Supplementary information for: High oxygen-vacancy diffusivity predicted for perovskite oxide $Ca_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{2.5}$ †

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The interactions in our molecular dynamics (MD) calculations are described with a short-range potential of the Buckingham form and a long-range coulombic term:

$$V_{ij}(r) = A_{ij}\exp\left(-\frac{r}{\rho_{ij}}\right) - \frac{C_{ij}}{r^6} + \frac{q_i q_j}{4\pi\varepsilon_0 r}$$

The empirical parameters A_{ij} and ρ_{ij} represent Pauli repulsion; C_{ij} describes dispersion (van der Waals interactions); q_i and q_j are the formal ion charges, and ε_0 is the vacuum permittivity. We employ a set of parameters that has been used previously to examine BSCF materials.[1,2] We expand the model to include Ca^{2+} by using parameters derived for the same oxygen–oxygen potential. The potential parameters are given in Table S1. All MD calculations were carried out in the LAMMPS code[3,4], with periodic boundary conditions, in an isothermal-isobaric (NpT) ensemble ($p_{\mathrm{damp}} = 1\,\mathrm{ps}$, $T_{\mathrm{damp}} = 0.1\,\mathrm{ps}$) at $p = 1013.25\,\mathrm{mbar}$, and a timestep of $\delta t = 1\,\mathrm{fs}$. Metropolis Monte Carlo calculations[5] were carried out as implemented in the LAMMPS code.

Table S1 Buckingham type potential parameters.

Interaction	A_{ij} / eV	ρ_{ij} / Å	C_{ij} / eVÅ^{-6}	source
Ca^{2+}– O^{2-} (I)	1340.18	0.3214	0.0	6
Ca^{2+}– O^{2-} (II)	1090.4	0.3437	0.0	7
Ba^{2+}– O^{2-}	1214.4	0.3522	0.0	1,8
Sr^{2+}– O^{2-}	774.2	0.3538	0.0	9
Co^{3+}– O^{2-}	1329.82	0.3087	0.0	9,10
Fe^{3+}– O^{2-}	1156.36	0.3299	0.0	10
O^{2-}– O^{2-}	22764.3	0.1490	43.0	9,10

Simulations at 500 K show a cubic cell parameter of $a(\mathrm{BSCF}) = 3.799\,\text{Å}$, exactly the same as Shiiba *et al.*[2], and $a(\mathrm{CSCF}) = 3.744\,\text{Å}$, slightly lower due to $r_{Ca^{2+}} < r_{Ba^{2+}}$.

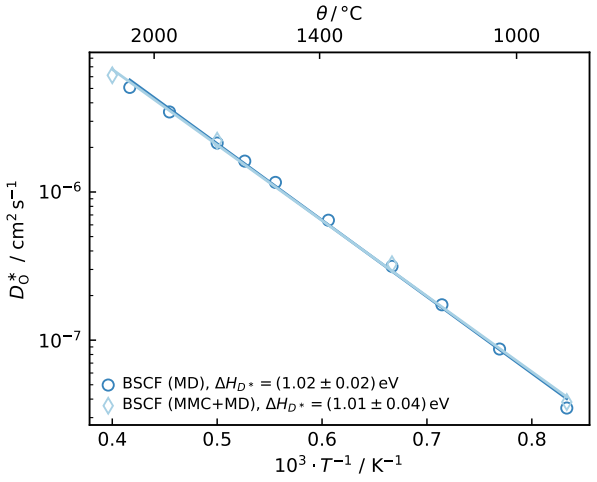

Fig. S1 Oxygen tracer diffusion coefficients versus inverse temperature for cubic $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{2.5}$, obtained from MD simulations, with and without preceeding MMC simulations.

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