**Supplementary Information**

# Linear and Ring Polypeptides Complexed with Oppositely Charged Surfactants. The Cohesion of the Complexes as Revealed in Atomistic Simulations

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Our analysis of polypeptide conformations at different temperatures revealed distinct changes in their structures. The R18 chain exhibited a transition from a compact stacking of loops to a dumbbell-shaped conformation and finally to a coiled structure as temperature increased. Similarly, the R36 chain transformed from a ring-like conformation to a worm-like shape and eventually to an extended coil. The R72 chain maintained a slightly deformed ring structure with a large central cavity throughout the temperature range. For the K18 chain, we observed a stacking of loops within a space bounded by a triangular base, with the chain predominantly wandering along one direction. As the temperature increased, the conformation displayed coiled and spiral parts, resulting in an angle or half of a rectangular frame. The K36 chain resembled a flattened dumbbell shape, with elongated halves connected by a short bridge. While the overall conformation of the K36 chain showed little variation with temperature, there was an increased localization of atoms at the chain ends. The K72 chain exhibited small loops aligned along helical or sinusoidal trajectories, forming elongated coils with adjacent coils oriented at an angle. The chain adopted a zigzag arrangement, with links lying in different planes.

These findings highlight the temperature-dependent conformational changes in polypeptide chains. The observed transformations provide valuable insights into the structural properties of these chains and their responses to temperature variations. Such knowledge can be beneficial for the design and development of biomaterials with controlled conformational characteristics. Further studies in this field could contribute to the advancement of bioactive materials with tailored properties for various applications, including tissue engineering and drug delivery.

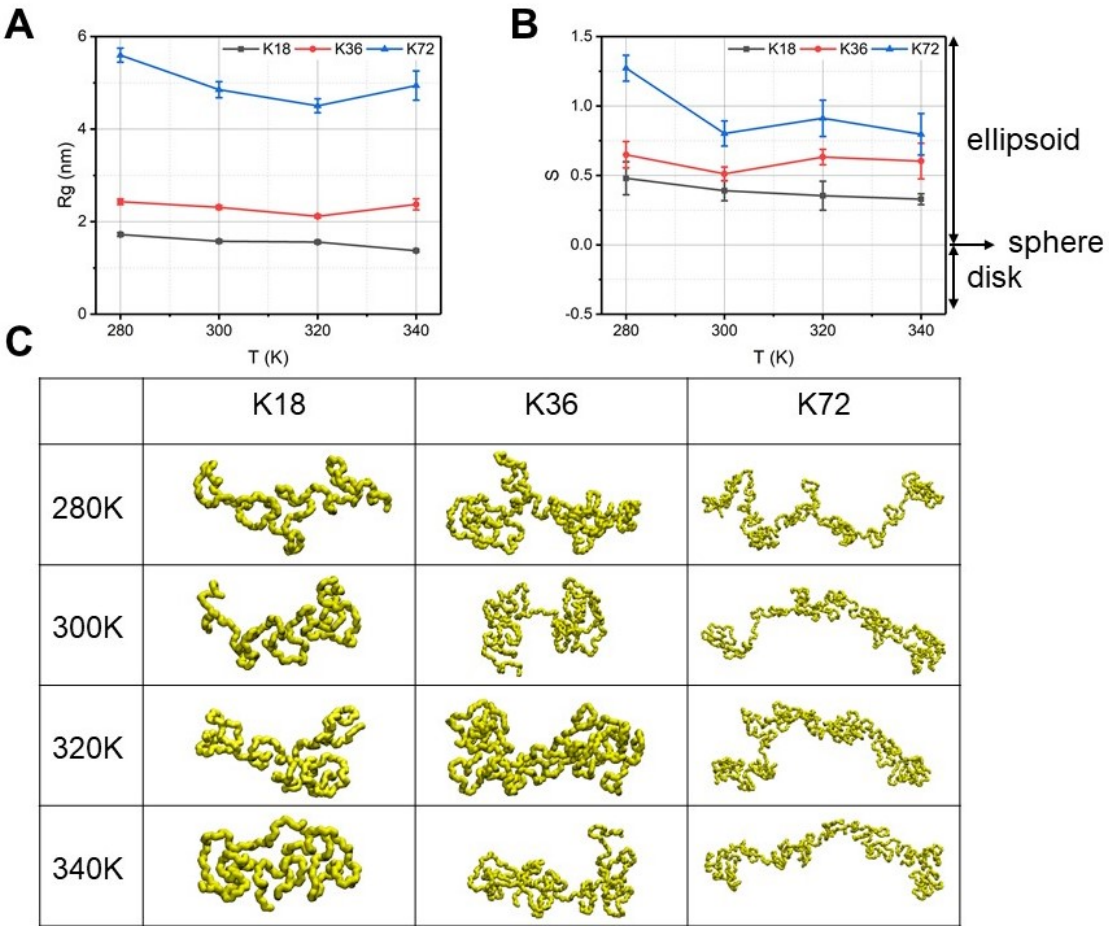


**Figure S1**. Plot of the dependence of the radius of gyration of a linear polypeptide chain (A) and elongation (B) on the temperature. Snapshots of the main chain of equilibrated linear chains (C).

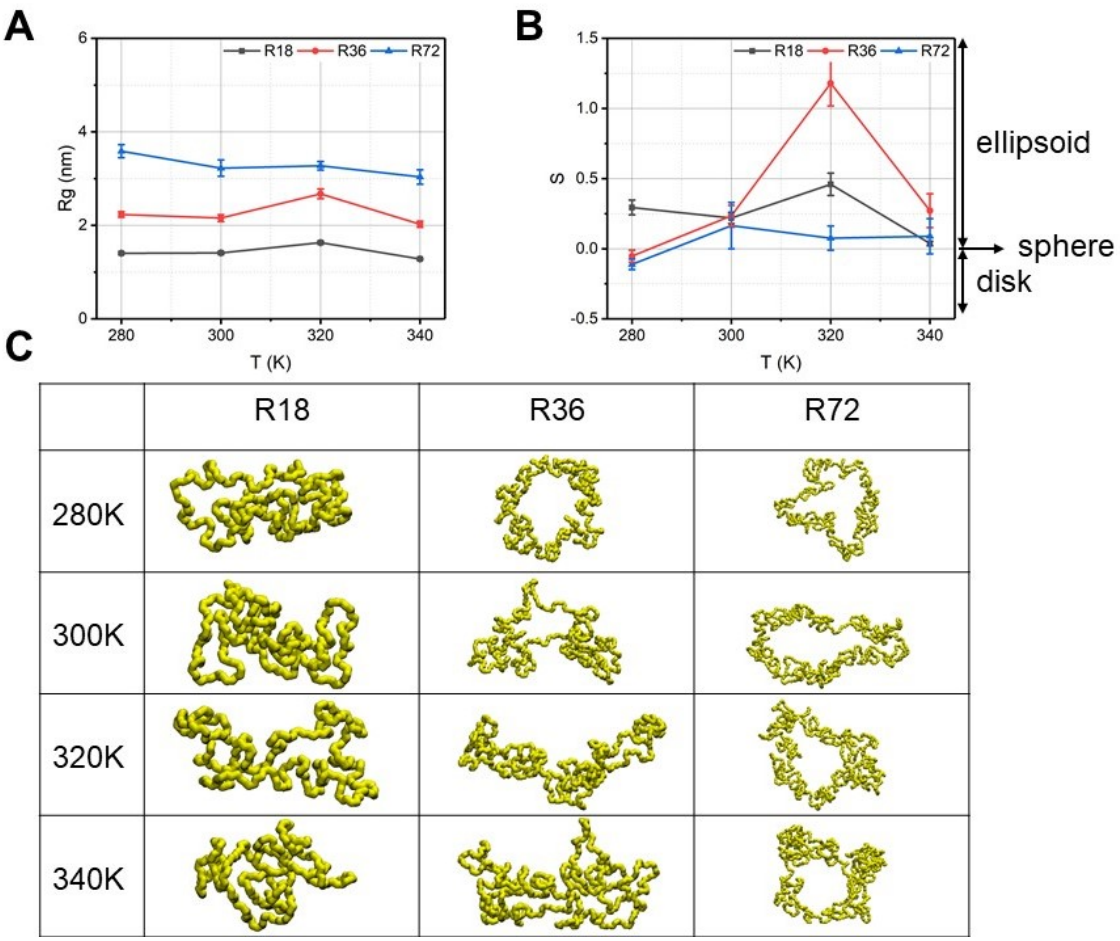


**Figure S2**. Plot of the dependence of the radius of gyration of the ring polypeptide chain (A) and elongation (B) on the temperature. Snapshots of the main chain of equilibrated ring chains (C).

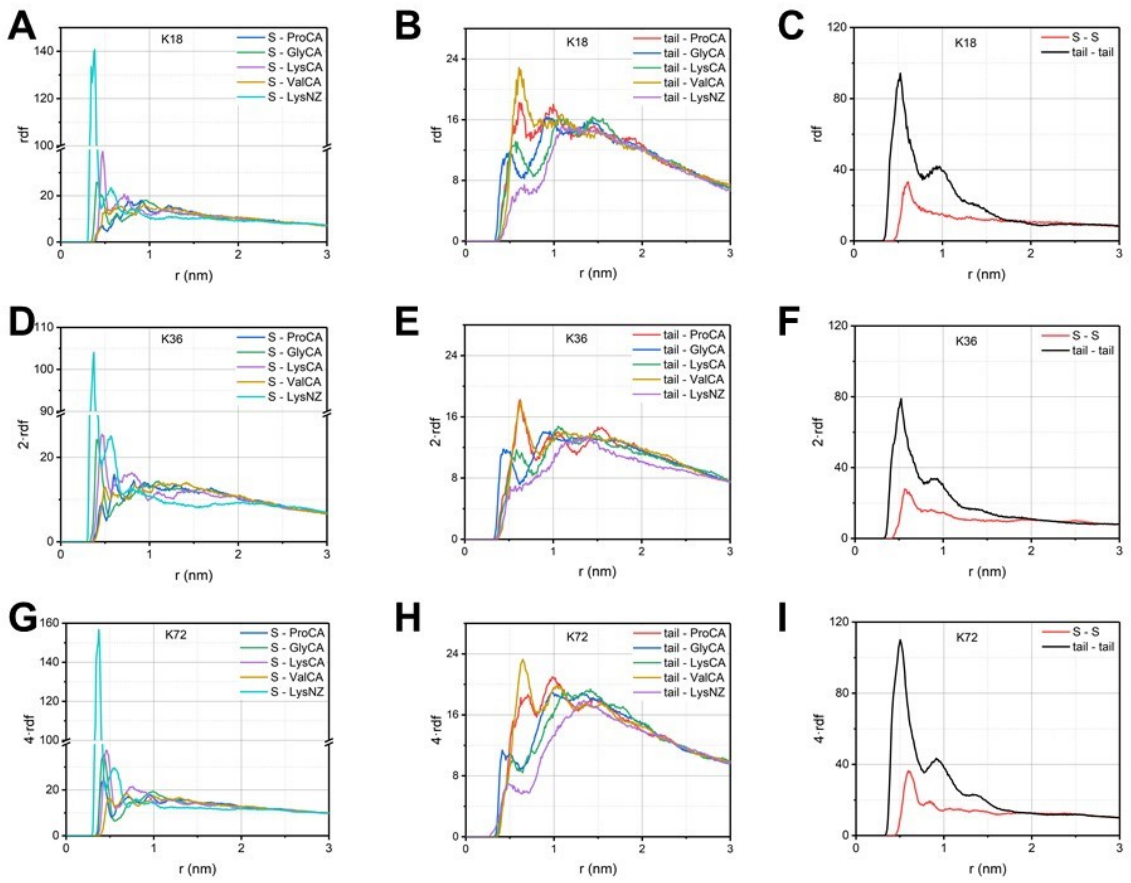


**Figure S3**. Radial distribution functions of individual atoms of amino acid residues of linear polypeptide chains relative to the sulfur atom of the surfactant head (A,D,G), the middle carbon atom of the surfactant tail (B,E,H), and the sulfur atoms of the head/tail carbon atom relative to the same atoms of other molecules SDBS (C,F,I) for three polypeptide lengths: (A,B,C) K18, (D,E,F) K36, (G,H,I) K72.

**Figure S4**. Radial distribution functions of individual atoms of amino acid residues of linear polypeptide chains relative to the sulfur atom of the surfactant head (A,D,G), the middle carbon atom of the surfactant tail (B,E,H), and the sulfur atoms of the head/tail carbon atom relative to the same atoms of other molecules SDBS (C,F,I) for three polypeptide lengths: (A,B,C) R18, (D,E,F) R36, (G,H,I) R72.

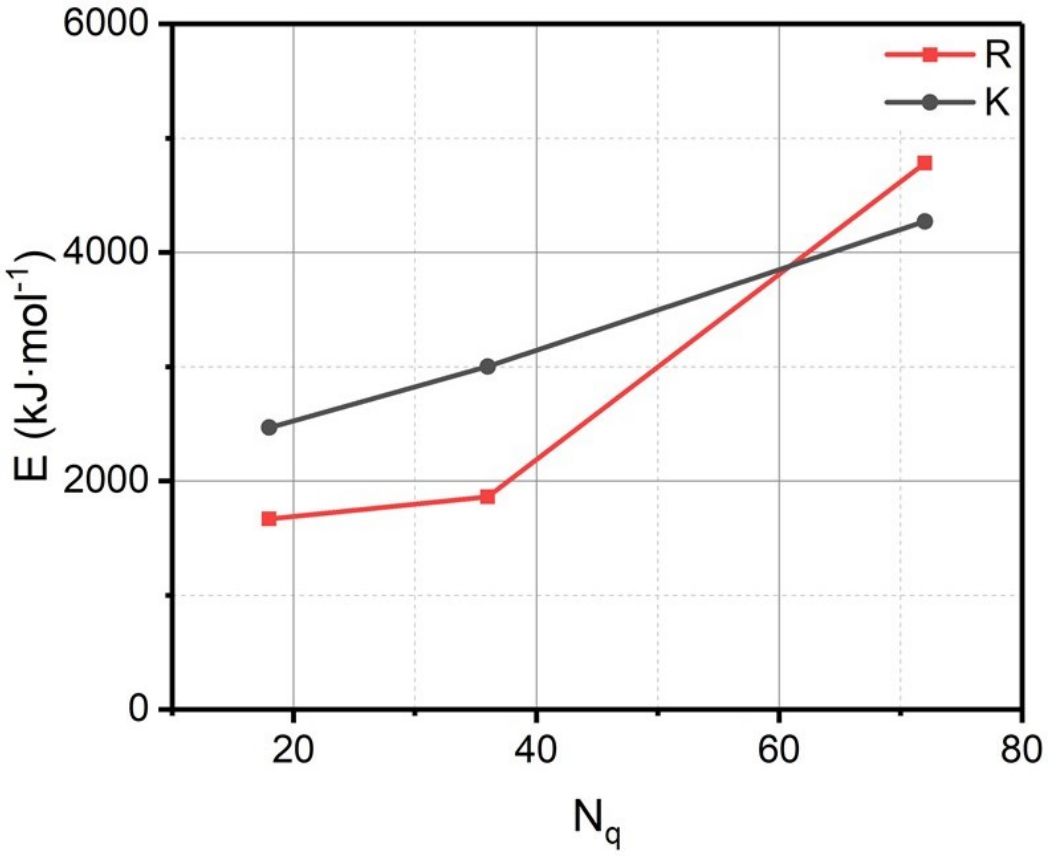


**Figure S5**. Plot of the dependence of the specific energy required to stretch the polypeptide chain from the complex by 10 nm on the length of the chain (in units of charged blocks).