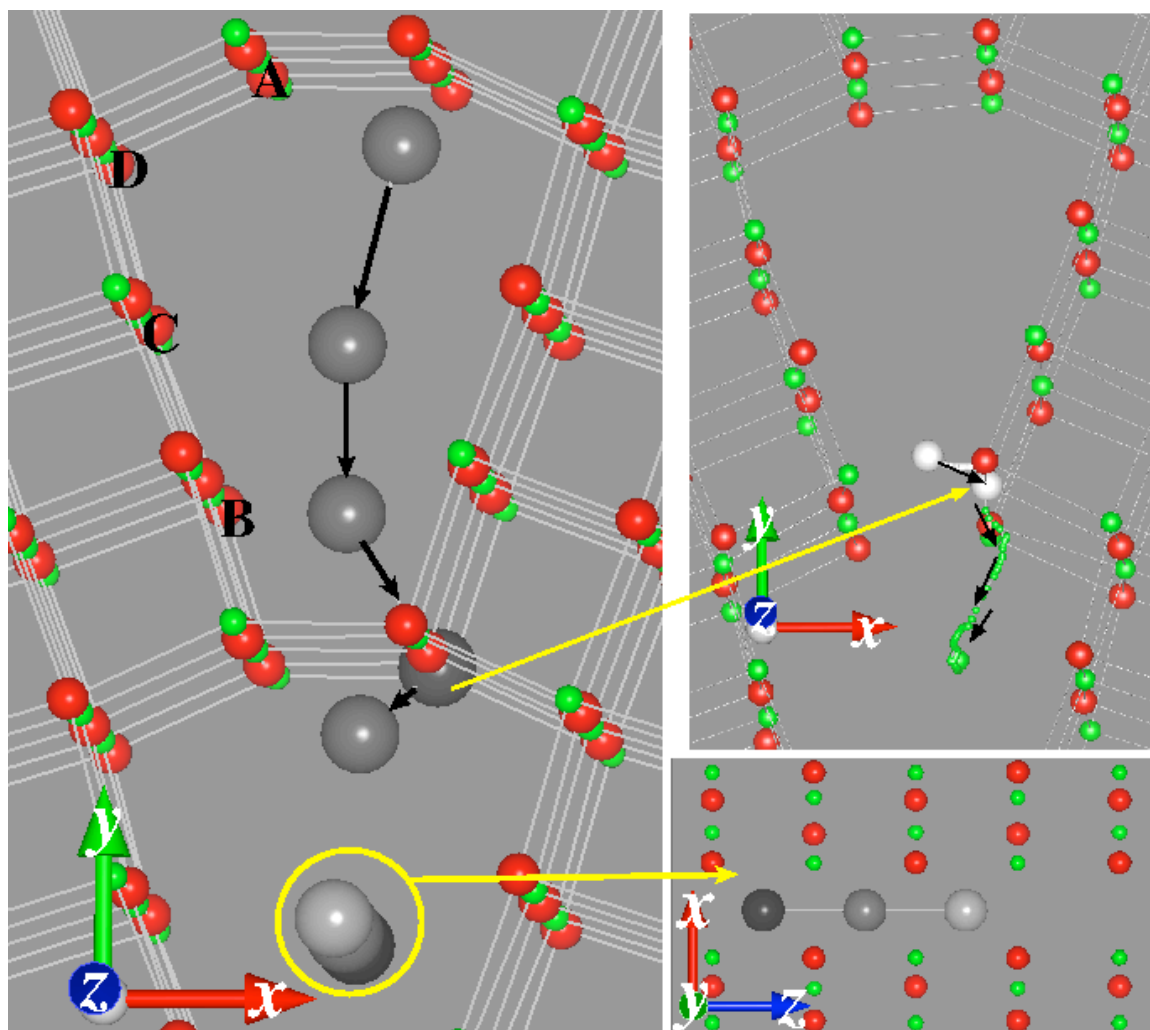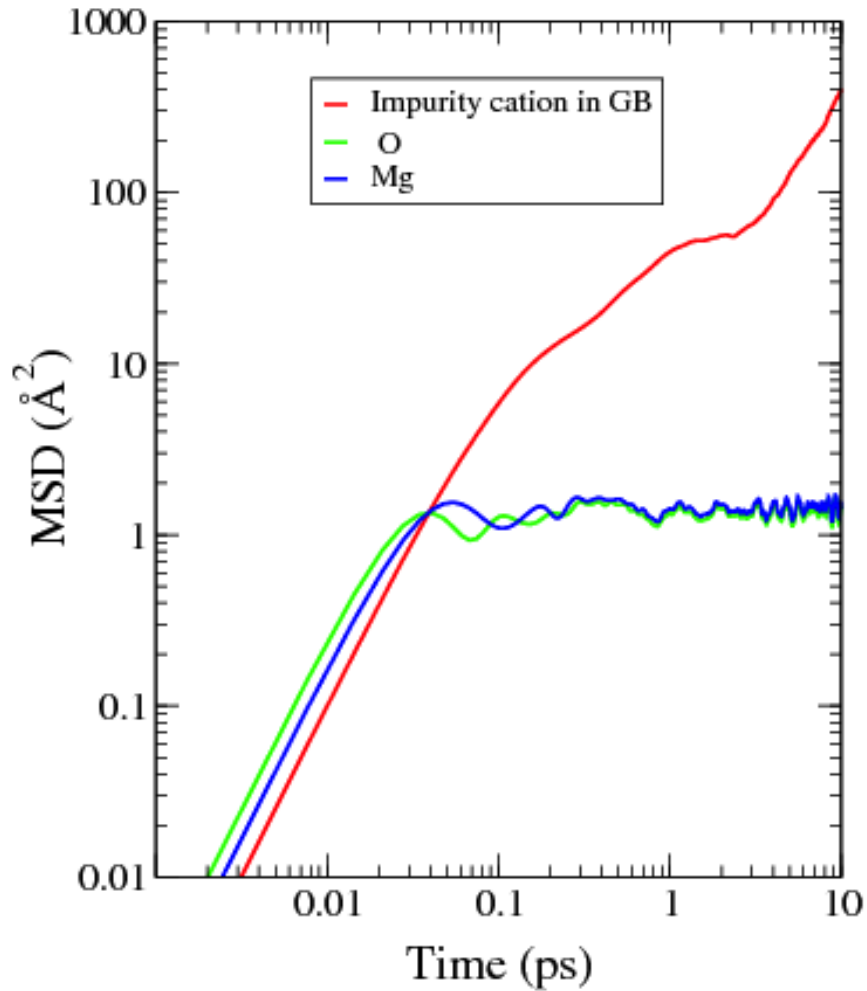# Supplementary Information

First-Principles Prediction of Pressure Enhanced Defect Segregation and Migration at MgO Grain Boundaries

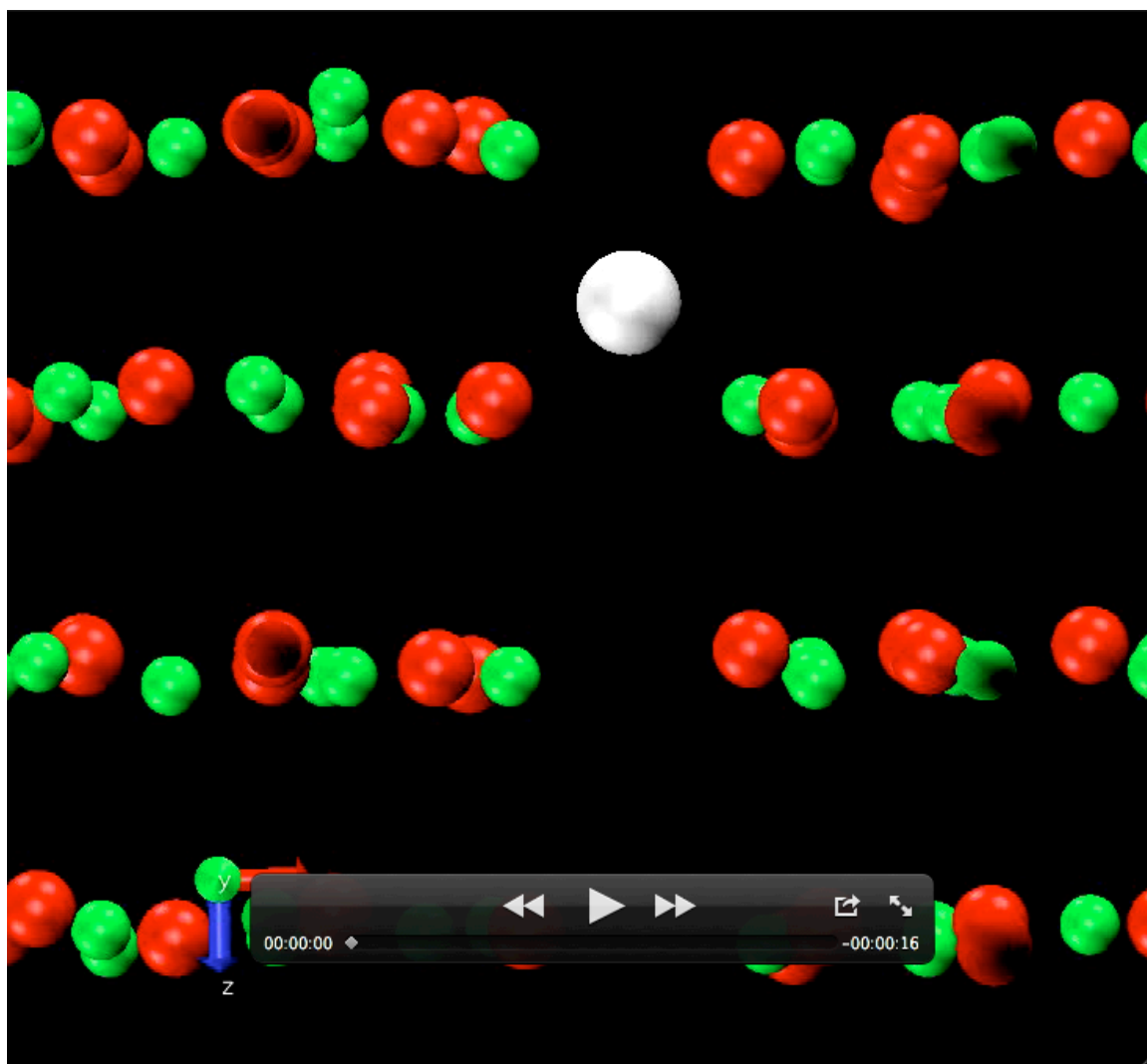Bijaya B. Karki, Dipta B. Ghosh, and Ashok K. Verma

S1: Possible paths for cation (Mg/Ca/Al) migration *along* (left image) and *down* (lower portion of left image, and bottom right image) at 0 GPa. The successive positions of the migrating ion are displayed by large sphere with gray color representing the *z*-coordinate. Top right image displays the substitution mechanism that occurs during the migration (i.e., the impurity ion knocking out the lattice Mg). The green (small) and red (large) spheres represent Mg and O, respectively.

S2: Mean square distance (MSD) plotted as a function of time for Ca impurity ion and host Mg and O ions at 1000 K and 0 GPa, calculated using position-time series from our first-principles molecular dynamics simulations of MgO tilt boundary. For the impurity ion, the MSD plot shows a clear diffusive regime where the MSD-time slope in the log-log scale is roughly equal to the unity. Also, the impurity ion has moved by several Angstroms during 10 picoseconds. However, both Mg and O ions do not show any significant movements.

2

S3: Visualization of first principles molecular dynamics simulation of MgO tilt symmetric boundary performed at 0 GPa and 1000 K containing an impurity Ca ion (shown as large white sphere) in the boundary region. The horizontal and vertical directions represent x ad z directions, respectively. The green (small) and red (large) spheres represent Mg and O ions, respectively. The simulation was run for 10 picoseconds (with a time step of 1 femtosecond) of which only one portion is covered in the video file **VideoS3.avi**. As one can see, the Ca ion moves downward by one full lattice constant within a couple of picoseconds.

S4: Possible paths for cation (Mg/Ca/Al) migration at 100 GPa from one GB core to next core. The successive positions of the migrating ion *along* (positions 1 to 6) and *down* (positions 7 to 10 via 6) the boundary are shown by large (numbered) sphere with gray-scale representing the *z*-coordinate. Note that two knockout events occur, one at the position 2 and the other at the position 6 for along (*y*-direction) migration. The *x-z* view is shown for the along (b) and down (c) migration. The green (small) and red (large) spheres represent Mg and O, respectively.