Institute of High Temperature Electrochemistry of the Ural Branch of the Russian Academy of Sciences IHTE

Ural Federal University named after the first President of Russia B.N.Yeltsin

A REVERSIBLE PROTON-CONDUCTING CELL WITH SYMMETRICALLY FORMED $Pr_2NiO_{4+\delta}$-BASED ELECTRODES

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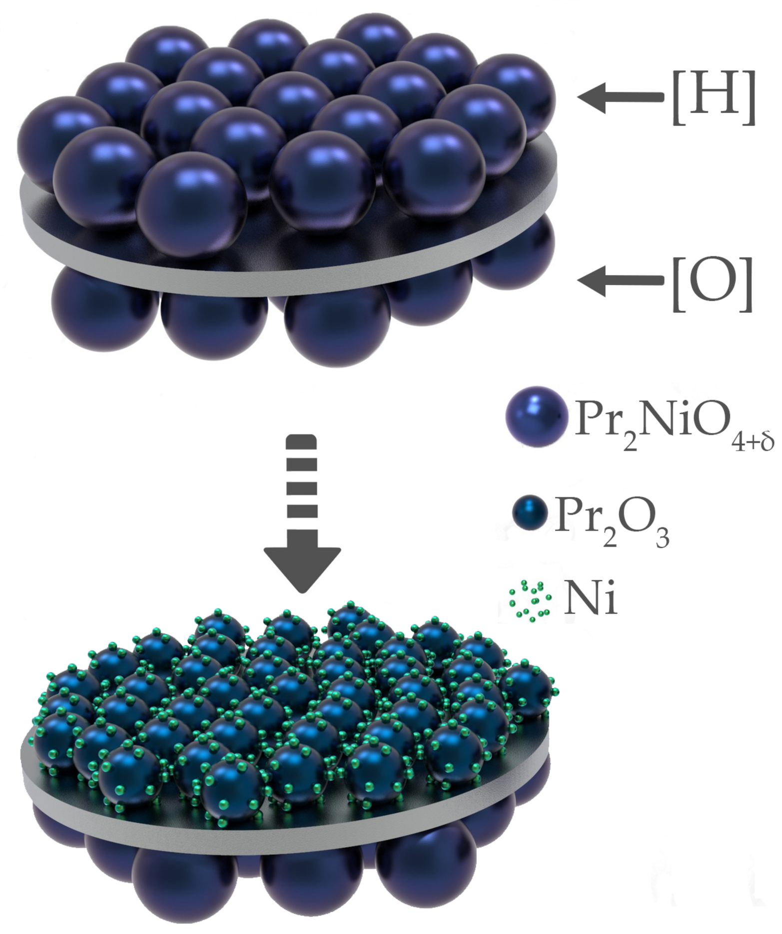
Introduction

Solid oxide systems with predominant protonic transport are considered as advanced applied materials for the energy sector. A particular interest is associated with their utilization as proton-conducting electrolytes in protonic ceramic cells, including reversible solid oxide cells that are used to convert different types of energy with high efficiency and no harmful impact. Recently, many efforts have been made to simplify technological processes involved in the fabrication of solid oxide devices based on oxygen-ionic or proton-conducting electrolytes. One of these efforts consists in designing electrochemical cells having symmetrical electrodes as an efficient strategy for reducing fabrication costs. Moreover, this strategy can help to resolve problems associated with thermal incompatibility and electrochemical degradation if the latter has a reversible nature.

In the present work, we propose to use a $Pr_2NiO_{4+\delta}$-based (PNO) oxide as symmetrical functional layers for a reversible protonic ceramic cell. This cell was fabricated from three functional materials, including $BaCe_{0.5}Zr_{0.3}Dy_{0.2}O_{3-\delta}$ (BCZDO) as a proton-conducting electrolyte layer, its mixture with nickel oxide as a substrate for supporting fuel electrode layer and $Pr_{1.9}Ba_{0.1}NiO_{4+\delta}$ as a basis for functional oxygen and functional fuel electrode layers. The used nickelate is stable under oxidizing conditions and almost completely decomposes at temperatures above 600 °C under H_2-containing atmospheres, with the formation of a Pr_2O_3/Ni cermet having mixed ionic-electronic behavior and, correspondingly, high electrocatalitical activity.

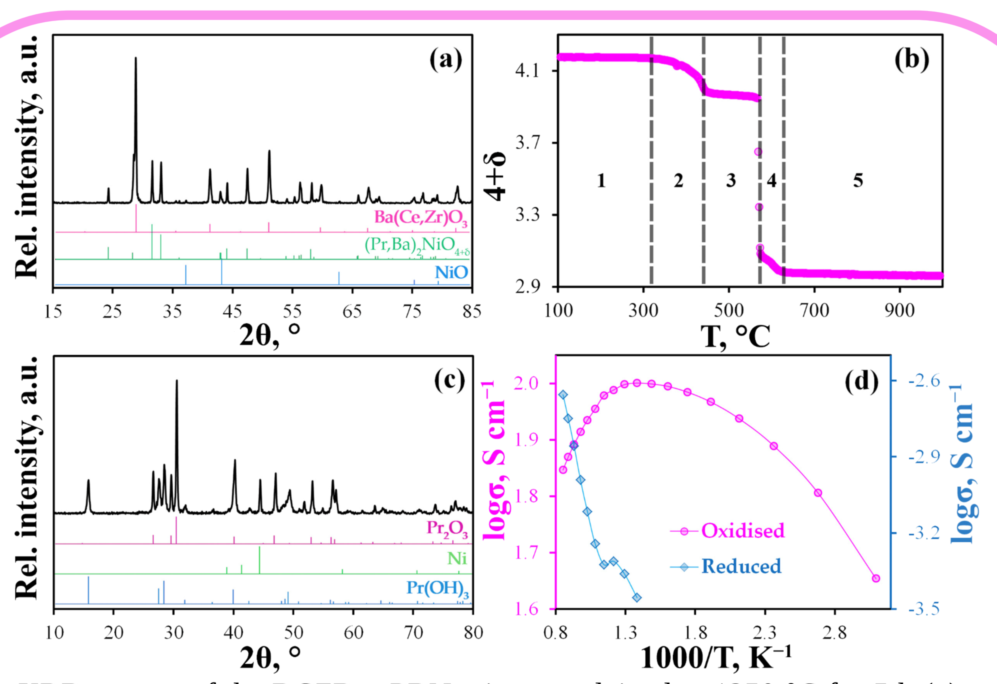


Methods

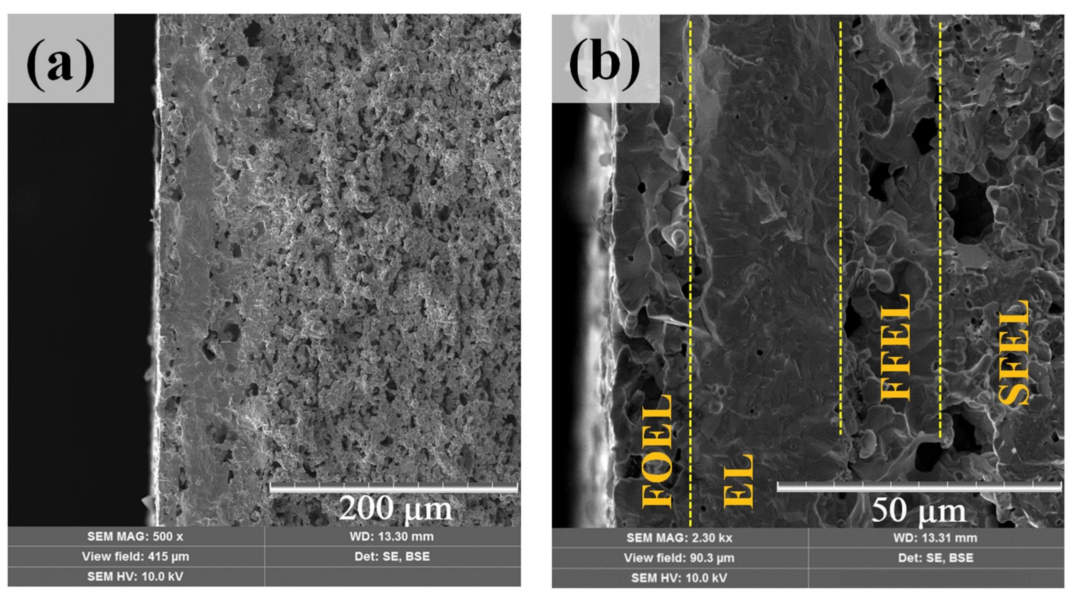
- Co-rolling films
- XRD
- 4-probe DC measurement
- TG-DSC
- Impedance (with DRT)
- SEM

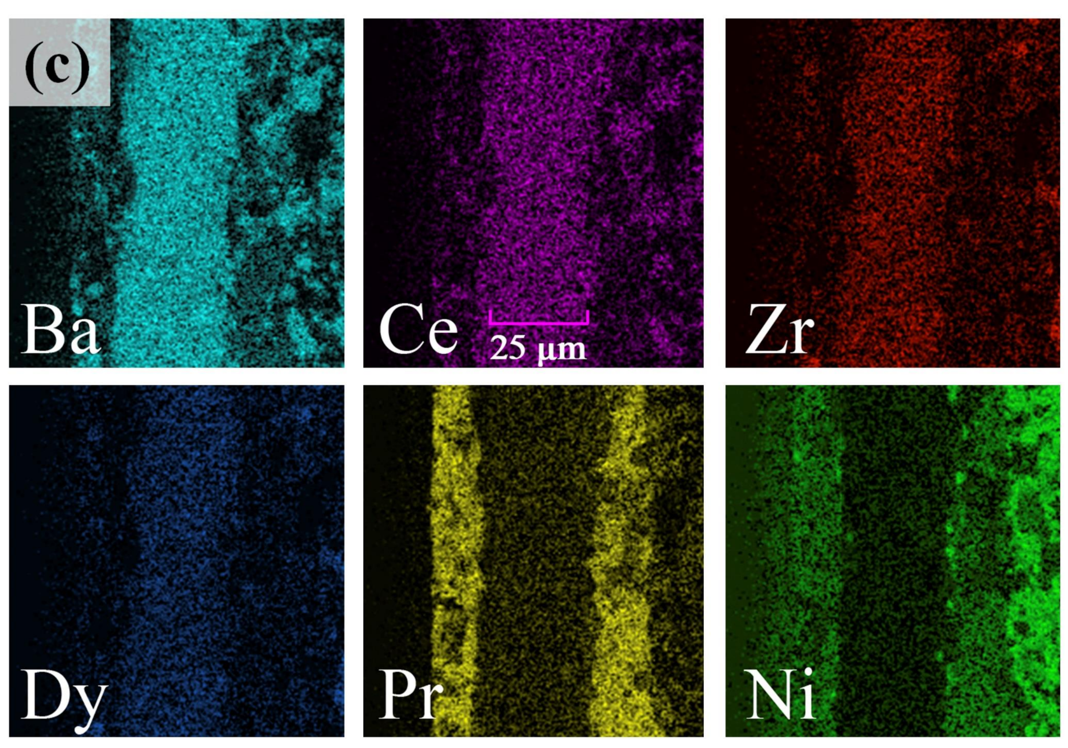
Experimental

Properties of $Pr_{1.9}Ba_{0.1}NiO_{4+\delta}$



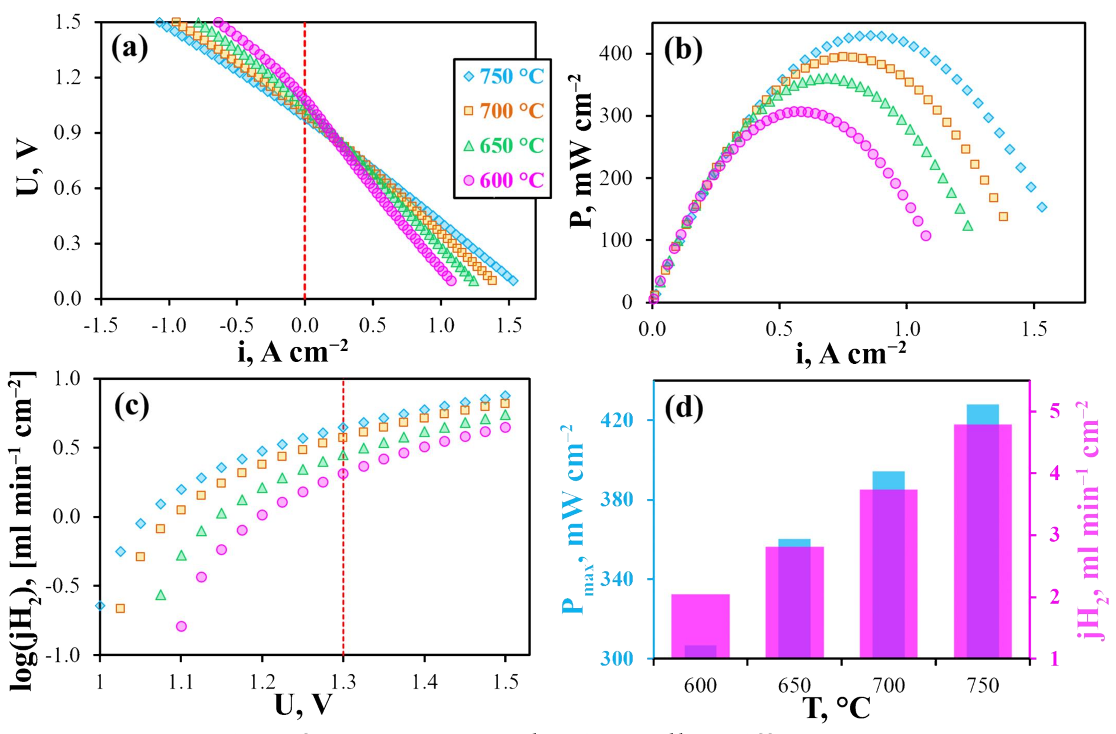
XRD pattern of the BCZD – PBN mixture calcined at 1350 °C for 5 h (**a**); TG curve obtained under reduction of the PBN powder in 50%H_2/N_2 atmosphere (**b**); XRD pattern of the reduced product of the PBN material (after TG analysis) (**c**); Conductivity of the PBN samples in oxidizing and reducing atmospheres (**d**).



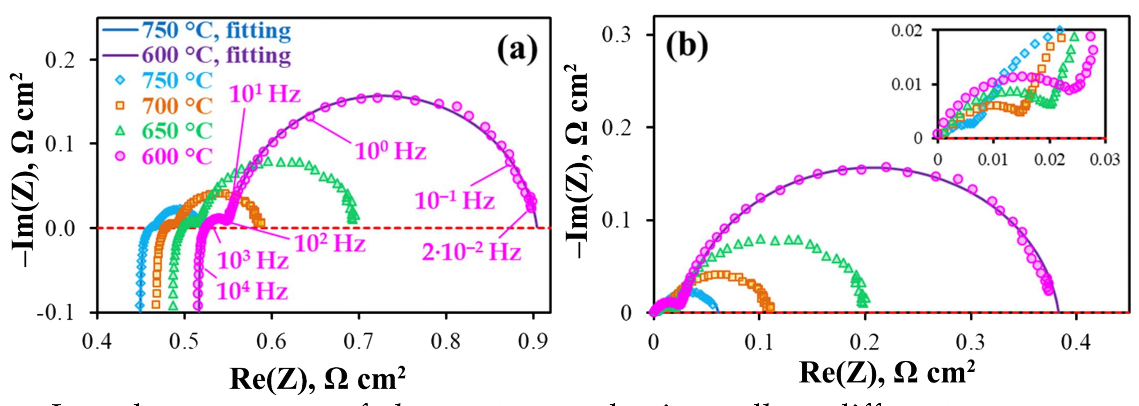


Cross-section images of the fabricated reversible proton-conducting cell at different magnification (**a**,**b**) and maps of the elements distribution (**c**). EL - electrolyte layer; SFEL - supporting fuel electrode layer; FFEL - functional fuel electrolyte layer; FOEL - functional oxygen electrode layer.

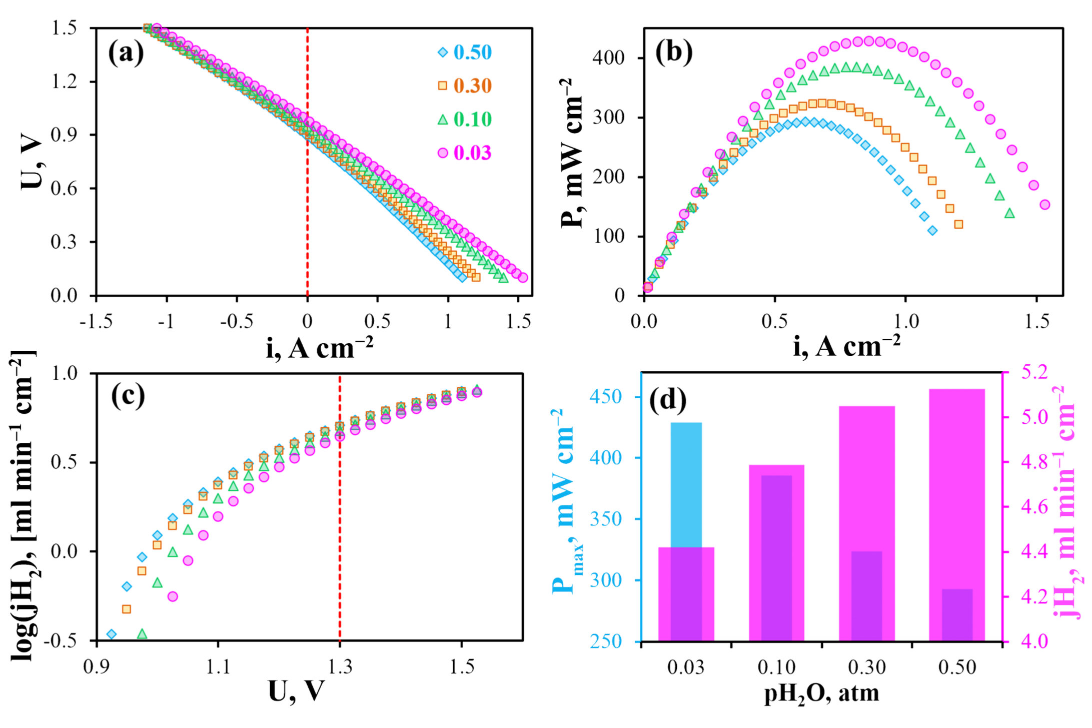
Characterization symmetrical cell as SOFC and SOEC



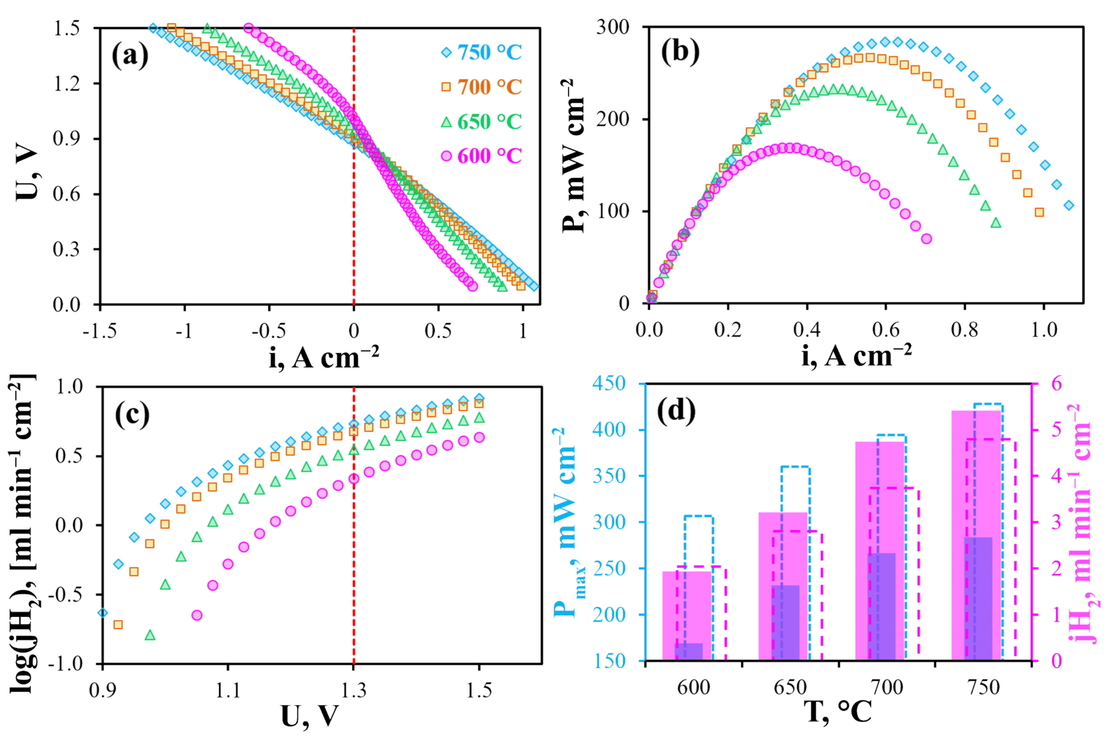
Reversible operation of the proton-conducting cell at different temperatures under 3%H_2O/H_2 – 3%H_2O/air conditions: volt-ampere curves (**a**), power density characteristics (**b**), maximal achievable hydrogen flux density (**c**), maximal power density and hydrogen flux density at U = 1.3 V depending on temperature (**d**).



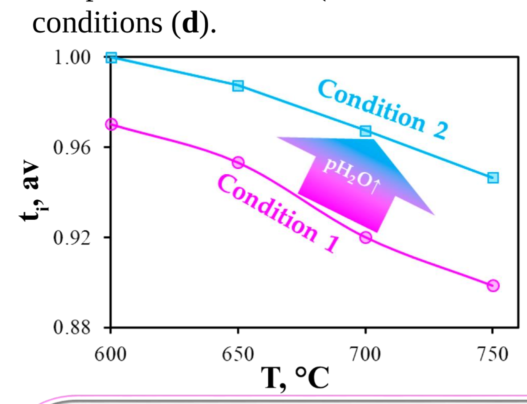
Impedance spectra of the proton-conducting cell at different temperatures under 3%H_2O/H_2 – 3%H_2O/air and OCV conditions: original spectra (**a**) and ones obtained after subtracting the ohmic resistance (**b**).



Reversible operation of the proton-conducting cell at 750 °C depending on different pH_2O in wet air with the constant fuel gas composition (3%H_2O/H_2): volt-ampere curves (**a**), power density characteristics (**b**), maximal achievable hydrogen flux density (**c**), maximal power density and hydrogen flux density at U = 1.3 V depending on temperature (**d**).



Reversible operation of the proton-conducting cell at different temperatures under 50%H_2O/H_2 – 50%H_2O/air conditions: volt-ampere curves (**a**), power density characteristics (**b**), maximal achievable hydrogen flux density (**c**), maximal power density and hydrogen flux density at U = 1.3 V depending on temperature compared with those (dashed columns) obtained under 3%H_2O/H_2 – 3%H_2O/air conditions (**d**).



Temperature dependences of the average ionic transference numbers of the BCZD electrolyte membrane in the current-free mode of the rsible proton-conducting cell under condition 1 ($p'H_2O = p''H_2O$ = 0.03 atm) and condition 2 ($p'H_2O = p''H_2O$ = 0.5 atm).

Conclusion

The fabricated cell based on a 25 μm-thick $BaCe_{0.5}Zr_{0.3}Dy_{0.2}O_{3-\delta}$ (BCZD) proton-conducting electrolyte demonstrated as high output characteristics at 600 °C as ~300 mW cm^{-2} in fuel mode of operation and ~300 mA cm^{-2} in electrolysis mode of operation at thermoneutral conditions. This cell was tested under different humid conditions in order to evaluate electrode and electrolyte performance. It was found that the PBN–BCZD oxygen electrode determined the overall electrode performance at 750 °C, operating as a dual conducting ($O^{2-}/h^{\bullet}$) system because of a negative electrochemical response towards gases humidification.

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