

Electronic Supplementary Information: Mechanochemical Synthesis of Glycine Oligomers in a Virtual Rotational Diamond Anvil Cell

Brad A. Steele,[†] Nir Goldman,^{†,‡} I-Feng W. Kuo,[†] and Matthew P. Kroonblawd^{*,†}

[†]*Physical and Life Sciences Directorate, Lawrence Livermore National Laboratory,
Livermore, CA 94550, United States*

[‡]*Department of Chemical Engineering, University of California, Davis, California 95616,
United States*

E-mail: kroonblawd1@llnl.gov

1 Supplemental Figures

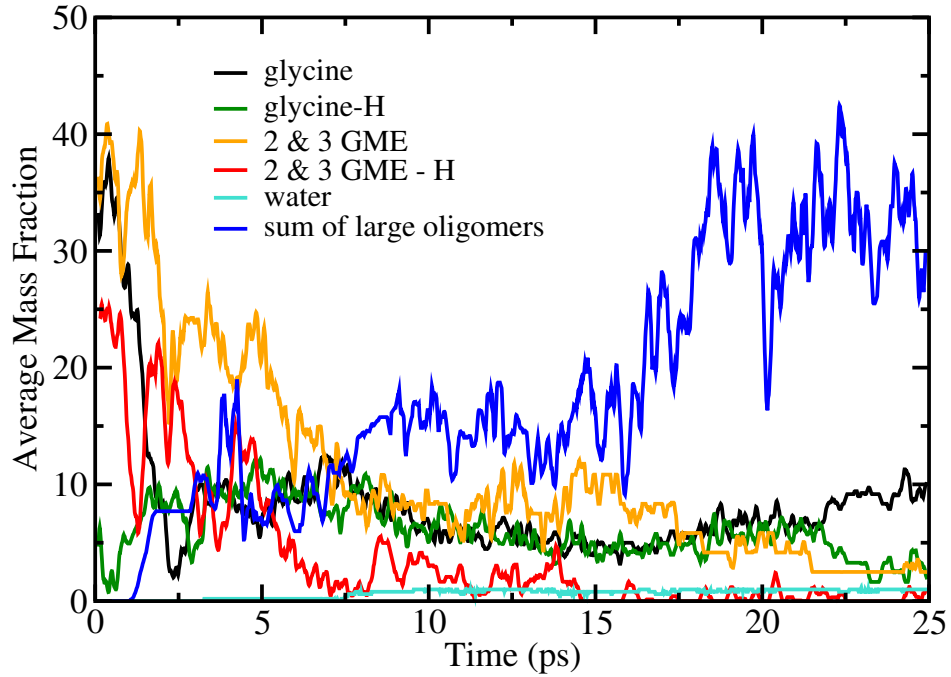


Figure 1: The average mass fraction during shear at 15.6 GPa ($L_z = 8 \text{ \AA}$) and anvil velocity $V_x = 1.0 \text{ \AA ps}^{-1}$. In this analysis, bond distance cutoffs were reduced to 1.6 \AA for non-H covalent bonds and 1.2 \AA for covalent H bonds. Using these short cutoffs the mass fractions generally decrease, but their trend with time is qualitatively similar to those with larger cutoffs discussed in the manuscript.

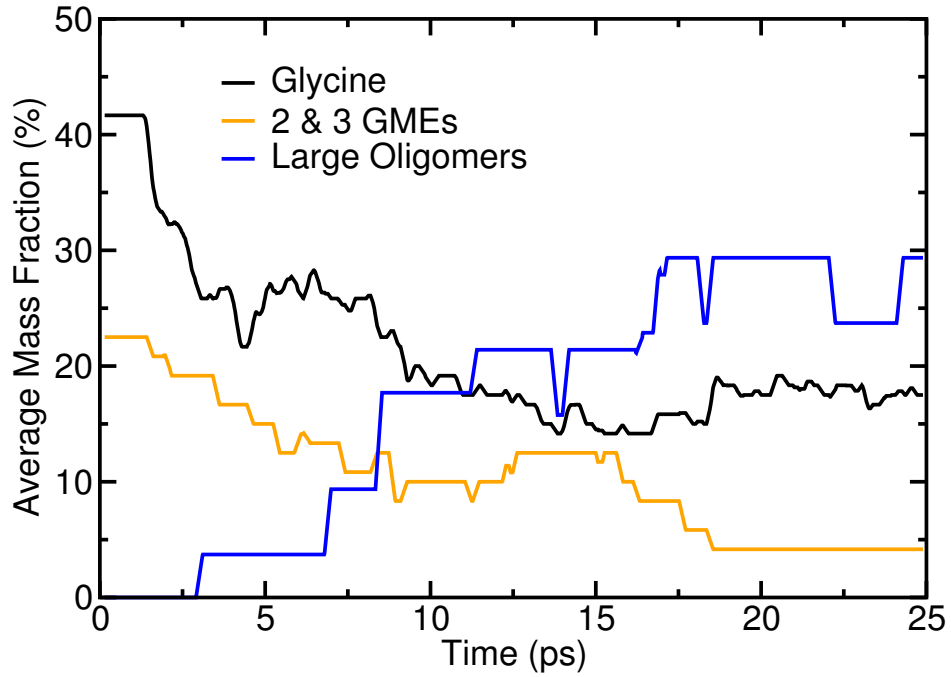


Figure 2: The average mass fraction during shear at 15.6 GPa ($L_z = 8 \text{ \AA}$) and anvil velocity $V_x = 1.0 \text{ \AA ps}^{-1}$. In this analysis, the hydrogen atoms were removed to analyze the chemistry occurring in the CNO backbone structure more accurately. The analysis shows that backbone-altering reactions are not an artifact of the cutoffs for hydrogen and are qualitatively similar to those with hydrogen discussed in the manuscript.

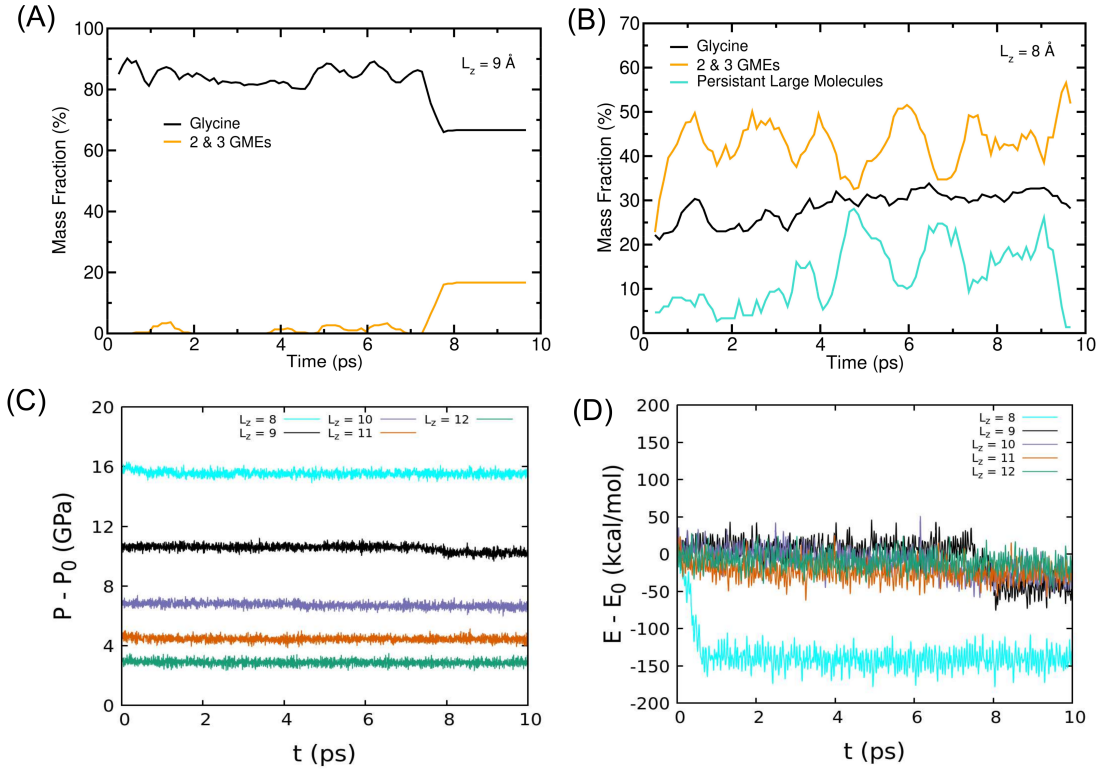


Figure 3: Time histories for the initial 10 ps *NVT* equilibration simulations at static pressure including (A) chemical analysis at 10.2 GPa, (B) chemical analysis at 15.6 GPa, (C) system pressure for all five L_z values, and (D) change in energy for all five L_z values. Chemical analysis shows the appearance of 2 & 3 GMEs at the second highest pressure and a fluctuating concentration of 2 & 3 GMEs and large oligomers at the highest pressure. System pressure and energy are approximately constant, with any abrupt changes being clearly correlated to chemical events.

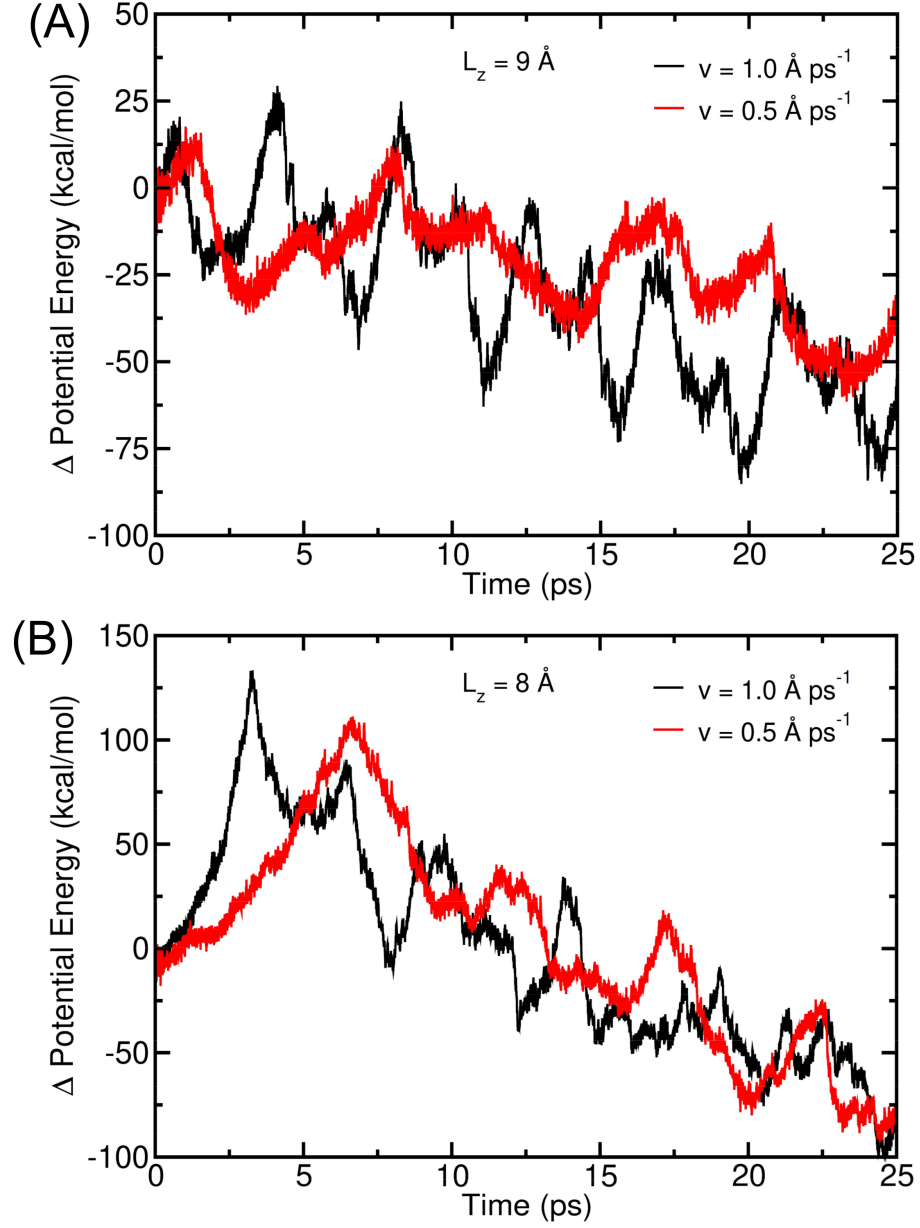


Figure 4: Ensemble-average change in potential energy during shear at (A) 10.2 GPa and (B) 15.6 GPa for both shear rates.

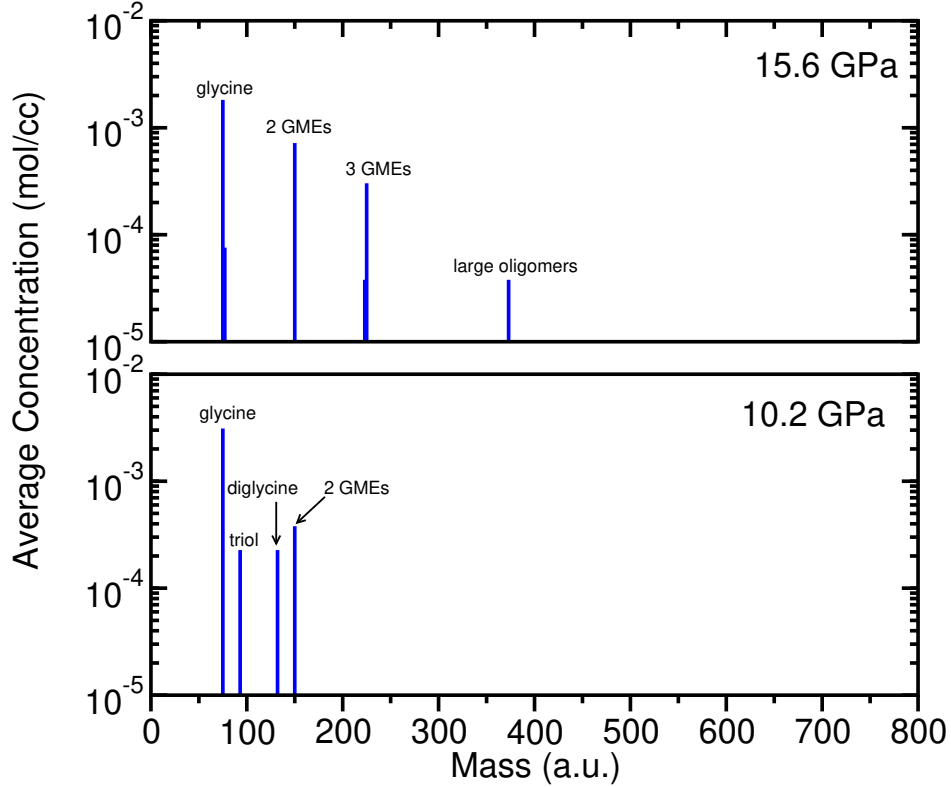


Figure 5: The mass spectrum (on a log scale) of recovered products for the control case where no shear is applied at the two highest pressures. The spectra were obtained from the ensembles of the final *NVT* equilibration simulations at ambient conditions. Dividing the concentrations by $3.7 \times 10^{-5} \text{ mol cm}^{-3}$ yields the molecule count in all ten simulations. Only ten unique molecules were identified across the contributing 20 simulations, which indicates a significantly less diverse set of products compared to those formed under shear. The triol species found in the 10.2 GPa case is the same glycine-like triol intermediate identified in Figure 7 of the main article.

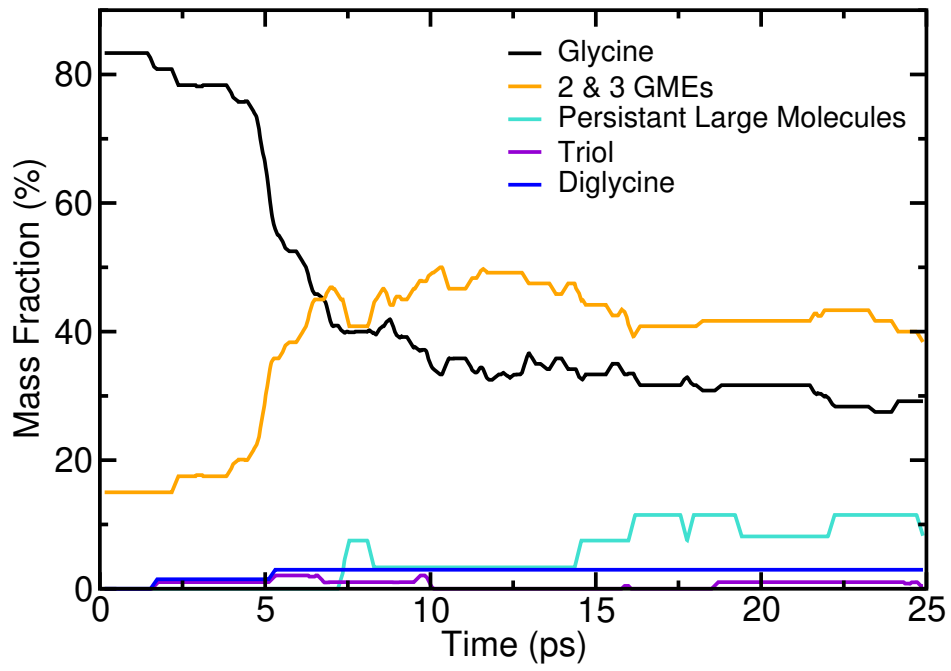


Figure 6: The ensemble-average mass fraction during shear at 10.2 GPa and with the slower diamond anvil shear velocity of 0.5 \AA ps^{-1} . In this simulation, 3 glycine molecules react into diglycine and triol without the formation of molecular water.

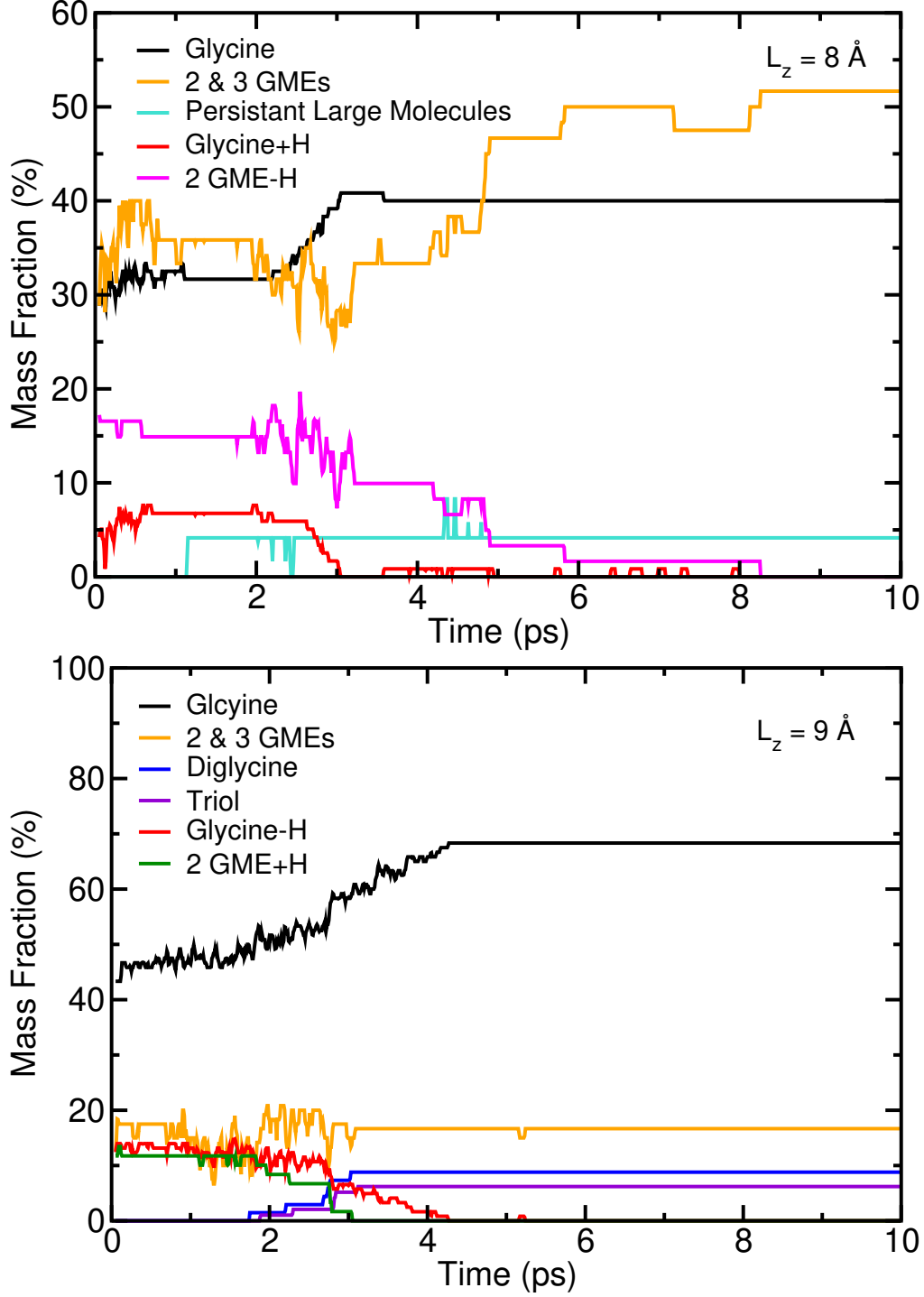


Figure 7: Ensemble-average mass fractions of species found during pressure unloading for the two no-shear cases ($V_x = 0$) including 15.6 GPa ($L_z = 8 \text{ \AA}$) and 10.2 GPa ($L_z = 9 \text{ \AA}$). Diglycine and the triol molecule found in the 10.2 GPa no-shear case are formed during unloading and not while held at static pressure. Final populations of large molecules are substantially lower in both no-shear ensembles than in any of the sheared ones at a given pressure.