Supplementary Figure S1. $3_{10}$-helix content in the resultant ensembles with (A) $\tau = 2$ ps and (B) $\tau = 0.2$ ps. The conditions $t_{\text{att}} = 0.001, 0.005, 0.01, 0.1, \text{ and } 1$ ps are shown in red, green, blue, cyan, and purple, respectively. The error bars indicate the standard errors among 20 sub-trajectories generated by uniformly dividing the last 9 ns of the original trajectory.
Supplementary Figure S2. Radius of gyration ($R_g$) in the resultant ensembles with (A) $\tau = 2$ ps and (B) $\tau = 0.2$ ps. The conditions $t_{\text{att}} = 0.001$, 0.005, 0.01, 0.1, and 1 ps are shown in red, green, blue, cyan, and purple, respectively. The error bars indicate the standard errors among 20 sub-trajectories generated by uniformly dividing the last 9 ns of the original trajectory.
Supplementary Figure S3. Cumulative average of radius of gyration ($R_g$) calculated with (A, C, E, G, I) $\tau = 2$ ps and (B, D, F, H, J) $\tau = 0.2$ ps for each $N_{\text{rep}}$ condition: (A, B) $N_{\text{rep}} = 8$, (C, D) $N_{\text{rep}} = 12$, (E, F) $N_{\text{rep}} = 16$, (G, H) $N_{\text{rep}} = 20$, and (I, J) $N_{\text{rep}} = 32$. The conditions $t_{\text{att}} = 0.001, 0.005, 0.01, 0.1,$ and $1$ ps are shown in red, green, blue, cyan, and purple, respectively.
Supplementary Figure S4. Potential energy distributions in ensembles at 300 K with (A, C) $\tau = 2$ ps and (D, E, F) $\tau = 0.2$ ps for each $N_{\text{rep}}$ condition: (A, D) $N_{\text{rep}} = 12$, (B, E) $N_{\text{rep}} = 16$, and (C, F) $N_{\text{rep}} = 20$. See also Figure 3.
Supplementary Figure S5. Temperature distributions in ensembles at 300 K with (A, C) $\tau = 2$ ps and (D, E, F) $\tau = 0.2$ ps for each $N_{\text{rep}}$ condition: (A, D) $N_{\text{rep}} = 12$, (B, E) $N_{\text{rep}} = 16$, and (C, F) $N_{\text{rep}} = 20$. See also Figure 4.
**Supplementary Figure S6.** Ratios of the ensemble average of temperatures to the target temperature at $k$-th thermostat $\langle T \rangle_k / T_k$ with (A, B) $\tau = 2$ ps and (C, D) $\tau = 0.2$ ps for each $N_{\text{rep}}$ condition: (A, C) $N_{\text{rep}} = 12$, and (B, D) $N_{\text{rep}} = 20$. The results with for $t_{\text{att}} = 0.001$, 0.005, and 0.1 ps are shown in red, green and cyan, and those with $t_{\text{att}} = 0.01$ and 1 ps are not shown for clarity. See also Figure 5.