Investigation of oxygen vacancy migration energy in yttrium doped cerium

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Abstract. Using the statistical moment method, the analytic expression of vacancy migration energy in yttria doped cerium (YDC) is derived within the fourth order moment approximation. This expression depends strongly on the interaction potentials between the diffusing oxygen ion and the surrounding cations. Our results show the predominant direction of oxygen vacancy migration and the influence of cation barriers on oxygen vacancy diffusion. The dependences of the migration energy on temperature and dopant concentration are evaluated in detail.
Calculated results are compared with other theoretical results.

1. Introduction
Ceria (CeO\textsubscript{2}) is not good ionic conductor. However, doping of ceria with divalent or trivalent cations creates oxygen vacancies, and increases the concentration of oxygen vacancies compared to pure ceria. In yttrium doped cerium (YDC), there are many vacancies generated in the anion sublattice to maintain overall charge neutrality in crystal lattice [1-4]. Because of the high diffusion coefficient, YDC is one of the most popular electrolytes in solid oxide fuel cells [5].

To study the oxygen vacancy diffusion in YDC, it is necessary to calculate vacancy migration energy. Using density-functional theory (DFT), M. Nakayama \textit{et al.} [6] showed that oxygen ions hop dominantly along the <100> direction connecting two nearest neighbor oxygen sites. They concentrated on five types of nearest neighbor cation configurations around the diffusing oxygen ion. Their results indicated that the increase of Y\textsuperscript{3+} concentration causes a decrease of the vacancy migration energy, and vice versa. The vacancy migration energy in CeO\textsubscript{2} was calculated in detail using statistical moment method (SMM) [7]. Therein, the oxygen vacancies are thermally generated and the vacancy migration energy equal eighth the interaction potential of an oxygen ion.

In YDC, the most oxygen vacancies are generated due to doping and the dopant ions influence strongly on the hopping of oxygen vacancies. In this study, we will perform the expressions to calculate the vacancy migration energy. These expressions depend on cation configuration surrounding the diffusing oxygen ion. The predominant direction of oxygen vacancy migration and the influence of cation barriers on oxygen vacancy diffusion is evaluated in detail. Our results also show the dependences of the vacancy migration energy on temperature and dopant concentration.
2. Theory approach

2.1. Free energy
In the fluorite structure of YDC, Ce\textsuperscript{4+} ions occupy a face-centered cubic (fcc) lattice and O\textsuperscript{2−} ions occupy the tetrahedral in interstitial sites forming a simple cubic sublattice of length a/2. Free energy of CeO\textsubscript{2} with the fluorite structure was studied by V.V. Hung et al. by SMM formulation [8,9]. The free energy of YDC can be determined by concentration distribution

$$
Ψ = C_{Ce}Ψ_{Ce} + C_{Y}Ψ_{Y} + C_{O}Ψ_{O} - TS_c,
$$

where $C_{Ce}$, $C_{Y}$, $C_{O}$ denote concentrations of Ce\textsuperscript{4+}, Y\textsuperscript{3+} and O\textsuperscript{2−} ions, respectively, and $Ψ_{Ce}, Ψ_{Y}, Ψ_{O}$ are the total Helmholtz free energies of Ce\textsuperscript{4+}, Y\textsuperscript{3+} and O\textsuperscript{2−} ions, respectively, and $S_c$ the configurational entropy. The total free energies of Ce\textsuperscript{4+}, Y\textsuperscript{3+} and O\textsuperscript{2−} ions are defined by

$$
Ψ_{Ce} = U_{0}^{Ce} + Ψ_{0}^{Ce} + 3N_{Ce} \left\{ \frac{θ^2}{k_{Ce}} \left[ \frac{γ_{Ce}^2}{2} x_{Ce}^2 x_{Ce} coth x_{Ce} - \frac{1}{3} a_{1}^{Ce} \right] \right\} + \frac{2θ^2}{k_{Ce}} \left\{ \frac{4}{3} (γ_{Ce}^2) a_{1}^{Ce} x_{Ce} coth x_{Ce} - 2 \left( (γ_{Ce}^2) + 2 (γ_{Ce}^2) a_{1}^{Ce} (2a_{1}^{Ce} - 1) \right) \right\},
$$

$$
Ψ_{Y} = U_{0}^{Y} + Ψ_{0}^{Y} + 3N_{Y} \left\{ \frac{θ^2}{k_{Y}} \left[ \frac{γ_{Y}^2}{2} x_{Y}^2 x_{Y} coth x_{Y} - \frac{1}{3} a_{1}^{Y} \right] \right\} + \frac{2θ^2}{k_{Y}} \left\{ \frac{4}{3} (γ_{Y}^2) a_{1}^{Y} x_{Y} coth x_{Y} - 2 \left( (γ_{Y}^2) + 2 (γ_{Y}^2) a_{1}^{Y} (2a_{1}^{Y} - 1) \right) \right\},
$$

$$
Ψ_{O} = U_{0}^{O} + Ψ_{0}^{O} + 3N_{O} \left\{ \frac{θ^2}{k_{O}} \left[ \frac{γ_{O}^2}{2} x_{O}^2 coth x_{O} - \frac{1}{3} a_{1}^{O} \right] \right\} + \frac{2θ^2}{k_{O}} \left\{ \frac{4}{3} (γ_{O}^2) a_{1}^{O} x_{O} coth x_{O} - 2 \left( (γ_{O}^2) + 2 (γ_{O}^2) a_{1}^{O} (2a_{1}^{O} - 1) \right) \right\} + \frac{θ^2 β_{O}}{9K^3} \left\{ \left( \frac{2a_{1}^{O}}{9K^2} \right)^{1/2} - \frac{β_{O} a_{1}^{O}}{9K^2} \right\} + \frac{θ^2 K_{O} a_{1}^{O}}{9K^3} \left\{ \left( x_{O} coth x_{O} - 1 \right) \right\},
$$

where

$$
k_{Ce,Y} = \frac{1}{2} \sum_{i} \left( \frac{∂^2 Ψ_{Ce,Y}}{∂u_{i,j}^2} \right), \quad x_{Ce,Y} = h \sqrt{\frac{k_{Ce,Y}}{m}}, \quad a_{1}^{Ce,Y} = 1 + \frac{1}{2} x_{Ce,Y} coth x_{Ce,Y},
$$

$$
γ_{1}^{Ce,Y} = \frac{1}{48} \sum_{i} \left( \frac{∂^4 Ψ_{Ce,Y}}{∂u_{i,j}^4} \right), \quad γ_{2}^{Ce,Y} = 6 \sum_{i} \left( \frac{∂^4 Ψ_{Ce,Y}}{∂u_{i,j}^4} \right), \quad γ_{Ce,Y} = 4 (γ_{1}^{Ce,Y} + γ_{2}^{Ce,Y}),
$$

$$
k_{O} = \frac{1}{2} \sum_{i} \left( \frac{∂^2 Ψ_{O}}{∂u_{i,j}^2} \right) = mω_{O}^2, \quad x_{O} = h \sqrt{\frac{k_{O}}{m}}, \quad a_{1}^{O} = 1 + \frac{1}{2} x_{O} coth x_{O},
$$

2
\[
\beta_O = \frac{1}{2} \sum_i \left( \frac{\partial^3 \varphi_{\alpha 0}^O}{\partial u_{i\alpha} \partial u_{i\beta} \partial u_{i\gamma}} \right)_{eq}, \quad K = k_O - \frac{\beta^2_O}{3 \gamma_O},
\] (8)

\[
\gamma_1^O = \frac{1}{48} \sum_i \left( \frac{\partial^4 \varphi_{\alpha 0}^O}{\partial u_{i\alpha}^4} \right)_{eq}, \quad \gamma_2^O = \frac{6}{48} \sum_i \left( \frac{\partial^4 \varphi_{\alpha 0}^O}{\partial u_{i\beta}^4} \right)_{eq}, \quad \gamma^O = 4 (\gamma_1^O + \gamma_2^O),
\] (9)

where \(\alpha \neq \beta \neq \gamma = x, y, \) or \(z,\) and \(\varphi^X_{\alpha 0} (X = Ce^{4+}, Y^{3+}, O^{2-})\) is the interaction potential between the zero-th \(X\) and the \(i-th\) ions, and \(U_0^X\) represents the sum of effective pair interaction energies for \(X\) ion at the equilibrium position.

2.2. The vacancy migration energy

Due to \(Y^{3+}\) ions of lower charge than the host cations, oxygen vacancies are generated in the anion sublattice to maintain overall charge neutrality in crystal lattice. For every two \(Y^{3+}\) ions, one oxygen vacancy is formed. If \(N\) is the number of cations in the crystal lattice and \(x\) is the concentration of \(Y^{3+}\) ions then the numbers of \(Ce^{4+}, Y^{3+}, O^{2-}\) ions and the oxygen vacancies are \(N_{Ce} = N (1-x), N_Y = N x, N_O = 2 N (1-x/4),\) and \(N_{va} = N x/2,\) respectively. The vacancy migration energy is determined as [10]

\[
E_m = \Psi_1 - \Psi_2,
\] (10)

with \(\Psi_1, \Psi_2\) are the total 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.

a. The total interaction potentials of the ions at the initial state

The total interaction potential of \(Ce^{4+}\) ions is determined by the average potential energy of a \(Ce^{4+}\) ion, \(u_{Ce}\), for the interaction between it and surrounding ions

\[
u_{Ce} = u_{Ce-Ce} + u_{Ce-Y} + u_{Ce-O},
\] (11)

where \(u_{Ce-Ce}, u_{Ce-Y}, \) and \(u_{Ce-O}\) are the average interaction potentials between a \(Ce^{4+}\) ion and the surrounding \(Ce^{4+}, Y^{3+}, O^{2-}\) ions.

One considers the \(i-th\) nearest-neighbor sites relative to a certain single \(Y^{3+}\) ion (denoted as \(Y^*\)). The number of occupied \(i-th\) nearest-neighbor sites by \(Ce^{4+}\) ions is \(b_i^{Ce-Ce}\) then the number of \(Ce^{4+}\) ions occupying the \(i-th\) nearest-neighbor sites of \(Y^*\) ion can be given by

\[
b_i^{Ce-Ce} = b_i^{Ce-Ce} \frac{N_{Ce}}{N-1},
\] (12)

Thus, the number of \(Ce^{4+}\) ions that have \(Y^*\) ion at the \(i-th\) nearest-neighbor sites is also \(c_i^{Ce-Ce}\). From the number of the associations of \(N_{Ce}\) \(Ce^{4+}\) ions with surrounding \(Ce^{4+}\) ions at the \(i-th\) nearest-neighbor sites, we obtain the average number of the associations of a \(Ce^{4+}\) ions with other \(Ce^{4+}\) ions at the \(i-th\) nearest-neighbor sites

\[
h_i^{Ce-Ce} = b_i^{Ce-Ce} \frac{N_{Ce} - 1}{N-1},
\] (13)

Accordingly, the expression of \(u_{Ce-Ce}\) is given by

\[
u_{Ce-Ce} = \frac{N_{Ce} - 1}{N-1} \sum_i b_i^{Ce-Ce} \varphi_{\gamma_i Ce-Ce}.
\] (14)
In the same way, we also obtain the expressions of $u_{Ce-Ce}$, $u_{Ce-O}$ and lead to the expression of $U_{0}^{Ce}$

$$
U_{0}^{Ce} = \frac{N_{Ce}}{2} \left( \frac{N_{Ce} - 1}{N - 1} \sum_{i} b_{i}^{Ce-Ce} \varphi_{i0}^{Ce-Ce} + \frac{N_{Y}}{N - 1} \sum_{i} b_{i}^{Ce-Y} \varphi_{i0}^{Ce-Y} + \left( 1 - \frac{N_{va}}{2N} \right) \sum_{i} b_{i}^{Ce-O} \varphi_{i0}^{Ce-O} \right). (15)
$$

Similar to the way of calculation for $u_{Ce}$, we obtain the expressions of $u_{Y}$ and $u_{O}$. The total interaction potentials of $Y^{3+}$ and $O^{2-}$ ions are derived from $u_{Y}$ and $u_{O}$

$$
U_{0}^{Y} = \frac{N_{Y}}{2} \left( \frac{N_{R}}{N - 1} \sum_{i} b_{i}^{Y-R} \varphi_{i0}^{Y-R} + \frac{N_{Y} - 1}{N - 1} \sum_{i} b_{i}^{Y-Y} \varphi_{i0}^{Y-Y} + \left( 1 - \frac{N_{va}}{2N} \right) \sum_{i} b_{i}^{Y-O} \varphi_{i0}^{Y-O} \right). (16)
$$

$$
U_{0}^{O} = \frac{N_{O}}{2} \left( \frac{N_{R}}{N} \sum_{i} b_{i}^{O-R} \varphi_{i0}^{O-R} + \frac{N_{Y}}{N} \sum_{i} b_{i}^{O-Y} \varphi_{i0}^{O-Y} + \left( 1 - \frac{N_{va}}{2N - 1} \right) \sum_{i} b_{i}^{O-O} \varphi_{i0}^{O-O} \right). (17)
$$

where $b_{i}^{X-Ce}$ (or $b_{i}^{X-Y}$, or $b_{i}^{X-O}$) is the number of the $i$-th nearest-neighbor sites relative to $X$ ion ($X = Ce^{4+}$, $Y^{3+}$, $O^{2-}$) that $Ce^{4+}$ (or $Y^{3+}$, or $O^{2-}$) ions could occupy, respectively, $\varphi_{i0}^{X-Ce}$ (or $\varphi_{i0}^{X-Y}$, or $\varphi_{i0}^{X-O}$) is the interaction potential between the $0$-th $X$ ion and an ion $Ce^{4+}$ (or $Y^{3+}$, or $O^{2-}$) at the $i$-th nearest-neighbor sites relative to this $X$ ion, respectively.

b. The total interaction potentials of the ions at the saddle-point state

```
A   B   C
  o   X   □
```

- o : Oxygen ion
- □ : Oxygen vacancy

**Figure 1.** An $O^{2-}$ ion hops from the lattice site A, across the saddle point B and occupies an adjacent vacant site C.

Figure 1 presents the movement process of an oxygen ion from the lattice site A, along $<100>$ direction and to the saddle point B. The average interaction potentials of a $Ce^{4+}$ ion, a $Y^{3+}$ ion and an $O^{2-}$ ion at the saddle-point state are determined as

$$
\begin{align*}
&u_{x}^{B} = u_{x}^{A} + \Delta u_{x}^{A}, \\
&u_{y}^{B} = u_{y}^{A} + \Delta u_{y}^{A},
\end{align*}
$$

where $u_{x}^{A}$ and $u_{y}^{A}$ are the average interaction potentials of a $Ce^{4+}$ ion and a $Y^{3+}$ ion at the initial state, respectively, and $\Delta u_{x}^{A}$, $\Delta u_{y}^{A}$ are the changes in the average interaction potentials of a $Ce^{4+}$ ion and a $Y^{3+}$ ion due to the oxygen migration

$$
\begin{align*}
&\Delta u_{x}^{Ce} = \frac{\varphi_{x}^{B-O-Ce} - \varphi_{x}^{B-Ce}}{N_{Ce}}, \\
&\Delta u_{y}^{Y} = \frac{\varphi_{y}^{B-O-Y} - \varphi_{y}^{B-Y}}{N_{Y}},
\end{align*}
$$

with $\varphi_{x}^{A,B}$ and $\varphi_{y}^{A,B}$ are the interaction potentials between the diffusing oxygen ion and surrounding $Ce^{4+}$ (or $Y^{3+}$) ions at the sites A and B, respectively. The expressions of $\varphi_{x,y}^{A,B}$ depend strongly on the configurations of the neighboring cations around the diffusing oxygen ion.
The average interaction potential of an $O^{2-}$ ion at the saddle-point state is given by

$$u_B^O = u_O + \Delta u_O^* + \Delta u_O^{O-O},$$  \hspace{1cm} (20)$$

where $u_O$ is the average potential energy of an $O^{2-}$ ion at initial state, $\Delta u_O^*$ (or $\Delta u_O^{O-O}$) is the change in the average potential energy of an $O^{2-}$ ion for the interaction between it and the surrounding cations (or $O^{2-}$ ions) due to the oxygen hopping. Because of the reciprocal interactions between $O^{2-}$ ions and Ce$^{4+}$, Y$^{3+}$ ions, $\Delta u_O^*$ can be determined through $\Delta u_{Ce}^*$ and $\Delta u_{Y}^*$. From Eqs. (18) - (20), we obtain the expressions of the total interaction potentials of Ce$^{4+}$, Y$^{3+}$ and $O^{2-}$ ions at the saddle-point state

$$U_{yn}^R = U_0^R + \frac{\varphi_B^R - \varphi_A^R}{2},$$  \hspace{1cm} (21)

$$U_{yn}^Y = U_0^Y + \frac{\varphi_B^Y - \varphi_A^Y}{2},$$

$$U_{yn}^O = U_0^O + \frac{N_O}{2} \left( \Delta u_O^* + \Delta u_O^{O-O} \right).$$  \hspace{1cm} (22)

3. Results and Discussions

The interaction potential in YDC is Buckingham potential [11]

$$\varphi_{ij} = \frac{q_i q_j}{r} + A_{ij} e^{-\frac{r}{B_{ij}}} - C_{ij} r^2,$$  \hspace{1cm} (23)$$

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 by Ref. [12]. In this study, we use the Wolf method extended by C.J. Fennell et al. [13] to turn the Coulomb interaction effectively into spherically symmetric potentials with relatively short-ranges [14]

$$u_{ij}(r) = q_i q_j \left[ \frac{\text{erfc}(ar)}{r} - \frac{\text{erfc}(aR_c)}{R_c} + \left( \frac{\text{erfc}(aR_c)}{R_c^2} + \frac{2a}{\pi^{1/2}} \frac{\text{erfc}(-a^2R_c^2)}{R_c^2} \right)(r - R_c) \right], \hspace{0.5cm} r \leq R_c. \hspace{1cm} (24)$$

Here, we find the optimum values of the damping parameter $\alpha$ and the cutoff radius $R_c$ as $\alpha = 0.31 \text{ Å}^{-1}$, $R_c = 11.715 \text{ Å}$.  

![Figure 2](image-url)  

Figure 2. Three conceivable migration Paths of an oxygen vacancy in the fluorite structure.

Fig. 2 shows three conceivable migration directions of an oxygen vacancy in the fluorite structure of CeO$_2$: $<100>$ direction (Path 1), $<110>$ direction (Path 2) and $<111>$ direction.
The migration of the oxygen vacancy corresponds to an opposite oxygen hopping along these directions. The results of vacancy migration energy along three paths are presented in Table 1. One can see that the oxygen vacancy could hop along Path 1 and Path 2 but with Path 3, this migration is forbidden. Due to $E_{Path1}^m < E_{Path2}^m$ so the oxygen vacancy hops mainly along Path 1. M. Nakayama et al. [6] showed that for Path 3, the oxygen ion hops via a large interstitial space which is coordinated by eight oxygen ions. The electrostatic repulsion by surrounding anions could prevent the migration of the oxygen ion. Other authors [15,16] also indicated that main path for the migration of the oxygen vacancy in YDC is $<100>$ direction.

| Path | Path 1 | Path 2 | Path 3 |
|------|--------|--------|--------|
| $E_{m}^m$(eV) | 1.097  | 7.912  | 11.442 |

In YDC, the oxygen vacancy also hops dominantly along the $<100>$ direction. The expressions of $\varphi_{O-Ce,Y}^{A,B}$ in Eq. (21) depend on the different configurations of the neighboring cations around site A and point B. These configurations give rise to the three possible types of cation barriers for the oxygen migration, namely, the Ce$^{4+}$ - Ce$^{4+}$, Ce$^{4+}$ - Y$^{3+}$ and Y$^{3+}$ - Y$^{3+}$ barriers (Fig. 3). The calculated results for the vacancy migration pass three cation barriers are presented in Table 2. One can see that the attendance of Y$^{3+}$ ions to the cation barriers restricts the hopping of the oxygen vacancy and the migration through the Y$^{3+}$ - Y$^{3+}$ barrier is forbidden. Our results are in good agreement with DFT calculations by M. Nakayama et al. [6].

![Diagram](image)

**Figure 3.** Three configurations of neighboring ions around the diffusing oxygen ion.

The calculated results of the vacancy migration energy at various dopant concentrations and temperatures are presented in Fig. 4. One can see that the vacancy migration energy increases with the increase of the temperature and the dopant concentration. There are two reasons for this property: (i) The vibrations of ions at the lattice sites restrict the vacancy migration due to the effect of temperature; (ii) The higher dopant concentration causes the increased possible
Table 2. The oxygen vacancy migration energy $E_m$ (eV) across the cation barriers.

| Method | Ce$^{4+}$ - Ce$^{4+}$ | Ce$^{4+}$ - Y$^{3+}$ | Y$^{3+}$ - Y$^{3+}$ |
|--------|-----------------------|----------------------|---------------------|
| SMM    | 0.4193                | 0.603                | -0.467              |
| DFT [6]| 0.479                 | 0.533                | 0.795               |

existence of cation barriers with higher migration energy, such as the Ce$^{4+}$ - Y$^{3+}$, Y$^{3+}$ - Y$^{3+}$ barriers. Our calculations are performed at the temperature $T = 400 – 1400$ K and the dopant concentration $x = 0.05 – 0.4$. The obtained results of the vacancy migration energy are in the range $E_m = 0.23 – 0.77$ eV. One can see that our results are in good agreement with the calculated results by S.B. Adler et al. [17] ($E_m = 0.49$ eV) and by D.Y. Wang et al. [18] ($E_m = 0.87$ eV). The comparison between the calculated results for YDC and those for YSZ [19] shows that the vacancy migration in YDC is more easily than those in YSZ.

4. Conclusions
We have performed the analytic expressions to calculate the vacancy migration energy in YDC using SMM. The oxygen vacancy hops mainly along direction $<100>$ from the lattice site. The energy for vacancy migration through the Ce$^{4+}$ - Ce$^{4+}$ barrier has the smallest value compared to the other barriers and the presence of dopant ions in the cation barriers restricts the vacancy hopping. The migration energy are calculated as a function of the temperature and dopant concentration. Notably, the increasing dopant concentration causes an increase of the migration energy due to the increased possible existence of the cation barriers with higher migration energy. This study can be developed to calculate the ionic conductivity of the oxide materials with fluorite structure.

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Figure 4. The dependences of the vacancy migration energy on the temperature (a) and dopant concentration (b).
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