Supporting Information

Modeling fission product nucleation in molten NaCl using universal machine-learning potentials

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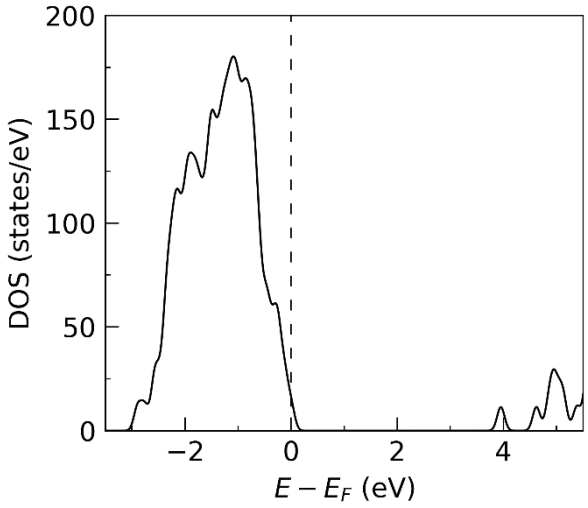

Figure S1. DFT-calculated density of states (DOS) of pure NaCl after 5 ps of equilibration. Gaussian broadening = 0.07 eV.

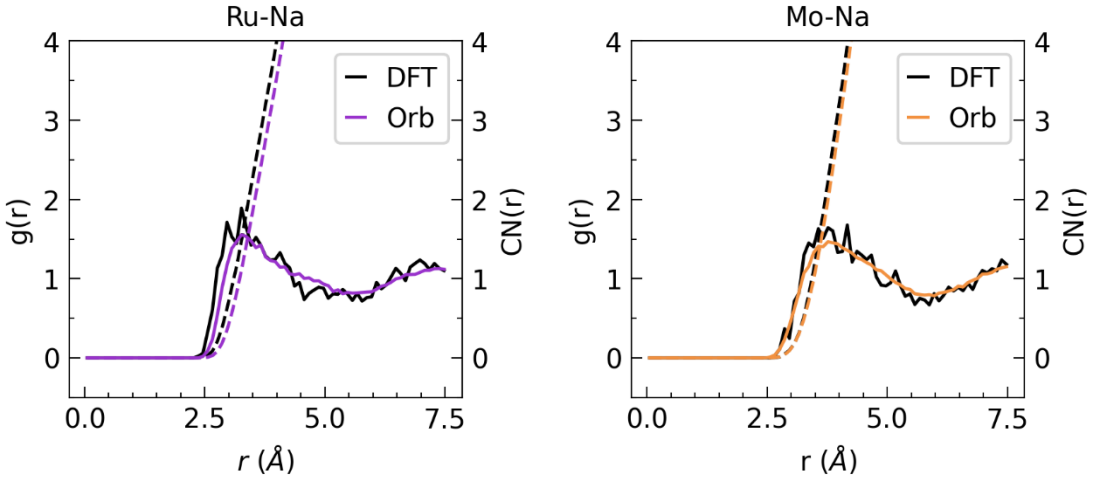

Figure S2. Partial radial distribution functions g(r) (solid lines), and coordination numbers CN(r) (dashed lines), for the Ru-Na and Mo-Na correlations. Results from DFT are represented with black lines. Orb results are indicated in purple (Ru) or orange (Mo).

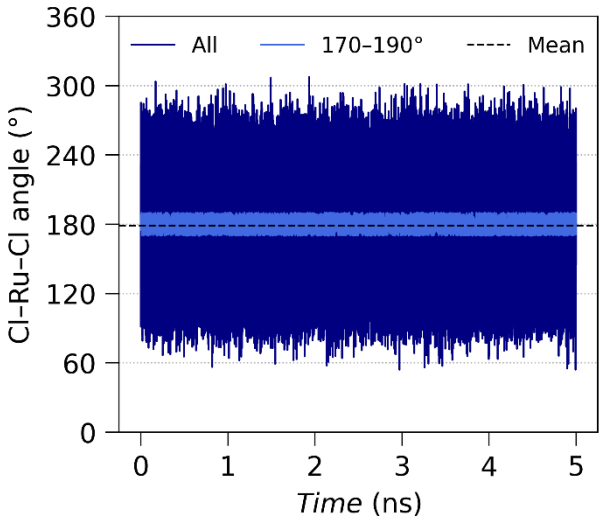

Figure S3. Time evolution of the Cl–Ru–Cl angle computed with the Orb model over a 5 ns MD trajectory. The distinction between acute (<180°) and obtuse (>180°) angles is defined using an arbitrary sign convention, chosen such that a reference configuration corresponds to an acute angle. Although a wide variety of coordination numbers and corresponding angles is sampled during the trajectory (minimum of 52.6° / 307.4°), configurations with angles close to 180° are observed across the entire trajectory, and the average angle is 178°, consistent with the predominance of twofold-coordinated Ru species.

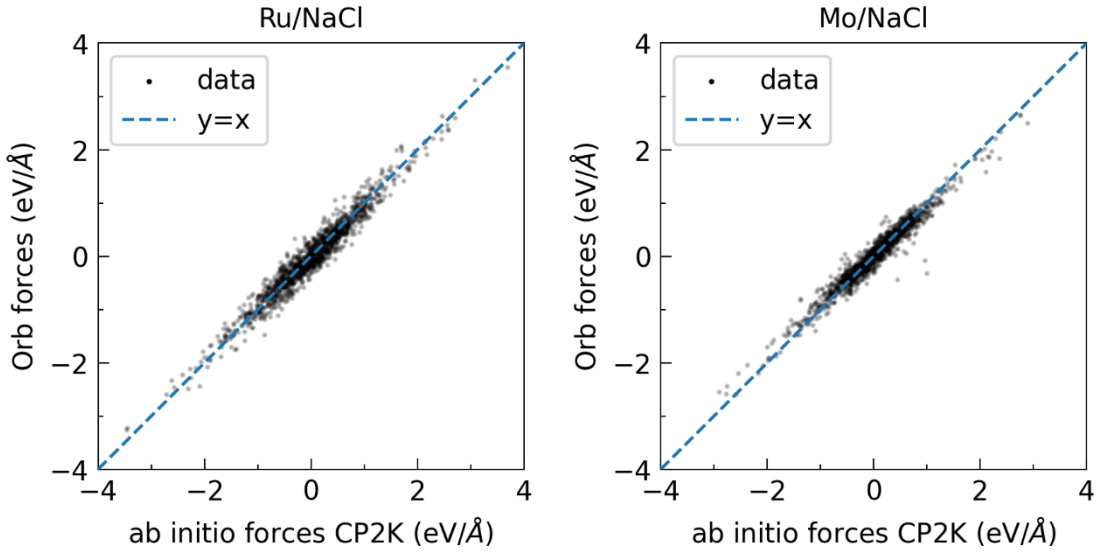

Figure S4. Parity plots illustrating the correlation between the forces calculated using Orb (*y*-axis) and DFT (*x*-axis) for the same molecular configurations. Left panel: total forces on Ru; right panel: total forces on Mo.

Table S1. Summary of literature-reported ground-state bond lengths, binding energies, and magnetic moments of Ru_2 and Mo_2 dimers, compared with the values obtained in this work.

Method	Bond length (Å)	Binding energy (eV)	Magnetization (μB)	Reference
		Ru		
CASTP2	2.10	−3.93	4	[1]
BLYP	2.10	−3.64	4	[1]
B3LYP	2.23	−2.22	6	[1]
r^{2}SCAN	2.22	−3.00	6	This work
PBE	2.09	−3.67	4	This work
Orb v3	2.10	−2.76	-	This work
		Mo		
MS-CASTP2	1.95	−4.41	0	[2]
CCSD(T)	1.93	−3.94	0	[3]
Photolysis (experimental)	1.93	−4.10	-	[4]
r^{2}SCAN	2.06	−2.16	0	This work
PBE	1.94	−3.36	0	This work
Orb v3	1.91	−3.45	-	This work

Table S2. Comparison of Orb-calculated cohesive energies and lattice parameters with experimental values from the literature.

Metal	Orb			Experimental		
	E_{coh} (eV)	a (Å)	c (Å)	E_{coh} (eV)	a (Å)	c (Å)
Mo	6.20	3.1610		6.80 [5]	3.1471 [6]	
Ru	6.26	2.7203	4.2903	6.74 [5]	2.7058 [6]	4.2816 [6]

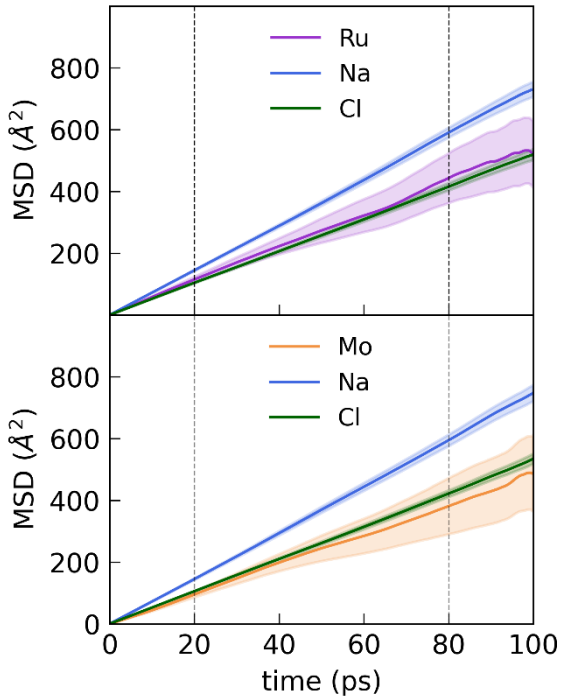

Figure S5. Mean square displacement (MSD) as a function of time for Ru (top) and Mo (bottom) atoms in molten NaCl at 900 °C, computed from 5ns NVT MD simulations by block averaging (with blocks of 100 ps), using the Orb model. Solid lines represent the block-averaged MSD, while shaded areas indicate the associated 95% confidence interval. Dashed vertical lines delimit the time window used for linear regression in the Einstein relation to extract diffusion coefficients.

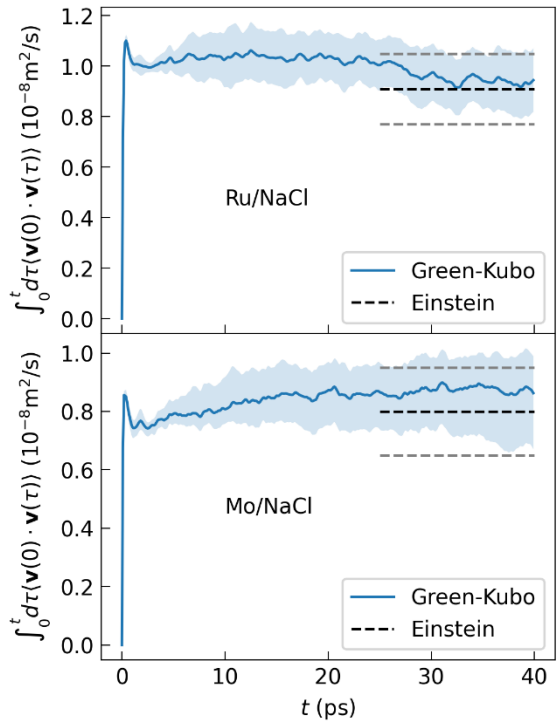

Figure S6. Green-Kubo approach for the diffusion coefficient calculation of metallics in NaCl: cumulative integrals of the velocity autocorrelation function (blue lines) as a function of time for Ru (upper panel) and Mo (lower panel). The uncertainty (shaded areas) is calculated as the 95% confidence interval by splitting the original trajectory into ten blocks. The Green-Kubo approach is compared with the results from the Einstein mean square displacement analysis (mean values represented with dashed black lines, mean ± uncertainty in grey).

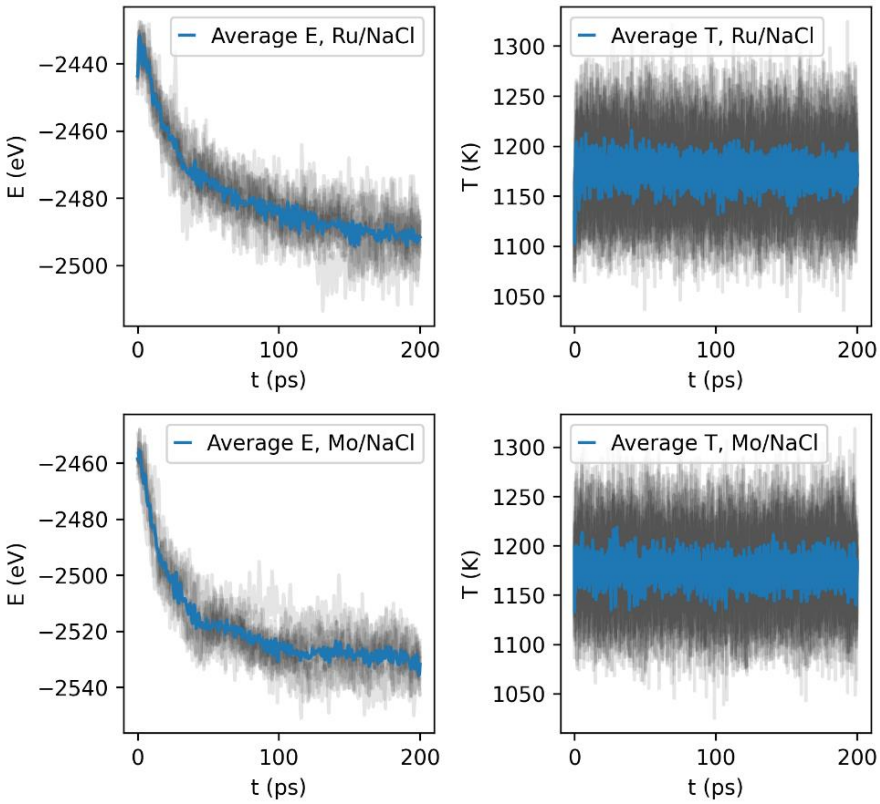

Figure S7. Energy and temperature dynamics in nucleation simulations. Energy (left column) and temperature (right column) are reported as a function of simulation time for Ru/NaCl (upper row) and Mo/NaCl (lower row). Results for each replica are indicated in grey, while the average over replicas is represented in blue.

References

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