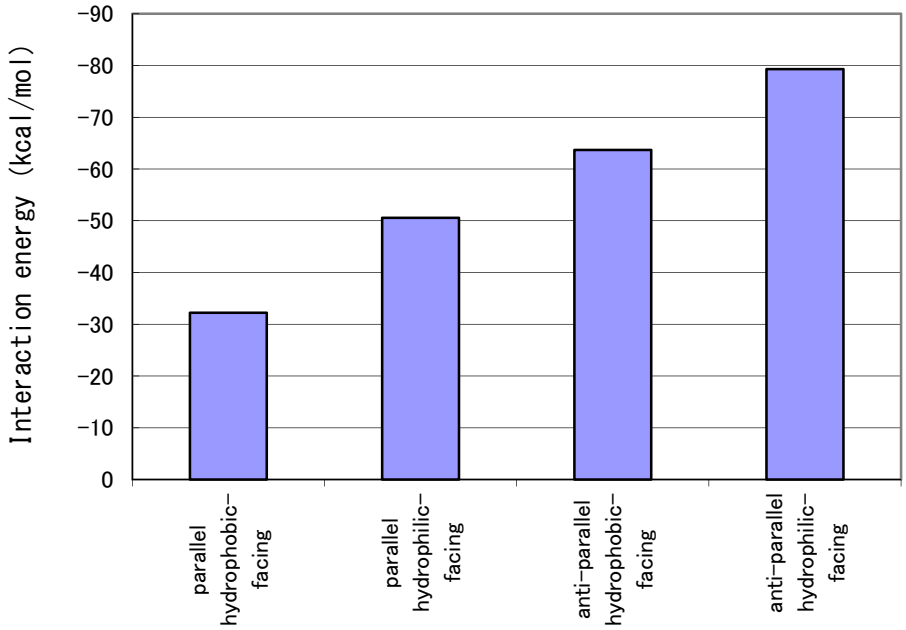
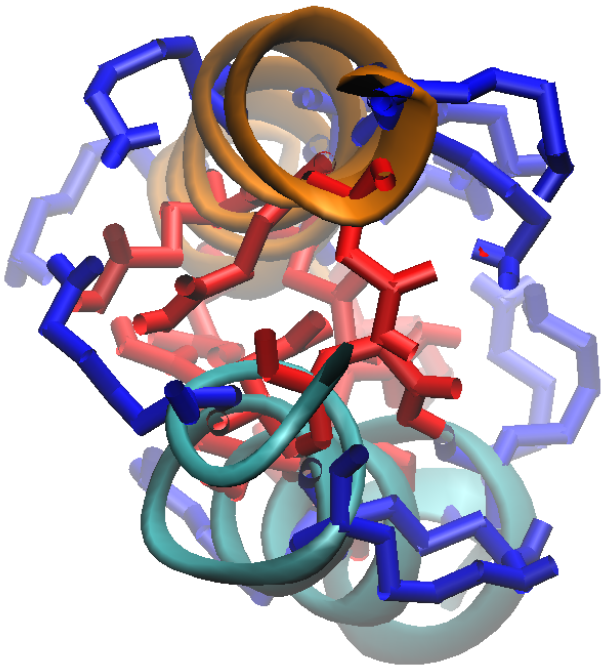
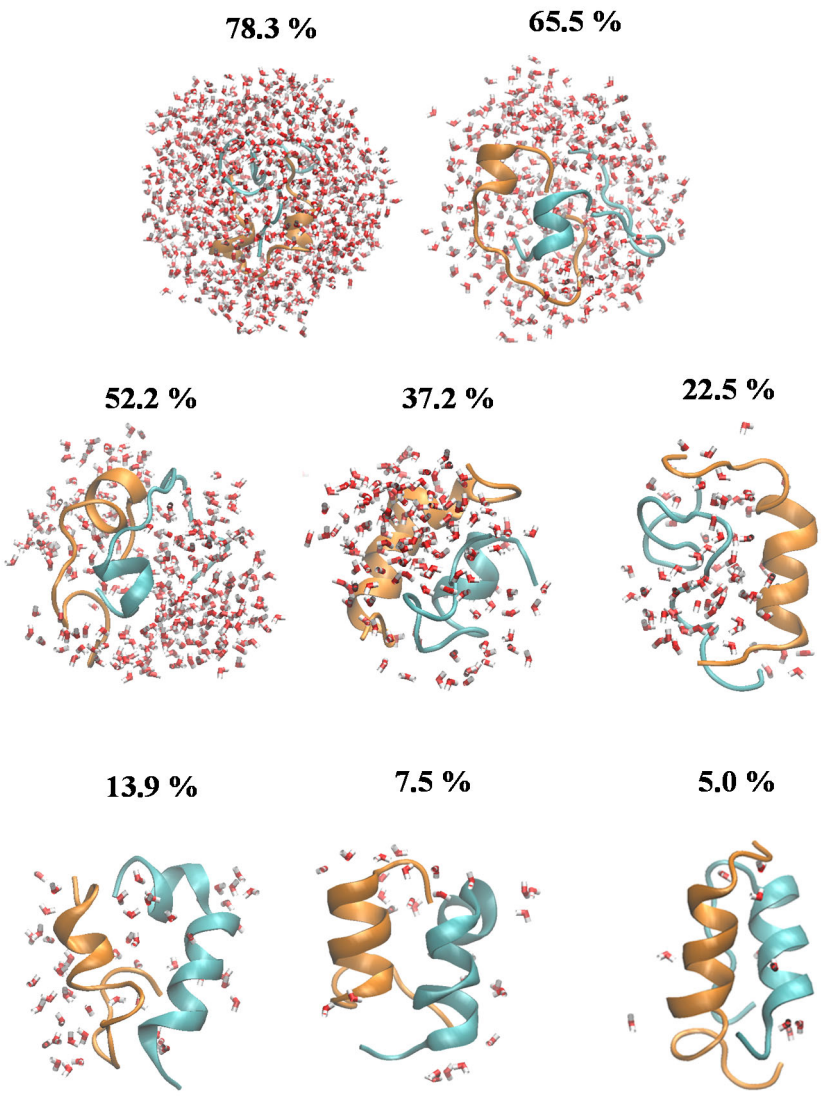


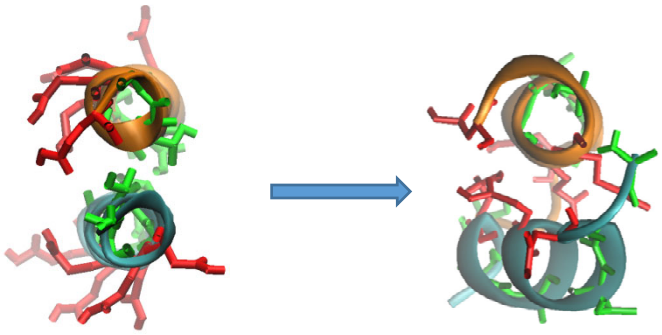
Supplementary Figure S1. The interaction energies ΔE between two α-helical chains in the four different arrangements. ΔE was obtained by subtracting the energies of the infinitely-separated two chains from that of their complexed state.



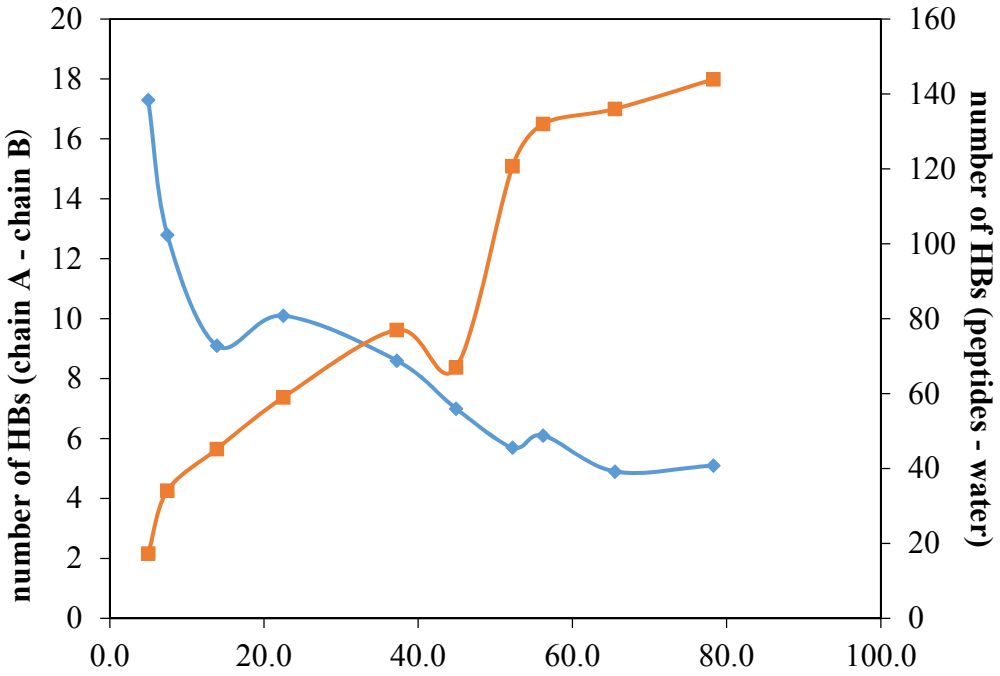
Supplementary Figure S2. The structure of the anti-parallel hydrophilic model at the water content of 22.5 wt%. For clarity, water molecules are deleted. The red and blue stick models represent the side chains of acidic (Asp and Glu) and basic (Lys) amino acid residues, respectively.



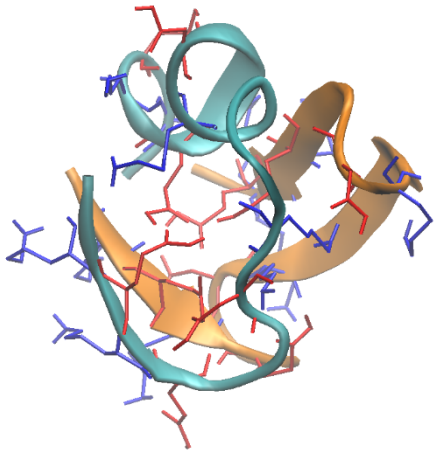
Supplementary Figure S3. Representative structure of the anti-parallel hydrophobic-facing model in each water content level (in wt%).



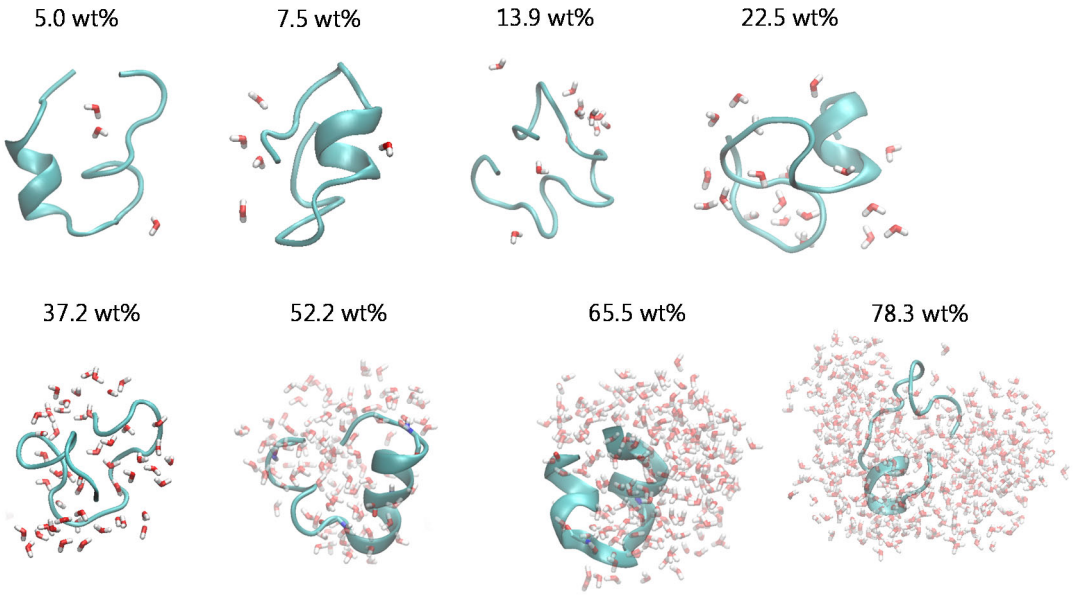
Supplementary Figure S4. Structural transition from hydrophobic-facing to hydrophilic-facing, as was observed during the REMD simulation for the antiparallel hydrophobic-facing model. Left: The initial structure where the side chains of the neutral residues (Ala and Thr) are oriented to the interface between the two helices. Right: The post-REMD structure where each helix was rotated about each helical axis to form the hydrophilic-facing structure.



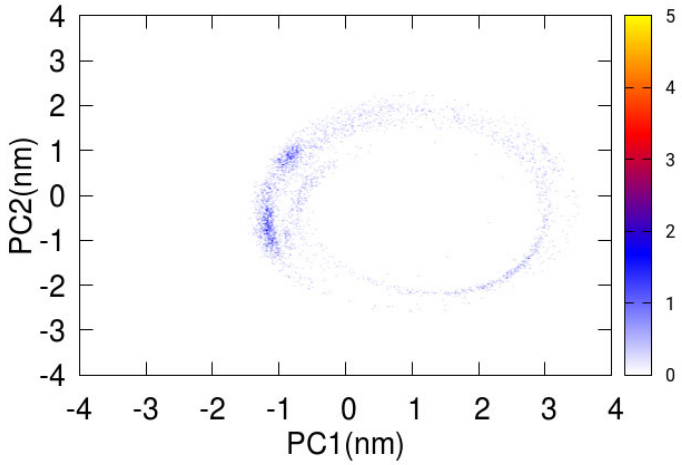
Supplementary Figure S5. Water content dependence of the number of interchain hydrogen bonds (blue) and that of the peptide-water hydrogen bonds (red) in the anti-parallel hydrophobic-facing model.



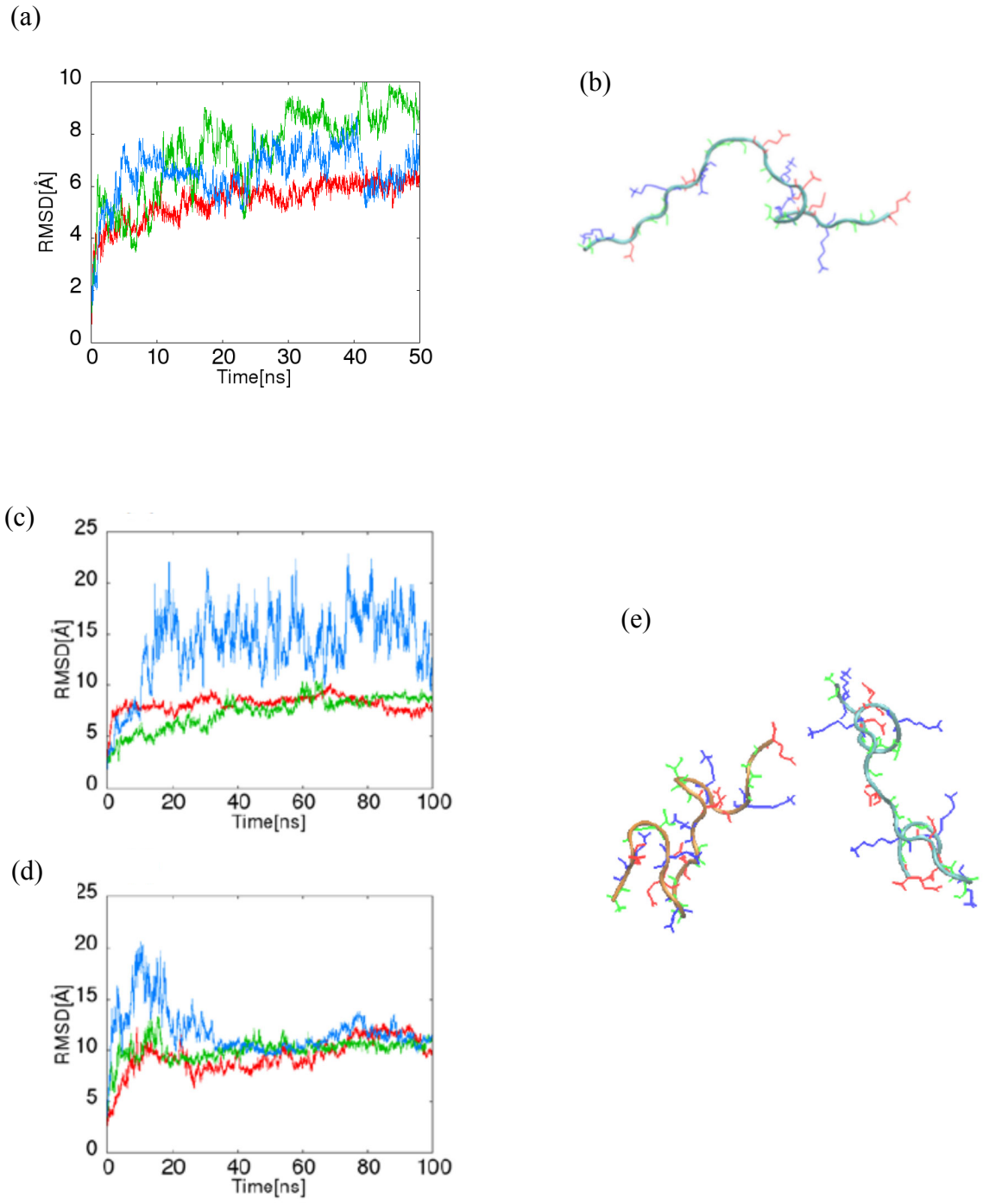
Supplementary Figure S6. A representative structure of the anti-parallel model of the scrambled peptide in a water content of 5 wt%. The two chains aggregated where the rate of random coil was more than 40%.



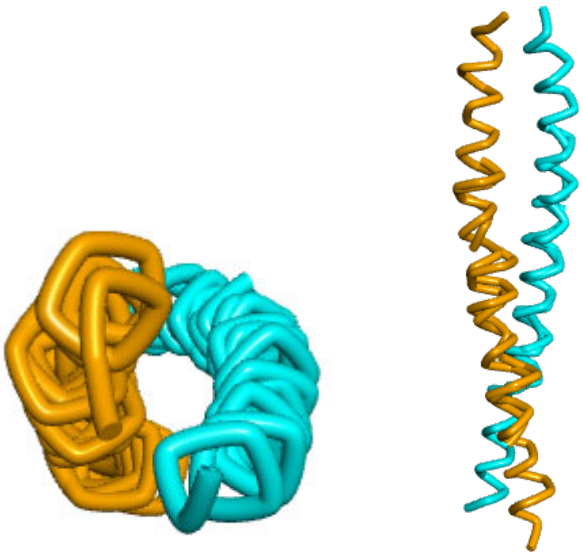
Supplementary Figure S7. Representative structures of the single chain of PvLEA-22 at different water content levels.



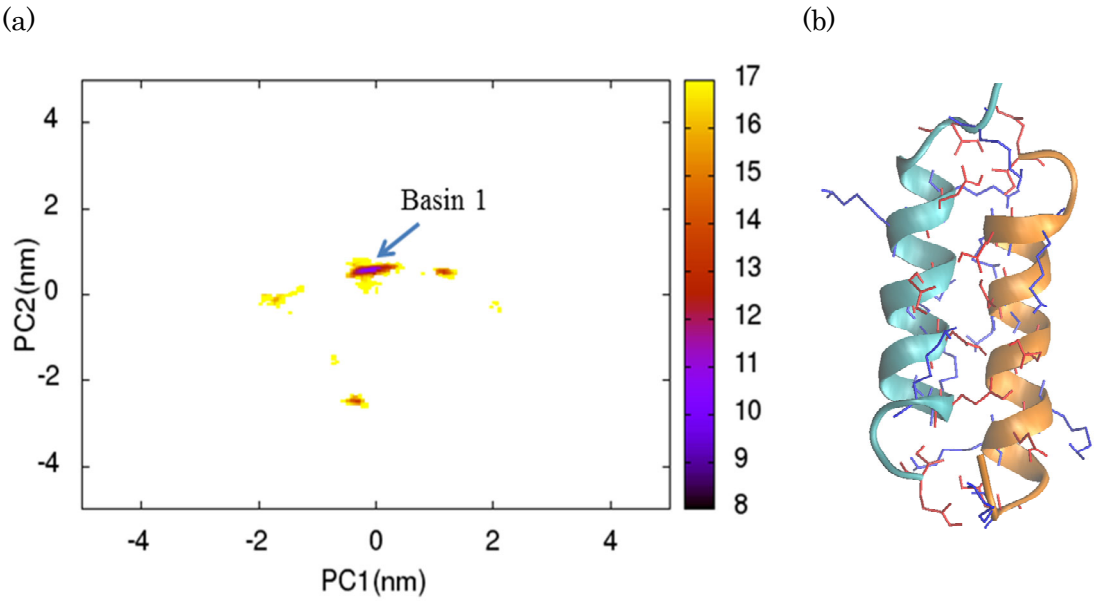
Supplementary Figure S8. Free energy maps obtained from the PCA analysis for the single-chain model of PvLEA-22 at the water content level of 5 wt%. The energy values (kJ/mol) are indicated by the color code.



Supplementary Figure S9. RMSD results of three conventional MD simulation runs (red, green and blue) for (a) single chain, (c) anti-parallel hydrophobic-facing, and (d) anti-parallel hydrophilic-facing models. (b) and (e) are representative snapshots at 50 ns for (a) and (c), respectively.



Supplementary Figure S10. Coiled coil structure of PvLEA-22. Two sets of the double-bundled α-helical strands that are shown in Figure 6 were jointed with partial overlap along the helix axis. Left: top view along the helix axis, right: side view.



Supplementary Figure S11. Analysis of the REMD trajectory at 400 K for the PvLEA-22 solution with a water content of 7.5 wt%. (a) Result of the PCA analysis. (b) A representative snapshot at Basin 1.