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Intermediate-Temperature SOFCs with All-Perovskite Materials, Part One: Interactions between LSGM Electrolyte and LSCM Anode

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Technical trends in SOFC development and deployment include: (1) reducing operating temperatures by (a) using new electrolyte materials, doped lanthanum gallate or doped ceria, for example; or (b) minimizing electrolyte resistance (using electrode as support, thin film electrolyte); (2) increasing cell/stack performance and reliability; (3) lowering costs; (4) modularity and fuel flexibility. This study intends to advance the SOFC technology by using a more compatible, all-perovskite materials' system: $La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{2.8}$ (LSGM) electrolyte, $La_{0.75}Sr_{0.25}Cr_{0.5}Mn_{0.5}O_3$ (LSCM) anode and $La_{0.6}Sr_{0.4}Fe_{0.8}Co_{0.2}O_3$ (LSFC) cathode.

As one of the few available electrolyte materials, strontium- and magnesium-doped lanthanum gallate (LSGM) has drawn much attention due to its high ionic conductivity (0.12-0.17 S/cm at 800^{o}C) over a wide range of oxygen partial pressures (1-10^{-20} atm) [1-3]. The conductivity is higher by a factor of over two compared to the existing yttria doped zirconia (YSZ) (0.052 S/cm [6]). The LSGM materials, however require very high temperature, 1450-1550ºC for example, to be fully densified [4]. Three issues associated with these high firing temperatures are: (1) the LSGM electrolyte reacting with the currently used nickel-based anode during sintering, forming low conductive compounds, for example $LaNiO_3$ [5-8]; (2) Ga depletion; and (3) decreasing anode porosity [9]. Because of these reasons, LSGM based SOFCs are limited, at this time, to the LSGM electrolyte supported configuration, which is usually prepared by densifying the LSGM electrolyte first and then applying the anode and cathode layers afterwards.

Although a thin and dense LSGM layer with support substrate can be fabricated using various techniques, attempts to fabricate LSGM thin film SOFCs have been without much success. For example, electrostatic spraying could produce a crystallized LSGM layer after annealing at 900^{o}C for 2 h; however the LSGM layer was porous [10]. Joseph *et al.* [11] successfully obtained a 0.5 μm LSGM layer using pulsed laser ablation on quartz substrate, however, this must be done under high vacuum and high temperature conditions, and there was no density data from that report. Mathews *et al.* [12] prepared a dense film of 4 μm on Pt using electrophoretic deposition after five repetitions of deposition and sintering at 1400^{o}C. Majewski *et al.*'s study [13] concluded that only at temperatures higher than 1400^{o}C could a dense single phase LSGM structure be obtained.

One possible approach to use electrode supported LSGM thin electrolyte configuration is to use a plasma spray technique to apply the LSGM layer onto the fired electrode (anode or cathode) support. Feasibility studies [14] showed difficulties in putting down very thin (less than 50 μm) dense LSGM layers, and furthermore, the use of plasma spray is also costly. Our approach is to employ new electrode materials that are compatible to the LSGM, especially during high temperature firing, and to fabricate the thin LSGM electrolyte cells using conventional processes.

On the anode side, attempts to composite LSGM and Co, Ni or Fe were reported to be unsuitable due to the interactions [7]. The focus is then turned to $LaCrO_3$-based materials. $LaCrO_3$-based materials have been mainly used as interconnect materials for SOFCs [15, 16] due to their high electrical conductivity, stability in both fuel and oxidant atmospheres at high temperatures [17] and compatibility with other fuel cell components. Dopants on A- and B-sites, for example with Sr and Mn, respectively, showed significant reduction of the polarization resistance (from 10 to <1 Ωcm^2) and increase in electrical conductivity (to 1.5 S/cm in 5% H_2 at 900^{o}C) [18, 19]. In addition to good chemical compatibility with the YSZ electrolyte, when using methane as a fuel, the LSCM anode requires only ambient humidification (~3%) to prevent coke formation, compared to about 50% steam requirement for Ni-YSZ anodes for the same purpose (direct oxidation).

On the cathode side, a number of mixed electric and ionic conductors were explored for possible use as cathodic materials with the LSGM electrolyte. These included $La_{0.6}Sr_{0.4}CoO_3$ (LSC) [20, 21], $Sm_{0.5}Sr_{0.5}CoO_3$ [22], $La_{0.9}Sr_{0.1}Ga_{0.5}Ni_{0.5}O_3$ (LSGN) [23], and $La_{0.6}Sr_{0.4}Fe_{0.8}Co_{0.2}O_3$ [24, 25].

Interfacial studies have primarily focused in the cell operating temperature range, and very little information is available in relation to the compatibilities within the cell fabrication (LSGM sintering) temperature range. The new system, proposed with the LSGM electrolyte, LSCM anode and LSCF cathode, possesses an all-perovskite structure. It is our anticipation that all-perovskite structured and multilayered ceramic materials could increase the electrochemical and thermomechanical compatibilities of the SOFC system and eliminate the interaction between the electrolyte and electrode at the cell operating and fabricating temperatures (800-1500^{o}C). This paper will report the first part of our series study, the interactions between the LSCM anode and LSGM electrolyte in the cell fabricating temperature range (up to 1500^{o}C).

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