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Supporting Information

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Exploration of Amorphous V_2O_5 as Cathode for Magnesium Batteries

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Supporting Information for "**Exploration of Amorphous V_2O_5 as Cathode for Magnesium Batteries**"

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Table S1. Description of hyperparameters and their optimized values used in the moment tensor potentials (MTPs) constructed in this work.

Hyperparameter	Value	Description
R_{min}	automatic (or default)	Minimum cut-off radius
R_{cut}	7 Å	Maximum cut-off radius
N_Q	8	Maximum order of Chebychev polynomials used to describe radial components
lev_{max}	16	Maximum level of moment tensors used
Weight on total energies	1	Weight assigned to total energies during training in the loss
Weight on atomic forces	0.01	Weight assigned to atomic forces during training in the loss
Weight on stress tensors	0.001	Weight assigned to structural stress during training in the loss

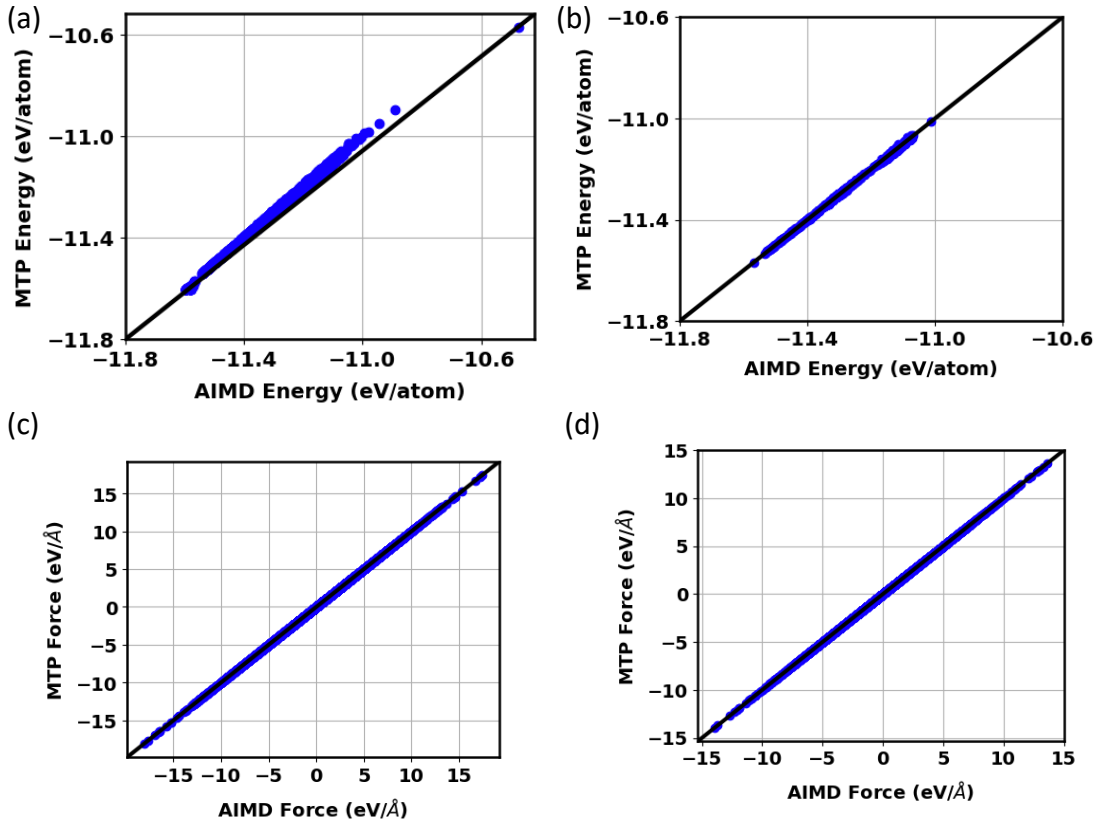

Figure S1. Parity plots of energies (panels a and b) and atomic forces (panels c and d), as predicted by MTP against ab initio molecular dynamics (AIMD) simulations within the training (panels a and c) and test (panels b and d) subsets. Solid black lines represent parity lines.

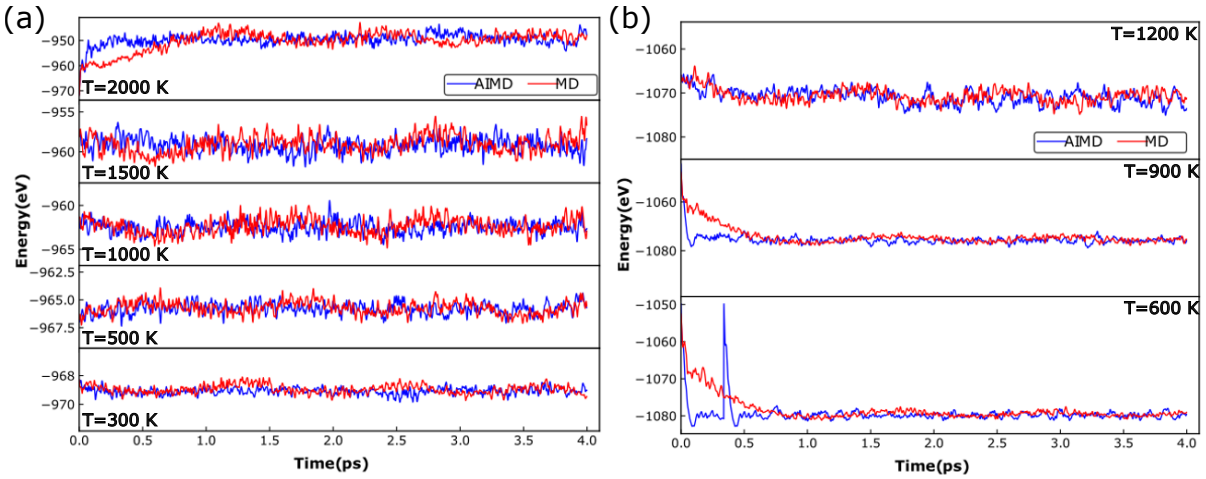

Figure S2. Comparison of AIMD-calculated (blue lines) and MTP-predicted (red lines) potential energies in V_2O_5 (panel a) and MgV_2O_5 (panel b) at different temperatures in a 1×2×3 supercell.,

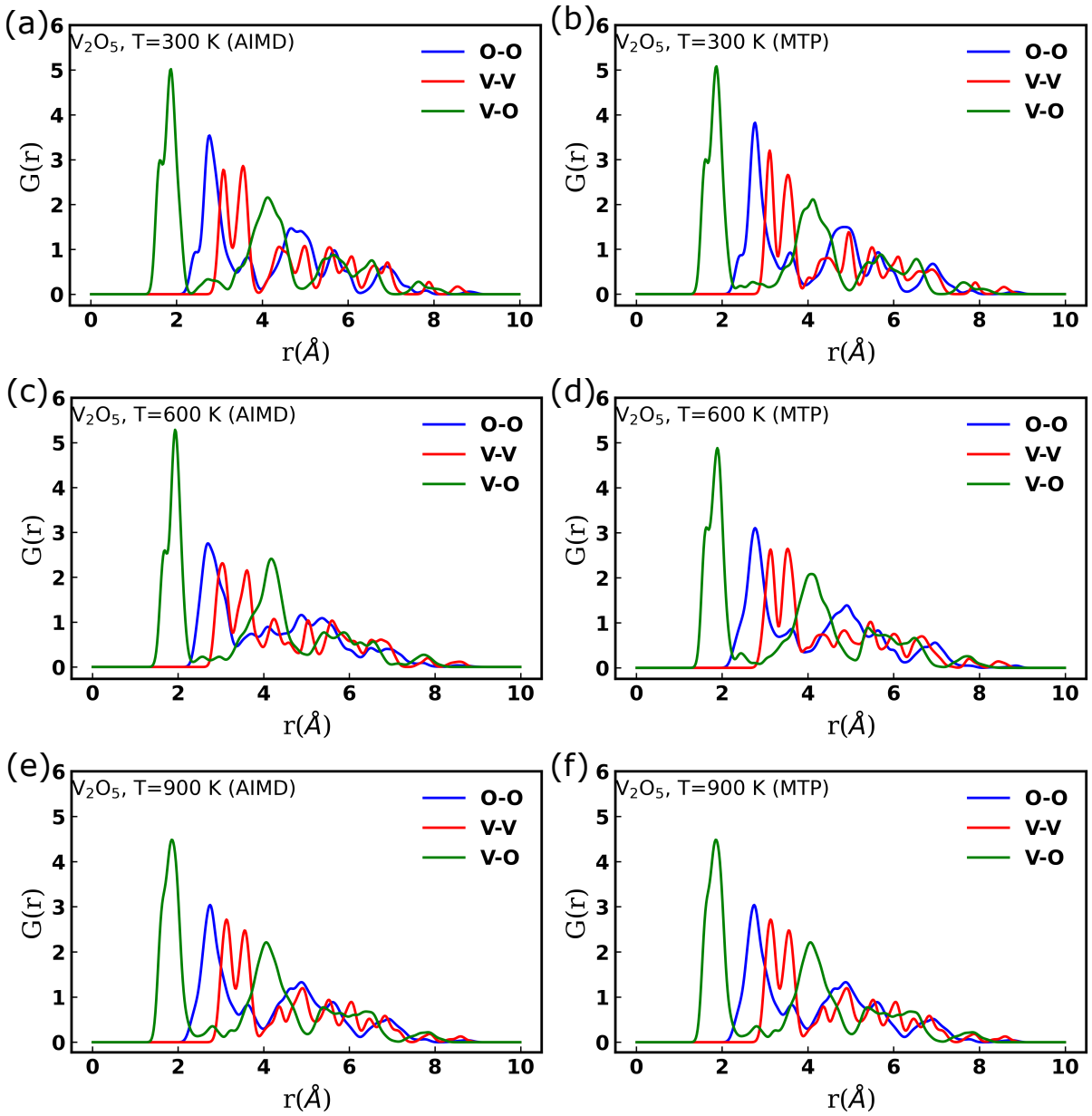

Figure S3. AIMD/MTP calculated radial distribution functions (RDFs) of V_2O_5 at 300 K (panels a and b), 600 K (panels c and d), and 900 K (panels e and f) in a 1×2×3 supercell. Panel a, c, and e are from AIMD simulations after 4 ps, while panels b, d, and f are from MTP simulations after 50 ps. Blue, red, and green lines in all panels represent O-O, V-V, and V-O bonds, respectively.

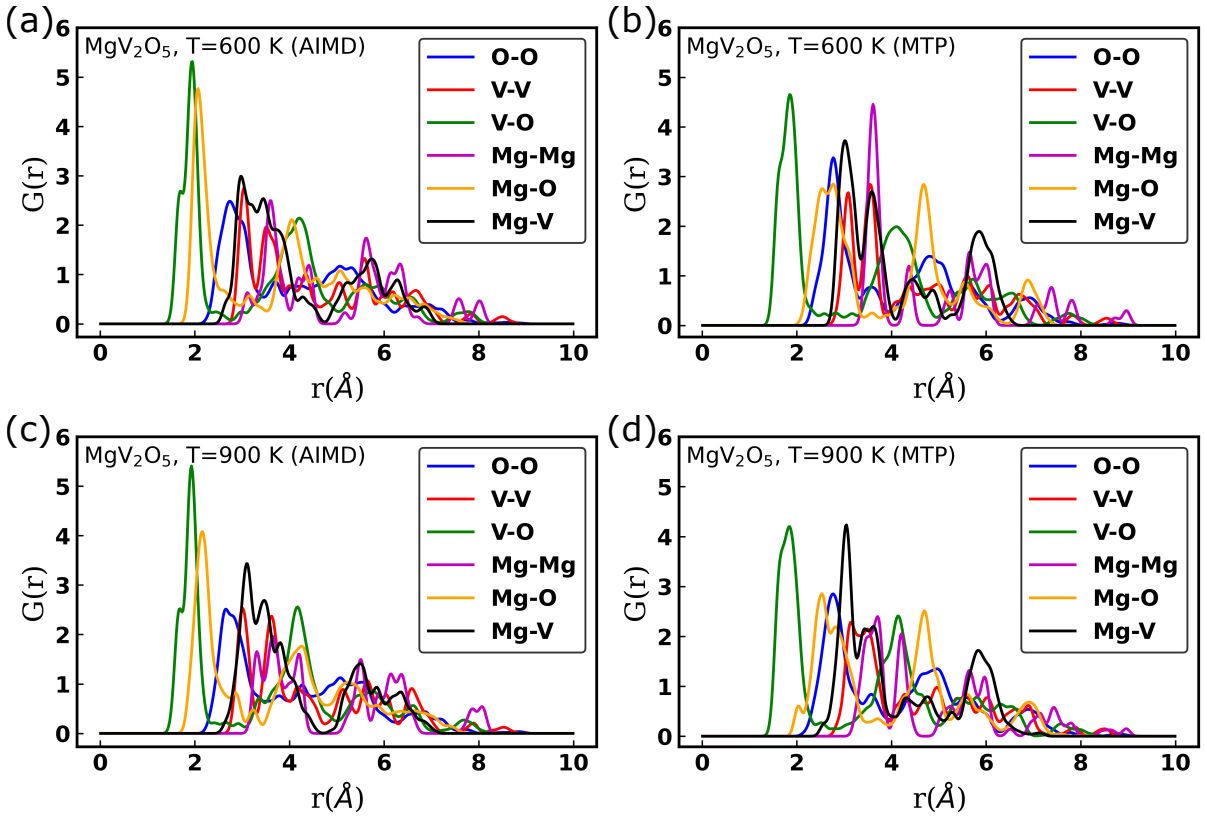

Figure S4. AIMD/MTP calculated RDFs of MgV_2O_5 calculated at 600 K (panels a and b), and 900 K (panels c and d) in a 1×2×3 supercell. Panels and c are AIMD-simulated after 4 ps, while panels b and d are MTP-simulated after 50 ps. Blue, red, green, purple, orange, and black lines in all panels represent O-O, V-V, V-O, Mg-Mg, Mg-O, and Mg-V bonds, respectively.

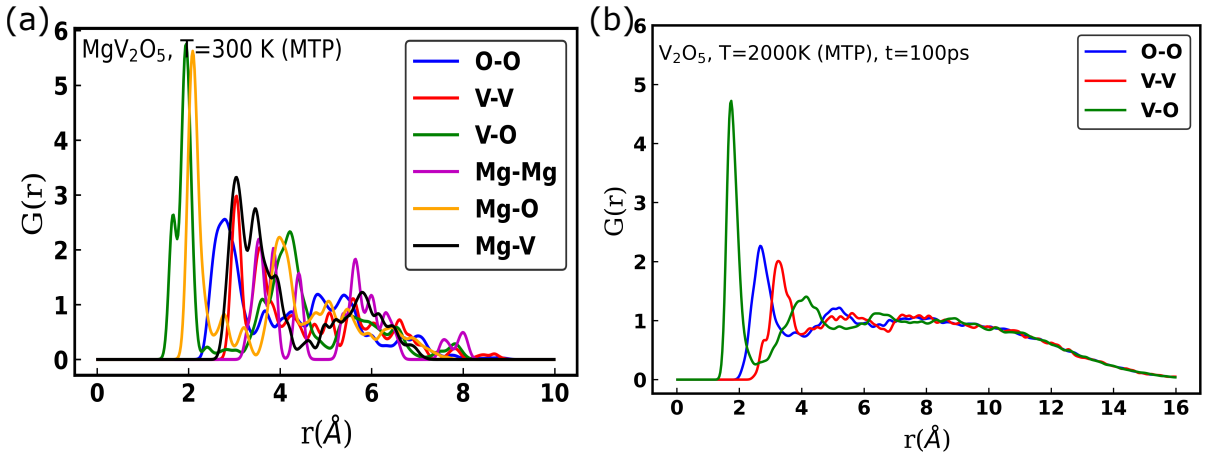

Figure S5. MTP-calculated RDFs of (a) MgV_2O_5 at 300 K after 50 ps in a 1×2×3 supercell and (b) V_2O_5 at 2000 K after 100 ps in a 2×4×6 supercell. Notations used in both panels are identical to **Figures S3** and **S4**.

Table S2. PyLRO-calculated temperature-dependent long-range disorder values in different directions in MTP-generated MgV_2O_5 1×2×3 supercell structures after 20 ps.

Temperature (K)	Minimum	Maximum	Along [100]	Along [010]	Along [001]
300	0.01	0.11	0.09	0.05	0.05
600	0.02	0.12	0.09	0.06	0.04
900	0.05	0.21	0.1	0.14	0.12
1200	0.07	0.34	0.16	0.25	0.14

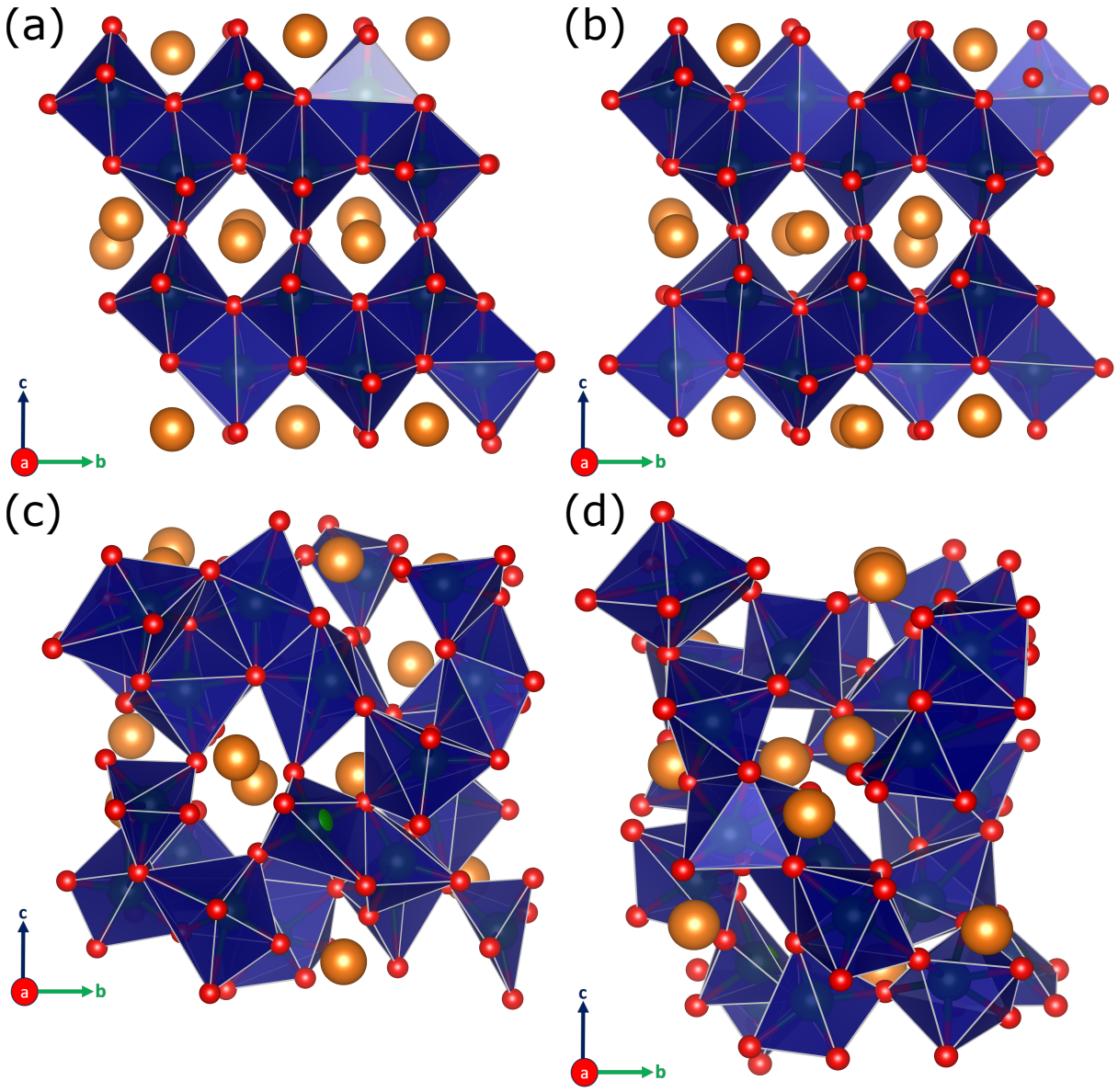

Figure S6. MTP-generated MgV_2O_5 structures at (a) 300 K, (b) 600 K, (c) 900 K, and (d) 1200 K used for PyLRO calculations (**Table S2**). Blue polyhedra represent VO_5/VO_6 units, while Mg atoms are depicted as orange spheres.

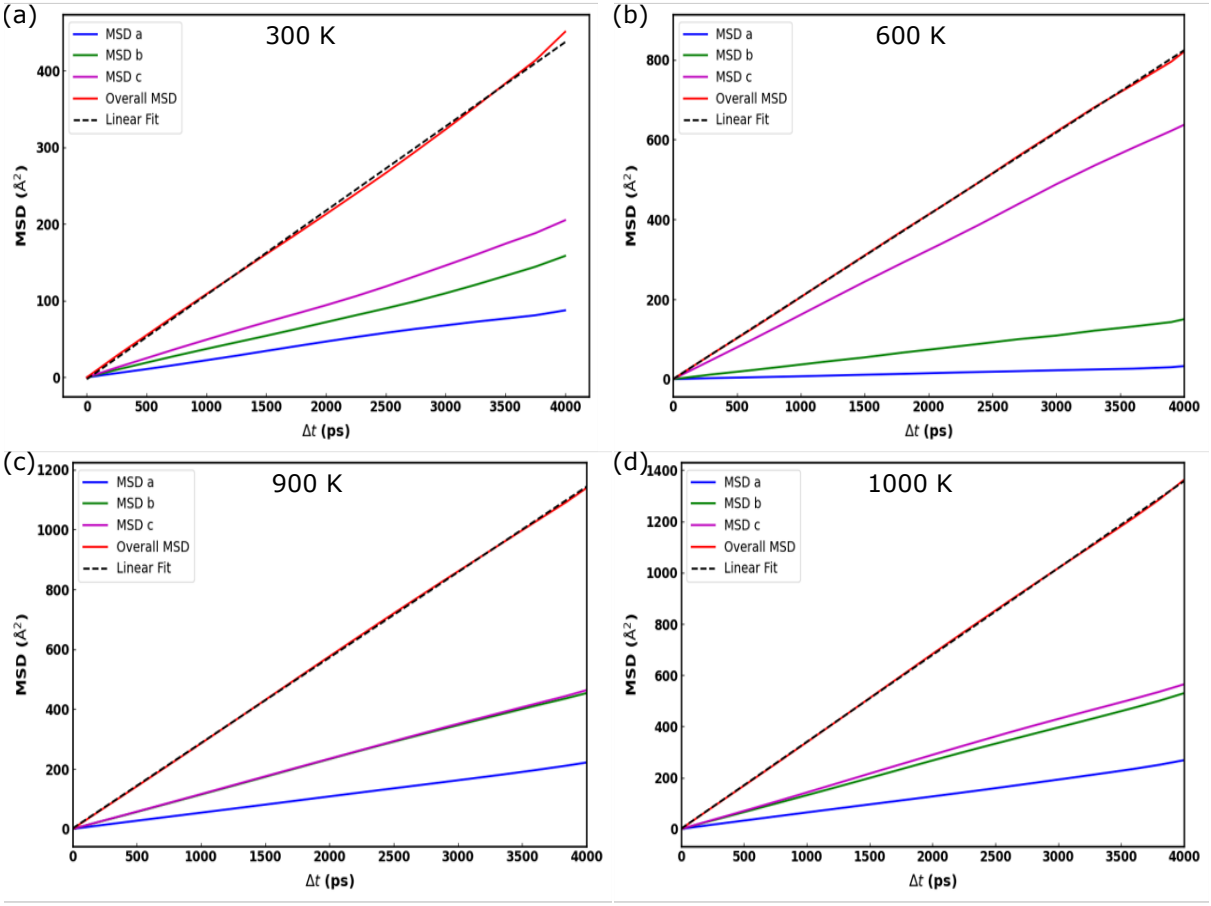

Figure S7. MTP-calculated mean square displacement (MSD) of Mg atoms as a function of Δt using a 2×4×6 MgV_2O_5 supercell over 4000 ps at (a) 300 K, (b) 600 K, (c) 900 K, and (d) 1000 K. Blue, green, and purple lines in each panel represent MSD along the a, b, and c lattice directions, respectively, within the MgV_2O_5 structure. Red and dashed black lines indicate the overall MSD and its associated linear fit, respectively.

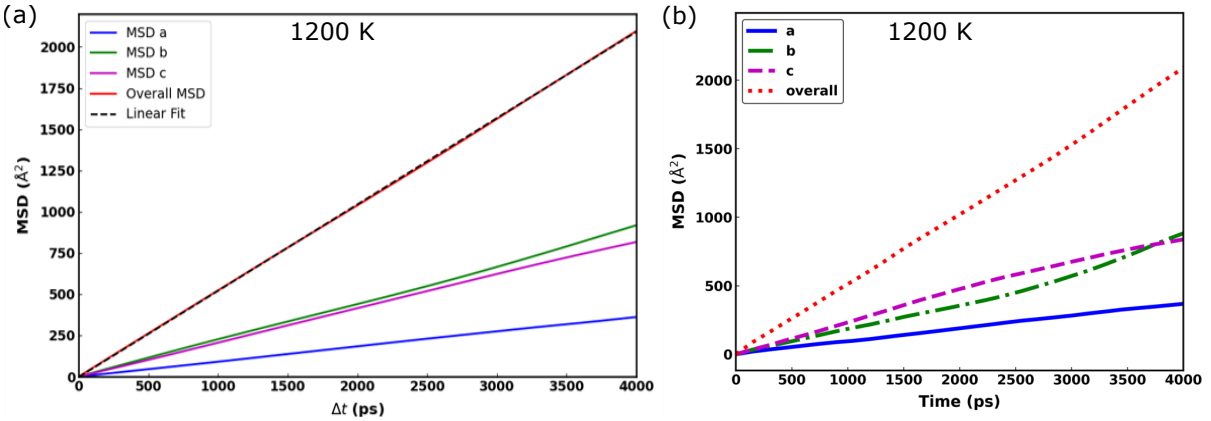

Figure S8. MTP-calculated (a) MSD vs. Δt and (b) MSD vs. t curves in a 2×4×6 MgV_2O_5 supercell at 1200 K over 4000 ps. Notations used in panel a are identical to **Figure S7**. Blue, green, dashed purple, and dashed red lines in panel b represent MSD along a, b, and c lattice directions and the overall MSD, respectively.

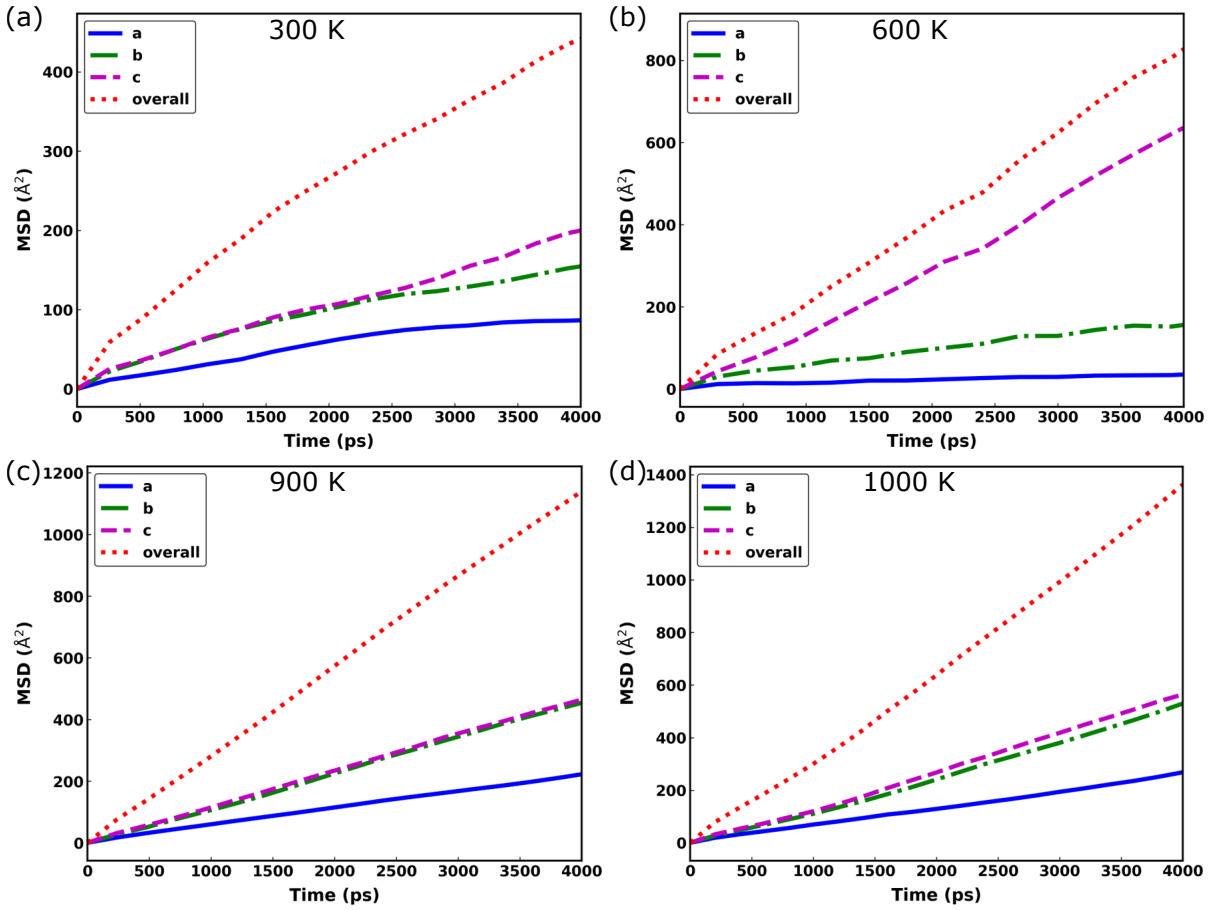

Figure S9. MTP-calculated MSD vs. t curves in 2×4×6 MgV_2O_5 supercells over 4000 ps at (a) 300 K, (b) 600 K, (c) 900 K, and (d) 1000 K. Notations used in each panel are identical to **Figure S8b**.

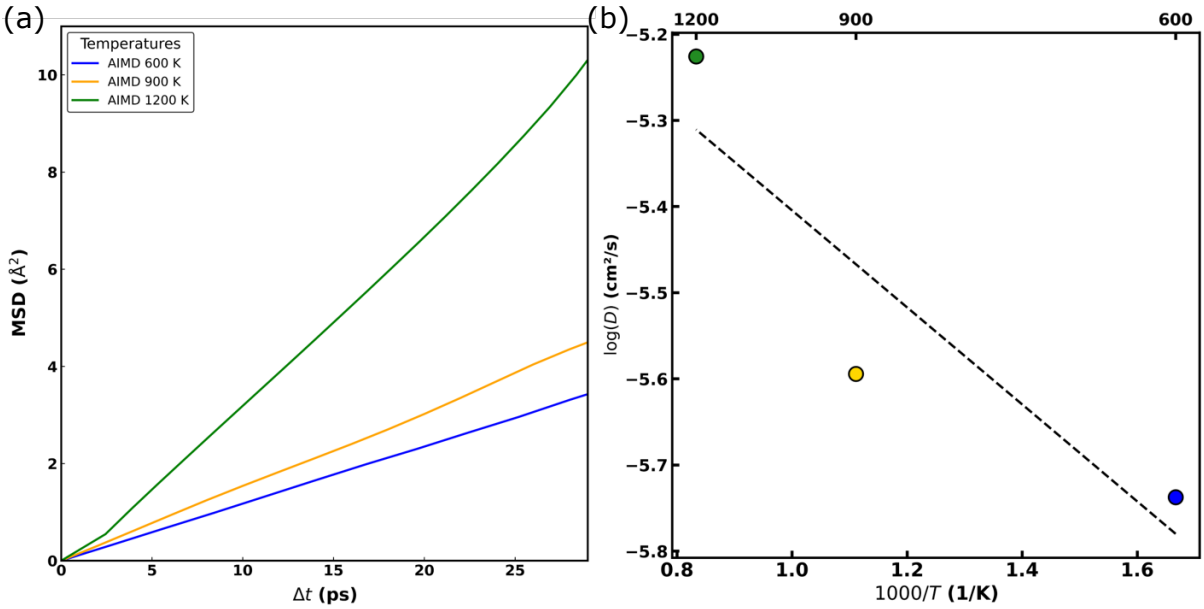

Figure S10. (a) AIMD calculated MSD vs. Δt curves for a 1×2×3 MgV_2O_5 supercell over 30 ps. Green, yellow, and green lines represent MSD at 600 K, 900 K, and 1200 K, respectively. (b) Arrhenius plot involving the logarithm of diffusivity (D in cm^2/s), as obtained from the AIMD data of panel a, as a function of temperature (T in K), plotted as $\log D$ vs. $1000/T$. The top x-axis indicates temperature. Colors of each data point indicates the temperature and is identical to the colors used in panel a. Dashed black line indicates a linear fit. We estimate the activation barrier (E_a) to be 0.11 eV.

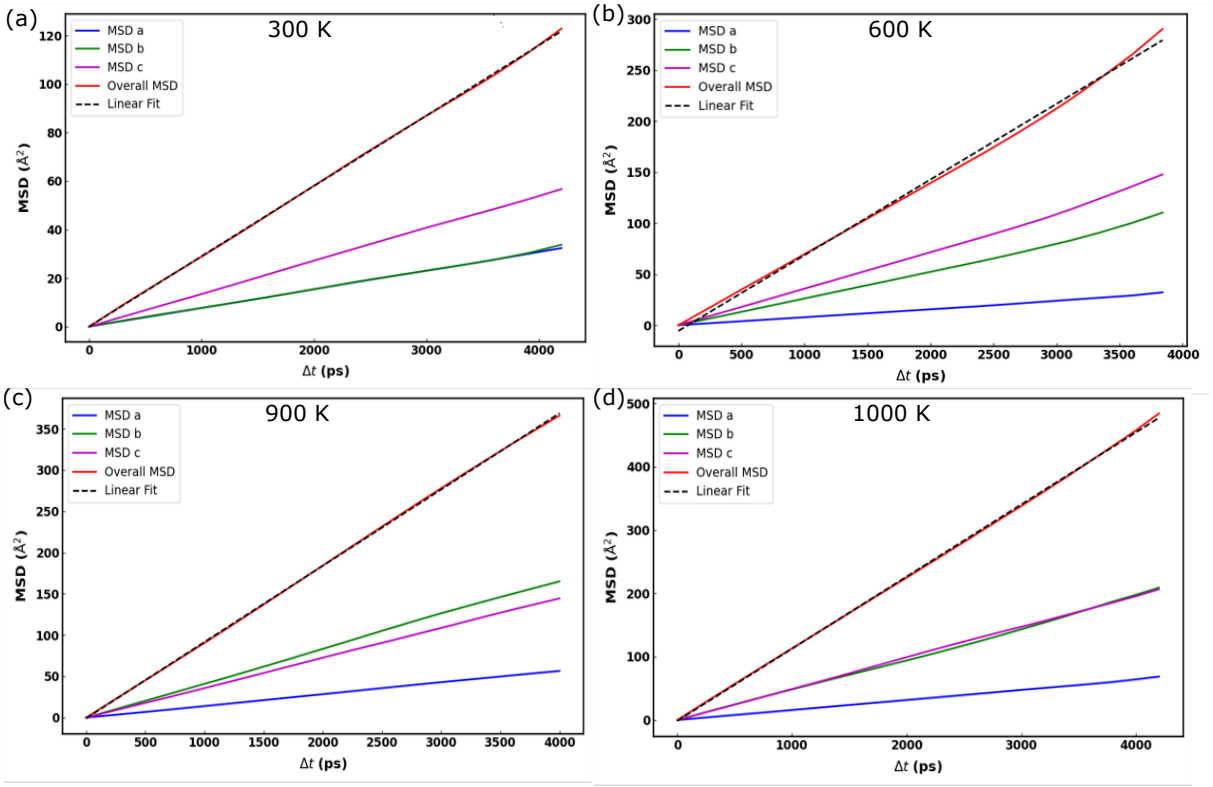

Figure S11. MTP-calculated MSD vs. Δt curves for a 1×2×3 MgV_2O_5 supercell over 4000 ps at (a) 300 K, (b) 600 K, (c) 900 K, and (d) 1000 K. Notations used in each panel are identical to those used in **Figure S7**.

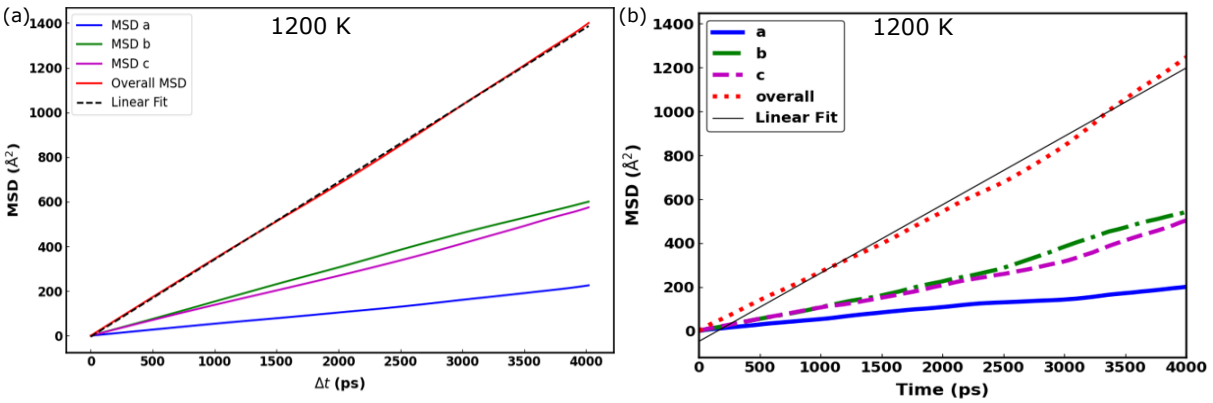

Figure S12. MTP-calculated (a) MSD vs. Δt and (b) MSD vs. t curves at 1200 K for a 1×2×3 MgV_2O_5 supercell over 4000 ps. Notations used in each panel are similar to those used in **Figure S8**.

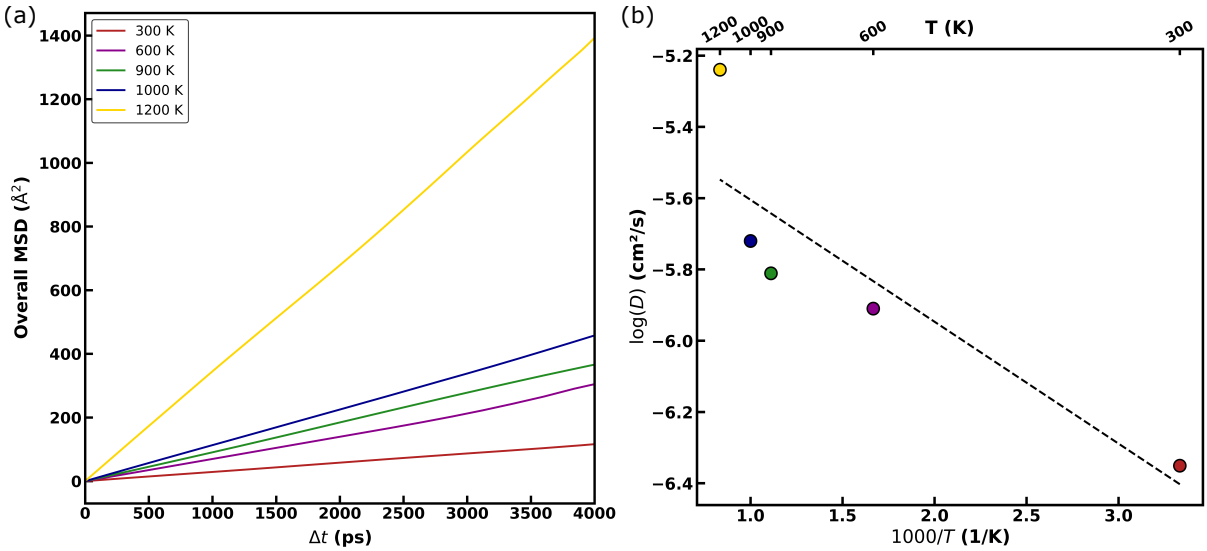

Figure S13. (a) MTP-calculated overall MSD vs. Δt curves at different temperatures for a 1×2×3 MgV_2O_5 supercell over 4000 ps. Red, purple, green, blue, and yellow lines indicate MSD data at 300 K, 600 K, 900 K, 1000 K, and 1200 K, respectively. (b) Arrhenius plot of log D (with D in cm^2/s) plotted against $1000/T$ (top x-axis is T), obtained from the data of panel a. Colors of data points indicate their temperatures and are identical to the colors used in panel a. Dotted black line indicates a linear fit, which yields an E_a of 0.068 eV.

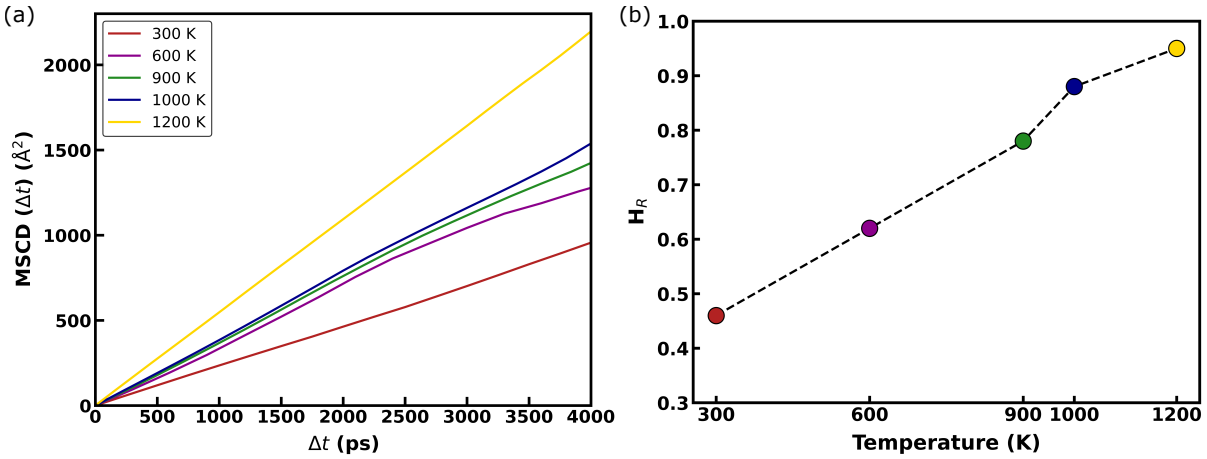

Figure S14. Mean square displacement of the centre-of-mass (MSCD) of Mg atoms versus Δt calculated using MTP-MD in a 1×2×3 MgV_2O_5 supercell over 4000 ps at different temperatures. (b) Haven's ratio (H_R) calculated using the data of panel a and **Figure S13a**, as a function of temperature. Colors used in both panels are identical to those used in **Figure S13**.

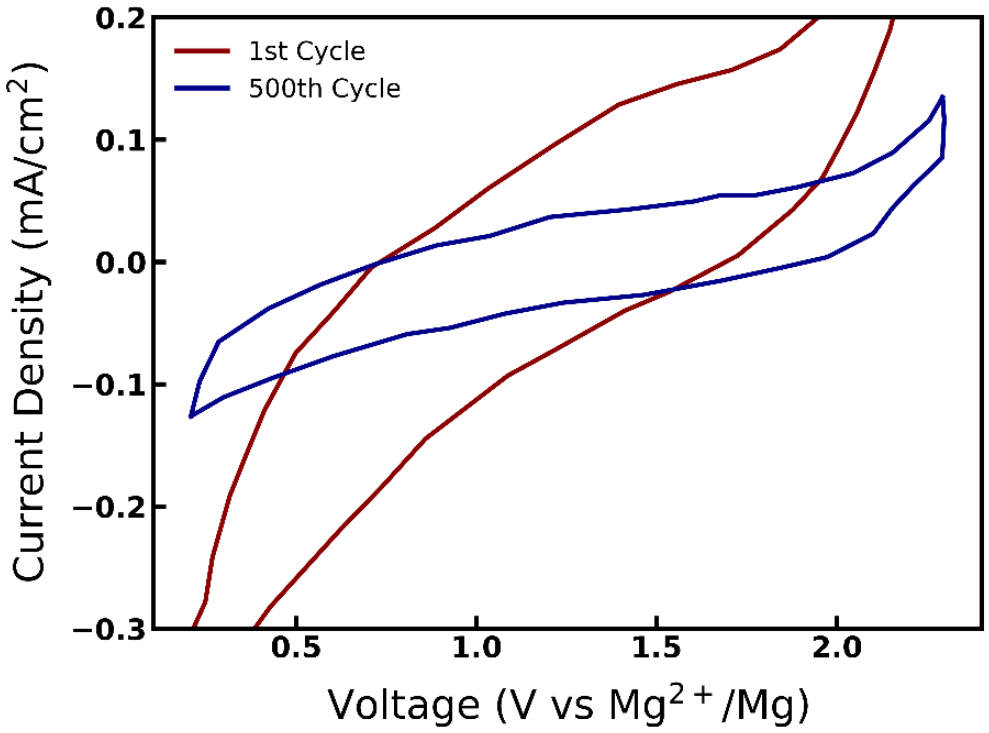

Figure S15. Digitized Mg cyclic voltammetry (CV) performance of the 12 h Na_2S-V_2O_5-P_2O_5 glassy composite electrode measured over 500 consecutive cycles at room temperature. The CV scans were performed from 0.2 to 2.3 V vs Mg^{2+}/Mg at a scan rate of 0.2 V s^{-1}. The brown curve corresponds to the 1st cycle, while the blue curve represents the 500th cycle. The CV data is reproduced with permission from Wally et al., [J. Alloys Compd. 895, 162644 (2022)], which is cited as Ref. 46 in the main text. All copyrights are reserved with 2022 Elsevier B.V.