Machine-learning potentials for nanoscale simulations of deformation and fracture: example of TiB_2 ceramic

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Supplementary Materials

- **Finite-temperature equilibration of TiB_2 supercells.** Fig. S1 illustrates a procedure used to evaluate equilibrium lattice parameters of TiB_2 (a 720-atom supercell with x, y, z axes chosen as $x \parallel [10\bar{1}0]$, $y \parallel [\bar{1}2\bar{1}0]$, $z \parallel [0001]$) at 300 and 1200 K. The supercell is employed to produce the initial training data (Section "1 Training procedure and fitting initial MLIPs") and to carry out all atomic scale tensile and shear tests (Section "2 MLIPs' validation against atomic scale tensile tests", "5 Other loading conditions and MLIP's transferability").

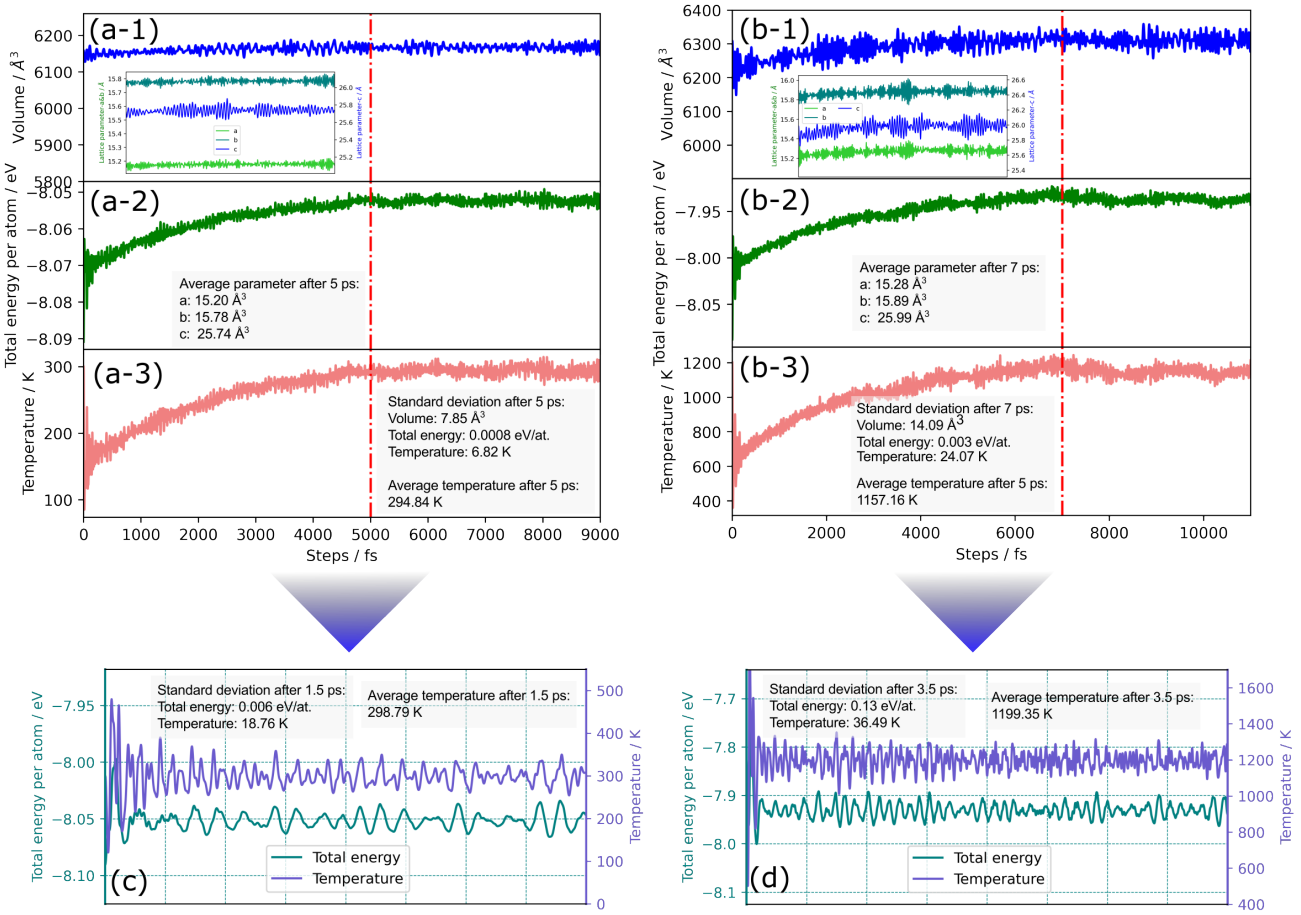

Figure S1. The volume, total energy, and temperature evolution during an AIMD simulation with an NPT ensemble, using a 720-atom TiB_2 supercell and temperature of (a) 300 and (b) 1200 K. Total energy variation during a subsequent NVT simulation with time-averaged lattice parameters—calculated based on the NPT data after the red vertical line (satisfactory convergence of volume, total energy, and temperature)—at (c) 300 K and (d) 1200 K. Specifically, equilibration is reached after 5 ps: steady trends in property *vs*. time. Thus, equilibrium lattice parameters (a, b, and c) are calculated as time-averages after 5 ps till the end of the simulation.

- **Validation of "intermediate" MLIPs (MLIP-[1-3]) against atomic scale AIMD tensile tests and their performance at the nanoscale.** Fig. S2 compares AIMD and ML-MD stress/strain curves of TiB_2 subject to [0001], [$10\bar{1}0$], and [$\bar{1}2\bar{1}0$] tensile loading, including representative snapshots of the simulation cells close to the fracture point (indicating that AIMD fracture pathways are correctly reproduced). The ML-MD results are obtained obtained using MLIP-[1-3]. As discussed in the manuscript (Section "3 MLIPs' up-fitting for nanoscale tensile tests"), these are "intermediate" MLIPs applicable at the atomic scale but not yet sufficient to simulate TiB_2's fracture at the nanoscale. Besides high extrapolation grades (please read Section "3 MLIPs' up-fitting for nanoscale tensile tests"), this is further illustrated in Fig. S3 depicting one of our nanoscale supercells (*S*1) close to the fracture point. The usage of MLIP-[1–3] leads to lost atoms (and MD simulation crushing abruptly), while MLIP-[4] (described and validated in the manuscript) allows to continue the simulation until full fracture (while exhibiting reasonably low extrapolation grades).

- **Training set analysis: comparison between MLIP-[1] and MLIP-[4]**. In the manuscript (Section "3 MLIPs' up-fitting for nanoscale tensile tests"), we discuss qualitative differences between the training set of MLIP-[1], TS(MLIP-[1]), and the training set of MLIP-[4], TS(MLIP-[4]), where the latter is applicable to simulate TiB_2's fracture subejct to tensile loading at the nanoscale, in contrast to the former, which can be only used at the atomic scale. Fig.S4 depicts the bond angle and bond length distribution for the two training sets, indicating only minor differences.

- **Transferability of MLIP-[4] to atomic scale tensile tests at 1200 K**. Fig. S5 presents validation of MLIP-[4]—in terms of stress/strain curves and fracture mechanisms—against atomic scale AIMD tensile tests at 1200 K (see Section "5 Other loading conditions and MLIP's transferability").

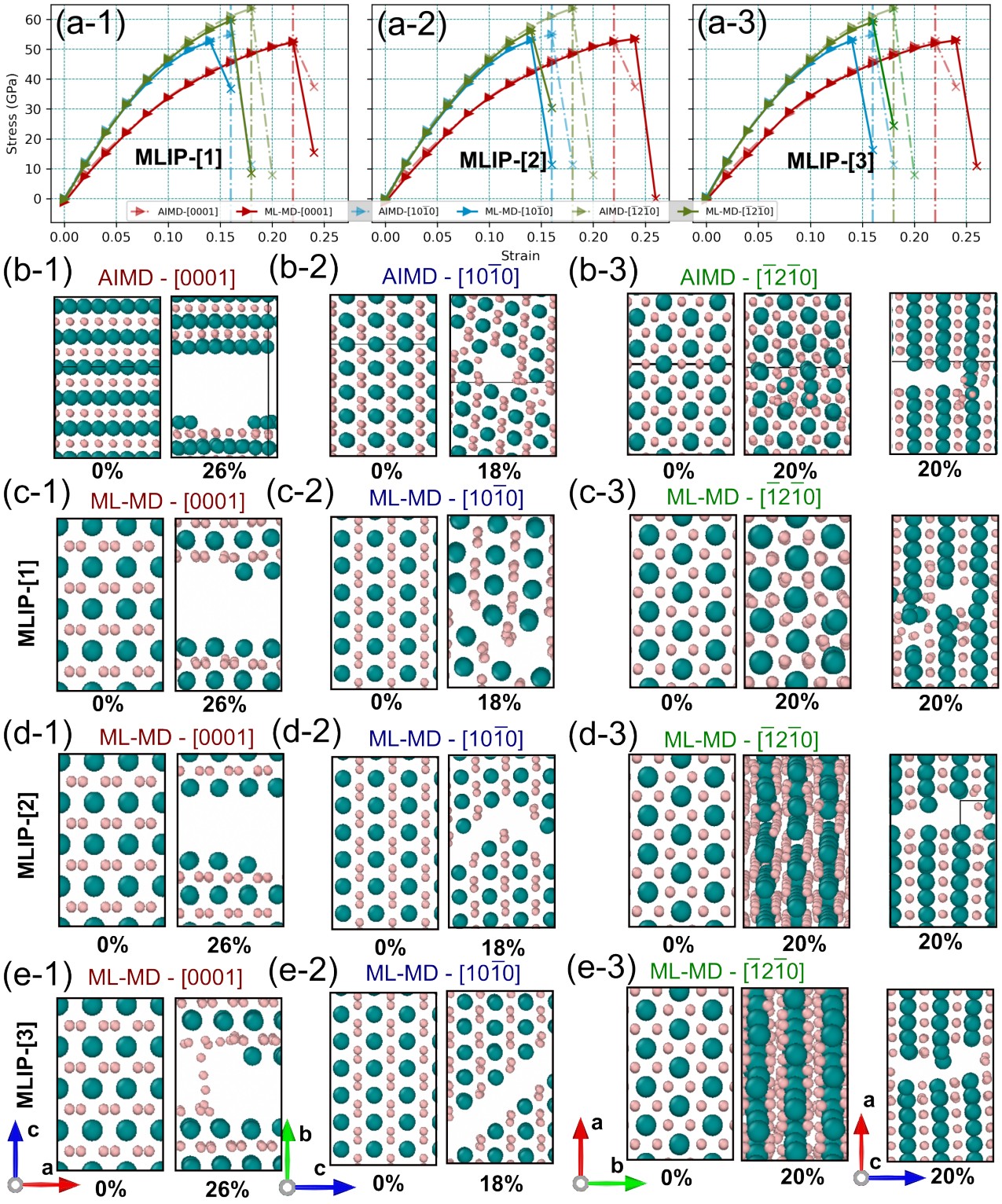

Figure S2. Validation of "intermediate" MLIPs (MLIP-[1-3]) against atomic scale AIMD tensile tests (see Section "3 MLIPs' up-fitting for nanoscale tensile tests"). (a) Comparison of AIMD and ML-MD stress/strain curves for TiB_2 subject to [0001], [10$\bar{1}$0], and [$\bar{1}$2$\bar{1}$0] tensile deformation at 300 K. All simulations were carried out in a 720-atom TiB_2supercell. Only the stress component in the loaded direction is plotted. Snapshots of the supercell close to the fracture points in (b) AIMD and ML-MD simulations using (c) MLIP-[1], (d) MLIP-[2], and (e) MLIP-[3].

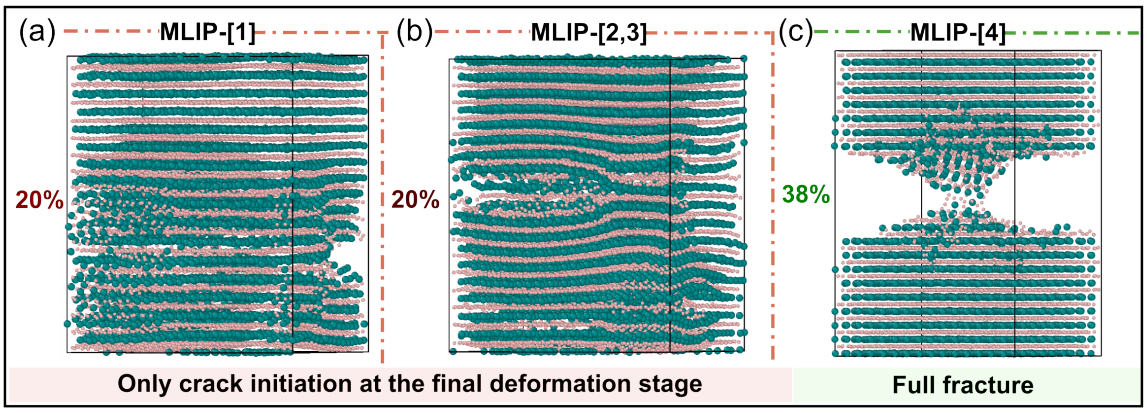

Figure S3. Snapshots of the *S*1 supercell during nanoscale [0001] tensile test close to the fracture point, taken from ML-MD simulations using different MLIPs: (a) MLIP-[1], (b) MLIP-[2,3], (c) MLIP-[4]. As detailed in the manuscript (Section "3 MLIPs' up-fitting for nanoscale tensile tests"), only MLIP-[4] allows to capture the deformation until full fracture, while MLIP-[1–3] only show very initial stages of crack growth.

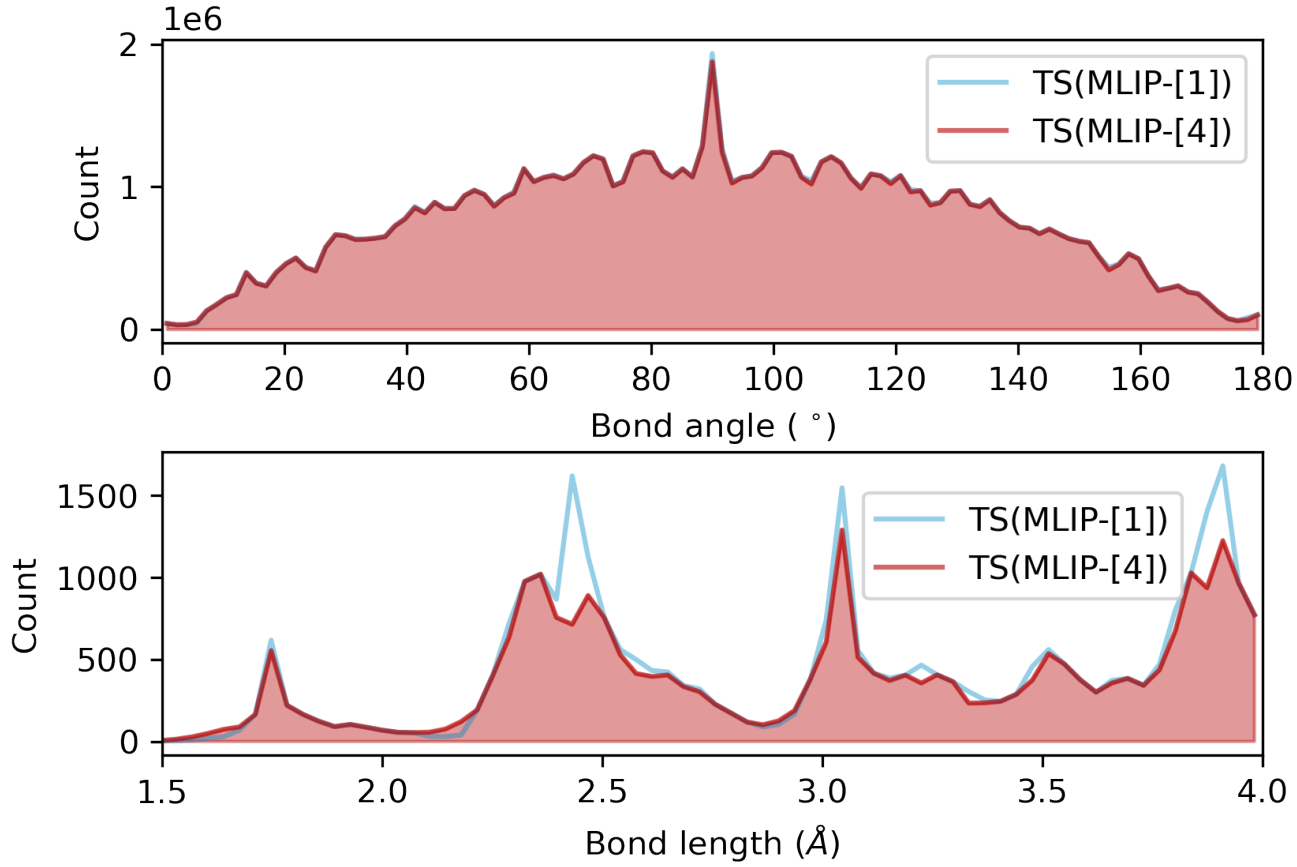

Figure S4. (a) Bond angle and (b) bond length distribution in the training sets of MLIP-[1], TS(MLIP-[1]), and MLIP-[4], TS(MLIP-[4]).

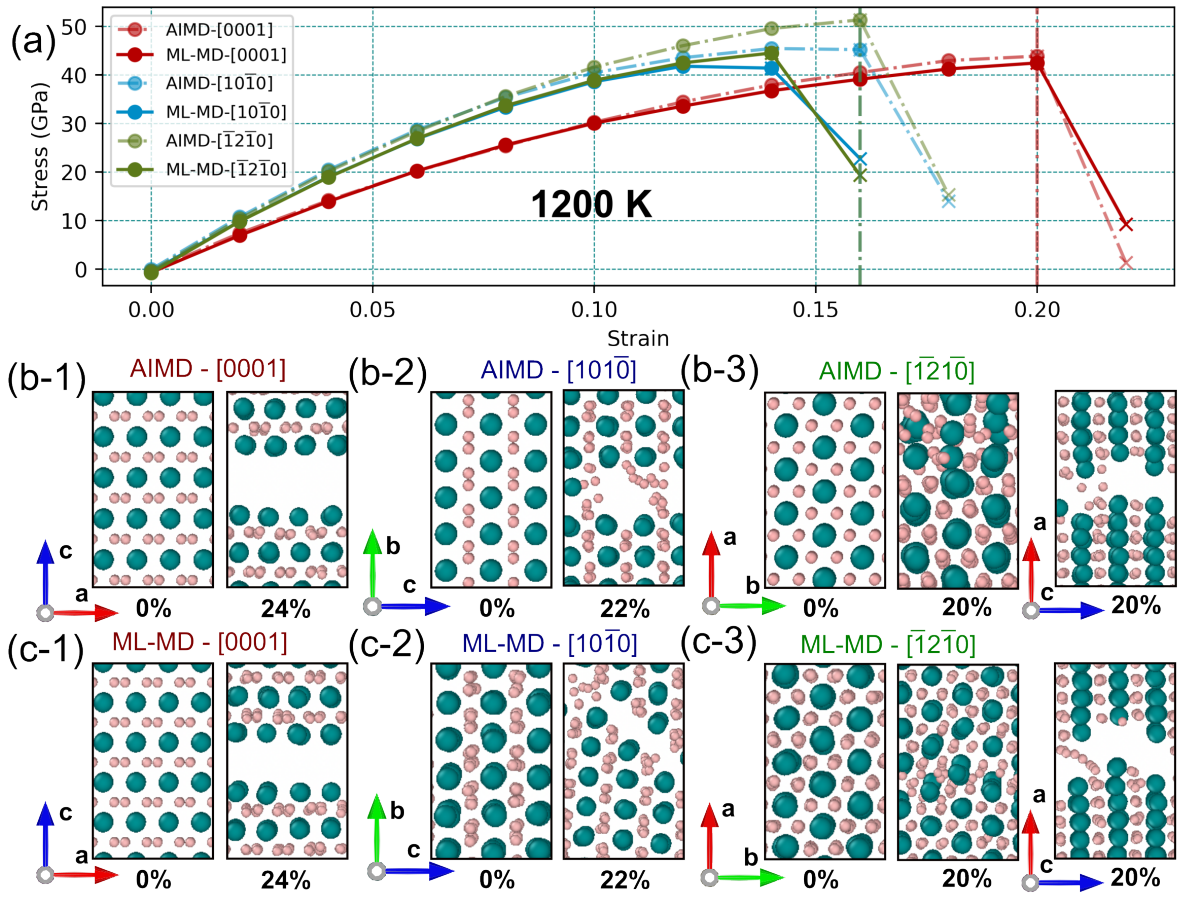

Figure S5. Validation of MLIP-[4] against atomic scale AIMD tensile tests (see Section "5 Other loading conditions and MLIP's transferability"). (a) Comparison of AIMD and ML-MD stress/strain curves for TiB_2 subject to [0001], [10$\bar{1}$0], and [$\bar{1}$2$\bar{1}$0] tensile deformation at 1200 K. All simulations were carried out in a 720-atom TiB_2supercell. Only the stress component in the loaded direction is plotted. Snapshots of the supercell close to the fracture points in (b) AIMD and(c) ML-MD simulations.