

20. ENERGY CALCULATIONS AND MOLECULAR DYNAMICS

Table 20.1.3.3. Root-mean-square fluctuations of polypeptide backbone and ψ dihedral angles ($^{\circ}$) for the different molecules using different time-averaging periods

The bottom row shows the averages over all four protein molecules.

Molecule	400–800 ps φ/ψ	400–1200 ps φ/ψ	400–2000 ps φ/ψ
1	18.4/22.9	19.5/23.7	31.0/33.6
2	17.2/17.0	18.6/18.7	23.6/26.8
3	18.5/20.4	25.6/26.3	35.3/37.5
4	19.7/18.8	19.4/20.3	21.6/28.8
All	26.1/26.2	28.0/28.6	35.2/38.1

only certain dihedral-angle flips induce large movements that determine the RMSD value.

20.1.3.7. Water diffusion

In Fig. 20.1.3.9, the number of water oxygen atoms with a given atomic root-mean-square position fluctuation (RMSF) are plotted. The time evolution and the shapes of these curves are similar to those obtained for bulk water, a Gaussian distribution with the maximum at larger RMSF values and larger standard deviations when using longer averaging times. Using a diffusion constant of bulk SPC water at 300 K of $3.9 \times 10^{-3} \text{ nm}^2 \text{ ps}^{-1}$ (Smith & van Gunsteren, 1995), the root-mean-square position fluctuation for an average water molecule would be 1.25 nm for a 400 ps period, 1.77 nm for a 800 ps period, and 2.5 nm for a 1600 ps period. Comparison of these values with the distributions in Fig. 20.1.3.9 indicates that the motion of most of the crystal water molecules is restricted by the crystalline environment. A number of water molecules adopt well defined positions for periods of the order of some 100 ps. Several water molecules were also observed to visit the same location repeatedly. This indicates that, although not remaining fixed in space, they stay close to a crystallographically determined site which they revisit periodically, alternating with other water molecules.

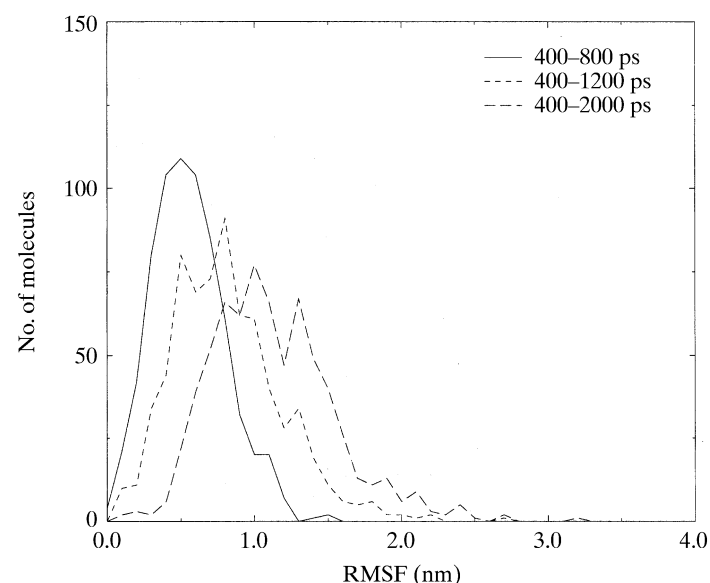


Fig. 20.1.3.9. The number of water molecules with a given root-mean-square oxygen-position fluctuation (RMSF) in nm are shown for different averaging periods: 400–800 ps (solid line), 400–1200 ps (short-dashed line), 400–2000 ps (long-dashed line).

Table 20.1.3.4. Number of protein-backbone dihedral-angle transitions per 100 ps for the different molecules using different time periods

Dihedral angles with threefold and sixfold potential-energy wells are distinguished. The bottom rows show the averages over all protein molecules.

(a) 120° transitions

Molecule	400–800 ps	400–1200 ps	400–2000 ps
1	46.5	45.4	47.7
2	40.5	41.5	47.3
3	50.5	57.1	51.3
4	44.8	46.4	46.4
All	45.6	47.6	48.2

(b) 60° transitions

Molecule	400–800 ps	400–1200 ps	400–2000 ps
1	245.5	246.6	289.3
2	271.5	272.1	261.3
3	381.5	381.0	348.3
4	356.8	325.4	325.4
All	313.8	306.3	306.1

(c) All transitions

Molecule	400–800 ps	400–1200 ps	400–2000 ps
1	292.0	292.0	336.9
2	312.0	313.7	308.6
3	432.0	438.1	399.6
4	401.5	371.8	371.8
All	359.4	353.9	354.2

20.1.4. Conclusions

In the present molecular-dynamics simulation, fast convergence in energy, within about 100 ps, was observed. Other properties, such as dihedral-angle fluctuations and backbone atom-position fluctuations, converged on an intermediate timescale of hundreds of picoseconds. Root-mean-square deviations of the simulated protein molecules from the starting X-ray structure required of the order of 1 ns to reach a plateau. Longer simulations would be necessary to obtain convergence for all molecular properties. The convergence of quickly relaxing properties of the system, such as the energies, is not a reliable indicator of the degree of global convergence in such a molecular-dynamics simulation.

The GROMOS96 force field used in this simulation largely reproduces the secondary structure and the relative internal mobility of ubiquitin. The simulation does, however, overestimate the magnitude of the fluctuations in the most mobile regions of the protein. The different protein molecules were observed to translate and rotate relative to one another during this simulation. This indicates that the force field would not be able to reproduce the experimental melting temperature of this crystal under the conditions simulated.

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