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Chemical solution deposition and characterization of the $\text{La}_{1-x}\text{Sr}_x\text{ScO}_{3-\alpha}$ thin films on $\text{La}_{1-x}\text{Sr}_x\text{MnO}_{3-\alpha}$ substrate[☆]

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ABSTRACT

The development of solid oxide fuel cells (SOFC) is one of the priority directions for the creation of alternative energy sources. High operating temperatures of these devices lead to the complexity of the constructions and active diffusion processes between the functional materials. The use of thin-film proton electrolytes is a promising way to reduce the operating temperature of SOFC, while the conductivity level and, as a result, the effective power, may be maintained due to the smaller thickness of the film.

Based on LaScO_3 proton-conducting oxides with a perovskite structure, having high chemical stability to water vapor, are promising proton electrolytes for SOFC, but they are poorly studied in the form of thin films. Lanthanum-strontium manganite is one of the most common materials for the SOFC cathode. The aim of this work is to study the effect of the $\text{La}_{0.6}\text{Sr}_{0.4}\text{MnO}_{3-x}$ cathode substrate composition on the properties, including the electrical conductivity of $\text{La}_{1-x}\text{Sr}_x\text{ScO}_3$ (0.01, 0.05 and 0.10) thin-film proton electrolytes, obtained by simple centrifugation of the film-forming solution. The properties of $\text{La}_{1-x}\text{Sr}_x\text{ScO}_3$ in the form of ceramic and thin-film samples are compared.

The experiment showed that the films $\text{La}_{1-x}\text{Sr}_x\text{ScO}_3$ at 5 ÷ 30-fold deposition on cathode substrates form continuous coatings with a grain size of 50–200 nm, which do not contain transverse pore. These results have a fundamental importance for the development of SOFC with ultra-thin film electrolyte on a supporting electrode. It is established that under dry and wet air the electrical conductivity of $\text{La}_{0.6}\text{Sr}_{0.4}\text{MnO}_{3-x}/\text{La}_{1-x}\text{Sr}_x\text{ScO}_3/\text{Pt}$ cells is bulk conductivity and it rises with increasing atmospheric humidity, which indicates an increase in the contribution of proton conductivity. In this case, the grain-boundary resistance of the material and the polarization resistance of the electrodes are practically not realized. The conductivity of LSS films is 1–2 orders of magnitude higher than the bulk conductivity of ceramic samples of similar composition and has a low activation energy. The observed differences in the conductive properties of films are explained by the interaction of related perovskites of the scan date and lanthanum manganite.

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The data obtained may be of interest to specialists in the fields of hydrogen energy, electrochemistry, materials science, the development of electrochemical devices: sensors, fuel cells.

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Nomenclature

List of notations and abbreviations

gb	Grain boundary
Ea	Activation energy, eV
pH ₂ O	Partial pressure of water vapor, kPa
LSS	La _{1-x} Sr _x ScO _{3-α}
LSS1	La _{0.99} Sr _{0.01} ScO _{3-α}
LSS5	La _{0.95} Sr _{0.05} ScO _{3-α}
LSS10	La _{0.9} Sr _{0.1} ScO _{3-α}
Sr _{La'}	The acceptor defect of substitution by ions of strontium of lanthanum ions
T	Temperature, °C, K
tot	total
Vo, V _O ^{••}	Oxygen vacancy
α	Nonstoichiometry coefficient
σ	Conductivity, S/cm
BET	Brunauer–Emmett–Teller method
XRD	X-ray diffraction analysis
PVB	Polyvinyl butyral
SEM	Scanning electronic microscopy
TCLE	Thermal coefficient of linear expansion
SOFC	Solid oxide fuel cell

Introduction

Currently, a solid oxide fuel cells (SOFC) development, especially operating at relatively low temperature, has become one of the most prioritized directions for creating alternative power sources. A key part of these devices is a proton-conductive membrane deposited as a thin film on a supportive electrode. Electrolyte has to possess chemical stability and rather high proton conductivity at low operating temperatures [1–5]. An effective fuel cell should have a low internal resistance thus the usage of thin-film electrolytes instead of bulk ceramics could be one of the most perspective development paths for the SOFC advance.

A great amount of research works related to studying the thin-film electrolytes and methods of their application are fulfilled by scientific community by now [6–8]. In SOFC film deposits on one of the electrodes from which follows number of questions on the interaction of materials, mutual diffusion of components and its influence on the properties of the film electrolyte and the system as a whole. Influence of the electrodes material increases along with reducing the membrane

thickness. There are many physical-chemical approaches for obtaining thin films with a very wide range of cost and implementation complexity. However, the simplest deposition methods are spin- and deep-coating. These techniques are easy way to get films of multicomponent oxides with average layer thickness lying between 20 and 100 nm [9–12].

Proton-conducting oxides based on the LaScO₃ with a perovskite type structure are promising electrolytes for SOFC, due to a significant level of proton conductivity and high chemical stability to the water vapors. These materials are promising for application in SOFC as electrolyte membrane [13–20], but they are poorly studied as thin films.

Most perspective materials for SOFC cathodes are the same perovskite -type oxides as for electrolyte membranes [21,22]. This similarity is helpful for deposition of proton-conducting films on cathode. But, on the other hand, the same structural type leads to increasing of interdiffusion. The question is, whether or not the interdiffusion has a significant influence on the electrolyte.

One of the most common materials for the SOFC cathode is lanthanum-strontium manganite [23–25] and the purpose of the research is to establish the influence of La_{0.6}Sr_{0.4}MnO_{3-α} substrate on properties, including electrical conductivity, of thin-film proton electrolytes La_{1-x}Sr_xScO₃, where x = 0.01, 0.05 и 0.10, which were obtained by simple spin-coating method. These perovskites differ only in one cation and the processes of mutual influence must be maximal. For comparison, the properties of La_{1-x}Sr_xScO₃ in the form of ceramic and film samples are compared.

Theoretical analysis

Technology of films deposition from precursor solutions include following stages

- preparation of precursor solutions;
- application of a solution to the substrate surface;
- drying, pyrolysis of organic constituents and the formation of an amorphous film by low-temperature heat treatment;
- formation of the crystal film of required composition by high-temperature heat treatment.

To successfully implement the technology of deposition of films from precursor solutions, the following conditions must be fulfilled:

- sufficient solubility of precursors in a solvent;
- Good wetting ability of the solution with respect to the substrate material;

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