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Protein-based Polymers: Mechanistic Foundations for Design and Processing

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Thesis: Minimization of apolar-polar repulsive free energy of hydration, ΔG_{ap} , dominates tertiary and quaternary structure formation of amphiphilic polymers in water and even modulates secondary structure. Consequently, ΔG_{ap} represents the dominant force underlying processing of amphiphilic polymers in water. Hydration of polar and hydrophobic groups is exothermic.¹ The result is competition for hydration that causes hydrophobic and polar (e.g., charged) groups to reach out for water unperturbed by the other, i.e., to be repulsed. In bioproduction (protein biosynthesis), nucleic acid sequence determines protein sequence, irrespective of different high free energies due to ΔG_{ap} between side chains of the protein product. The free energies can then be modulated to achieve folding, assembly, and function that dictate processing. ΔG_{ap} is the modulatable component that gives rise to changes in Gibbs free energy for hydrophobic association, ΔG_{HA} , which may be evaluated from the heat and temperature of an inverse temperature transition for hydrophobic association. During function as well as processing, energy of deformation is stored by the damping of internal chain dynamics within individual chains. These mechanistic foundations, derived through design and characterization of elastic-contractile model proteins, can be argued to provide function in protein-based motors of biology,² for example, Complex III/Rieske Iron Protein,³ mitochondrial ATP synthase,⁴ the myosin II motor of muscle contraction,³ and kinesin of microtubular transport.⁵

With the above mechanistic development comes design and processing of elastic protein-based polymers for improved materials. Formation of nanoporous membranes and bilayer membranes can result with proper processing of anionic or of cationic polymers of regular specified sequences. Also, the repulsive ΔG_{ap} values in the cationic polymers and the anionic polymers become lowered on ion-pairing between polymers. This provides the driving force for alignment preceding chemical cross-linking of suitably extruded fibers. The result can be improved elastic moduli, break stresses and break strains.