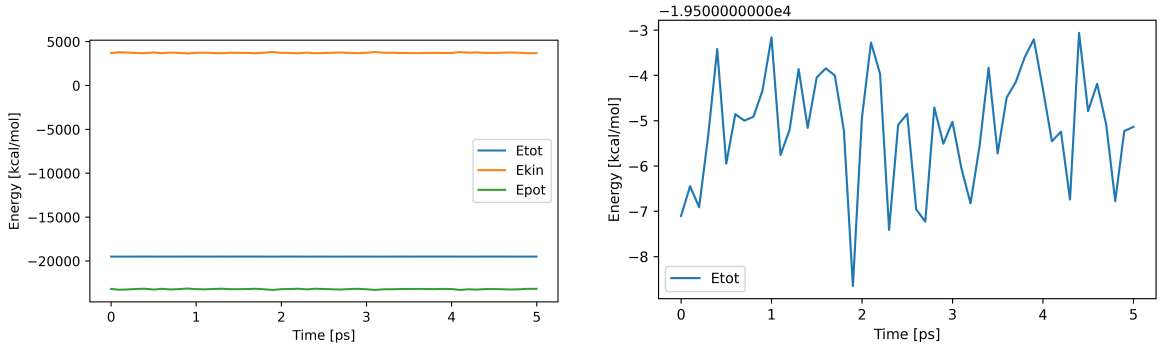
Supporting Information

April 14, 2023

1 Additional analysis DMOF-1

1.1 Energy conservation

The method was tested concerning energy conservation in an NVE run for 5 ps using the open pore form of the 4x4x4 NC as a test system. The reference volume corresponded to the current volume of the test system, and the force constant was set to 0.002 $kcal \cdot mol^{-1} \cdot \mathring{A}^{-6}$. The kinetic energy, the potential energy, and the total energy are shown in Figure 1. The total energy is constant during the simulation, which verifies the energy conservation of the method.



(a) The kinetic energy, the potential energy, and the total energy.

(b) Close up of the total energy.

Figure 1: Testing energy conservation using the 4x4x4 NC during a 5 ps simulation in the NVE ensemble. The total energy is constant during the simulation, which verifies the energy conservation of the method.

1.2 Umbrella sampling: Overlap of the respective Windows

To confirm that the selected windows have a sufficient overlap required for a valid free energy profile, the distributions of the collective variable (CV) were illustrated by histograms as seen in Figure 2. The overlap between adjacent histograms of the reference volume is sufficient. A

more extensive overlap could be archived by additional windows or a smaller force constant allowing more volume fluctuations.

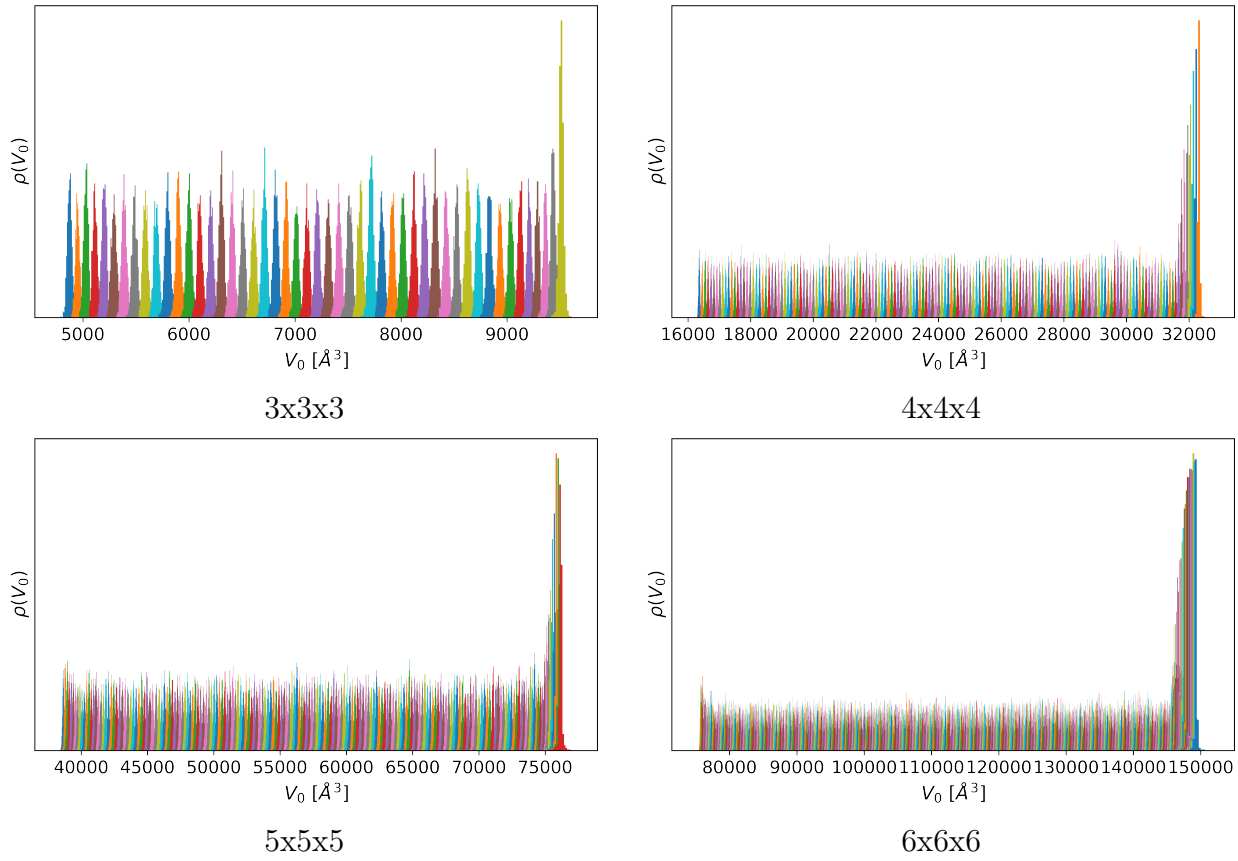


Figure 2: Probability distributions of the CV trajectories for all investigated NC of sizes. The histograms for each reference volume sufficiently overlap with the adjacent histogram.

2 Additional analysis DUT-128

2.1 Umbrella sampling: Overlap of the respective windows

To confirm that the selected windows have a sufficient overlap required for a valid free energy profile, the distributions of the collective variable (CV) were illustrated by histograms as seen in Figure 3. The overlap between adjacent histograms of the reference volume is sufficient. A more extensive overlap could be archived by additional windows or a smaller force constant allowing more volume fluctuations.

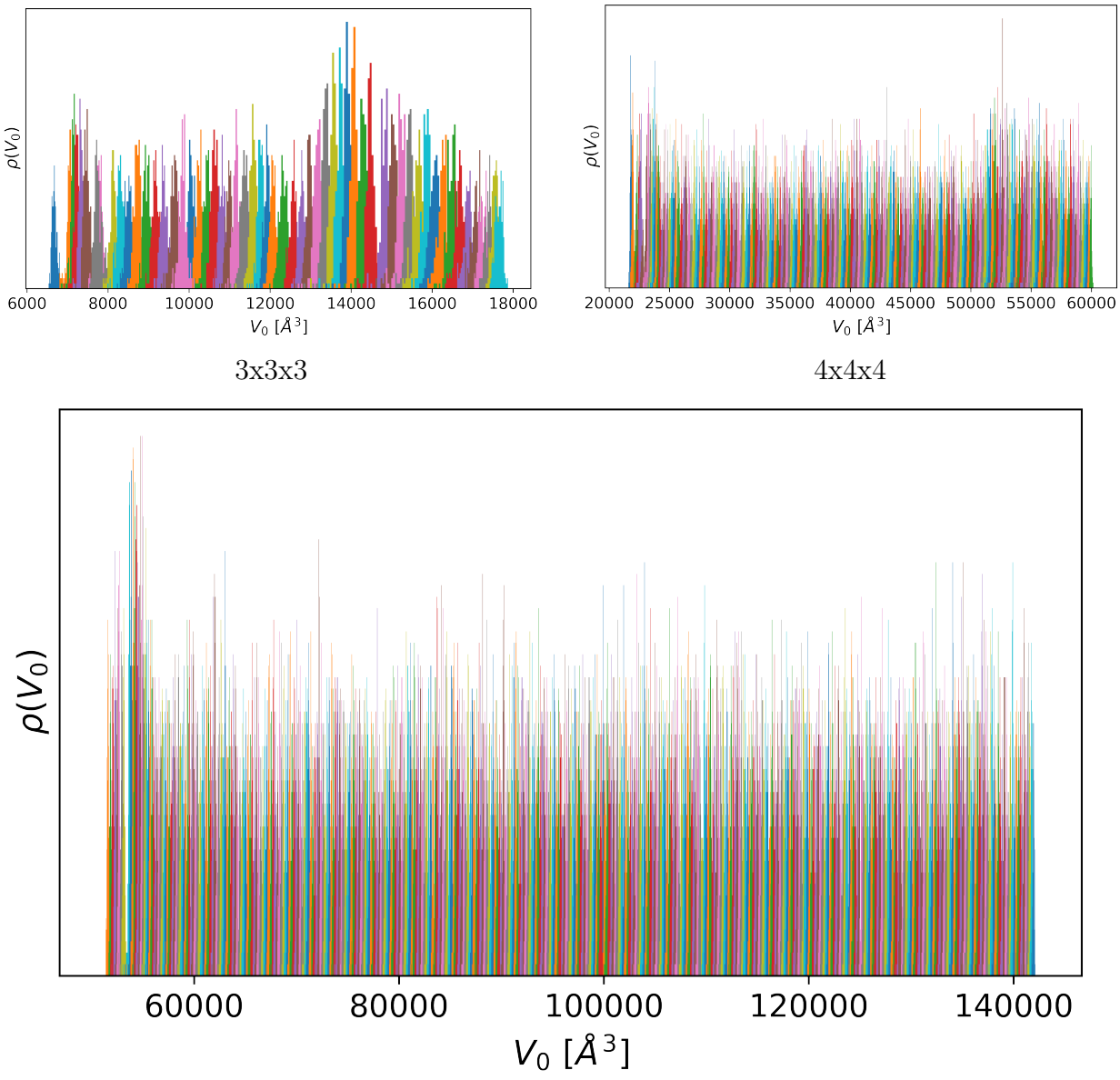


Figure 3: Probability distributions of the CV trajectories for all investigated NC of sizes. The histograms for each reference volume sufficiently overlap with the adjacent histogram.