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Random sequential adsorption of polydisperse spherical particles: An integral-equation theory

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Abstract

Our previously developed integral-equation theories were applied to incorporate the effect of polydispersity in the study of the random sequential addition of spherical particles. By using the simplest uniform size distribution, we found that results from theories were in consistence with the Monte Carlo simulation results. Some deviations were seen, which resulted from the exclusion effects of polydisperse particles. It was found in the simulations that with increasing densities, small particles adsorbed preferentially and the size distribution skewed towards the smaller particles. Therefore, to accurately predict the correct radial distribution functions, the more appropriate size distributions are needed. For all size ranges, which were 0.40*d*–1.60*d*, 0.75*d*–1.25*d*, and 0.90*d*–1.10*d*, the radial distribution functions from theory at number densities of 0.2, 0.4 and 0.65 were in good agreements with those from the simulations.

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1. Introduction

For the adsorption of large molecules such as proteins [1] and colloids [2] on solid surfaces, the interactions between a molecule and the surface are strong. Multiple bonds may be formed very quickly, so that desorption and surface diffusion become negligible and adsorption may be reasonably considered an irreversible process. This is often modeled by the random sequential adsorption (RSA). In this model, rigid molecules are sequentially added onto a surface at random, with the only condition that they do not overlap with one another. Once successfully inserted, a molecule is frozen in position and can neither move on the surface nor be removed. RSA gives rise to the configurations which are different from their equilibrium counterparts. The kinetics of RSA has been studied extensively and the model has also been modified to add more realistic features. Instead of random arrivals, specific transport mechanisms for the arrival of particles from the bulk to

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the vicinity of the surface has been considered, including ballistic deposition [3] where an external field such as gravity plays a role, and diffusional deposition, where Brownian motion [4] and hydrodynamic interactions [5] are incorporated. Studies which consider interactions between adsorbates [6] and of adsorbates with free particles near to (but not adsorbed onto) the surface [6], actually revert the RSA model to an equilibrium situation. Some of these studies involve desorption from the surface [3], and also surface diffusion [7]. Among these works, we have proposed a model called sequential quenching (SQ) [8], in which the surface diffusion of each added particle follows its arrival to the surface. After a particle gets equilibrated with other previously quenched ones, the particle itself is quenched in place and the new addition begins. For the situations where there is no attractive interaction among particles and strong interaction between particles and the surface, the deposition of particles will grow a monolayer film, whose structure can be obtained by methods based on statistical thermodynamics [8].

The three-dimensional (3D) fluid structures have long been investigated by equilibrium liquid theories such as Ornstein–Zernike integral-equation theory. Subsequently, the 2D fluid structures were studied and the application of integral-equation theory yielded structures of monolayer film. Madden and Glandt [9] and later Given and Stell [10] developed the application of integral equation theories to systems where at least one of the components was not in equilibrium but quenched in an imposed configuration. The Replica Ornstein–Zernike (ROZ) integral equations [11] have been applied to the description of fluids adsorbed within quenched disordered matrices. In a SQ process each newly arriving particle samples the space on the surface within the disordered matrix formed by the already quenched ones and is thus a particular case of a quenched–annealed mixture.

In our previous work, we applied the set of integral equations (modified Ornstein–Zernike equations) to a study of the simple SQ of hard particles of the same size, whose resulted structures were equivalent to ones grown through the RSA [8]. The equivalence arises from the fact that a newly added, annealed hard particle performs a uniform sampling from the free volume of the sequentially quenched system. Therefore, it becomes irrelevant whether it is or is not allowed to undergo equilibration and the result of the addition is identical to that of a successful insertion in RSA. We also extended our work to study the effect of short-range attractive forces among particles on the final structures of the films [12]. In all previous works, we assumed size monodispersity. However, particles in nature such as colloidal- and bio-particles are not monodisperse—their sizes vary considerably, resulting in different patterns of structures. Researches on nanoparticle film structure of colloidal lithography made use of variation of feature size, shape and spacing of particle [13,14]. Those structures were formed under the influence of electrostatic particle–particle interactions, which could be controlled by adjusting the ionic strength of the particle suspensions, and were described by the RSA model. The effect of size polydispersity on the monolayer film structures grown through self-assembly was mainly studied to obtain their jamming limits and their kinetics which were different from the monodisperse case. A study to obtain the monolayer structure by thinning the liquid multilayer films also reported that original films with size polydispersity were less stable than the monodisperse system [15].

Most theoretical work on structures of polydisperse systems are for equilibrium situations. The integral-equation theories were proposed to study Yukawa fluids [16], colloidal suspensions [17], and fluid mixtures of adhesive colloidal particles [18], by using the continuous Schulz distribution for size polydispersity. Some works explained equilibrium structures of monolayer films [19]. Influence of polydispersity on RSA has also been investigated by kinetic equations and the structures obtained from simulations were consistent with ones from experiments [20]. Unlike those works, in this study we firstly aim to apply our modified Ornstein–Zernike equations, which was successful before in the study of monodisperse RSA, to the RSA of polydisperse spherical particles, together with the Percus–Yevick (PY) approximation which is often used in the study of fluid structures. In order to check the validity of our theories, the effect of polydispersity is added by simply randomizing the sizes in a specified range with uniform probability.

In Section 2, the integral-equation theory based on the multi-component treatment is explained. In Section 3 the detail of the simulation is given. Section 4 gives preliminary results when the theories are applied using the uniform distribution function. The size exclusion effect is discussed in Section 5 and the final results along with the discussions are provided in Section 6.

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