Commun. Comput. Phys. Vol. **33**, No. 1, pp. 1-21
doi: 10.4208/cicp.OA-2021-0244 January 2023

Dynamical Demixing of a Binary Mixture Under Sedimentation

André S. Nunes[1,2,3], Rodrigo C. V. Coelho[1,2], Vasco C. Braz[1,2], Margarida M. Telo da Gama[1,2] and Nuno A. M. Araújo[1,2,*]

[1] *Centro de Física Teórica e Computacional, Faculdade de Ciências, Universidade de Lisboa, P-1749-016 Lisboa, Portugal.*
[2] *Departamento de Física, Faculdade de Ciências, Universidade de Lisboa, P-1749-016 Lisboa, Portugal.*
[3] *Electric Ant Lab, Science Park 106, 1098 XG, Amsterdam, The Netherlands.*

Received 3 December 2021; Accepted (in revised version) 19 April 2022

Abstract. We investigate the sedimentation dynamics of a binary mixture, the species of which differ by their Stokes coefficients but are identical otherwise. We analyze the sedimentation dynamics and the morphology of the final deposits using Brownian dynamics simulations for mixtures with a range of sedimentation velocities of both species. In addition, we use the lattice Boltzmann method to study hydrodynamic effects. We found a threshold in the difference of the sedimentation velocities above which the species in the final deposit are segregated. The degree of segregation increases with the difference in the Stokes coefficients or the sedimentation velocities above the threshold. We propose a simple analytical model that captures the main features of the simulated deposits.

AMS subject classifications: 82-08, 76Dxx, 76Txx

Key words: Sedimentation, Brownian dynamics, lattice Boltzmann method.

1 Introduction

The process of sedimentation, where particles in suspension settle in the presence of a gravitational field, is ubiquitous over a wide range of length scales [1–5]. For example, sedimentation plays a relevant role in natural water transport, affecting the chemical composition of the seabed [6] and the water quality in reservoirs [7, 8]. At the other end of the scale, sedimentation by ultracentrifugation is used as an analytical tool in medical, biological and pharmaceutical applications, where the constituents of a suspension are

*Corresponding author. *Email address:* `nmaraujo@fc.ul.pt` (N. A. M. Araújo), `and.sousanunes@gmail.com` (A. S. Nunes), `rcvcoelho@fc.ul.pt` (R. C. V. Coelho), `vasco_braz@hotmail.com` (V. C. Braz), `mmgama@fc.ul.pt` (M. M. Telo da Gama)

 ©2023 Global-Science Press

separated by molecular weight [9, 10]. At the fundamental level, sedimentation experiments were developed and used extensively in statistical physics and colloidal science to evaluate the equation of state of hard spheres [11] and to study the phase diagram of colloidal particles [12].

Studies of the sedimentation of mixtures of particles that differ in their buoyant mass revealed a rich phase stacking diagram under thermodynamic equilibrium conditions [13, 14]. The structure of the final deposit depends not only on the difference in buoyant masses but also on the particle-particle interactions [15–19]. The roughness of the particle surface is known to affect the hydrodynamics of the surrounding fluid, e.g., alters the lubrication film thickness [20, 21]. These conditions could be realized in an experiment with particles composed by a rigid core covered by an elastic surface layer, which would affect the hydrodynamics while the pairwise interactions remain dominated by the rigid cores. In order to shed light on the role of the hydrodynamic radius on the sedimentation dynamics, we consider a binary mixture of particles that differ only through their Stokes coefficient when using molecular dynamics. Following a method developed previously [22], we consider that the particles differ by their Stokes coefficients only, and are identical otherwise. Thus, the thermodynamic phase is perfectly mixed and demixing, if it occurs, is dynamically driven. Hydrodynamic interactions are know to be relevant in different limits, triggering, for example, a number of different instabilities [23, 24]. To account for hydrodynamic effects, we consider a complementary set of simulations by modelling the fluid-particle interaction with the lattice Boltzmann method (LBM).

When thermal fluctuations are negligible, colloidal particles in solution are expected to sediment with a sedimentation velocity that depends only on the strength of the gravitational field and their Stokes coefficient. Thus, distinct Stokes coefficients imply different sedimentation velocities. In what follows, we show that the morphology of the final deposit depends crucially on the ratio of the sedimentation velocities. Above a certain threshold, which will be quantified below, the particles are segregated in the final deposit, as they arrive at the substrate at different rates and do not have time to relax to the thermodynamic equilibrium mixed state. We investigate this segregation and discuss its dependence on the model parameters.

The paper is organized in the following way. In Section 2, we describe the model and the details of the simulations. Results from the particle-based simulations (Brownian dynamics), LBM and an analytical model are discussed in Section 3. Finally, we draw some conclusions in Section 4.

2 Model and simulations

2.1 Molecular dynamics

We consider a binary mixture of identical spherical particles where the two species are characterized by distinct Stokes coefficients. The particles are in a uniform gravitational field along the vertical direction (y-direction) and inside a rectangular two-dimensional