Ab initio calculation of the migration free energy of oxygen diffusion in pure and samarium-doped ceria

Supplemental Material

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Migration edge	Initial state	Transition state
	(Å)	(Å)
Ce-Ce	4.076	4.237
	4.098	4.249
	4.116	4.257
Ce-Sm	4.066	4.235
	4.089	4.246
	4.107	4.254
Sm-Sm	4.059	4.231
	4.083	4.244
	4.102	4.256

Table S1: Distances between migration-edge atoms in relaxed structures using in QHA calculations.

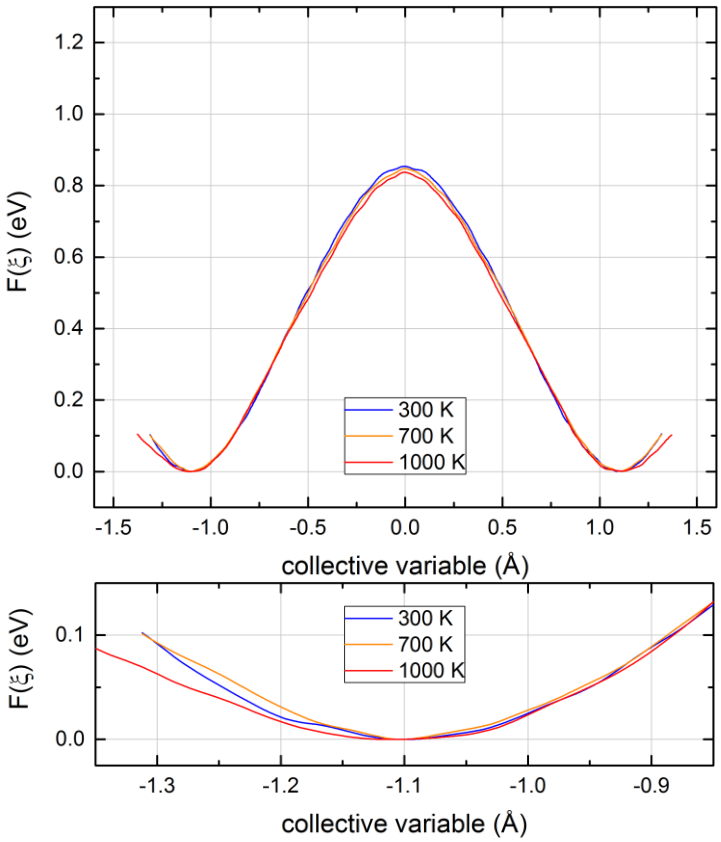

Figure S1: Free energy profiles for the jump of a migrating oxygen ion for the Ce-Sm edge for different temperatures, *T* = 300 K, 700 K and 1000 K. In the bottom figure, an excerpt of the upper data is shown.

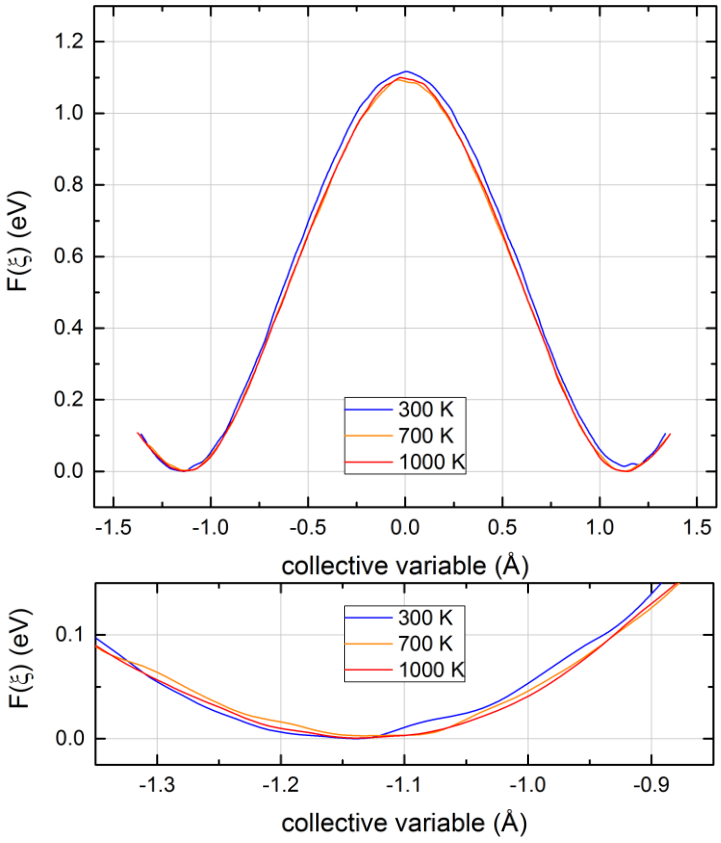

Figure S2: Free energy profiles for the jump of a migrating oxygen ion for the Sm-Sm edge for different temperatures, *T* = 300 K, 700 K and 1000 K. In the bottom figure, an excerpt of the upper data is shown.

In this section, we provide histograms for the reaction coordinate ξ, which were computed using molecular dynamics for the reactant configuration of the oxygen ion migration in pure and doped ceria. The values of $P(\xi) = \frac{\langle \delta(\xi_a - \xi) \rangle}{\langle \Theta(\xi_b - \xi) \rangle}$ were used in our free-energy calculations discussed in Section 4.3 in the main text.

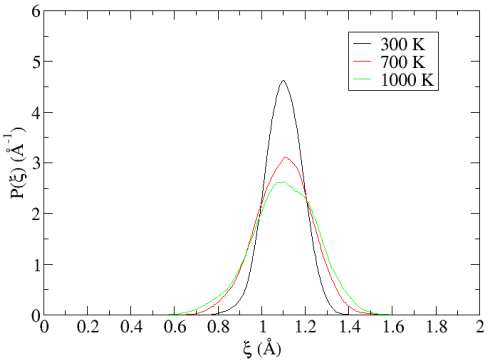

Figure S3: Probability density for the reaction coordinate (ξ) computed for the reactant configuration of the Ce-Sm edge.

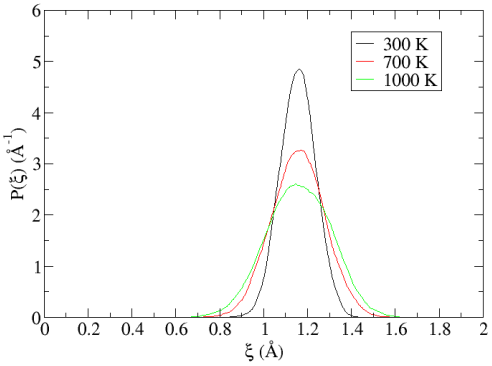

Figure S4: Probability density for the reaction coordinate (ξ) computed for the reactant configuration of the Sm-Sm edge.