined at 1200 °C for 6 h.

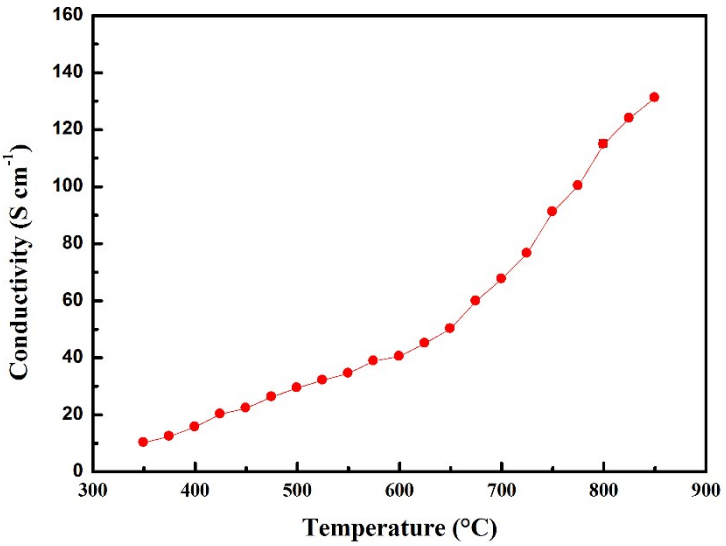

Fig. S2 The conductivity of $CuCo_2O_4$ at temperature ranging from 350 to 850 °C.

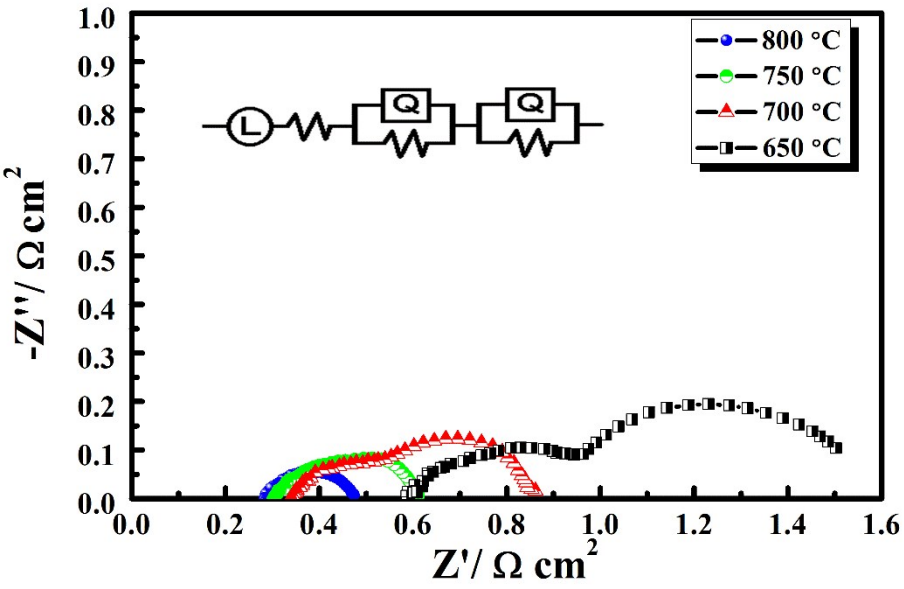

Fig.S3 The EIS of the Ni-SSZ|SSZ| $CuCo_2O_4$ at different temperatures.