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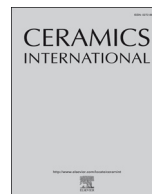
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Structure-conductivity relationship of PrBaMnMoO_{6-δ} through *in-situ* measurements: A neutron diffraction study



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ABSTRACT

The structural and electrochemical properties of the double perovskite-type oxide, PrBaMnMoO_{6-δ}, was investigated using neutron diffraction with *in-situ* conductivity measurement under a dry Argon atmosphere from 25 °C to 700 °C. A Rietveld refinement of the neutron diffraction data confirmed monoclinic symmetry in the $P2_1/n$ space group. Rietveld refinement also confirms the unit cell parameters of $a = 5.6567$ (1) Å, $b = 5.6065$ (2) Å, $c = 7.9344$ (1) Å and $\beta = 84.43^\circ$ with reliable atomic positions and refinement factors (R-factors). Neutron diffraction data refinement shows two minor phases (< 5%), an orthorhombic AB₂O₅ type phase of PrMn₂O₅ in the *Pbam* (No. 32) space group with unit cell parameters, $a = 7.9672$ (1) Å, $b = 8.9043$ (2) Å and $c = 5.8540$ (1) Å and a scheelite phase of BaMoO₄ in the tetragonal $I4_1/a$ (88) space group with the unit cell parameters, $a = b = 5.9522$ (1) Å, and $c = 12.3211$ (2) Å. Morphological images revealed a porous and intertwined microstructure. *In-situ* conductivity measurement shows that the total conductivity of this material was 130.84 Scm⁻¹ at 700 °C.

1. Introduction

The solid oxide fuel cell (SOFC) is considered one of the most viable energy conversion devices due to its high energy efficiency, fuel flexibility, and near-zero-emission operation [1,2]. SOFCs generate electricity through electrochemical reactions, directly from the oxidation of fuels [3,4]. It can use hydrogen, hydrocarbon [5], natural gas (methane), syngas [6,7], and biogas [8,9] as fuel and is not restricted by the Carnot cycle. It has also become a promising technology for a wide range of uses, especially in stationary situations; for example, they can be used to produce combined heat and power (CHP) for residential households [10,11] and in medium-scale distributed electricity generation [12]. The research in optimizing common materials for use in SOFCs is a vital topic in materials science in order to improve long-term stability, reduce costs, and also provide more material alternatives. A lot of emphasis is placed upon anode research because the performance of the SOFC is heavily-dependent upon it as the anode is where fuel is fed into the SOFC [13,14]. Conventionally, Ni-based composites are most commonly-used to construct SOFC anodes as these composites yield high activity for pure H₂ oxidation and they are also good current

collectors [15]. An example of such would be Ni-substituted yttria-stabilized zirconia (YSZ) cermet. However, Ni-YSZ results in poor redox cycling constancy as Ni is deactivated in NiO by rapid oxidation [16] in addition to its sensitivity to sulfur poisoning [17,18]. As such, many types of materials have been investigated to find a suitable replacement material for SOFC anode construction. Generally, SOFC requires mingled electronic-ionic conducting oxides for both anode and cathode materials [19].

Many novel perovskite materials (ABO₃) have already been developed for the SOFC electrode [20–22] to overcome the problems of Ni-YSZ. One of the significant advantages of perovskite materials is high electrical conductivity. In this paper, the double perovskite material (AA'BB'O₆) [23,24] is the main focus due to its structural versatility, chemical-, physical-, and thermal stability during SOFC applications. B and B' sites are occupied with different cations based on their valence and ionic radii [25].

Various rare-earth cations have been introduced at the A-site to obtain enhanced performance. Among them, the Pr-containing materials are suitable because of high stability and chemical compatibility with electrolytes [26–29]. A mixture of rare-earth and alkaline-earth A-

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sites enables mixed-valence states in double perovskites and this demonstrates higher electronic conductivity. As an example, Pr^{3+} and Ba^{2+} cations have alternating layers that make them ideal as electrode materials for SOFCs [30]. The oxygen atoms in the PrO_x plane create many oxygen-vacant sites in the crystalline structure which facilitates the ionic and electronic conductivity [31].

In this work, we have performed *in-situ* conductivity-structure measurements of $\text{PrBaMnMoO}_{6.8}$ to understand the changes of structure and conductivity with temperature. Structure determination was conducted by neutron powder diffraction (NPD) and conductivity was measured by Electrochemical Impedance Spectroscopy (EIS). Neutron powder diffraction (NPD) is a potent procedure that is used for its ability to detect light molecular weight elements [32] such as hydrogen and oxygen in order to analyze the structure of ion-conducting solids. Also, neutron diffraction can be used to detect the scattering from oxygen which is shown as additional peaks and consequently, the changes to the crystalline system. In the case of oxide materials, this scattering occurs due to oxygen anions deflecting the neutrons as they penetrate deep into the material structure [33]. For *in-situ* measurements, electronic-ionic conductivity was measured during neutron diffraction data collection at different temperatures. The time-of-flight (TOF) neutron data was collected and analyzed to obtain a complete diffraction pattern with a small scattering angle, which enabled the rapid collection of long-range data. Based on the structural analysis, the electrical properties of the crystal structure is essential for understanding the process of oxygen transport.

Materials development for SOFCs is an on-going process to get better performance. However, *in-situ* conductivity-diffraction measurement is a unique process to understand the underlying mechanism and very few works have been performed until now. The purpose of this work is to investigate the temperature-dependent changes on the crystalline structure as a result of changes in electronic-ionic conductivity and to form a correlation between the electrochemical and structural properties [34]. As far as we know, *no such study has been piloted*, in which the *in-situ* technique is applied to understand the process occurring in the SOFC on this double perovskite composition.

2. Methods

The double perovskite $\text{PrBaMnMoO}_{6.8}$ material was synthesized using a solid-state synthesis process [35–38]. Stoichiometric amounts of Pr_6O_{11} ($\geq 99.99\%$, Aldrich), SrCO_3 ($\geq 99.90\%$, Aldrich), MnO ($\geq 99.50\%$, Aldrich), and MoO_3 ($\geq 99.50\%$, Aldrich) were mixed in an agate mortar & pestle using ethanol as the mixing medium. The well-mixed powders were then pre-sintered in a furnace for 10 h at 650°C . The powders were re-ground using the agate mortar & pestle, pressed into small discs and heated at 900°C for 12 h. The pellets were re-fired at 1200°C and finally, at 1400°C for 12 h each. The sample was heated in $5\% \text{H}_2$ at 1000°C for 5 h to check the stability of the sintered product at reducing atmosphere. The heating and cooling rate was $5^\circ\text{C}/\text{min}$ for all heating cycles and conducted under Argon atmosphere [2].

The phase structure was first determined using X-ray powder diffraction by a Bruker axS-D8 Advance diffractometer. The data was collected in the 2θ range from 10° to 79.995° with increments of 0.02° per second and was analyzed by Rietveld analysis using Fullprof software [39]. The background was demonstrated using the 6-coefficient polynomial function and the peak shape was deliberated as pseudo-Voigt.

The crystalline structure and conductivity of $\text{PrBaMnMoO}_{6.8}$ were studied using *in-situ* Neutron Diffraction and impedance spectroscopy cell at the ISIS Neutron and Muon Source at the STFC Rutherford Appleton Laboratory in Oxfordshire, UK using the POLARIS time-of-flight (TOF) diffractometer [40,41]. The sample was loaded into 8 mm diameter cylindrical vanadium can, which was bound in a quartz tube and sealed with a Cu gasket. The measurements were performed at different temperatures (ranging from 25°C to 700°C). The temperature

was recorded using a thermocouple (similar setup was shown by Kinyanjui et al.) [42]. The sample was heated up to 700°C by two resistive coils above and beneath the sample container. The experiment was carried out at 1 mbar pressure, controlled by an inlet and outlet valve in a vacuum. The TOF data was analyzed by Rietveld refinement with GSAS-II software [43] using the data collected at bank 2 (up to 7 \AA). The background parameters, cell parameters, scale factor, profile parameters (type 3, as applied in GSAS), site occupancy factors (SOF), atomic displacement factors (ADP), atomic coordinates and phase fractions were refined to get complete structural information at different temperature.

The morphological structure was analyzed using scanning electron microscopy (SEM) to determine its porosity. A carbon layer was coated onto the sample to avoid overcharging as the sample was very conductive. The images (both surface and cross-sectional) of the pellet were taken with a JSM-7610F (Japan Electron Optics Laboratory Co. Ltd., Japan) [44]. The SEM images provided an excellent, high contrast view of the pellet.

In-situ conductivity was measured using a Solartron impedance analyzer coupled with a Frequency Response Analyzer (FRA) in the frequency range of $0.1 \text{ Hz} - 5 \text{ MHz}$ at AC amplitude of 100 mV . The electrochemical impedance software Z-view was used to analyze the impedance data. The measurements were carried out in a dry Argon atmosphere (Ar gas was passed through paraffin). The impedance measurement jig, which was designed and built at ISIS [34], was mounted vertically in the furnace and placed into the sample tank. The temperature was set in regular intervals from 25°C to 700°C (25°C , 100°C , and 200°C , up to 700°C) and the sample remained at each temperature for 2 h to allow for thermal equilibrium. The neutron diffraction data and impedance spectra were then collected at the same time to determine whether there were any changes in structure and conductivity. Fig. 1 shows the picture and schematic diagram of the *in-situ* experimental setup.

3. Result and discussions

The double perovskite-type polycrystalline $\text{PrBaMnMoO}_{6.8}$ was prepared using solid-state reaction method using final sintering temperature of 1400°C under Argon atmosphere. Then the sample was heated at 1000°C in reducing atmosphere ($5\% \text{H}_2/\text{Ar}$) for 5 h. The preliminary structural determination was carried out by X-ray powder diffraction (XPD). Indexing and cell refinement of the XRD data showed that the material crystallizes in two different phases namely, double perovskite ($\text{PrBaMnMoO}_{6.8}$) and scheelite (BaMoO_4). The Rietveld analysis of XPD data shows a monoclinic structure of $\text{PrBaMnMoO}_{6.8}$ in the space group $P2_1/n$ (no. 14) and a tetragonal structure of BaMoO_4 in the space group $I4_1/a$ (no. 88) [45]. However, neutron diffraction is more powerful tool to get accurate structural information for its high sensitivity for light atoms, scattering factor and non-absorbance in atomic electron cloud. Neutron diffraction is useful for studying compounds that contain heavy atoms that strongly absorb X-ray radiation.

The Rietveld refinement of the NPD data shows one additional orthorhombic phase of PrMn_2O_5 in the *Pbam* space group. The main phase was monoclinic $\text{PrBaMnMoO}_{6.8}$ in the $P2_1/n$ space group which comprises more than 95% of the material. So, the final structure from neutron diffraction show three distinct phases: (i) monoclinic structure for $\text{PrBaMnMoO}_{6.8}$, orthorhombic structure for PrMn_2O_5 and tetragonal structure for BaMoO_4 . The lattice parameters of the main phase was as follows: $a = 5.6567 (1) \text{ \AA}$, $b = 5.6065 (2) \text{ \AA}$, $c = 7.9344 (1) \text{ \AA}$ and $\beta = 84.4300^\circ$ (monoclinic, $P2_1/n$ space group). This cell parameters are related to the basic perovskite as $a \approx \sqrt{2}a_p$, $b \approx \sqrt{2}a_p$, $c \approx 2a_p$, $\beta \approx 90^\circ$. Table 1 shows the atomic positions, isotropic thermal parameters and fractional occupancies of the primary phase. The refinement of the 2nd phase (PrMn_2O_5) shows that this phase crystallizes in the AB_2O_5 type structure which can be refined in the orthorhombic *Pbam* (No. 32) space group. The unit cell parameters were, $a = 7.9672 (1) \text{ \AA}$, $b = 8.9043 (2)$

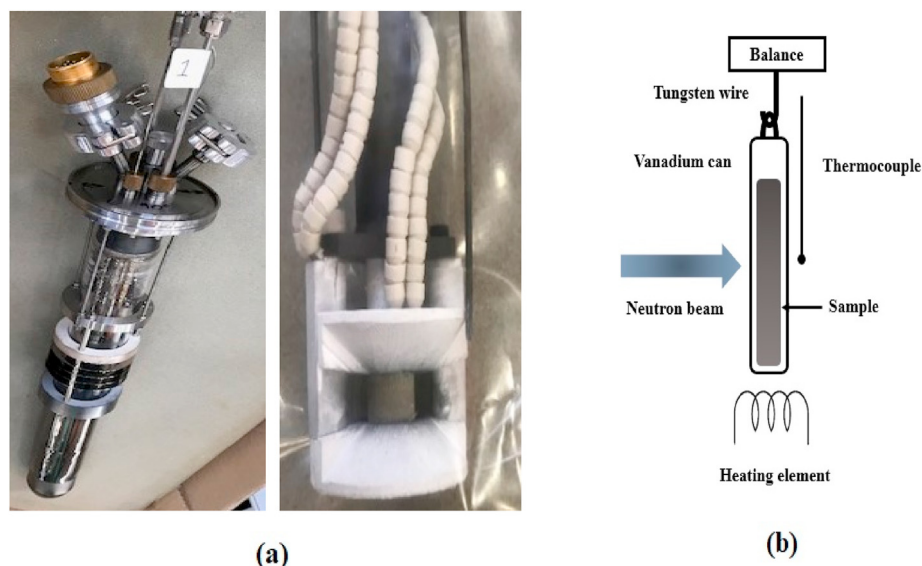


Fig. 1. (a) In-situ sample environment for PrBaMnMoO_{6.8} in an inert atmosphere. (b) Schematic illustration of the in-situ experimental setup at ISIS.

Table 1

List of atomic positions, isotropic temperature factors, and fractional occupancies for the main phase PrBaMnMoO_{6.8} (space group, $P2_1/n$) from neutron diffraction data at room temperature.

Atomic	x	y	z	U _{iso}	Fractional occupancy
Pr	0.9490 (2)	0.4729 (1)	0.1195 (2)	0.0057 (3)	1.000
Ba	0.9490 (2)	0.4729 (1)	0.1195 (2)	0.0350 (4)	1.000
Mn	0.5000	0.0000	0.5000	0.0051 (4)	1.000
Mo	0.5000	0.0000	0.0000	0.2474 (1)	1.000
O1	0.2789 (1)	0.6370 (1)	0.2115 (1)	0.0616 (1)	1.896 (3)
O2	0.7358 (2)	0.3304 (2)	0.0151 (1)	0.1076 (2)	2.273 (2)
O3	0.2250 (1)	0.0625 (1)	0.9460 (3)	0.0219 (2)	1.886 (2)

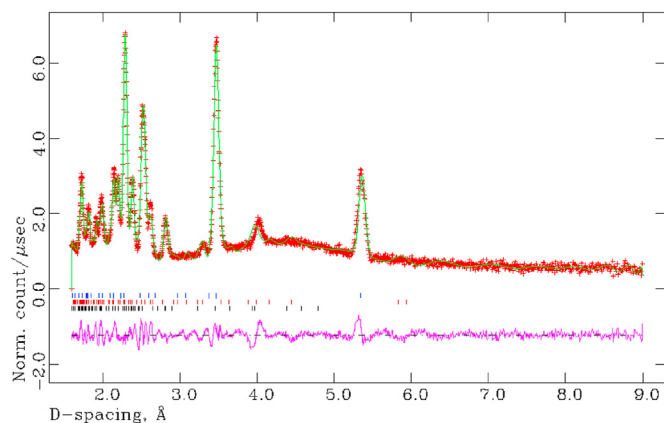


Fig. 2. Rietveld refinement profile of the neutron diffraction data collected at room temperature (295K) for PrBaMnMoO_{6.8}. Main phase: monoclinic, $P2_1/n$ space group, 2nd phase: orthorhombic, $Pbam$ space group, 3rd phase: tetragonal, $I4_1/a$ space group. Observed data (red + sign), calculated profile (green line), Bragg positions (vertical lines) and difference pattern (purple line in the bottom) are shown. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Å and $c = 5.8540$ (1) Å. The 3rd phase of BaMoO₄ was crystallized as a scheelite type structure in the $I4_1/a$ space group with the unit cell parameters, $a = b = 5.9522$ (1) Å, and $c = 12.3211$ (2) Å. All three phases were refined simultaneously to get reliable structural parameters and refinement factors (R-factors) where all peaks were indexed

with proper intensity matching, as shown in Fig. 2.

The preliminary refinement results demonstrated that Mn was situated at the 2a crystallographic site, while Mo originated from the 2c site. O2 displayed larger thermal displacements than O1 because of different oxygen sites. Fractional occupancies of cations were kept fixed due to small variations from their nominal occupancies. The Mn:Mo ratio was refined separately without applying any constraints which shows no mixing in their sites. Site occupancy factors (SOF), atomic displacement factors (ADP) were refined in different cycles due to their strong correlation. The oxygen vacancies are located in the praseodymium layers. A scheelite phase impurity is common in double perovskite type materials (e.g. AA'MnMoO₆) especially for the rare-earth-containing compounds as found in Ba₂MMoO₆ (M = Fe, Co, Mn, Ni) [46]. The unit cell volume depends on the Pr³⁺:Ba²⁺ ratio which is strongly correlated to the Mn²⁺:Mo⁶⁺ ratio [47]. Table 2 shows the cell parameters and refinement factors in elevated temperatures.

The presence of oxygen vacancies in the Pr layer was observed due to the mixed-valence state of Pr/Ba cations which also related to the mixed valence state of Mn and measure temperature. U_{iso} of all the atoms were refined isotropically. The refinement of oxygen occupancies for three sites improved the matching of the peak intensities. The Rietveld refinement of the neutron data collected at 200 °C, 400 °C and 700 °C (as shown in Fig. 3) confirmed that there were no structural phase changes at higher temperatures. However, there was a small increase in unit cell parameters. The variation of temperature has a significant effect on the physical properties of double perovskites, which is very important for fuel cell applications. At high temperature, the conductivity increases.

SEM imaging is a suitable method to categorize the microstructure

Table 2

Unit cell parameters, χ^2 values, and R-factors of PrBaMnMoO_{6.8} at selected temperatures.

Parameters	RT	200	400	700
Space group	$P2_1/n$	$P2_1/n$	$P2_1/n$	$P2_1/n$
a (Å)	5.65679 (1)	5.76210 (2)	5.68380 (1)	5.6646 (0)
b (Å)	5.62133 (2)	5.56072 (1)	5.48287 (1)	5.6474 (2)
c (Å)	7.94766 (2)	7.95738 (1)	7.86343 (2)	7.9931 (3)
β (°)	84.43000	83.00640	84.50310	84.43610
χ^2	4.28500	3.58800	3.03000	2.24200
R _p %	7.22000	7.28000	7.63000	7.29000
R _{wp} %	8.03000	8.89000	8.34000	7.34000

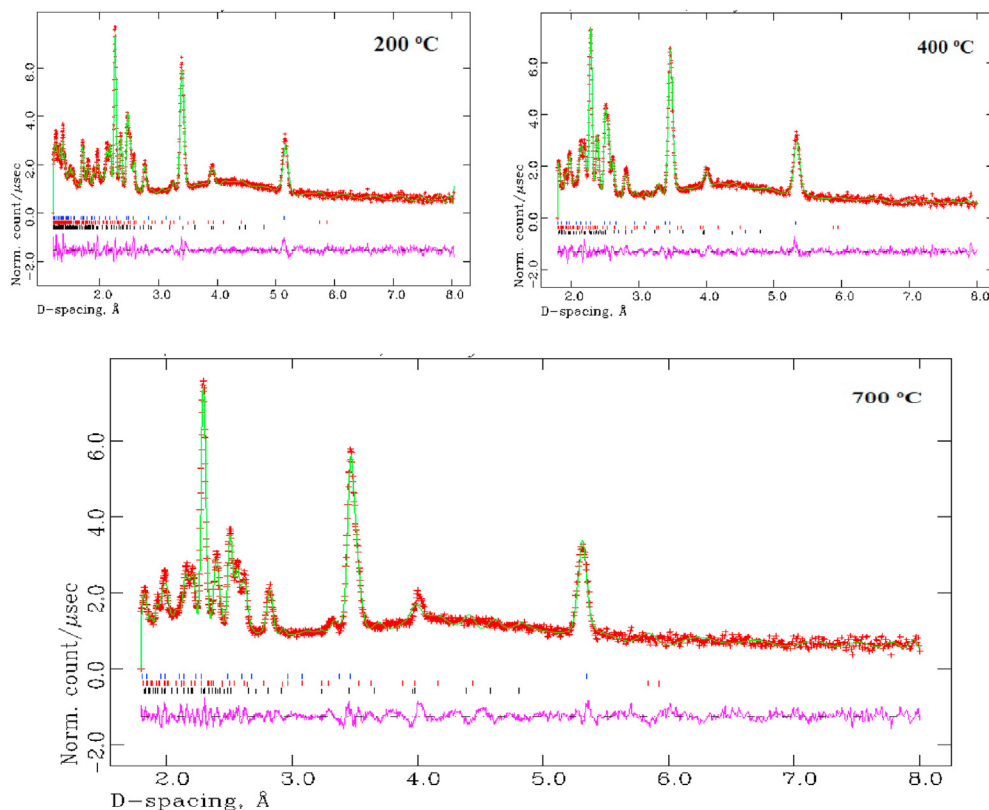


Fig. 3. Neutron diffraction refinement plots at 200 °C, 400 °C, and 700 °C, respectively for PrBaMnMoO_{6.8}. The main phase and impurity phases were same up to 700 °C. Measured data (red), a calculated pattern (green line), difference pattern (purple line), and Bragg positions (vertical lines) are observed in the model. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

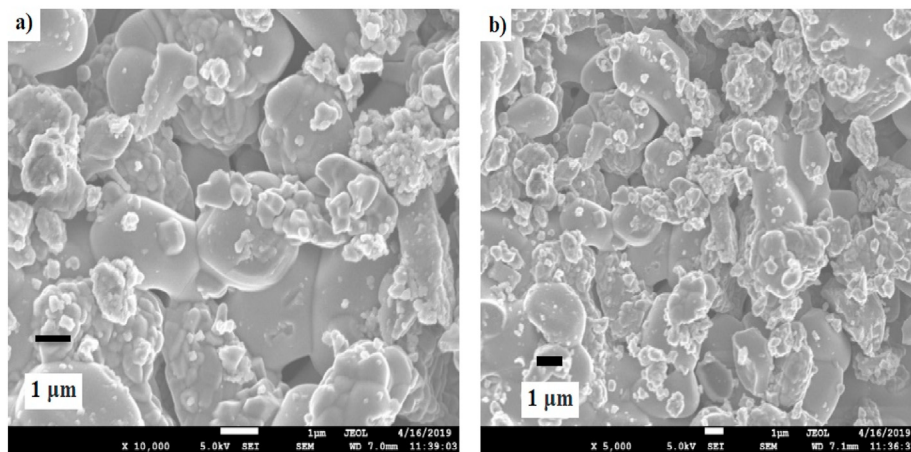


Fig. 4. SEM micrographs for PrBaMnMoO_{6.8} pellet at a voltage, 5.0 kV, (a) SEM images of the pellet surface with a working distance (WD) 7.0 mm, scale bar 1 μm with magnification 10,000X (b) SEM images of the cross-sectional pellet with working distance (WD) 7.1 mm, scale bar 1 μm with magnification 5,000X.

of perovskite-type oxide materials [48]. The microstructure of the ceramic materials is greatly affected by A-site and B-site cations as well as by the synthesis and sample preparation procedure [49]. Fig. 4 illustrates the SEM image of the pellet surface (a) and cross-section of the pellet (b) at different magnifications. Both images show a porous morphology. The secondary phases are well-distributed throughout the material with the average grain size approximately 1 μm. This is significant as the grain boundary facilitates and expedites the conduction of fuel during the three-phase reaction in a fuel cell anode [50].

In-situ conductivity allows the change in the crystal structure and conductivity to be measured simultaneously. As a result, the change in lattice parameters, as well as unit cell volume due to thermal expansion [48] and electron-ion movement, can be quantified. It can be understood from the literature that the rare-earth perovskite oxide materials show satisfactory electrochemical performance. The presence of bulk,

grain-boundary and electrode resistance in the prepared samples showed more than one semicircle appeared in electrochemical impedance measurements in a dry Argon atmosphere. The total conductivity can be measured using the following equation [51,52]:

$$\sigma = \frac{1}{\rho} = \frac{L}{RA} \quad (1)$$

where σ is the conductivity, ρ is the resistivity, L is the length, R is the resistance, and A is the cross-sectional area of the pellet.

It can be observed from electrochemical impedance spectroscopy that the conductivity of the sample increases with temperature (Fig. 5). The conductivity of PrBaMnMoO_{6.8} was calculated (fitted) from the impedance spectra by Z-view software modelled with an equivalent circuit. The value of R_s is the Ohmic resistance, while R_1 is the bulk resistance [53] in the fitted equivalent circuit (Fig. 5 inserted). R_2 and

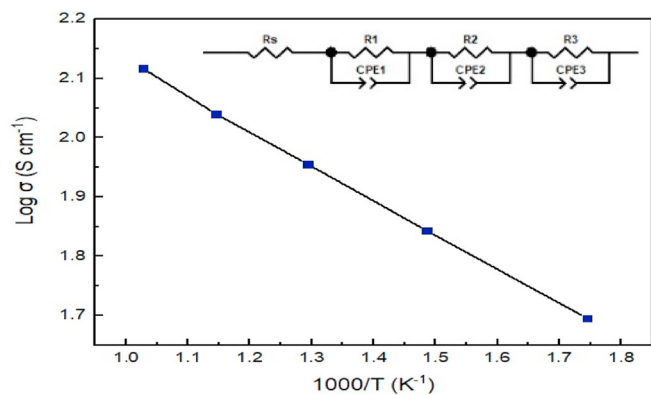


Fig. 5. Arrhenius plot of conductivity of PrBaMnMoO_{6.8} at 25–700 °C under dry Ar.

R₃ are resistances from the grain boundary and the electrode, respectively. A detailed analysis of the impedance spectra has been reported [54,55]. The conductivity values can be extracted from the data. Fig. 5 exhibits the Arrhenius plot of the PrBaMnMoO_{6.8}, where the activation energy of the sample was 0.116 eV measured under dry Argon environment.

The obtained conductivity values of PrBaMnMoO_{6.8} were 130.84, 109.69, 90.21, 69.71, and 49.59 Scm⁻¹ at 700 °C, 600 °C, 500 °C, 400 °C, and 300 °C, respectively. We observed that the total conductivity increased with temperature. Although a very small amount of impurity phases exists in the sample, the conductivity must be due to the contribution of the main phase of PrBaMnMoO_{6.8} (> 95%). Moreover, from the literature, it has been proven that scheelite structure BaMoO₄ shows very low conductivity [56,57]. *In-situ* structure-conductivity measurement simply allows to understand the properties in more realistic or noble conditions. The Argon gas was used to create a chemically-inert atmosphere to prevent oxidation of the electrodes. The phase fractions were extracted from the diffraction data collected from Rietveld refinement. This study proves that PrBaMnMoO_{6.8} is structurally stable and highly conductive at elevated temperatures.

4. Conclusions

This research demonstrated a novel approach of *in-situ* structure and conductivity measurement using neutron diffraction. Double perovskite-type PrBaMnMoO_{6.8} exhibits a monoclinic crystal structure in the P2₁/n space group and there were no distinct phase changes observed at higher temperatures. SEM analysis of the microstructure showed a significantly porous matrix with distinctive grain borders. The analysis of impedance spectra revealed that the total conductivity of PrBaMnMoO_{6.8} considerably increased with temperature. The maximum conductivity of 130.84 Scm⁻¹ was achieved at 700 °C. Through these observations, we were able to successfully determine the phase stability, porosity and conductivity of PrBaMnMoO_{6.8}. These characteristics can contribute to the high potential of the double perovskite being utilized as electrode materials in SOFCs. The authors recommend the use of *in-situ* studies to further enhance the development of electrodes for SOFCs.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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