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Formation of Ordered Arrays of Proteins on Surfaces

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Roth, C. M., B. L. Neal and A. M. Lenhoff, "Van der Waals Interactions Involving Proteins", *Biophys. J.*, **70**, 977-987 (1996).

Abstract: Van der Waals (dispersion) forces contribute to interactions of proteins with other molecules or with surfaces, but because of the structural complexity of protein molecules, the magnitude of these effects is usually estimated based on idealized models of the molecular geometry, e.g., spheres or spheroids. The calculations reported here seek to account for both the geometric irregularity of protein molecules and the material properties of the interacting media. While the latter are found to fall in the generally accepted range, the molecular shape is shown to cause the magnitudes of the interactions to differ significantly from those calculated using idealized models, with important consequences. First, the roughness of the molecular surface leads to much lower average interaction energies for both protein - protein and protein - surface cases relative to calculations in which the protein molecule is approximated as a sphere. These results indicate that a form of steric stabilization may be an important effect in protein solutions. Underlying this behavior is appreciable orientational dependence, one reflection of which is that molecules of complementary shape are found to exhibit very strong attractive dispersion interactions. Although this has been widely discussed previously in the context of molecular recognition processes, the broader implications of these phenomena may also be important at larger molecular separations, e.g., in the dynamics of aggregation, precipitation and crystal growth.

Palkar, S. A., and A. M. Lenhoff, "Diffusion and Adsorption of Proteins in a Model Pore in the Surface Forces Apparatus", *Colloids Surf. A*, **110**, 119-127 (1996).

Abstract: When the energetic interactions between proteins and pore surfaces are favorable, both hindered diffusion and adsorption govern the overall transport rates in fine pores. To study these effects, we have used the narrow gap between mica surfaces in a surface forces apparatus (SFA) as a model slit-like pore that has a well-characterized geometry and surface chemistry. The optical technique of multiple beam interferometry used in the SFA for measuring surface separations also provides a means for determining protein concentrations inside the pore. The results obtained show the effect of ionic strength on the net transport rates for lysozyme diffusing in a mica-bounded pore, a consequence of the influence of electrolyte screening on the extent of adsorption. It is also seen how adsorption on apposing surfaces leads to pore blockage once the pore size decreases to 2-3 times the molecular diameter. Although quantitative determination of hindrance effects is potentially also possible using the method, an improved method for acquisition of the interferometry data must first be implemented.

Johnson, C. A., and A. M. Lenhoff, "Adsorption of Charged Latex Particles on Mica Studied by Atomic Force Microscopy", *J. Coll. Interf. Sci.*, **179**, 587-599 (1996).

Abstract: Atomic force microscopy (AFM) is used to study the localized adsorption of charged latex particles adsorbed to a mica substrate with varying adsorption times and solution ionic strengths. Images were obtained of these colloidal particles by employing tapping mode AFM with silane-treated probes to reduce the intrusive effect of the imaging process on physisorbed particles. The initial kinetics of the adsorption process are found to be diffusion-limited and the long-time asymptotic kinetics are found to resemble those of a random sequential adsorption (RSA) process. Double-layer screening of interparticle electrostatic repulsions is shown to have a controlling effect on the surface coverage at saturation and, to a lesser degree, on the diffusion-limited rate of adsorption. *In situ* images of adsorbed layers are compared with those of dried samples to reveal the morphological effects of film evaporation on the microstructure of adsorbed particles. Results show that particles in adsorbed layers of high surface coverage can rearrange into two-dimensional clusters during film evaporation -- a phenomenon attributed to capillary forces between particles in near-contact. Ordering of particles at the solid-liquid interface is analyzed statistically by producing two-dimensional radial distribution functions. A high degree of short-range order is found among adsorbed particles when the surface coverage approaches saturation, and a significant reduction in the range of ordering due to double-layer screening of interparticle repulsions is also reflected in the radial distributions.