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Porosity effects on oxygen ions diffusion in the yttria-stabilized zirconia (YSZ) by molecular dynamics simulation

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ABSTRACT

In this paper, the porosity effects on oxygen ions diffusion was investigated in the yttria-stabilized zirconia (YSZ) by molecular dynamics simulation (MD). Different models were simulated to elucidate the role of yttria (Y_2O_3) concentration (3 to 8 mol% Y_2O_3), number of pores and pores density and structure (cubic and tetragonal lattice) on the oxygen ions diffusion in YSZ system. MD simulation results showed that the diffusion coefficient of the tetragonal YSZ was smaller than that of the cubic YSZ, the maximum of oxygen ions diffusion coefficient exhibited in the 6 mol% Y_2O_3 among all the tetragonal YSZ systems. From MD simulation results, it was found that the pores in the YSZ system were retarding the oxygen ions diffusion. For constant porosity, smaller single pore volume resulted in lower *D*, while higher total porosity also led to smaller *D*.

1. Introduction

As one of the promising ceramic materials, Yttria-stabilized zirconia (YSZ) has potential applications for being used in thermal barrier coatings (TBCs). YSZ based TBCs are of interest due to their potential applications in gas-turbines and aero engines. TBCs protect parts/components of gas-turbines and aero engines from corrosion and oxidation during operation at elevated temperatures [1–3]. Yttria-stabilized zirconia (YSZ) is a promising top-coat material which offers high fracture toughness, high coefficient of thermal expansion (CTE) and low thermal conductivity [4–6]. Experimental studies show that the coatings produced through different spraying methods have different densities and microstructural characteristics, thus affecting the diffusion of oxygen ions in the YSZ lattice [7,8]. For a better understanding, the mechanism of oxygen ions diffusion in YSZ system needs to be studied by applying molecular dynamics (MD) approach. In previous studies, MD approach has been widely applied to investigate the diffusion of oxygen ions in the YSZ system [9–11]. However, all these simulation studies were carried out on cubic YSZ system. The simulation results studying the oxygen ions diffusion in the tetragonal YSZ system were rarely reported.

Previous literatures show that the diffusion behavior of oxygen ions in the cubic YSZ system with different composition have been studied by various groups [12,13]. For example, 8 mol% YSZ has important applications (as a common solid electrolyte) in devices such as oxygen sensors, oxygen pumps and solid oxide fuel cells (SOFCs) [14,15]. According to the phase diagram, the YSZ could crystallize in two crystal structures; (i) tetragonal (*t*-phase) structure and (ii) cubic (*c*-phase) structure [16]. In tetragonal YSZ, the diffusion of oxygen ions could be different due to different lattice constants/dimensions. It is worth to mention here that the 4 mol% YSZ (containing 4 mol% Y_2O_3) exhibit tetragonal structure and is widely being used for TBCs. So far, the work done on TBCs shows that the parametric rules (affecting the properties of TBCs) are not systematically defined. Further, the role of sintering behavior and microstructure on the oxidation behavior in YSZ system is still under debate. For example, Steil et al. [17] measured the top-coat resistance as a function of sintered temperature in YSZ system having different porosities. They found the decrease in porosity with the increase in sintering temperature. This reflected the decrease in diffusibility of the oxygen ions with the increase in porosity. However, our recent experimental study [18] showed that the resistance could be increased with the decrease in porosity. From experimental investigations, the role of pore size on the ions conductivity in YSZ is not clear. The aim of this theoretical work was to study the oxygen ions diffusion in the tetragonal YSZ TBCs. Molecular dynamics (MD) simulations were performed to study the parametric rules of oxygen ions diffusion in 4 mol% YSZ top-coat. MD simulations were run for tetragonal YSZ system by considering YSZ models with different porosity/configurations.

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Table 1
Lattice constants of cubic and tetragonal YSZ (taken from Ref. [16]) used for simulation runs.

Lattice constant (nm)	*a*	*b*	*c*
Cubic	0.512	0.512	0.512
Tetragonal	0.507	0.507	0.518

2. Simulation details

Six models of yttria-stabilized zirconia (YSZ) system with different concentrations (i.e. 3–8 mol%) of yttria (Y_2O_3) in zirconia (ZrO_2) were constructed to study the effect of Y_2O_3 doping on the diffusion of oxygen ions in YSZ. Both cubic and tetragonal systems were taken into account for 4 mol% YSZ and 8 mol% YSZ while for other concentrations, only tetragonal YSZ system was considered. MD simulation box with 10 × 10 × 10 unit cells containing maximum of 11,846 ions was chosen. Table 1 presents the lattice constants [16] of both (cubic and tetragonal) YSZ systems. To simulate the effect of pore volume and number of pores on the oxygen ions diffusion, different models (with different pore volumes) based on the tetragonal YSZ (4 mol% YSZ) were constructed (see Table 2). MD simulations were performed with constant [N,V,T] ensemble by applying periodic boundary (PB) conditions using LAMMPS codes [19]. The initial velocities of atoms were chosen at random but conformed to Maxwell-Boltzmann distribution at temperature of 1273 K. The temperature and volume of constructed YSZ system was controlled by Nose-Hoover method [20]. The equations of motion were solved by Velocity-Verlet algorithm with time step of 1 fs [21]. The system was first allowed to relax for 100 ps at chosen temperature (1273 K) and then production runs were performed for 400 ps at constant temperature (1273 K). The trajectories of oxygen ions and mean square displacement (MSD) were measured for every 1000 time steps. The kinetic, potential and total energy of the system were also recorded for every 1000 time steps. The sketch of YSZ model for sample B1 has been plotted in Fig. 1.

Long range interactions can be described by Coulomb's law and Ewald summations [22], whereas short range interactions are given by Born-Meyer- Buckingham pair potential [23] as follows:

$$V(r_{ij}) = -\frac{A_{ij}}{r_{ij}^6} + B_{ij}\exp(-C_{ij}r_{ij}) \quad (1)$$

where r_{ij} is the distance between the ions i and j; A_{ij}, B_{ij} and C_{ij} are the empirical parameters of the ion-ion interaction. For simulation runs, all required parameters were taken from Ref. [12].

For time $t \to \infty$, the MSD and diffusion coefficient, D of oxygen ion was found to follow a linear relation [24] as:

$$MSD = B + 6Dt \quad (2)$$

where B is a constant.

Table 2
Summary of tetragonal YSZ (4 mol% YSZ) systems (with different pore volume and pore numbers) taken for simulation runs at 1273 K.

Sample code/configuration	Single pore volume	Number of pores	Total porosity	D (m^2/s)
A0	0 × 0 × 0	0	0	5.498×10^{-11}
B1	1 × 1 × 1	9	9	4.267×10^{-11}
B2	1 × 1 × 1	18	18	3.497×10^{-11}
C1	3 × 1 × 1	1	3	5.118×10^{-11}
C2	3 × 1 × 1	2	6	4.876×10^{-11}
C3	3 × 1 × 1	4	12	4.646×10^{-11}
C4	3 × 1 × 1	6	18	4.353×10^{-11}
D1	3 × 2 × 1	3	18	4.554×10^{-11}
E1	3 × 3 × 1	2	18	4.622×10^{-11}
E2	3 × 3 × 2	1	18	5.205×10^{-11}

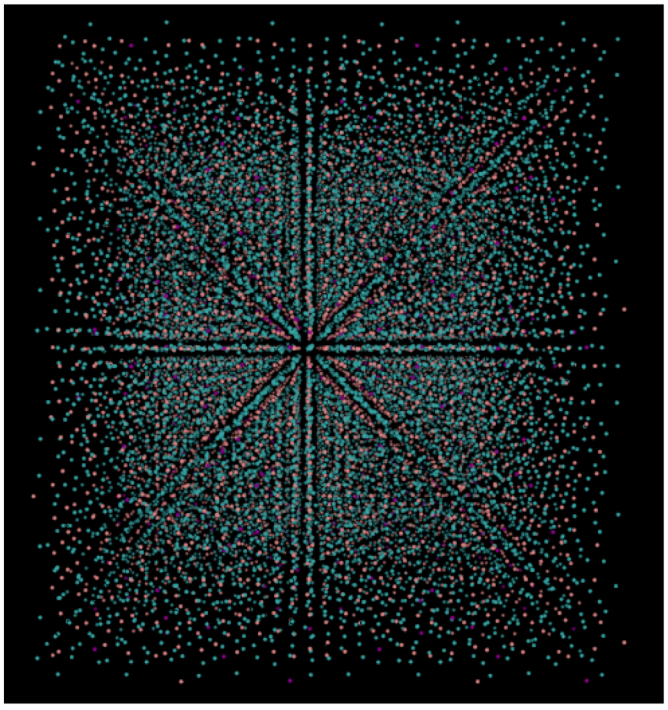

Fig. 1. The sketch of YSZ model for sample B1.

3. Results and discussion

Fig. 2 shows the diffusion coefficient, D of oxygen ions as a function of 1/T. It can be seen that the ln$D$ versus 1/T (Fig. 2) fits well and shows a linear decreasing trend for 8 mol% Y_2O_3 concentration in YSZ. The diffusion coefficient, D of oxygen ions, at given temperature T can be well described (Fig. 2) by Arrhenius equation of the form [25];

$$D = D_0\exp(-E_D/k_BT) \quad (3)$$

where D_0 is the pre-exponential factor, E_D is the activation energy of oxygen ions required for migration and k_B is the Boltzmann's constant. From Fig. 2, the slope of the line gives the activation energy (E_D = 54.35 kJ mol^{-1}) for oxygen ions migration. These results are found to be consistent with the results reported in literatures [26,27].

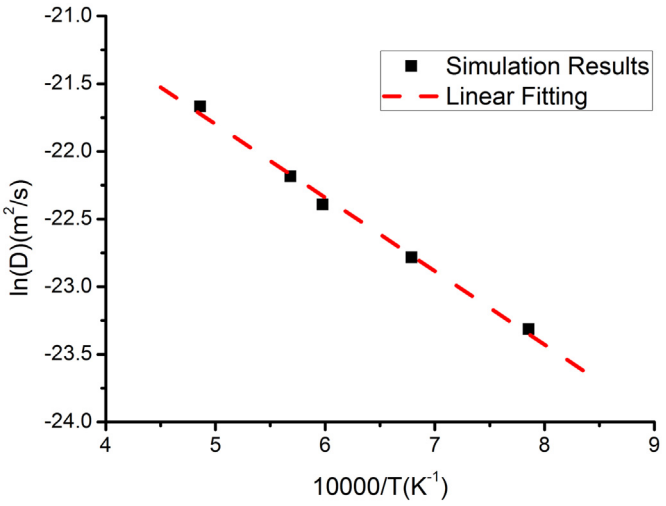

Fig. 2. Arrhenius plot showing the diffusion coefficient (D) of oxygen ions as a function of 1/T for 8 mol% Y_2O_3 concentrations in cubic yttria-stabilized zirconia.