



Short communication

Oxygen ion transference number of doped lanthanum gallate

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ABSTRACT

The transference numbers for oxygen ion (t_0) in several LaGaO₃-based materials are determined from oxygen concentration cells using the materials as the electrolyte, including La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-δ} (LSGM8282), La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.15}Co_{0.05}O_{3-δ} (LSGMC5) and La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.115}Co_{0.085}O_{3-δ} (LSGMC8.5). Analysis indicates that the accuracy in determination of oxygen ion transference number depends on the electrode polarization resistances of the concentration cell as well as the transport properties of the materials studied. For example, the ratio of open cell voltage to Nernst potential is a good approximation to the ionic transference number for LSGM8282. However, this approximation is no longer adequate for LSGMC5 and LSGMC8.5; the effect of electrode polarization resistances must be taken into consideration in estimation of the ionic transference numbers. In particular, the ionic transference number for LSGMC5 is as high as 0.99, suggesting that it is a promising electrolyte material for low-temperature solid-state electrochemical applications.

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1. Introduction

Low-temperature solid oxide fuel cells (LT-SOFCs) have attracted much attention due to their potential for high efficiency, long-term stability, and low cost. However, the ohmic resistance of electrolyte increases rapidly as the operating temperature is reduced, leading to a sharp decrease in cell performance. Thus, it is imperative to develop electrolyte materials that have high ionic conductivity at low temperatures. Cobalt-doped La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-δ} perovskites show superior ionic conductivity and good chemical stability at intermediate and low temperatures [1,2], and thus are regarded as a promising family of electrolyte for LT-SOFCs.

It is well known that any electronic conductivity of the electrolyte would degrade energy efficiency of SOFCs since the motion of electronic defects (e.g., electrons and holes) would translate to a short-circuiting current [3,4], resulting in a loss in cell voltage. Partial electronic conductivity in cobalt-doped LSGM8282 has been studied recently using ion blocking method. The electronic conductivity increases with increasing cobalt content because electronic

defects (electrons and holes) may be produced by the redox reactions involving doped Co [5–8].

Ionic transference numbers of cobalt-doped lanthanum gallate have also been measured using traditional electromotive force (EMF) method [2,9]. However, the accuracy of this method depends on the electrode polarization resistances of the concentration cell, and careful analysis and validation is necessary [10,11].

In general, when the electrode polarization resistances of a concentration cell are relatively small, the average ionic transference number of the electrolyte can be adequately approximated by the ratio of the open circuit voltage (OCV) of the cell to the Nernst potential imposed across the cell:

$$t_{0,1} = \frac{V_{oc}}{E_N} \quad (1)$$

Here V_{oc} is the OCV and E_N is the Nernst potential across the cell determined by the chemical conditions imposed on the cell.

When the electrode polarization resistances of a concentration cell are not negligible, however, the average ionic transference numbers can be approximated as follows [10,11]:

$$t_{0,2} = 1 - \frac{R_b}{R_T} \left(1 - \frac{V_{oc}}{E_N} \right) \quad (2)$$

where R_T and R_b are the total resistance and bulk resistance of the cell, respectively, and can be readily determined from impedance spectroscopy [11]. Clearly, Eq. (2) reduces to Eq. (1) as the electrode polarization resistance approaches 0 (or R_T approaches R_b). This approximation works very well for materials with predominant

Abbreviations: LSGM8282, La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3-δ}; LSGMC5, La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.15}Co_{0.05}O_{3-δ}; LSGMC8.5, La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.115}Co_{0.085}O_{3-δ}; MIEC, mixed ionic–electronic conductor; SOFC, solid oxide fuel cell; LT-SOFCs, low-temperature solid oxide fuel cells; EMF, electromotive force; OCV, open circuit voltage; SSC, Sm_{0.5}Sr_{0.5}CoO₃.

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Nomenclature

E_N	Nernst potential as defined by $E_N = (R_T/4F) \ln(P_{O_{2(c)}}/P_{O_{2(a)}})$
F	Faraday's constant (96,485 C mol ⁻¹)
n	number of electrons transferred in electrode reactions
$P_{O_{2(a)}}$	oxygen partial pressure at the anode/electrolyte interface
$P_{O_{2(c)}}$	oxygen partial pressure at the cathode/electrolyte interface
R	molar gas constant (8.314 J mol ⁻¹ K ⁻¹)
R_b	bulk resistance of a concentration cell
R_p	electrode polarization resistance of a concentration cell
R_T	total resistance of a concentration cell
ΔS	change in entropy
t_0	oxygen ion transference number
$t_{0,1}$	oxygen ion transference number determined using Eq. (1)
$t_{0,2}$	oxygen ion transference number determined using Eq. (2)
$t_{0,3}$	oxygen ion transference number determined using Eq. (3)
T	absolute temperature (K)
V_{oc}	open cell voltage

Greek letters

σ_0	conductivity due to the motion of oxygen vacancies or interstitials
σ_e	conductivity due to the motion of electrons or electron holes

Subscripts

O	oxygen ion
oc	open circuit
N	Nernst
b	bulk
T	total
P	polarization

ionic conduction (i.e., the ionic conductivity is much greater than the electronic conductivity). For materials with predominant electronic conduction (i.e., the electronic conductivity is much greater than the ionic conductivity), however, different approximations should be used [12].

When both the electronic and the oxygen ion conductivities of a material are measured in a wide range of oxygen partial pressures, the OCV of the concentration cell based on the material can be calculated using Wagner method, and thus the oxygen ion transference number can be estimated as follows [10]:

$$t_{0,3} = \frac{1}{E_N} \left(\frac{R_T}{4F} \right) \int_{P_1}^{P_2} \frac{\sigma_0}{\sigma_0 + \sigma_e} d \ln P_{O_2} \quad (3)$$

Here, σ_e and σ_0 are the electronic and oxygen ion conductivity of the electrolyte at a given partial pressure of oxygen, respectively; P_{O_2} is the local partial pressure of oxygen; P_1 and P_2 are the partial pressures of oxygen at the two electrode surfaces of the concentration cell, respectively.

In this study, the properties of LSGM8282, LSGMC5, and LSGMC8.5 are measured using concentration cells based on these materials and the results are analyzed using different approx-

imations, Eqs. (1)–(3), to estimate the oxygen ion transference numbers.

2. Experimental

La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.2}O_{3- δ} (LSGM8282), La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.15}Co_{0.05}O_{3- δ} (LSGMC5), and La_{0.8}Sr_{0.2}Ga_{0.8}Mg_{0.115}Co_{0.085}O_{3- δ} (LSGMC8.5) were prepared using a conventional solid-state reaction. The precursors used were La₂O₃ (99.99%), SrCO₃ (99.99%), MgO (99.99%), Ga₂O₃ (99.99%), and CoO (99.9%). Powders of the precursors in stoichiometric ratio were ball-milled in ethanol for 24 h. The mixture was then pre-calcined at 1273 K for 6 h. The resulting powders were then isostatically pressed into pellets (20 mm in diameter) at a pressure of 274.6 MPa for 10 min. The green pellets were then sintered at 1773 K for 6 h in air, followed by grinding and polishing down to a thickness of 1 mm. The phase compositions of the samples were verified using X-ray diffraction (Panalytical X'pert).

The densities of the LSGM8282, LSGMC5, and LSGMC8.5 samples were 6.39, 6.53, and 6.59 g cm⁻³, respectively, as measured using the Archimedes technique. The relative densities for all samples are higher than 98%.

Platinum paste or Ni8–Fe2–LSGMC8.5 [13] composite paste was screen printed on one side of the electrolyte as the anode. Platinum paste or Sm_{0.5}Sr_{0.5}CoO₃(SSC)–LSGMC5 [13] composite paste was applied to the other side of the electrolyte as the cathode. The geometry of the anode (5 mm in diameter) is identical to that of the cathode and the two electrodes were aligned as symmetrically as possible. The sintering temperature for Ni8–Fe2–LSGMC8.5 was 1473 K for 2 h, and that for Pt and SSC–LSGMC5 was 1173 K for 1 h. The test cells have a configuration of Ni8–Fe2–LSGMC8.5/electrolyte/SSC–LSGMC5 or Pt/electrolyte/Pt.

The anode/electrolyte/cathode assembly was then sealed between two alumina tubes using Pyrex glass to make a concentration cell. Pt mesh was covered on the electrodes using a spring system and connected to an electrochemical testing station using Pt wires. The anode was exposed to 100 ml min⁻¹ H₂ containing 3% water, and the cathode was exposed to 100 ml min⁻¹ pure oxygen or a mixture of high purity nitrogen and oxygen with a pre-determined oxygen partial pressure. Oxygen partial pressure of the gas for cathode was determined using an YSZ-based oxygen sensor.

The electronic and oxygen ion conductivities of LSGM8282, LSGMC5, and LSGMC8.5 are measured using four-probe impedance spectroscopy and Hebb–Wagner polarization method as reported elsewhere [8].

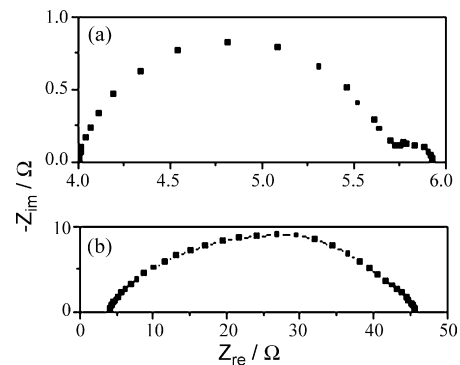


Fig. 1. Impedance spectra of cells with a configuration of (a) Ni8–Fe2–LSGMC8.5/LSGM8282/SSC–LSGMC5 and (b) Pt/LSGM8282/Pt cell at 1073 K under OCV with cathode exposed to pure oxygen and anode to hydrogen containing 3 vol.% water vapor.

Table 1

Electrode polarization resistance (R_p) and open circuit voltage (V_{oc}) measured at 1073 K of the cells based on electrolyte $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.2}\text{O}_{3-\delta}$ (LSGM8282), $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.15}\text{Co}_{0.05}\text{O}_{3-\delta}$ (LSGMC5), and $\text{La}_{0.8}\text{Sr}_{0.2}\text{Ga}_{0.8}\text{Mg}_{0.115}\text{Co}_{0.085}\text{O}_{3-\delta}$ (LSGMC8.5)

Materials	LSGM8282		LSGMC5		LSGMC8.5	
	Cell-1	Cell-2	Cell-1	Cell-2	Cell-1	Cell-2
R_p (Ω)	42.3	1.93	59.5	1.38	15.5	0.86
V_{oc} (V)	1.106	1.135	0.889	1.105	0.972	1.061
$t_{0,1}$	0.972	0.998	0.781	0.971	0.854	0.932

Also shown are the average oxygen ion transference numbers ($t_{0,1}$) of the electrolytes calculated using Eq. (1). Cell-1: Both cathode and anode are porous Pt electrodes. Cell-2—The cathode: porous SSC–LSGMC5; the anode: porous Ni8–Fe2–LSGMC8.5.

Open circuit voltages and impedances of the concentration cells were measured with a VMP/Z-40 (AMETECH) electrochemical testing station. The frequency for the impedance measurements was varied from 0.01 to 10^5 Hz, and the amplitude of the input sinusoidal signal was 10 mV.

3. Results and discussion

3.1. Approximation by the ratio of V_{oc} to E_N

Shown in Fig. 1(a) and (b) are the impedance spectra acquired at 1073 K under open circuit conditions for cells with a configuration of Ni8–Fe2–LSGMC8.5/LSGM8282/SSC–LSGMC5 and Pt/LSGM8282/Pt, respectively. The anode was exposed to humidified hydrogen (containing 3 vol.% water vapor) whereas the cathode to pure oxygen. The size of the impedance loop corresponds to the total electrode polarization resistance of the cell, and the intercept at high frequency corresponds to the bulk resistance of the electrolyte. Fig. 1 shows that the electrode polarization resistances of the cell with SSC–LSGMC5 cathode and Ni8–Fe2–LSGMC8.5 anode are much lower than those of the cell with Pt electrodes. The ohmic

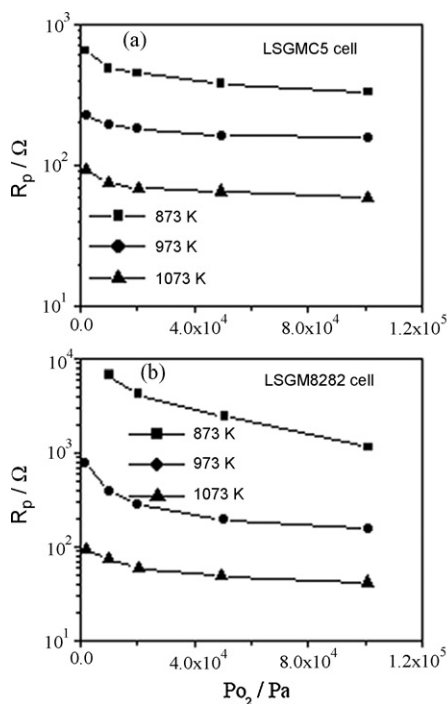


Fig. 2. Electrode polarization resistances of concentration cells with a configuration of (a) Pt/LSGMC5/Pt and (b) Pt/LSGM8282/Pt as a function of oxygen partial pressure in the cathode compartment at different temperatures.

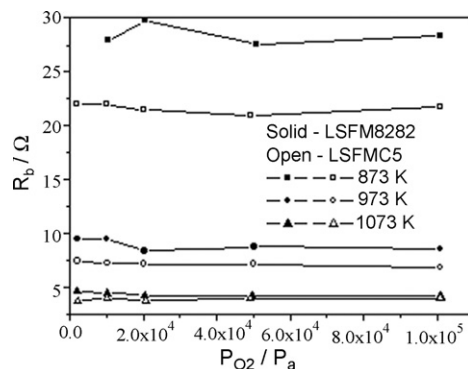


Fig. 3. Bulk resistances of the cells based on LSGM8282 and LSGMC5 as a function of oxygen partial pressure in the cathode compartment at different temperatures.

resistances of the two cells are similar, and are close to the theoretical value calculated from the conductivity of LSGM8282 [8], suggesting that the ohmic resistances of the cells are determined predominantly by the resistance of the electrolyte.

Shown in Table 1 are the electrode polarization resistances and OCVs of several cells measured at 1073 K, and the average oxygen ion transference numbers of the corresponding electrolytes as calculated using Eq. (1).

As can be clearly seen from the data shown in Table 1, the OCV of a concentration cell depends strongly on the electrode polarization resistances of the cell. The cell with a larger electrode polarization resistance has a lower OCV, thus leading to a smaller oxygen ion transference number. The effect of polarization resistance on OCV and on oxygen ion transference number is more pronounced for cobalt-doped lanthanum gallate.

3.2. Approximation by Eq. (2)

Oxygen ion transference numbers of LSGM8282, LSGMC5, and LSGMC8.5 were further estimated using Eq. (2) to take into consideration the effect of electrode polarization resistances on OCV under various testing conditions. Shown in Fig. 2(a) and (b) are the electrode polarization resistances, R_p , of the two concentration cells (with a configuration of Pt/LSGMC5/Pt and Pt/LSGM8282/Pt, respectively), as determined from impedance spectra acquired. The partial pressure of oxygen in the cathode compartment was systematically varied to observe its effect on cathode polarization resistance. As expected, the electrode polarization resistances decreased with increasing temperature and partial pressure of oxygen. The cells based on LSGMC5 electrolyte have a lower

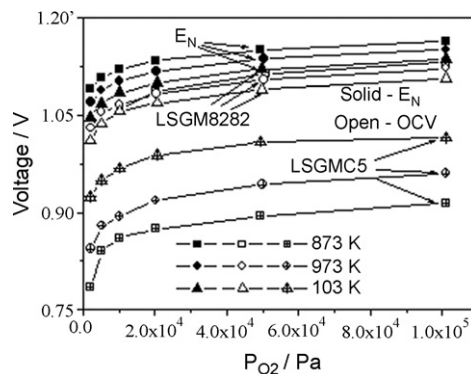


Fig. 4. Open circuit voltages and Nernst potentials of concentration cells, Pt/LSGMC5/Pt and Pt/LSGM8282/Pt, measured at different temperatures as a function of oxygen partial pressure in the cathode compartment.

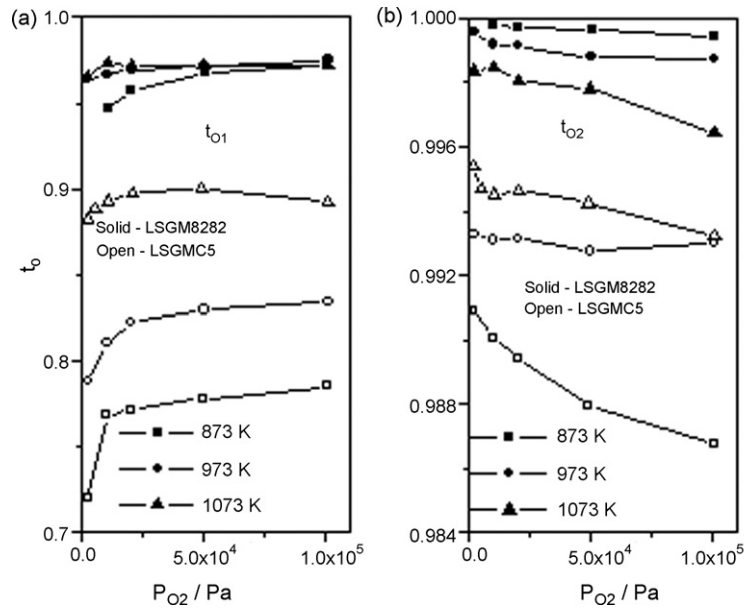


Fig. 5. Oxygen ion transference numbers of LSGM8282 and LSGMC5 calculated using Eq. (1) and (2) as a function of oxygen partial pressure in the cathode compartment.

polarization resistance under the same conditions, especially at low temperatures, due probably to the improved catalytic activity for oxygen reduction by the addition of cobalt to the electrolyte [9].

Shown in Fig. 3 are the bulk resistances of the two concentration cells, Pt/LSGM8282/Pt and Pt/LSGMC5/Pt, as a function of oxygen partial pressure in the cathode compartment at different temperatures. The bulk resistances of the cells decreased with increasing temperature, but were relatively insensitive to the change in partial pressure of oxygen. The bulk resistances of the cells based on LSGMC5 are slightly lower than those based on LSGM8282, consistent with the conductivities reported previously for these materials [8].

Shown in Fig. 4 are the measured OCVs of and the Nernst potential (E_N) imposed on the two concentration cells, Pt/LSGMC5/Pt and Pt/LSGM8282/Pt, under different testing conditions. The Nernst potential imposed on the cells can be calculated as follows:

$$E_N = \frac{R_T}{4F} \ln \frac{P_{O_{2(c)}}}{P_{O_{2(a)}}} \quad (4)$$

where $P_{O_{2(c)}}$ and $P_{O_{2(a)}}$ are the oxygen partial pressure of the gases in the cathodic and anodic compartment, respectively. Therefore, E_N increases with increasing partial pressure of oxygen in the cathode compartment, $P_{O_{2(c)}}$, while $P_{O_{2(a)}}$ was kept constant.

The dependence of E_N of a fuel cell on temperature under constant pressure can be expressed as [14]:

$$\left(\frac{\partial E_N}{\partial T} \right)_p = \frac{\Delta S}{nF} \quad (5)$$

Here, ΔS is the change in entropy, n is the number of electrons transferred in the reaction. For a H_2 – O_2 SOFC, the overall reaction

can be expressed as



Therefore, the change in entropy is negative, implying that the E_N decreases with temperature. The Nernst potentials and the OCVs of the cells based on a LSGM8282 electrolyte, as shown in Fig. 4, have the same trend as predicted by Eq. (5). However, the cells based on a LSGMC5 electrolyte showed a higher OCV at higher temperatures. Further, the OCV of the cell based on a LSGM8282 electrolyte was slightly lower than the E_N , whereas the OCV of the cell based on a LSGMC5 electrolyte is much lower than the E_N , suggesting a lower oxygen ion transference number for LSGMC5 than for LSGM8282.

With the electrode polarization resistances shown in Fig. 2, bulk resistances shown in Fig. 3, and the Nernst potential and OCV shown in Fig. 4, oxygen ion transference numbers for LSGM8282 and LSGMC5 are calculated using Eqs. (1) and (2) and plotted in Fig. 5(a) and (b), respectively. The values predicted from Eq. (1) are much smaller than those from Eq. (2), suggesting that electrode polarization resistances had significant effect on estimation of oxygen ion transference numbers. The $t_{0,1}$ from Eq. (1) for both LSGM8282 and LSGMC5 increased with increasing partial pressure of oxygen, while $t_{0,2}$ from Eq. (2) decreased with increasing partial pressure of oxygen. The electronic conductivity for both LSGM8282 and LSGMC5 increased with increasing partial pressure of oxygen within the oxygen partial pressure range studied as reported previously [8], suggesting that oxygen ion transference number should decrease with increasing partial pressure of oxygen. Therefore, the approximation using Eq. (2) is more reasonable considering the P_{O_2} dependences of oxygen ion transference number. Fig. 5(b) further shows that $t_{0,2}$ of LSGM8282 decreased with increasing temperature, while that

Table 2

Oxygen ion transference numbers of the three samples at 873, 973 and 1073 K calculated using Eq. (1) ($t_{0,1}$), Eq. (2) ($t_{0,2}$), and Eq. (3) ($t_{0,3}$)

	873 K		973 K			1073 K		
	$t_{0,2}$	$t_{0,1}$	$t_{0,3}$	$t_{0,2}$	$t_{0,1}$	$t_{0,3}$	$t_{0,2}$	$t_{0,1}$
LSGM8282	0.999	0.972	0.999	0.999	0.975	0.999	0.996	0.972
LSGMC5	0.987	0.785	0.996	0.993	0.834	0.997	0.993	0.892
LSGMC8.5	0.954	0.689	0.977	0.962	0.781	0.987	0.961	0.854

The concentration cells were exposed to 97% H_2 + 3 vol.% H_2O on the anode side and to oxygen on the cathode side.

of LSGMC5 increased with increasing temperature. This could be attributed to the additional electronic defects created by the redox reaction involving Co at low temperatures [5–8]. However, the oxygen ion transference number of LSGMC5 is still close to 0.99 even at 873 K, suggesting that it is a promising electrolyte for LT-SOFCs.

The apparent OCV of a concentration cell can be expressed as [10,11]:

$$V_{oc} = E_{NtO} \left[1 + \frac{R_p}{R_0 + R_e} \right]^{-1} \quad (7)$$

This equation implies that the OCV of a concentration cell increases as the electrode polarization resistance is reduced, approaching a constant value as R_p gets much smaller than the ionic and electronic resistance, i.e., $R_p \ll R_0 + R_e$. The effect of electrode polarization resistance on the measurement of oxygen ion transference number depends strongly on the oxygen ion and electronic conductivity of the material as well.

The oxygen ion transference numbers of LSGM8282, LSGMC5, and LSGMC8.5 estimated using both Eqs. (1) and (2) are summarized in Table 2. The anode was exposed to humidified H_2 (with 3 vol.% H_2O) whereas the cathode to pure oxygen. The results show that oxygen ion transference number of cobalt-doped lanthanum gallate decreases with increasing content of cobalt in the electrolyte. However, even LSGMC8.5 shows an oxygen ion transference number as high as 0.96.

3.3. Wagner's method

The oxygen ion transference numbers for the cobalt-doped lanthanum gallate electrolytes at 973 and 1073 K were also calculated using Eq. (3) (the Wagner's method) and summarized in Table 2. The oxygen partial pressure range in Eq. (3) varied from 97% H_2 with 3 vol.% water vapor (on the anode side) to pure oxygen (on the cathode side). The values calculated from Eq. (3) are very close to those obtained from Eq. (2), confirming that the cobalt-doped lanthanum gallates are promising electrolyte for LT-SOFCs as long as the cobalt is less than 8.5 mol%. The results also show that the approximation of the oxygen ion transference number by the ratio of V_{oc} to E_N is inadequate for these materials; the error in the approximation increases with increasing electronic conductivity of the materials and the electrode polarization resistance of the concentration cell. Also, it is noted that the use of Eq. (3) requires the electronic and

ionic conductivities of the material measured in a wide range of oxygen partial pressure. In contrast, the use of Eq. (2) requires much less information about the material; the required R_b and R_T can be readily determined from a single impedance spectrum under the conditions the OCV is measured.

4. Conclusions

The error in determining oxygen ion transference number using the ratio of V_{oc} to E_N depends sensitively on the electrode polarization resistance of the concentration cell. While the ratio of V_{oc} to E_N of a concentration cell is an adequate approximation to the oxygen ion transference number of LSGM8282, it is inadequate for that of LSGMC5 and LSGMC8.5, which can be properly determined only by using Eq. (2) or (3). The oxygen ion transference number of LSGMC5 is as high as 0.99, suggesting that it is a promising electrolyte material for LT-SOFCs.

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