Supplementary information for: A molecular-dynamics study of oxygen diffusion in polycrystalline (La,Sr)$FeO_{3-\delta}$ perovskite†

Alexander Bonkowski,[a] John A. Kilner,[b] and Roger A. De Souza*[a]

Supplementary Information

Table S1 Buckingham type potential parameters used for this project.

Interaction	A_{ij} / eV	ρ_{ij} / Å	C_{ij} / eV Å^6	lit.
La^{3+}–O^{2-}	1516.3	0.3525	0.0	1
Sr^{2+}–O^{2-}	774.2	0.3538	0.0	1
Fe^{3+}–O^{2-}	1156.36	0.3299	0.0	2
O^{2-}–O^{2-}	22764.3	0.1490	43.0	1

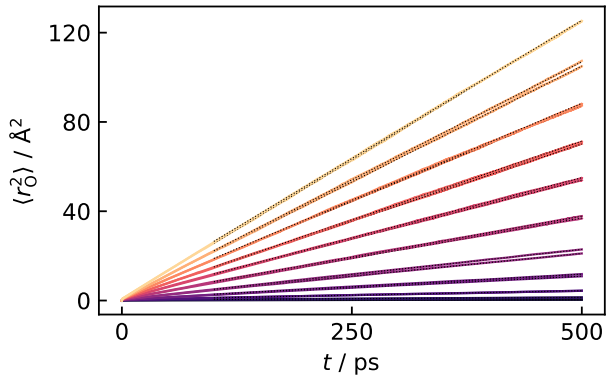

Fig. S1 Mean-squared displacement of oxygen ions $\langle r_O^2 \rangle$ obtained as a function of time from MD simulations of LSF10, $n_v = 1.66\,\%$. Results shown for various temperatures $1000 < T\,\mathrm{K} < 3000$; two simulations with different random initial kinetic energy distributions were carried out at each temperature.

[a] Institute for Physical Chemistry, RWTH Aachen University, 52074 Aachen, Germany. Fax: +49 241 80 92128; Tel: +49 241 80 94739; E-mail: desouza@pc.rwth-aachen.de

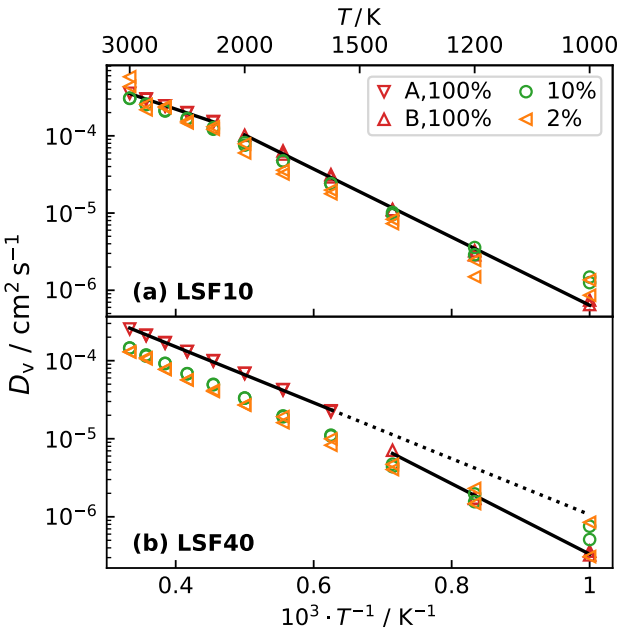

Fig. S2 Oxygen-vacancy diffusion coefficients D_v as a function of inverse temperature for (a) LSF10 and (b) LSF40: A, this study (orthorhombic); B, this study (cubic); C, oxygen vacancy concentration reduced to 10 %; D, oxygen vacancy concentration reduced to 2 %.

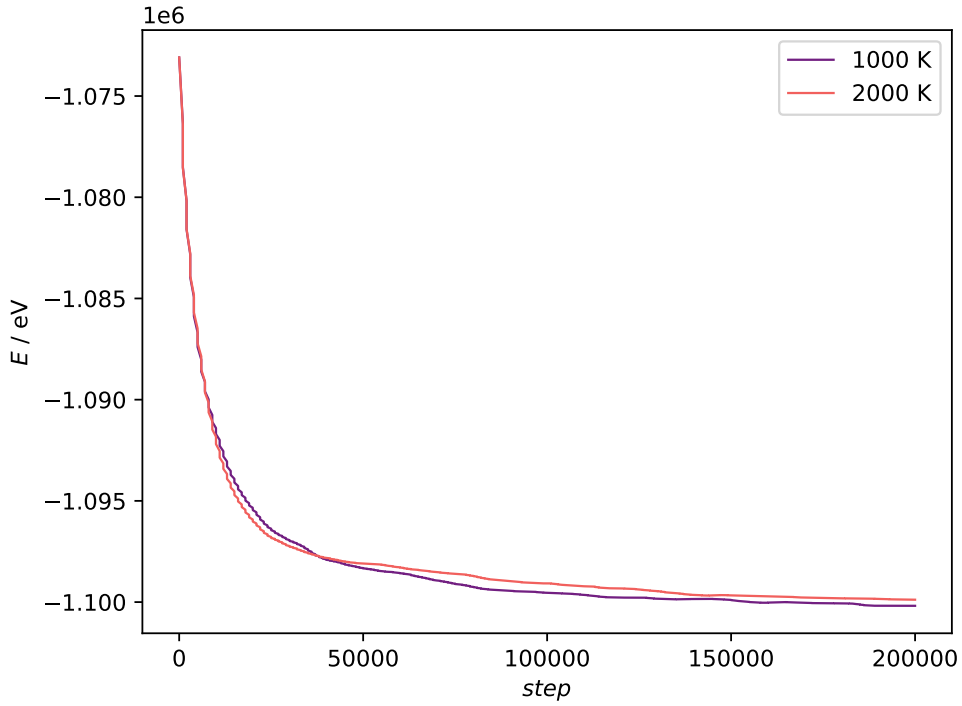

Fig. S3 Total energy of a representative polycrystalline cell (LSF10, 20 grains) during MMC swapping.

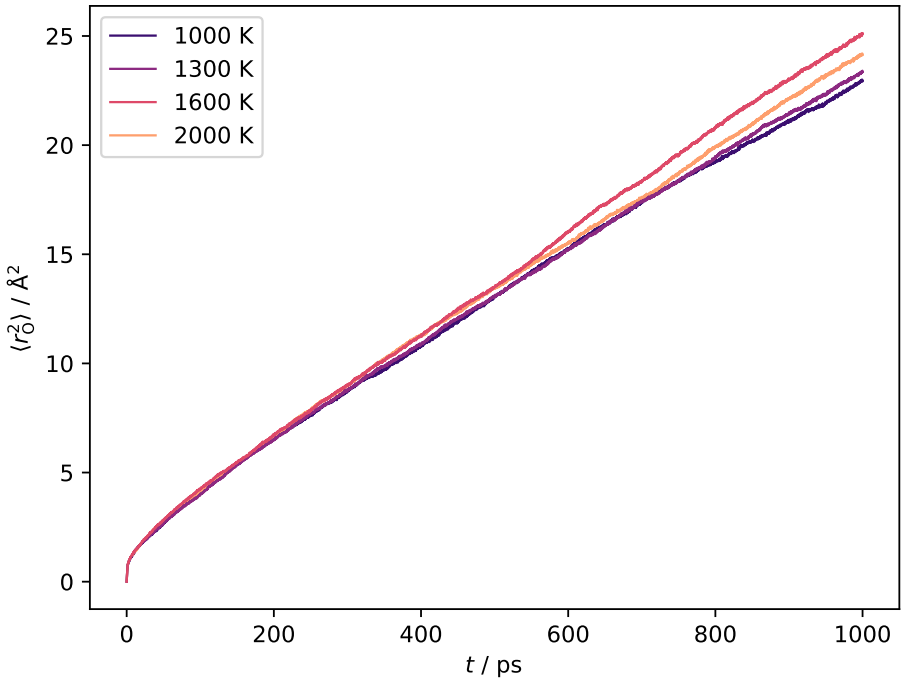

Fig. S4 Mean-squared displacement of oxygen ions $\langle r_O^2 \rangle$ obtained as a function of time from equilibration MD simulations of a polycrystalline LSF10 cell at 2000 K with 2 grains after MMC swapping at different temperatures.

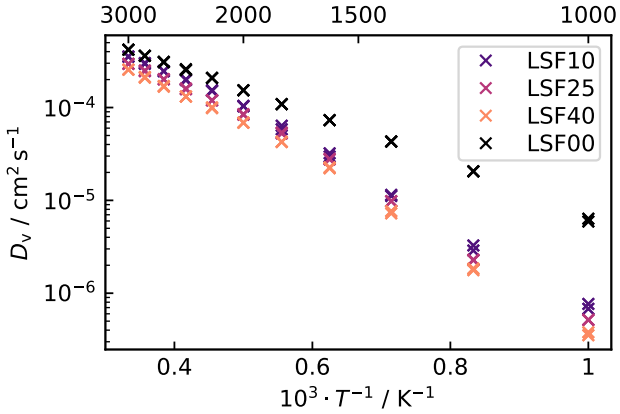

Fig. S5 Oxygen-vacancy diffusion coefficients D_v as a function of inverse temperature for different Sr'_{La} concentrations.

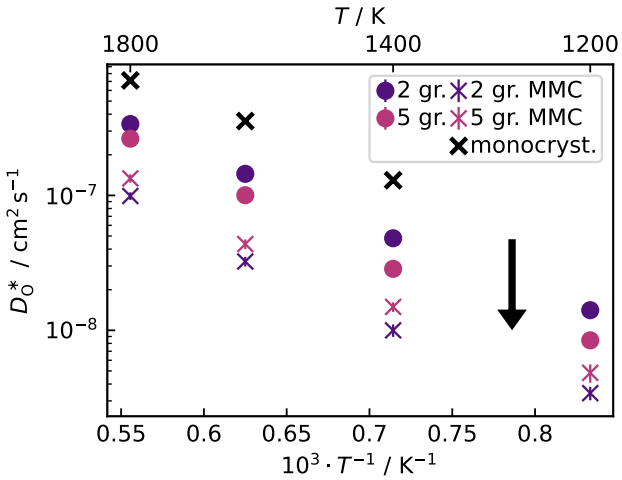

Fig. S6 Oxygen tracer diffusion coefficients in LSF10. The same figure for LSF40 can be found in the main text as Fig. 10.

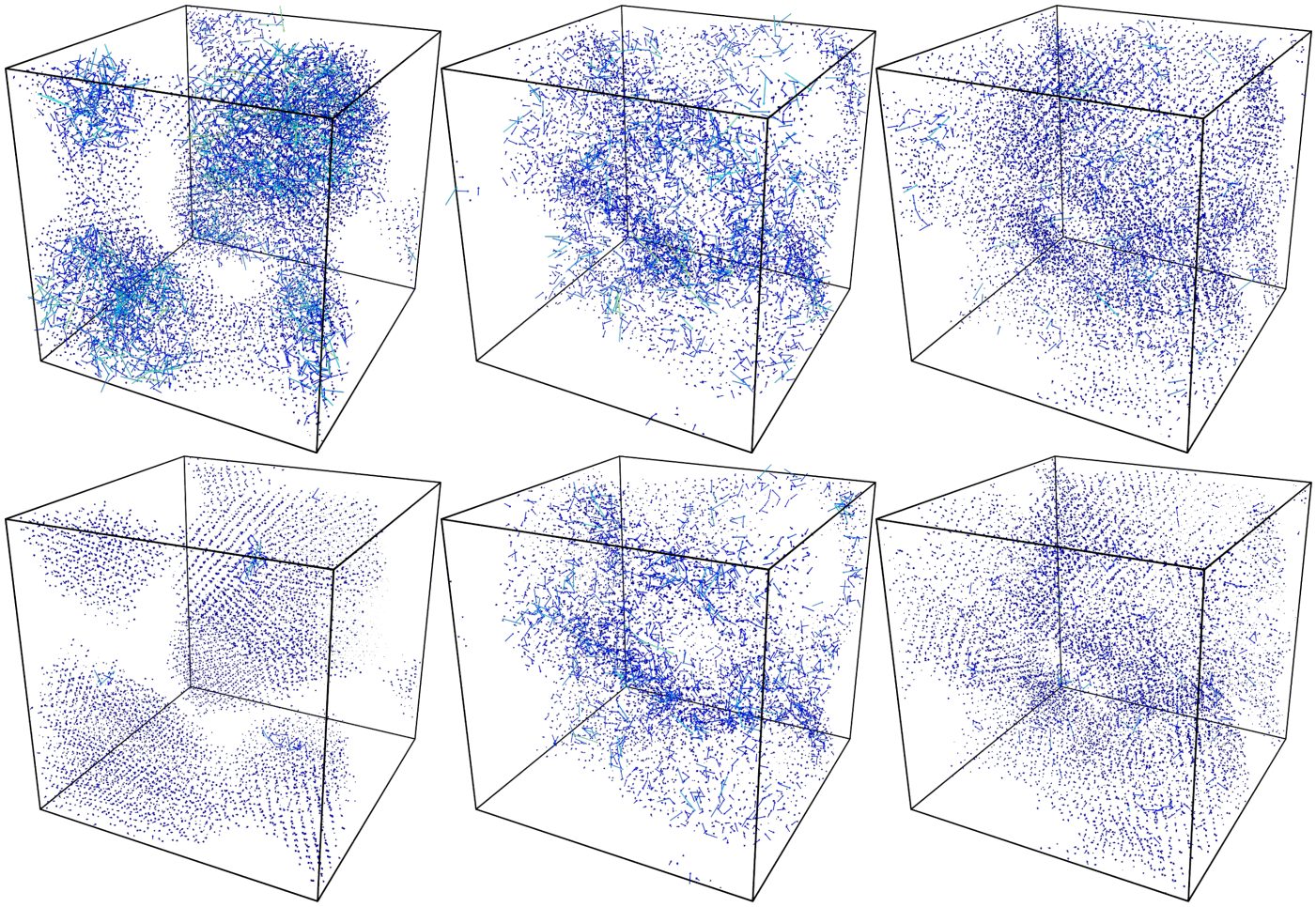

Fig. S7 Displacement vectors of oxygen ions a representative polycrystalline LSF10 cell with 2 grains after 1.0 ns at 1400 K. From left to right: bulk, grain boundary core, grain boundary edge. Upper row: 100 % oxygen vacancies; lower row: 10 % oxygen vacancies (relative to the stoichiometric oxygen vacancy concentration).

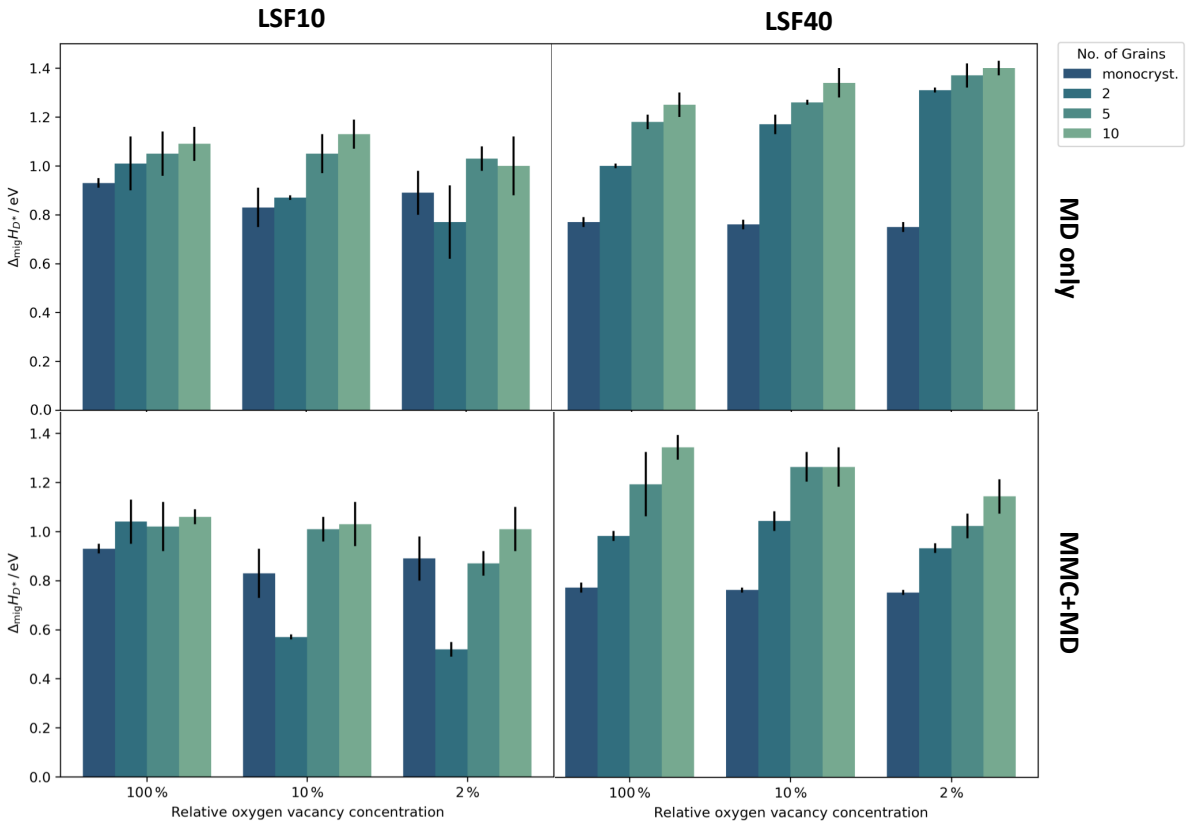

Fig. S8 Activation enthalpies of oxygen diffusion in LSF10 and LSF40, obtained from diffusion coefficients in Fig. 11 and 12. For LSF10 structures with low oxygen vacancy concentrations and 2 grains, the activation enthalpy of LFO is sampled (see Fig. S5) due to a nominally Sr free bulk and a few oxygen vacancies in the bulk regions.

Notes and references

1 M. Islam, M. Cherry and C. Catlow, *J. Solid State Chem.*, 1996, **124**, 230–237.
2 M. Cherry, M. Islam and C. Catlow, *J. Solid State Chem.*, 1995, **118**, 125–132.