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EFFECT OF TEMPERATURE ON ELECTRICAL PROPERTIES OF YTTRIA-DOPED CERIA AND YTTRIA-STABILIZED ZIRCONIA

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Abstract. Electrical properties of yttria-doped ceria and yttria-stabilized zirconia with fluorite structure have been investigated using statistical moment method. Lattice constants, vacancy activation energies, diffusion coefficients, ionic conductivities are calculated as a function of temperature. Numerical calculations have been performed using the Buckingham potential and compared with those of the experimental and other theoretical results showing the reasonable agreements. *Keywords:* Temperature, electrical property, yttria-doped ceria, yttria-stabilized zirconia, statistical moment method.

1. Introduction

Oxides with the cubic fluorite structure, e.g., ceria (CeO₂) and zirconia (ZrO₂) are important ionic conductors when they are doped with cations of lower valence than the host cations. Oxygen ion transport in the crystal lattice is mainly based on the vacancy hopping mechanism. By replacing R^{4+} ions (R^{4+} is a general symbol for Ce⁴⁺ and Zr⁴⁺ ions) Y^{3+} ions, oxygen vacancies are generated in the anion sublattice to maintain overall charge neutrality in crystal lattice [1, 2]. Yttria-doped ceria (YDC) and yttriastabilized zirconia (YSZ) have the high conductivities thus making them attractive electrolytes for solid oxide fuel cells (SOFCs) [3, 4]. Nowadays, SOFCs are used extensively due to high power density, high energy-conversion efficiency, low emissions and fuel flexibility [5, 6].

So far, a great large number of experimental and theoretical studies have been performed to investigate electrical properties of YDC and YSZ. Many approaches have been used to study YDC and YSZ such as the lattice dynamics (MD) [7], Monte-Carlo (MC) simulations [8], density functional theory (DFT) [9]. These studies showed the dependence of electrical properties on dopant concentration of YDC and YSZ. The diffusion

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coefficients decrease with the increasing dopant concentration. The ionic conductivities firstly increase up to about the yttrium concentration $x \approx 0.12$ in YDC and 7 - 8 % Y₂O₃ in YSZ and then decrease at higher values of the dopant concentration. Oxygen vacancy-dopant associations and oxygen vacancy migration limited across the cation barriers at high dopant concentration are responsible for the presence of the maximum in the ionic conductivities.

Up to now, however, the temperature dependence of the electrical properties of YDC and YSZ are not fully understood and previous calculations without including the thermal lattice vibration effects. The purpose of this study is to study the effect of temperature on the electrical properties of YDC and YSZ in within the statistical moment method (SMM) scheme in statistical mechanics. The lattice constants, vacancy activation energies, diffusion coefficients, ionic conductivities of YDC and YSZ will be considered talking into account the anharmonicity of thermal lattice vibrations.

2. Content

2.1. Theory

Compounds CeO₂ and ZrO₂ crystallize in the fluorite structure. At the atomic scale, the 8-coordinated R⁴⁺ ions form a face-centered cubic lattice with a cell parameter equal to about 5.4 Å for CeO₂ and 5.08 Å for ZrO₂. Oxygen ions occupy the tetrahedral sites forming a simple cubic sublattice. The open fluorite structure allows O²⁻ ions to hop through the lattice with relative ease. The models of cubic-fluorite YDC and YSZ are obtained by substituting *x* percent of R⁴⁺ ions in YDC and YSZ is denoted by *x* and there are *N* cations in the crystal lattice, then the numbers of R⁴⁺, Y³⁺ and O²⁻ ions and oxygen vacancies in YDC and YSZ are $N_R = N(1-x)$, $N_Y = Nx$, $N_O = N(2-x/2)$, $N_{va} = Nx/2$, respectively. Therefore, the general chemical formula of YDC and YSZ can be written as R_{1-x}Y_xO_{2-x/2}.

2.1.1. Helmholtz free energy

The general expression of Helmholtz free energy of CeO_2 (or ZrO_2) with the fluorite structure is given from the SMM as in Ref [10].

$$\Psi = C_R \Psi_R + C_O \Psi_O - TS_c, \qquad (1)$$

where C_R , C_o denote concentrations of \mathbb{R}^{4+} , \mathbb{O}^{2-} ions, respectively, S_c is the configuration entropy, and Ψ_R , Ψ_o are the Helmholtz free energies of \mathbb{R}^{4+} , \mathbb{O}^{2-} ions, respectively,

$$\Psi_{R} = U_{0}^{R} + \Psi_{0}^{R} + 3N_{R} \left\{ \frac{\theta^{2}}{k_{R}^{2}} \left[\gamma_{R}^{2} X_{R}^{2} - \frac{2\gamma_{R}^{1}}{3} a_{1}^{R} \right] + \frac{2\theta^{3} a_{1}^{R}}{k_{R}^{4}} \left[\frac{4}{3} \left(\gamma_{2}^{R} \right)^{2} X_{R} - 2 \left(\left(\gamma_{1}^{R} \right)^{2} + 2\gamma_{1}^{R} \gamma_{2}^{R} \right) \left(2a_{1}^{R} - 1 \right) \right] \right\},$$
(2)

69

Le Thu Lam, Vu Van Hung and Nguyen Thanh Hai

$$\begin{split} \Psi_{o} &= U_{0}^{o} + \Psi_{0}^{o} + 3N_{o} \left\{ \frac{\theta^{2}}{k_{o}^{2}} \left[\gamma_{o}^{2} X_{o}^{2} - \frac{2\gamma_{o}^{1}}{3} a_{1}^{o} \right] \right. \\ &+ \frac{2\theta^{3} a_{1}^{o}}{k_{o}^{4}} \left[\frac{4}{3} \left(\gamma_{2}^{o} \right)^{2} X_{o} - 2 \left(\left(\gamma_{1}^{o} \right)^{2} + 2\gamma_{1}^{o} \gamma_{2}^{o} \right) \left(2a_{1}^{o} - 1 \right) \right] \right\} \\ &+ 3N_{o} \left\{ \frac{\theta\beta}{6K\gamma_{o}} \left(\frac{k_{o}}{K} - 1 \right) + \frac{\theta^{2}\beta}{K} \left[\left[\left(\frac{2\gamma_{o} a_{1}^{o}}{3K^{3}} \right)^{\frac{1}{2}} - \frac{\beta a_{1}^{o}}{9K^{2}} + \frac{\beta k_{o} a_{1}^{o}}{9K^{3}} + \frac{\beta}{6Kk_{o}} \left(X_{o} - 1 \right) \right) \right] \right\}, (3) \end{split}$$

here, U_0^R , U_0^O represent the sums of effective pair interaction energies for \mathbb{R}^{4+} , \mathbb{O}^{2-} ions, respectively, and Ψ_0^R , Ψ_0^O denote the harmonic contributions of \mathbb{R}^{4+} , \mathbb{O}^{2-} ions to the free energies with the general formula as $\Psi_0 = 3N\theta \left[x + \ln \left(1 - e^{-2x} \right) \right]$. The parameters $k_{R,O}$, $x_{R,O}$, $a_1^{R,O}$, β , K, $\gamma_1^{R,O}$, $\gamma_{2,O}^{R,O}$, are defined as in Ref. [11].

In order to determine the Helmholtz free energy $R_{1-x}Y_xO_{2-x/2}$ system, we consider the change of Helmholtz free energy Ψ when $N_Y R^{4+}$ ions of system $RO_{2-x/2}$ are replaced by Y^{3+} ions. Firstly, the substitution of a R^{4+} ion by a Y^{3+} ion creates the change of the free Gibbs energy as

$$g_{\nu}^{f} \approx -u_{0}^{R} + \psi_{\gamma}, \qquad (4)$$

with u_0^R is the average interaction potential of a R⁴⁺ ion in RO_{2-x/2} system, ψ_Y is the Helmholtz free energy of a Y³⁺ ion in R_{1-x}Y_xO_{2-x/2} system. Because R_{1-x}Y_xO_{2-x/2} system is built by the substitution of N_Y Y³⁺ ions for N_Y R⁴⁺ ions in RO_{2-x/2} system, then the Gibbs free energy of R_{1-x}Y_xO_{2-x/2} system can be determined by the Gibbs free energy of RO_{2-x/2} system, G₀,

$$G = G_0 + N_Y g_v^f - TS_c^*$$

= $\Psi_{RO_{2-x/2}} + N_Y \left(-u_0^R + \psi_Y \right) + PV - TS_c^*.$ (5)

where *P* is the hydrostatic pressure, *V* is the volume of $R_{1-x}Y_xO_{2-x/2}$ system and S_c^* is the configuration entropies of $R_{1-x}Y_xO_{2-x/2}$ system. Using Eq. (5), the Helmholtz free energy of $R_{1-x}Y_xO_{2-x/2}$ system can be written as

$$\Psi = \Psi_{RO_{2-x/2}} + N_Y \left(-u_0^R + \psi_Y \right) - TS_c^*$$

= $\Psi_{RO_{2-x/2}} + \Psi_Y - N_Y u_0^R - TS_c^*,$ (6)

with Ψ_{Y} is the Helmholtz free energies of Y^{3+} ions in $R_{1-x}Y_{x}O_{2-x/2}$ system,

$$\Psi_{Y} = U_{0}^{Y} + \Psi_{0}^{Y} + 3N_{Y} \left\{ \frac{\theta^{2}}{k_{Y}^{2}} \left[\gamma_{Y}^{2} X_{Y}^{2} - \frac{2\gamma_{Y}^{1}}{3} a_{1}^{Y} \right] + \frac{2\theta^{3} a_{1}^{Y}}{k_{Y}^{4}} \left[\frac{4}{3} \left(\gamma_{2}^{Y} \right)^{2} X_{Y} - 2 \left(\left(\gamma_{1}^{Y} \right)^{2} + 2\gamma_{1}^{Y} \gamma_{2}^{Y} \right) \left(2a_{1}^{Y} - 1 \right) \right] \right\},$$
(7)

70

here, U_0^Y is the sum of effective pair interaction energy for Y^{3+} ions in $R_{1-x}Y_xO_{2-x/2}$ system. The expressions of k_Y , x_Y , a_1^Y , γ_1^Y , γ_2^Y , γ_Y have the same forms as those of k_R , , x_R , a_1^R , γ_1^R , γ_2^R , γ_R , respectively.

In addition, the average nearest-neighbor distance of YDC and YSZ at temperature T can be written as

$$r_1(T) = r_1(0) + C_R y_0^R + C_Y y_0^Y + C_O y_0^O,$$
(8)

where $r_1(0)$ is the distance r_1 at zero temperature which be determined from experiment or the minimum condition of the interaction potential of system, and y_0^R , y_0^Y , y_0^O are the average displacements of \mathbb{R}^{4+} , \mathbb{Y}^{3+} , \mathbb{O}^{2-} ions from the equilibrium position at temperature *T*, respectively. Here, y_0^R , y_0^O are defined as Ref. [11] and the expression of y_0^Y is the same form as that of y_0^R . The lattice constant of YDC and YSZ at temperature *T* can be calculated by using the relation $a_{lat}(T) = r_1(T) 4/\sqrt{3}$.

2.1.2. Diffusion coefficient and ionic conductivity

In the following presentation, we outline the calculation of the diffusion coefficients and ionic conductivities of YDC and YSZ within the SMM scheme basing on the general expression of the Helmholtz free energy of Eq. (6). In YDC and YSZ, the current is carried by oxygen ions that are transported by oxygen vacancies. So the diffusion coefficients and ionic conductivities are closely related to the vacancy formation and migration properties. The diffusion coefficient and ionic conductivity of the materials with fluorite structure are given by [12, 13].

$$D = \frac{D_0}{T} \exp\left(-\frac{E_a}{k_B T}\right), \text{ with } D_0 = n_1 f \nu r_1^2 \exp\left(\frac{S_\nu^f}{k_B}\right), \tag{9}$$

$$\sigma = \frac{\sigma_0}{T} \exp\left(-\frac{E_a}{k_B T}\right), \text{ with } \sigma_0 = \frac{nq^2 n_1 f \nu r_1^2 \exp\left(\frac{S_\nu}{k_B}\right)}{k_B}, \tag{10}$$

where n_1 is the number of O^{2^-} ions at the first nearest neighbour positions with regard to the oxygen vacancy, the factor f is correlation factor, v is the vibration frequency of the O^{2^-} ions, r_1 is the shortest distance between two lattice sites containing the O^{2^-} ions, S_v^f is entropy for the formation of a vacancy, n is the vacancy concentration and being

$$n = (8/a^3) \exp\left(-\frac{E_{ass}}{k_B}\right)$$
 for the fluorite structure, and E_a is vacancy activation energy.

For doped ceria and zirconia, E_a can be determined as the sum of vacancy-dopant association energy, E_{ass} , and vacancy migration energy, E_m , [1, 8]

$$E_a = E_{ass} + E_m. \tag{11}$$

The association energy E_{ass} between an oxygen vacancy and a Y^{3+} ion is the energy difference between the systems containing the oxygen vacancies and Y^{3+} ions at the

associated state and the systems containing the oxygen vacancies and Y^{3+} ions at the isolated state [3]. In this study, the expression of the vacancy-dopant association energy can be written as

$$E_{ass} = \left(\Psi_{R_{N_{R}}Y_{N_{Y}}O_{N_{O}}} + \Psi_{R_{N_{R}-2}Y_{N_{Y}+2}O_{N_{O}-1}}\right) - \left(\Psi_{R_{N_{R}-1}Y_{N_{Y}+1}O_{N_{O}}} + \Psi_{R_{N_{R}-1}Y_{N_{Y}+1}O_{N_{O}-1}}\right),$$
(12)

with $\Psi_{R_{N_R}Y_{N_V}O_{N_O}}$, $\Psi_{R_{N_R-2}Y_{N_Y+2}O_{N_O-1}}$, $\Psi_{R_{N_R-1}Y_{N_Y+1}O_{N_O}}$, and $\Psi_{R_{N_R-1}Y_{N_Y+1}O_{N_O-1}}$ are the Helmholtz free energies of the systems as $R_{N_R}Y_{N_V}O_{N_O}$, $R_{N_R-2}Y_{N_Y+2}O_{N_O-1}$, $R_{N_R-1}Y_{N_Y+1}O_{N_O}$, $R_{N_R-1}Y_{N_Y+1}O_{N_O-1}$, respectively, and they are calculated by using Eq. (6). The $R_{N_R}Y_{N_V}O_{N_O}$ system contains $N_{\nu a}$ oxygen vacancies and N_Y Y³⁺ ions at the associated state, while the $R_{N_R-1}Y_{N_Y+1}O_{N_O}$ system has more a Y³⁺ ion at the isolated state and the $R_{N_R-1}Y_{N_Y+1}O_{N_O-1}$ system has more an isolated oxygen vacancy. Similar with the former, the $R_{N_R-2}Y_{N_Y+2}O_{N_O-1}$ system also consists of $(N_{\nu a} + 1)$ oxygen vacancies associated with $(N_Y + 2)$ Y³⁺ ions. Here, it is noted that each oxygen vacancy is associated with two Y³⁺ ions because the substitution of Y³⁺ ions for R⁴⁺ is accompanied by the formation of an oxygen vacancy for every two Y³⁺ ions.

The vacancy migration energy E_m is given by

$$E_m = \Psi_0 - \Psi_{saddle}.$$
 (13)

with Ψ_0 , Ψ_{saddle} are the Helmholtz free energies of the crystal lattice before an oxygen migration from the lattice site (called as initial energy), and after the oxygen ion diffusion to the saddle point (so-called saddle point energy), respectively. Eq. (6) enables us to calculate the initial energy and the saddle point energy of the crystal lattice based on the total interaction potentials U_0^R , U_0^Y , U_0^O of R^{4+} , Y^{3+} , O^{2-} ions, respectively, at the initial and saddle point states [14].

2.2. Result and discussion

The ionic interaction in YDC and YSZ with fluorite structure is divided into Coulomb long-range interaction (summated by the Wolf method) and short-range interactions described by the Buckingham function [7, 13]

$$\varphi_{ij}\left(r\right) = \frac{q_i q_j}{r} + A_{ij} \exp\left(-\frac{r}{B_{ij}}\right) - \frac{C_{ij}}{r^6},$$
(14)

where q_i and q_j are the charges of the *i*-th and the *j*-th ions, *r* is the distance between them and the parameters A_{ij} , B_{ij} and C_{ij} are empirically determined (listed in Table 1).

Firstly, we present the lattice constants of YDC (Figure 1a) and YSZ (Figure 1b) calculated at the different temperatures by the SMM formalism, together with experimental data [15, 16] [in the case of pure CeO₂]. It is noted that the relation between the yttria concentration y and the yttrium concentration x is y = x/(2-x) [17]. Overall good agreements between the calculation and experimental results are obtained for a wide temperature range. One can see that the lattice constants increase smoothly

with an increase of temperature due to the thermal expansion. Our results at the different dopant concentrations also show that the lattice constant of YDC decreases with an increase of the dopant concentration, while that of YSZ increases with the increasing dopant concentration.

Material	Interaction	A_{ij} /eV	B_{ij} / $\overset{ m o}{ m A}$	C_{ij} / eV. (Å) ⁶
YDC [15]	$0^{2} - 0^{2}$	9547.96	0.2192	32.00
	$Ce^{4+}-O^{2-}$	1809.68	0.3547	20.40
	$Y^{3+} - O^{2-}$	1766.4	0.3385	19.43
YSZ [16]	$O^{2-}O^{2-}$	9547.96	0.224	32
	$Zr^{4+}-O^{2-}$	1502.11	0.345	5.1
	$Y^{3+} - O^{2-}$	1366.35	0.348	19.6

Table 1. The parameters of the Buckingham potential in YDC and YSZ



Figure 1. The temperature dependence of the lattice constants of YDC (a) and YSZ (b)at the various dopant concentrations

Using Eqs. (12) and (13), we can determine the vacancy-dopant association energy and the migration energy as a function of the temperature T. From these results, the activation energy E_a can be easily calculated based on Eq. (11). In Figure 2, we show the theoretical calculations of the activation energies of YDC (a) and YSZ (b) at the various dopant concentrations. When the temperature increases, the ions vibrate more strongly to restrict the movement of the oxygen vacancies and lead to an increase of the migration energy. Because the migration energy increases quickly with the increasing temperature, the activation energy also increases with an increase in the temperature. Moreover, one can see that the activation energy of YDC is slightly smaller than that of YSZ at the same temperature and dopant concentration.

Le Thu Lam, Vu Van Hung and Nguyen Thanh Hai



Figure 2. The temperature dependence of activation energies of YDC (a) and YSZ (b) at the various dopant concentrations



Figure 3. The temperature dependence of the diffusion coefficients and ionic conductivities of YDC (a,c) and YSZ (b,d) at the various dopant concentrations

The diffusion coefficient D and ionic conductivity σ are the specific electrical quantities of the ionic conductors. Their dependence on the temperature is showed in Figures 3a and 3c for YDC and Figures. 3b and 3d for YSZ. The experimental results of the diffusion coefficients and ionic conductivities of YDC and YSZ [18, 21] are also displayed for comparison. Although an increase in the temperature will increase the values of activation energies, the migration velocity of oxygen vacancies is strongly affected by the temperature. Therefore, both the diffusion coefficients and ionic

conductivities increase with the increasing temperature. Moreover, Figure 3 shows the larger values of diffusion coefficients at the smaller dopant concentrations. We predict that the diffusion coefficients decrease with the increasing dopant concentration. The calculated results are consistent with the experimental data [18-21].

3. Conclusions

The SMM calculations are performed using the Buckingham potential for YDC and YSZ with fluorite structure. The quantities related electrical properties of YDC and YSZ are calculated as a function of the temperature. The activation energies increase with the increasing temperature but the diffusion coefficients, ionic conductivities increase with an increase in temperature. This is originating from the strong dependence of vacancy migrate velocity on the temperature. The calculated lattice constants, activation energies, diffusion coefficients, ionic conductivities are compared with the theoretical and experimental results. The dopant concentration-dependence of these quantities will be discussed in our next report.

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