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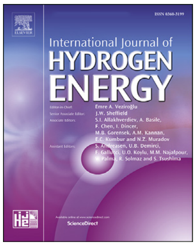

Development and evaluation of a 3-cell stack of metal-based solid oxide fuel cells fabricated via a sinter-joining method for auxiliary power unit applications

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ABSTRACT

Recently, metal-based solid oxide fuel cells (SOFCs) receive much attention for potential application in auxiliary power units (APUs). In this study, a sinter-joining method with a silver bonding layer is proposed. This method enables the fabrication of metal-based SOFCs by joining metal plates and conventional ceramic cells using a silver bonding layer. This sinter-joining method has the advantage of full-sintering of the cathode at 1100 °C, which facilitates a lower area specific resistance (ASR) of the cathode. Furthermore, the entire manufacturing process is conducted under air atmosphere. A 5×5 cm^2 metal-based cell is successfully fabricated by the sinter-joining method, and a maximum power density of 433 mW cm^{-2} and a low polarization resistance of 0.12 Ω cm^2 is obtained. Using the metal-based cells, a prototype 3-cell SOFC stack is developed considering mechanical robustness and diesel reformate fuel supply for future APU system applications. The stack exhibits a maximum power density of 100 mW cm^{-2} and is tested for 120 h. After the test, a post-mortem analysis is conducted, and the causes of the low electrochemical performance and degradation issue are investigated. In the conclusion, the sinter-joining method is considered as one of the methods for metal-based SOFCs.

Introduction

A solid oxide fuel cell (SOFC) is an energy conversion device that can generate electricity from hydrogen. SOFCs are considered to be promising technology because of their high energy conversion efficiency and fuel flexibility [1–8]. For this reason, numerous efforts have been made to utilize SOFCs in various energy systems, such as large-scale power plants, transportation, and mobile applications [9–11]. However, the applications of SOFCs in transportation and mobile devices have been limited because of their mechanical characteristics.

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SOFCs are composed of brittle ceramic materials such as yttria-stabilized zirconia (YSZ), Gd-doped CeO_2 (GDC), and $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}$ (LSCF), and hence, these cells can be easily fractured by external impacts. To overcome this limitation, metal-based SOFCs have been proposed [12–16]. A metal-based SOFC is composed of ceramic-based electrodes, electrolyte, and metal plate for mechanical support. For this reason, metal-based SOFCs possess a higher mechanical strength and demonstrate better resistance to vibrations and impacts than conventional SOFCs [17,18]. In addition, metal-based SOFCs can be sealed through the welding of the metal plates and interconnects. This method is more reliable and robust than those based on conventional glass or glass-ceramic composite sealants. Because of these advantages, metal-based SOFCs have been widely studied and adopted for scenarios involving movement, such as automobile applications. Among their various applications, auxiliary power units (APUs) have been attracting considerable attention [19–21]. In many countries, APUs are used as supplementary power systems for heavy-duty freight vehicles to reduce the emission of greenhouse gases and pollutants generated by diesel engines while idling for heating, cooling, or rest. According to US legislation, truck drivers must rest for several minutes to hours after long periods of driving for safety reasons. Therefore, the demand for APUs has been growing rapidly in many countries, and metal-based cells are considered to be promising candidates for use in APU power generation systems.

Various studies have been conducted on metal-based SOFCs for various applications including APU systems. The most significant issue hindering the development of metal-based SOFCs is the formation of a dense electrolyte. A metal-based SOFC is fabricated by coating electrodes and electrolytes on a rigid metal plate. However, the rigid plate prevents the formation of a dense electrolyte, resulting in pinholes and cracks [22]. To overcome this issue, a high sintering temperature of at least 1000 °C is required. However, such a high sintering temperature causes severe oxidation of the metal plate [23,24]. Furthermore, the interdiffusion of Ni, Cr, Fe occurs at the anode and the metal plate, leading to the formation of Ni–Cr–Fe alloy, which can degrade electrochemical performance and decrease thermal cycling stability because of the high thermal expansion coefficient of the Ni–Cr–Fe alloy [23,25]. To prevent the oxidation and degradation, various thin-film coating processes, such as plasma spray coating or physical vapor deposition, have also been widely studied [21,26–30]. However, these thin-film processes require expensive manufacturing equipment and a long process time. Therefore, conventional inexpensive coating processes such as screen printing or spin coating are also used in some research groups. However, the coated film deposited by conventional coating processes must be sintered under a reducing atmosphere. Sintering under a reducing atmosphere also requires additional process equipment and time, which again increases the manufacturing cost. In addition, the metal-based SOFCs are also subject to limitations related to cathode sintering at operating temperature of SOFCs in an approach known as un-sintered cathode or in-situ cathode. To prevent metal oxidation, metal-based SOFCs are fabricated without high temperature sintering of cathodes, which exhibit lower electrochemical performance, higher degradation rate, and more vulnerability to chromium poisoning than that of conventional fully sintered cathodes [23,31–34]. To improve cathode performance and durability, the infiltration of cathodic materials into a porous electrolyte backbone has recently been reported, and it was found that this approach allowed the sintering temperature to be reduced below 1000 °C [25,35]. However, this technique was implemented and tested only at the single-cell level, and further studies on long-term durability and the scaling up of the process to large-area cells must still be conducted before the cells produced using this method can be commercialized.

To overcome the limitations of metal-based SOFCs, a sinter-joining method has been developed in the past several years. To fabricate metal-based cells by conventional sinter-joining method, a conventional ceramic cell is bonded with a metal plate for mechanical support using bonding paste, which is composed of a mixture of stainless steel, Ni, and YSZ [36,37]. This process enables the fabrication of metal-based cells with high mechanical strength and thermal conductivity by metal plate bonding to the ceramic cell. Using the sinter-joining method, 10 × 10 cm^2 large-area cells are successfully fabricated in our group [37]. In addition, for integration with an APU system with a reformer, 5 × 5 cm^2 metal-based cells are tested with diesel reformate gas as fuel for 1000 h with a low degradation rate of 4%/1000 h, and the optimum operating conditions are proposed [38,39]. The fundamental electrochemical characteristics and heat and mass transfer characteristics of unit cells and stacks have been studied through computational modeling and simulations [40–42]. However, this sinter-joining method is still subject to the limitations of un-sintered cathodes, which result in a low performance of the metal-based cells [37]. Therefore, several studies have been conducted with the intent of improving the electrochemical performance of un-sintered cathodes through the development of cathode materials that can be sintered at low temperature and the optimization of the cathode microstructure based on impedance spectrum analyses [31,32,43,44]. However, the fundamental limitations associated with un-sintered cathodes, such as low power density, durability and cathode delamination at the interface with the electrolyte, have remained an issue.

In this study, a metal-based SOFC is proposed using a sinter-joining method with silver as the bonding material, and a 3-cell stack was developed for future integration into an APU system with a diesel reformer. To overcome the limitations of conventional metal-based SOFCs, metal-based cells were fabricated via a sinter-joining method that joins the metal plates and ceramic cell using a silver bonding layer instead of the conventional stainless steel-based bonding paste. This method has the advantage of processing all electrochemically active layers conventionally including sintering of the cathode at 1100 °C in air before sinter-joining with the metal plate at 950 °C. As a result, sintering under an air atmosphere and the full sintering of the cathode at a high temperature of 1100 °C were achieved, which had rarely been possible in previously developed manufacturing processes for metal-based cells because of metal plate oxidation. In addition, metal-based cells fabricated using a sinter-joining method have a simple fabrication process, good scalability, and high mechanical strength. To test the feasibility of the new configuration, a

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