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Short communication

Novel Al_2O_3-based compressive seals for IT-SOFC applications

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Abstract

Novel compressive Al_2O_3-based seals were developed and characterized under simulated intermediate temperature solid oxide fuel cell (IT-SOFC) environment. The seals were prepared by tape casting, mainly composed of fine Al_2O_3 powder with various contents of fine Al powder addition. The leakage rates were determined at 800 °C under 0.14–0.69 MPa compressive stresses, and the stabilities were evaluated at 750 °C under constant 0.35 MPa compressive stress. The leakage rates at 800 °C were in range of 0.2–0.01 sccm cm^{-1}, decreasing with increasing the compressive stress and Al content; Al addition significantly improved the stability, the leakage rate with 20 wt% Al addition was as low as 0.025 sccm cm^{-1} at 800 °C under 0.35 MPa compressive stress with a gauge pressure of 6.9 kPa, and exhibited good stability at 750 °C. Single cell test also confirmed the effectiveness of the tape cast Al_2O_3-based seal for planar IT-SOFC applications.

Keywords: SOFC; Compressive seal; Al_2O_3; Al; Leak rate

1. Introduction

Featured by reduced operation temperature, high power density, simple configuration, easy assembly, metallic interconnects and low cost manufacturing, the planar IT-SOFCs have attracted intensive attention since the end of last century [1]; however, one of the most significant challenges in the planar design is to develop effective high temperature seals to prevent fuel/oxidant gas leakage and provide electrical insulation within the stack [2,3]. The seal needs to have sufficient structural/chemical stabilities itself and thermal/chemical compatibilities with adjacent cell and interconnect components under the SOFC operating conditions.

So far, majority of SOFC seals development have been focused on rigid glass or ceramic-glass materials [2], deformable metallic materials [4,5] and mica-base seals [3,6–10]. The advantage of glass based seals is that their compositions can be tailored to optimize the required physical properties, such as the coefficient of thermal expansion (CTE); nevertheless, they tend to change in phases and react with the cell component materials and interconnects under SOFC operating conditions in a long run, because of the intrinsic thermodynamical instability [11,12]. The plain mica is virtually incompressible; a considerable compressive stress is required to obtain satisfied sealing effect, which may cause cell breakages. The hybrid mica-based seals [7–9] have achieved quite low leakage rate, but the sealing processes become more complex and other issues, such as stability and compatibility, have remained unsolved. The application of the deformable metallic seals is limited by its high electronic conductivity. In general, more robust sealing materials are desired to overcome the sealing difficulty in the planar SOFC design.

In the paper, novel Al_2O_3-based compressive seals and associated fabrication technique were reported. The seals were mainly composed of fine Al_2O_3 powder with fine Al powder addition, and prepared by tape casting. The effectiveness of the seals was confirmed by simulated out-of-cell and cell tests; and discussed in terms of the microstructural characterizations.

2. Experimental

2.1. Seal preparation

The characteristics and providers of the staring materials used in this investigation were summarized in Table 1. Fig. 1

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Table 1
The used raw materials and providers

Component	Material	Provider
Powder	Al_2O_3, $d_{10} = 1.2$ μm, $d_{50} = 3.5$ μm, $d_{90} = 7.1$ μm	Kunming Institute of Precious Metals
	Al, $d_{10} = 0.7$ μm, $d_{50} = 1.5$ μm, $d_{90} = 2.4$ μm	Beijing Mountain Technical Development Center for Non-ferrous Metals
Solvent	Ethanol/dimethylbenzene, volume ratio: 1:2	Tianjin No.3 Chemical Reagent Factory
Dispersant	Menhaden fish oil	Richard E. Mistler Inc.
Binder	Polyvinyl butyral-76 (PVB-76)	Solutia Inc.
Plasticizer	Butyl benzyl phthalate (BBP)	Wuhan Organic Synthesis Factory
	Polyalkylene glycol (PAG)	Richard E. Mistler Inc.

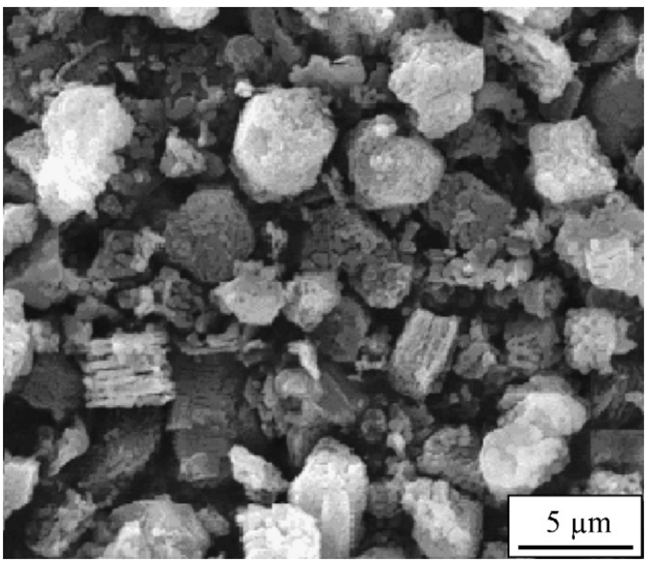

Fig. 1. SEM photograph of the used Al_2O_3 powders.

is the scanning electron microscope (SEM, QUANTA 200, FEI) photograph of the raw Al_2O_3 powders. The compressive Al_2O_3-based seals were prepared by tape casting, and the organic additives and their quantities used in the tape casting process are shown in Table 2. The thickness of the compressive seals is around 0.38 mm, and no defects were observed on the surfaces. One important characteristic of the compressive seals is that they have good flexibility at room temperature, and can be pressed on the sealing surfaces easily under moderate compressive stresses. Fig. 2 is the thermogravimetric curves of the cast tapes in air for the designated AS, AS1 and AS2 samples, respectively, obtained by using a DT-A7 thermal analysis system of Perkin-Elemer Inc. with a heating rate of 10 °C min^{-1}, which was used to guide the temperature profile during heating for the sealing tests.

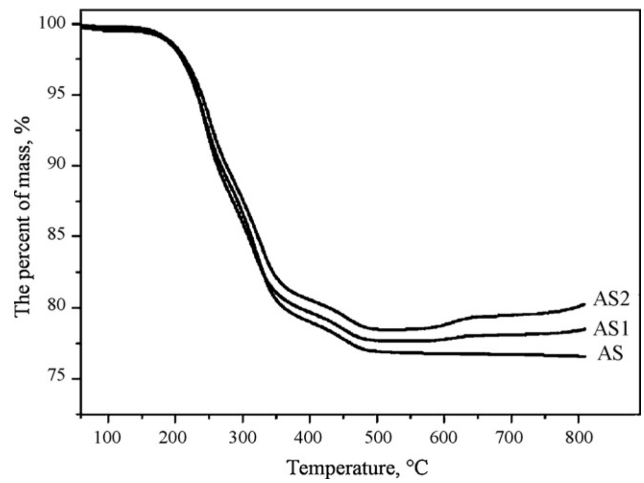

Fig. 2. Thermogravimetric graph of the tape cast AS, AS1 and AS2 samples.

2.2. Out-of-cell leak test and post-test characterization

The leak test samples were prepared as a window frame with an outside dimension of 7 cm by 7 cm and an inside dimension of 5 cm by 5 cm, as shown in Fig. 3. The leak tests were conducted by an in-house designed set-up, schematically shown in Fig. 4. The window frame specimen was place in between two polished stainless steel plates under a compressive load. The volume of gas reservoir was 200 cm^3, and the pressure in the gas reservoir was adjusted and stabilized at 3.5, 6.9 and 10.3 kPa, respectively. The flow reading from the rotometer (FL-310, OMGEA engineering Inc., USA) represented the leakage of the seals, once the equilibrium of the flow was reached. The leak rate was determined by the flow reading divided by the inside perimeter of the window frame samples.

According to the thermogravimetric analysis of the cast tapes, the samples were heated in a half furnace at a heating rate of 2 °C min^{-1} to 200 °C, and dwelled at 200 °C for 90 min followed by further heating at 3 °C min^{-1} up to 750 °C or 800 °C. The

Table 2
Tape casting formulations for the Al_2O_3-based seals

Sample	Al_2O_3 powder	Al powder	Solvent	Fish oil	P PVB-76	BBP	PAG
AS	100		42	2	8.5	8	8.5
AS1	90	10	42	2	8.5	8	8.5
AS2	80	20	42	2	8.5	8	8.5