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Multilayered Electrolyte-supported SOFC Based on NEVZ-Ceramics Membrane.

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SOFC batteries development is currently thriving in Russia (1). TVEL Fuel Company is a division of Rosatom State Corporation that promotes the SOFC technological platform, including cell, stacks, and integrated stack module for different applications. The technology development is based on R&D consortium between Institute of Solid State Physics Russian Academy of Sciences (ISSP RAS) with NEVZ-Ceramics (Novosibirsk, Russia) and includes materials synthesis and cost effective ceramic manufacturing methods for electrolyte supported flat cells with multilayered electrodes assembling for stack with metallic interconnectors from stainless steel Crofer 22H. Here we report on manufacturing and electrochemical tests of multilayered electrolyte-supported SOFC based on NEVZ-ceramics membrane. 10Sc1CeSZ membranes of 50x50 mm^2 area and 250 microns thick were chosen as a support. NiO-10Sc1CeSZ and $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$-10Sc1CeSZ multilayered electrodes were deposited by print-screen technique. Electrochemical tests in two-chamber cell were evaluated in wide temperature range (700-900^oC) and different oxygen atmospheres.

Introduction

Among the basic designs of solid oxide fuel cells, the electrolyte-supported SOFCs are extensively investigated because they exhibit higher advantage for stack manufacturing (2, 3). Significant improvement of cell performance has been achieved in recent years through the optimization of materials and microstructures of multilayer electrodes. NEVZ-Ceramics (Novosibirsk, Russia) is a company specialising in ceramic technologies that has a production line of electrolyte membranes for SOFC. In cooperation with NEVZ-Ceramics team whose work is concentrated on the development of a tape casting method (4) for manufacturing of planar 10Sc1CeSZ solid electrolytes with high durability and flatness, the ISSP RAS team has the task to develop the laboratory method for manufacturing of the cell with multilayered electrodes and to perform electrochemical tests of multilayered electrolyte-supported SOFC based on NEVZ-ceramics membrane.

Experimental

Solid electrolyte plates manufacturing process consists of several stages, which includes preparation of the ceramic slurry by mixing organic and inorganic components; ceramic tape casting the line, KYOKO Slovenia; drying of ceramic tape; lamination of stacks; removal of bundles; and roasting. Basing on the above process the technology of solid electrolyte plate casting method for moving the tape using the organic solvent tape casting of ceramic was perfected using the equipment by KEKO Ltd (Slovenia). Injection molding machine for the production of ceramic tape is a high-performance line for mass production (Figure. 1).

Figure 1. Tape casting line KEKO.

Bundles and removal processes were carried out in Nabertherm furnaces sintering for high temperature accuracy and energy efficiency. During firing the material acquires mechanical strength and all the necessary properties. Optimum sintering temperature is 1500°C because maximum density is reached at that temperature (Figure 2). When the temperature increases further the density decreases and porosity increases.

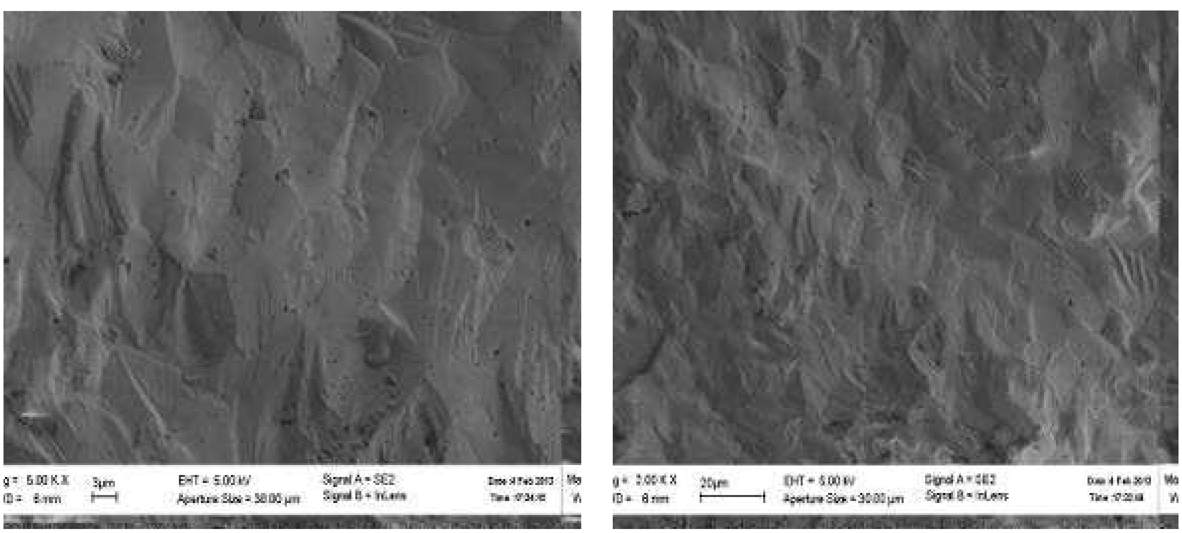

Figure 2. Electron-microscopic images of cross sections of ceramic plates 10%Sc_2O_3-1% 89% ZrO_2-CeO_2 sintered at a temperature of 1500°C.

Figure 3. shows the plates of solid electrolyte with composition of 10%$Sc_2O_3$1%CeO_2-ZrO_2 89% and the size of 50x50 mm^2 and thickness of 250 microns.

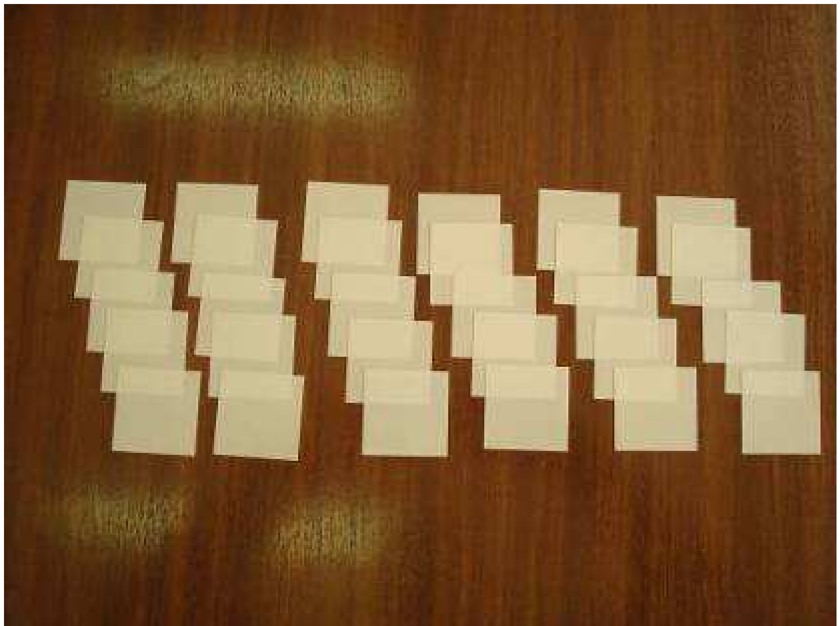

Figure 3. Samples of ceramic plates with size 50x50 mm^2 and a thickness of 250 microns.

In this work investigation of the electrochemical properties was carried out on two cell types: square-fitted 50×50 mm^2 cells and button cells of 21 mm diameter. Both electrolyte-supported cell types based on the electrolyte membranes of 250 um thickness were produced by NEVZ-ceramics.

Print-screenting anode and cathode pastes were prepared by mixing corresponding amount of powders and commercial binder HERAEUS V-006. Both electrodes were double-layered. Anode functional layer (AFL) with thickness of 15 um and 40/60 mass ratio of NiO/10Sc1CeSZ and anode current collector layer (ACCL) with thickness of 30 um and 60/40 mass ratio of NiO/10Sc1CeSZ were prepared in single firing at 1380^{o}C. SEM-image of anode side cross-section is shown in Figure 4.

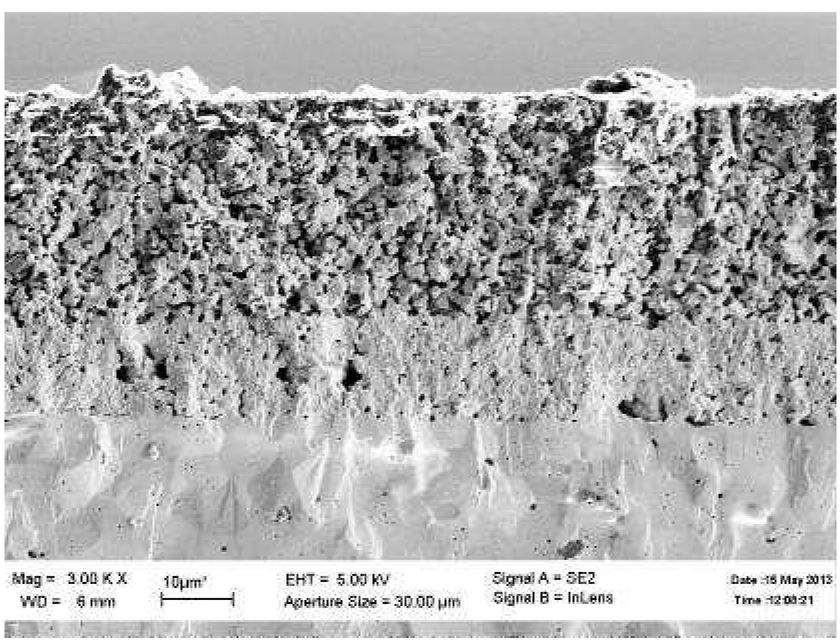

Figure 4. SEM-image of the anode side cross-section.

Than cathode electrode was deposited on the opposite surface of the membrane. Cathode functional layer (CFL) with thickness of 15 um and 60/40 mass ratio of $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$/10Sc1CeSZ and pure $(La_{0.8}Sr_{0.2})_{0.95}MnO_3$ cathode current collector layer (CCCL) with thickness of 30 um were also prepared in one step firing at 1100^{o}C. SEM-image of cathode side cross-section is shown in Figure 5. The areas of both electrodes were 40×40 mm^2 and 1.5 cm^2 for square-fitted and button cell respectively.

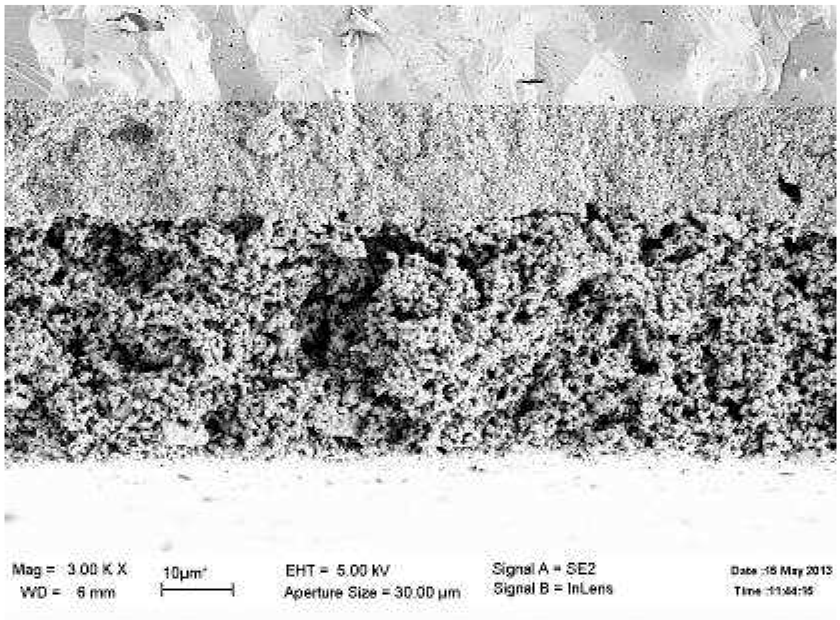

Figure 5. SEM-image of the cathode side cross-section.

Electrochemical tests of square-fitted cells were carried out by means of laboratory test station (Figure 6).

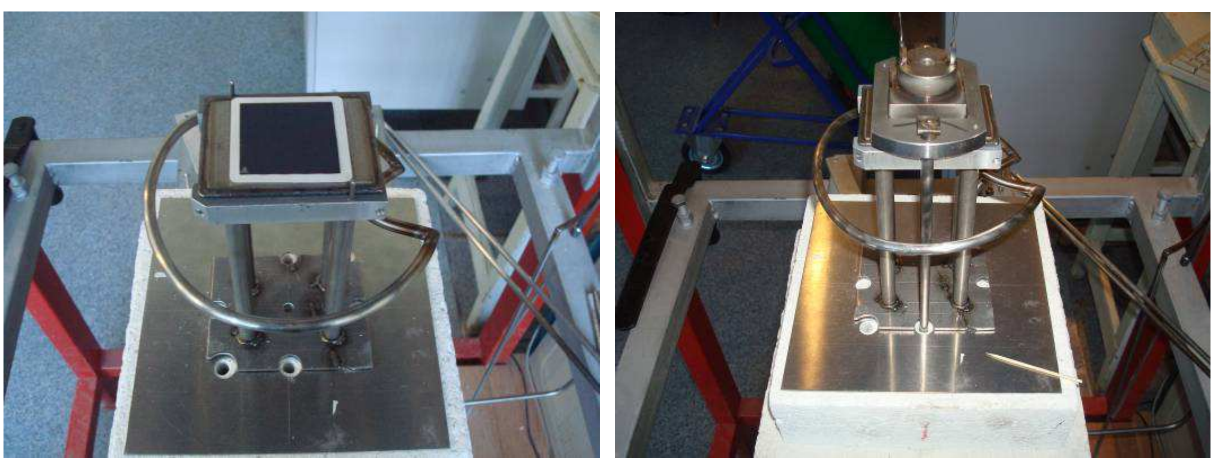

Figure 6. Laboratory test station with installed square-fitted fuel cell (left) and mounted mechanical load head (right).

Laboratory test station is equipped with heater, mass-flow controllers, electronic load and steel housing and allow to control composition and flow rate of fuel and oxidizing atmospheres, working temperature and conduct electrochemical tests of fuel cells. Separation of anodic and cathodic gas spaces was made without sealants by means polished surfaces.

Button cells test procedure see elsewhere (5).

Results and Discussion

Results of the electrochemical tests of square-fitted cells are shown in Figure 7. As can be seen on the picture the OCV value exceeds more than 1.1 V. That is the evidence of high quality of anodic and cathodic gas spaces separation by means of polished surfaces. It should be noted that the total output energy is less than 0.5 W for cell with 16 cm^2 active electrode area. To clarify the reason of such insufficient result the impedance measurements were made. Impedance spectrum measured at 800°C is presented in Figure 8. As can be seen on the picture ohmic and polarization part of the impedance spectrum are of the same order as expected. So, poor electrochemical characteristics are the consequence of the poor electric contact on one of the electrodes but not the quality of membrane or electrodes. To check that assumption the electrochemical tests were made on the button cell produced through the same procedure as a square-fitted one.

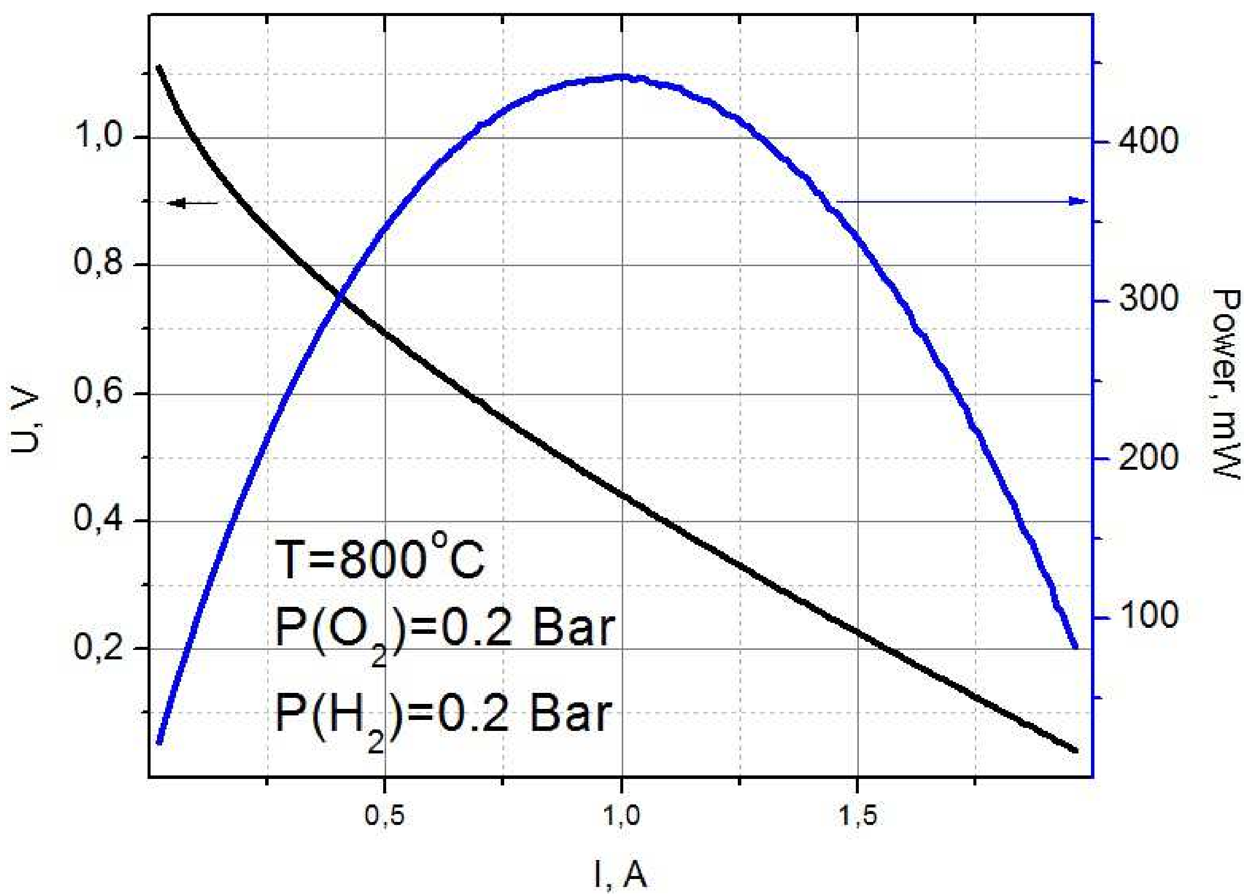

Figure 7. I-V and power curves of square-fitted fuel cell measured at 800°C.

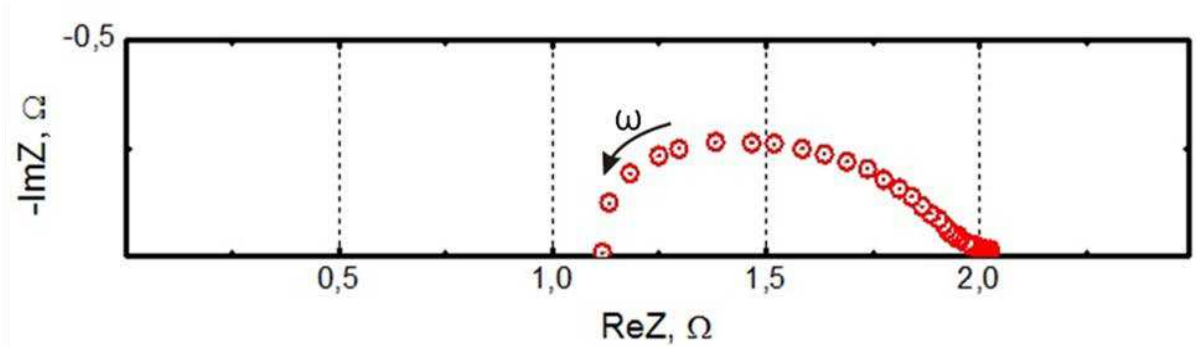

Figure 8. Result of the impedance measurements of square-fitted cell.

Results of button cell electrochemical tests are presented in Figure 9. OCV value exceeds more than 1V in all temperature range. It should be noted that output power density exceeds value of 0.6 W/cm^2 at 900^{o}C. Power density at 850^{o}C (target value for developed stack) and 0.7V is more than 0.35 W/cm^2. Thereby insufficient output power value for square-fitted cell is not a consequence of membrane or electrodes quality but active area reduction due to test station assemble problems.

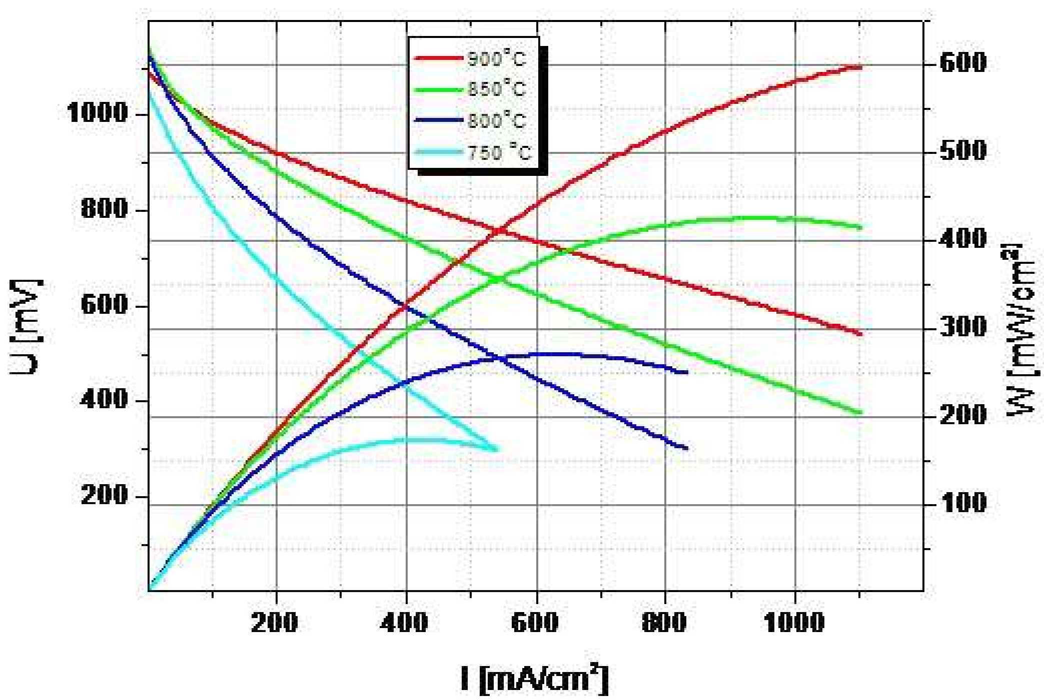

Figure 9. I-V and power curves of button fuel cell measured at 750-900^{o}C.

Conclusion

Here we report on manufacturing and first electrochemical tests of multilayered electrolyte-supported SOFC based on NEVZ-ceramics membrane. Ionic membrane (50x50 mm^2 area and 250 microns thick) was made through ordinary TLC route by NEVZ-CERAMICS. Multilayered composite electrodes were deposited by print-screen technique and fired at 1380 and 1100^{o}C for anode and cathode respectively. Electrochemical measurements of square-fitted cell showed insufficient results with only 30mW/cm^2 at 800^{o}C. Additional investigation of the impedance spectra and button cell characteristics indicated that poor electrochemical characteristics were the consequence of the test station assemble problem but not membrane or electrode quality. Further development of the fuel cells based on the NEVZ-Ceramics membranes is in progress.

References

1. Viktor Emel'yanov, *Fuel Cells Bulletin*, **2004**, 3, 2 (2004).
2. Norbert H. Menzler, Frank Tietz, Sven Uhlenbruck, Hans Peter Buchkremer an d Detlev Stöver, *Journal of Materials Science*, **45**, 12, 3109 (2010).
3. Nikolai Trofimenko, Mihails Kusnezoff, Alexander Michaelis, *ECS Transactions*, **35**, 1, 315 (2011).
4. Mistler R. E., Twiname E. R., *Tape casting: theory and practice*, American ceramic society (2000).
5. Burmistrov I., O. A. Drozhzhin, S. Ya. Istomin, V. V. Sinitsyn, E. V. Antipov, S. I. Bredikhin, *J. Electrochem. Soc.*, **156**, B1212 (2009).