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Electronic Supplementary Information: Synthesis of Functionalized Nitrogen-containing Polycyclic Aromatic Hydrocarbons and other Prebiotic Compounds in Impacting Glycine Solutions

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In order to validate our force matched DFTB model, detailed comparison was made with one of the simulations discussed in our manuscript to our DFT-MD simulation of the water glycine mixture at 2.5 g cm^{-3} and 3000 K. Analysis of all possible radial distribution functions (RDFs), denoted $g(R)$, shows close agreement between the two simulations (Fig. S1). In general, differences in peak heights are small and correspond to small differences in free energies. For instance, the deviation in the first peak of the C-H RDF corresponds to a free energy change of only $\sim 1 \text{ kcal mol}^{-1}$. These deviations can in part be attributed to the short time and length scales of the simulations, and due to their uncorrelated initial positions and velocities.

We determine the specific chemical reactivity in our simulations using a pre-established methodology of optimal bond cutoff distances and lifetimes.¹⁻⁵ The optimal value for r_c to distinguish between bonded and nonbonded atomic sites is given by the first minimum in the corresponding pair radial distribution function $g(R)$, which corresponds to the maximum of the potential of mean force through $W(R) = -k_B T \ln[g(R)]$, for all possible bonding pairs.¹ In addition, in order to avoid counting species that were entirely transient and not chemically bonded,² we also chose a lifetime cutoff τ_c of 20 femtoseconds for C-H, O-H, and N-H bonds and 50 femtoseconds for all others. No H-H bonds were detected in any of our simulations. This criteria is intuitive since bonds with this lifetime could conceivably be detected spectroscopically. As a result, atom pairs were considered to be bonded only if they resided within a distance of each other of r_c for a time of greater than τ_c . The concentrations of species at high pressure and temperature can have some dependence on bond and lifetime criteria (as expected), which has been shown for other hot dense materials.^{1,4,6} We found that the overall conclusions of this work were independent of these parameters.

We have validated the chemical reactivity from our DFTB model by computing the total number of C-C, C-N, and C-O bonds over the first 5 ps of one of our simulations, with comparison to results from DFT (Fig. S2). Given the short chemical lifetimes and complex reactivity in our simulations, this viewpoint allows for a detailed chemical picture under these conditions without relying on the fate of specific compounds and any intermediates they might form. In general, we find strong agreement between both sets of results. Initial differences between DFTB and DFT for C-C bond chemistry can again be attributed to the small spatial and time scales of the simulations and their uncorrelated

initial conditions. We note that the DFTB results exhibit closer agreement with DFT after several ps of simulation time.

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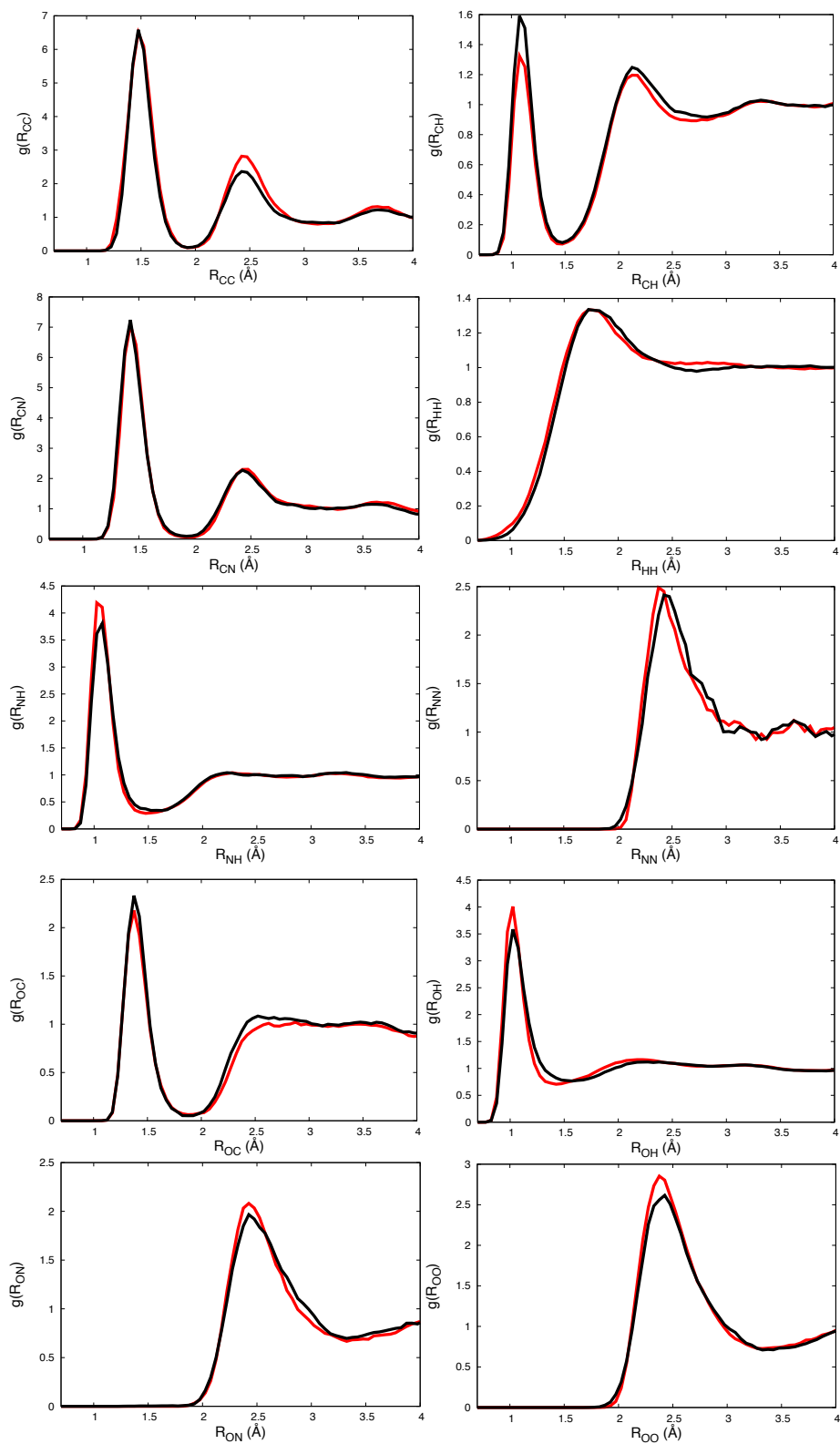


Fig. S1 Radial distribution functions computed for all ten atomic pair types from both DFT (black lines) and one of our forced matched DFTB simulations (red lines), averaged over the first 5 ps of simulation time.

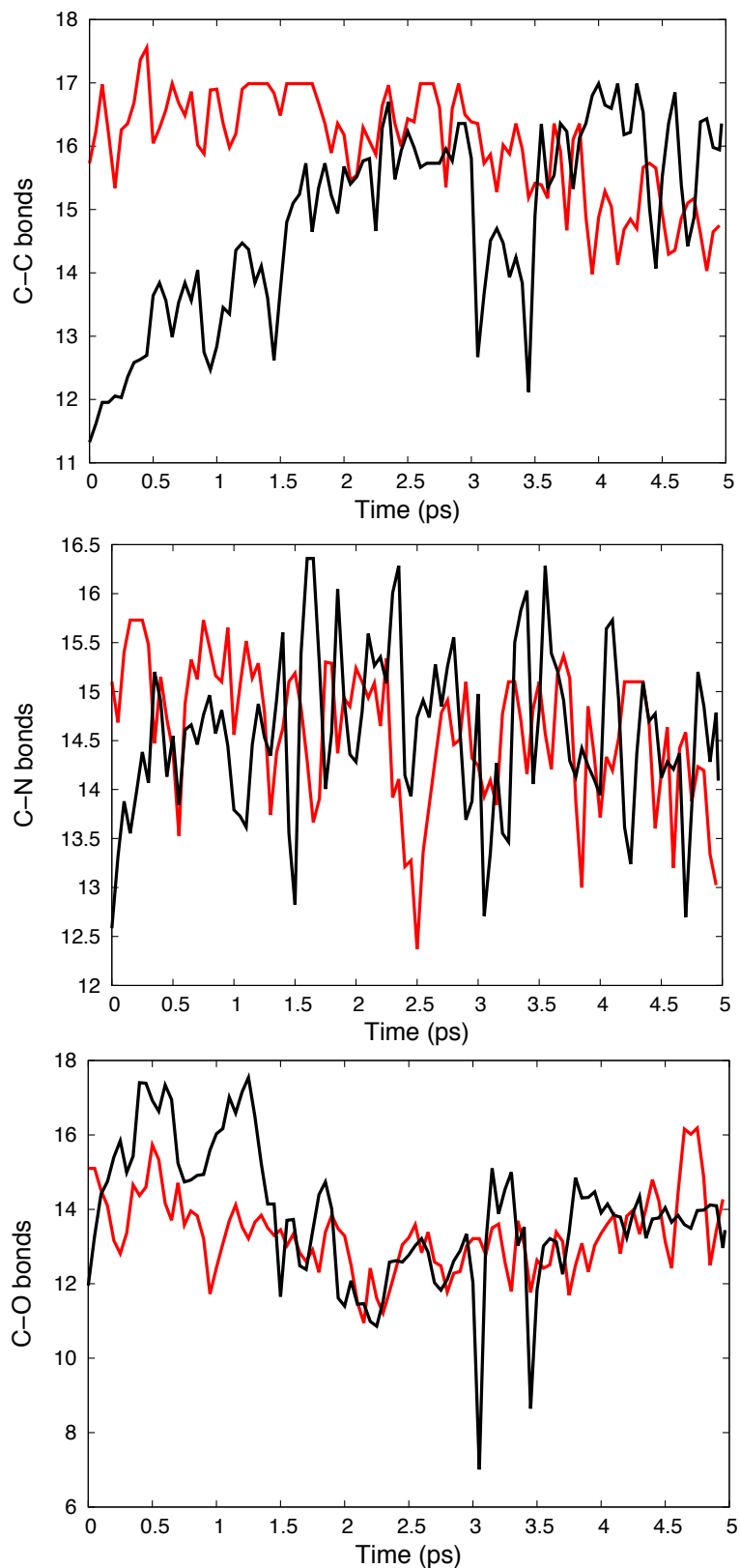


Fig. S2 Time histories for the number of C–C, C–N, and C–O bonds over the first 5 ps from both DFT (black lines) and one of our forced matched DFTB simulations (red lines), averaged over the first 5 ps of simulation time. Numbers of bonds were averaged over non-overlapping 50 fs windows.