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Electrostatics of Patchy Surfaces

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Abstract

In the study of colloidal, biological and electrochemical systems, it is customary to treat surfaces, macromolecules and electrodes as homogeneously charged. This simplified approach is proven successful in most cases, but fails to describe a wide range of heterogeneously charged surfaces commonly used in experiments. For example, recent experiments have revealed a long-range attraction between overall neutral surfaces, locally charged in a mosaic-like structure of positively and negatively charged domains ("patches"). Here we review experimental and theoretical studies addressing the stability of heterogeneously charged surfaces, their ionic strength in solution, and the interaction between two such surfaces. We focus on electrostatics, and highlight the important new physical parameters appearing in the heterogeneous case, such as the largest patch size and inter-surface charge correlations.

Keywords: Heterogeneously charged surfaces, ionic solutions, surface forces, hydrophobic surfaces, Poisson-Boltzmann theory

Contents

1. Introduction

Electrostatic interactions are paramount in the study of numerous colloidal, biological, and electrochemical systems. In aqueous media, some of the surface charges of macromolecules may dissociate whereas ions from the solution can bind to the macromolecules [1, 2]. Both processes result in a net surface charge leading to a long-range Coulombic interaction, mediated by ions in the solution. The interplay between electrostatics and the ion entropy of mixing is described by the Poisson-Boltzmann (PB) theory. Combining the universal van der Waals (vdW) interaction with PB theory yields the well-known Derjaguin-Landau-Verwey-Overbeek (DLVO) theory [3, 4].

Traditionally, DLVO theory is used to study the interaction between homogeneously charged surfaces [5, 6]. Homogeneity is an idealization, as charges are distributed discretely on the molecular level, and surfaces can also be heterogeneously charged over mesoscopic length scales (nanometers to micrometers), either spontaneously or by design [1, 5]. The latter has stimulated many experimental and theoretical works in the past few decades, addressing surface-charge heterogeneity on microscopic and mesoscopic levels, under a wide range of physical conditions. Different aspects of inhomogeneity have been investigated, including the stability of surface-charge heterogeneities, counterion distribution at the surface proximity, and interactions between two such surfaces across an ionic solution.

The study of heterogeneously charged ("patchy") surfaces has gained a growing interest during the last decade due to novel experiments, which measured a long-range attraction between hydrophobic surfaces across an aqueous solution [7–12]. Although these neutral surfaces were initially homogeneous during preparation, it was observed that they transform into mosaic-like structures of positively and negatively charged patches. These patchy surfaces remain stable during experimental times, and it has been established [7–9, 11, 12] that the

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