



Electrochemical performance of solid oxide fuel cells having electrolytes made by suspension and solution precursor plasma spraying



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HIGHLIGHTS

- SOFC electrolytes produced by plasma spraying from 3 suspensions and 1 solution.
- Evaluated for microstructure, permeability and electrochemical performance.
- Peak power density of 599 mW cm⁻² and open circuit voltage of 1.00 V at 750 °C.
- First reported electrochemical data on SOFCs with SPPS electrolytes.

ARTICLE INFO

Article history:

Received 2 April 2013

Received in revised form

13 June 2013

Accepted 14 June 2013

Available online 27 June 2013

Keywords:

Solid oxide fuel cell (SOFC)

Electrolyte

Yttria-stabilized zirconia (YSZ)

Suspension plasma spray (SPS)

Solution precursor plasma spray (SPPS)

ABSTRACT

Yttria-stabilized zirconia (YSZ) electrolytes were deposited by suspension plasma spraying (SPS) and solution precursor plasma spraying (SPPS). The electrolytes were evaluated for permeability, microstructure, and electrochemical performance. With SPS, three different suspensions were tested to explore the influence of powder size distribution and liquid properties. Electrolytes made from suspensions of a powder with $d_{50} = 2.6 \mu\text{m}$ were more gas-tight than those made from suspensions of a powder with $d_{50} = 0.6 \mu\text{m}$. A peak open circuit voltage of 1.00 V was measured at 750 °C with a cell with an electrolyte made from a suspension of $d_{50} = 2.6 \mu\text{m}$ powder. The use of a flammable suspension liquid was beneficial for improving electrolyte conductivity when using lower energy plasmas, but the choice of liquid was less important when using higher energy plasmas. With SPPS, peak electrolyte conductivities were comparable to the peak conductivities of the SPS electrolytes. However, leak rates through the SPPS electrolytes were higher than those through the electrolytes made from suspensions of $d_{50} = 2.6 \mu\text{m}$ powder. The electrochemical test data on SPPS electrolytes are the first reported in the literature.

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1. Introduction

Metal-supported solid oxide fuel cells (SOFCs) are a promising technology for clean energy conversion. Cells with yttria-stabilized zirconia (YSZ) electrolytes produced by wet ceramic processing [1], vacuum plasma spraying [2], and atmospheric dry powder plasma spraying [3] have achieved open circuit voltages (OCVs) at or near the Nernst potential. Atmospheric plasma spraying with suspension- or solution-based feedstocks could potentially deposit thinner and more uniform electrolytes than those produced from a dry powder feedstock. Electrolytes have been produced by suspension plasma spraying (SPS) [4,5], though cell OCVs are not yet as high as those of cells with electrolytes produced by other methods. Solution precursor plasma spraying (SPPS) is worth investigating

because it offers reduced process complexity and higher compositional flexibility compared to SPS. However, to date, electrochemical testing of cells with SPPS electrolytes has not been reported in the literature.

With SPS, the size distribution of the particles in the feedstock suspension affects the properties of the resulting coating. After the suspension is injected into the plasma, it breaks down into droplets which each contain multiple powder particles. As the liquid evaporates from the droplets, the particles will either agglomerate together or disperse as individual particles [6]. The smaller the particle, the more rapidly it is heated and accelerated by the plasma plume [7]. However, smaller particles also cool faster as the plasma loses energy, so depending on plasma power and stand-off distance, smaller particles can re-solidify prior to reaching the substrate. Furthermore, small particles have low momentum and are more easily deflected by the plasma plume as they approach the substrate, which causes them to impact at an angle with low

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normal velocity [8]. This impact behaviour increases coating roughness and forms regions of concentrated porosity.

The choice of suspension liquid is also important with SPS. In this article, some suspensions were made with mixtures of water, ethanol, and ethylene glycol. Relative to an equivalent volume of water, 70% and 33% less energy is needed to heat from room temperature and evaporate ethanol and ethylene glycol, respectively. Additionally, these liquids can add energy to the plasma through combustion. The enthalpies of combustion are approximately -22 and -20 kJ ml⁻¹, respectively, for ethanol and ethylene glycol. With typical suspension feed rates, the corresponding combustion enthalpy is small relative to the plasma enthalpy. However, combustion does not occur until multiple centimetres from the torch nozzle exit after the evaporated liquids have sufficiently mixed with air [9]. The combustion energy may partially offset the cooling of the plasma plume to help maintain the YSZ droplets in a molten state at substrate impact. Therefore, the use of flammable suspension liquids may permit the production of electrolytes using lower energy plasmas than would be needed with entirely water-based suspensions. Lowering the plasma energy reduces electrolyte segmentation cracking [10].

Other liquid properties, including viscosity and surface tension, are also important for their effect on the breakup behaviour of the suspension after injection. The viscosity of water and ethanol are similar, while the viscosity of ethylene glycol is more than 1 order of magnitude higher. The surface tensions of ethanol and ethylene glycol are approximately 70% and 35% lower, respectively, than that of water. Surface tension has a stronger influence than viscosity on the final droplet size distribution, and suspensions with lower surface tensions (higher We numbers) have wider size distributions [11]. Accordingly, the atomized droplets from water/ethanol/ethylene glycol-based suspensions likely have a wider size distribution than those from water-based suspensions. It may be more challenging to optimize the spray configuration for a wider droplet size distribution.

At this time, SPPS has not been extensively used to produce SOFC electrolytes. Jia and Gitzhofer [12] produced ceria-based electrolytes by induction plasma spraying of a solution feedstock in a vacuum, though coatings were relatively porous and no electrochemical test data were obtained. One variant on SPPS is a hybrid SPS/SPPS spray technique, which could be used to incorporate various additives into YSZ coatings to improve their conductivity or other properties. VanEvery et al. [13] plasma sprayed a suspension of yttria-partially-stabilized zirconia particles in a solution containing Yb and Nd precursors. These dopants were incorporated into the resulting coating and reduced its thermal conductivity, as was intended. Regarding SOFC electrolytes, co-doped ScYSZ has higher conductivity than YSZ [14], so Sc precursors could be added to a suspension of YSZ particles. Transition metal oxides such as Ni, Co, Fe, and Mn have been found to lower the sintering temperature of YSZ [15], so adding these elements to YSZ suspensions could increase the density of plasma sprayed electrolytes, though conductivity may be reduced. In the present study, however, SPPS was used alone to produce YSZ electrolytes without other additives.

With SPPS, the feedstock for YSZ coatings is a solution of dissolved salts of zirconium and yttrium. For the production of dense SPPS coatings, appropriate precursors must be selected. Muoto et al. [16] used zirconium acetate and yttrium nitrate as precursors to make dense yttria-partially-stabilized zirconia coatings. The thermal decomposition processes for these precursors are generally exothermic, which promotes complete reaction of the precursors to the target material (YSZ) and high droplet temperatures at impact. Additionally, the use of high concentration solutions improves coating density by encouraging solute precipitation throughout the volume of the droplet rather than just at the surface, which increases the size of the molten YSZ droplets at impact [17].

For this article, SOFC electrolytes were deposited by liquid-feed plasma spraying to compare four different feedstocks: three suspensions and one solution. The electrolytes were evaluated for permeability, microstructure, and electrochemical performance as part of full cells. For the electrolytes, the key electrochemical test results of interest were cell open circuit voltage (OCV) and series resistance (R_s).

2. Experimental procedure

2.1. Plasma spray processing

Y_{0.15}Zr_{0.85}O_{1.925} electrolytes were produced from 4 different liquid-based feedstocks, which are described in Table 1. Suspensions 1 and 2 were made with powder from Inframat Advanced Materials (product no. 4039OR-8601, Manchester, CT), and suspension 3 was made with powder from Tosoh Corp. (product no. TZ-8YS, Tokyo, Japan). Suspension 1 was entirely water-based while suspensions 2 and 3 were made from a mixture of water, ethanol and ethylene glycol. The solution was made from zirconium acetate solution (product no. 413801, Sigma Aldrich Co., St. Louis, MO) and yttrium nitrate hexahydrate (product no. 12898, Alfa Aesar, Ward Hill, MA). These precursors were mixed in a ratio of 0.15 mol. Y to 0.85 mol. Zr.

The electrolytes were deposited using an Axial III DC plasma torch combined with a peristaltic pump-based liquid feed system (Northwest Mettech Corp., North Vancouver, BC, Canada). A schematic of the plasma spraying process is shown in Fig. 1. For each feedstock, electrolytes were deposited using 3 different plasma conditions, which were each used at 2 stand-off distances (Table 2). (Suspension 1 and the solution were not tested with the 102 kW plasma at 75 mm due to a limited number of substrates and anticipated lower deposition efficiencies. Therefore, 22 different feedstock/plasma condition/stand-off distance combinations were tested in total.)

The 160 kW plasma was used to achieve a high degree of particle melting at impact with all feedstocks. The 102 and 112 kW plasmas were tested to determine if the use of flammable suspension liquids, smaller powders, and/or solution-based feedstocks would enable the production of denser coatings using lower energy plasmas. The 102 kW condition used a smaller torch nozzle than the other conditions to produce a higher velocity plasma plume.

Table 1
Liquid feedstock characteristics.

Suspension details	Suspension 1	Suspension 2	Suspension 3	Solution
Powder d_{10} (μm)	0.8	0.8	0.2	Zirconium acetate + Yttrium nitrate Molar concentration = 2.2 mol l^{-1}
Powder d_{50} (μm)	2.6	2.6	0.6	
Powder d_{90} (μm)	5.7	5.7	1.5	
Powder vol.%	5	5	5	
H ₂ O vol.%	95	19	19	
Ethanol vol.%	0	38	38	
Ethylene glycol vol.%	0	38	38	

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