John White Symposium



Contribution ID: 7 Type: not specified

Protein interactions and deuteration effects in the bulk and at liquid surfaces: the legacy of John White from FIGARO

Thursday, 18 July 2024 18:10 (20 minutes)

It has been known for almost one hundred years that a lower surface tension can be achieved at the air—water interface by spreading protein from a concentrated solution than by adsorption from an equivalent total bulk concentration. Nevertheless, the factors that control this nonequilibrium process have not been fully understood. In the present work, we apply ellipsometry, neutron reflectometry, X-ray reflectometry, and Brewster angle microscopy to elaborate the surface loading of human serum albumin in terms of both the macroscopic film morphology and the spreading dynamics. We show that the dominant contribution to the surface loading mechanism is the Marangoni spreading of protein from the bulk of the droplets rather than the direct transfer of their surface films. The films can be spread on a dilute subphase if the concentration of the spreading solution is sufficient; if not, dissolution of the protein occurs, and only a textured adsorbed layer slowly forms. The morphology of the spread protein films comprises an extended network with regions of less textured material or gaps. Further, mechanical cycling of the surface area of the spread films anneals the network into a membrane that approach constant compressibility and has increased durability. Our work provides a new perspective on an old problem in colloid and interface science. The scope for optimization of the surface loading mechanism in a range of systems leading to its exploitation in deposition-based technologies in the future is discussed.

Nanoparticle-protein complexes comprising silica or cadmium sulfide nanoparticles with human proteins have been detected with high sensitivity at the air-water interface using X-ray and neutron reflectivity measurements and ellipsometry. For the interaction between β -casein and 8 nm silica particles the sensitivity of the reflectivity signal at the air-water interface has been shown by X-ray reflectometry to be pico-molar. This high sensitivity results from the nanoparticle-protein interaction which generates surface active complexes. The interaction between human serum albumin and cadmium sulfide nanoparticles has been detected using neutron reflectometry and tracked kinetically by ellipsometry. Here we have shown that lateral domains or aggregates in the surface monolayer change in size or dissipate over the course of several hours. The three methods of monitoring the protein-nanoparticle interaction during film evolution are discussed.

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Session Classification: Reflectometry