

Separation of colloidal particles in a packed column using depletion and structural forces



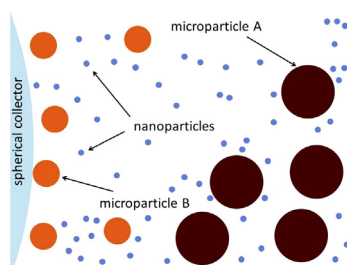
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HIGHLIGHTS

- A suspension of binary colloidal particles was passed through a packed bed.
- Suspension also contained highly charged nanoparticles.
- Nanoparticles created depletion and structural forces between colloids and bed.
- More of the smaller colloids were trapped in the bed, producing separation.
- Increased structural repulsion inhibited the large particle deposition.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 24 November 2013
 Received in revised form 20 January 2014
 Accepted 24 January 2014
 Available online 4 February 2014

Keywords:

Depletion forces
 Structural forces
 Particle separations
 Packed column

ABSTRACT

The depletion and structural forces produced by nonadsorbed nanoparticles were used to separate an equinumber binary dispersion of 1.5 and 0.82 μm polystyrene sulfate (PS) particles in a flow-through packed bed. Experiments consisted of injecting a pulse of the binary dispersion into a carrier fluid at the inlet of a cylindrical column packed with 0.5 mm silica collector beads. The carrier fluids used were either an aqueous electrolyte solution or a dispersion of silica nanoparticles. When the carrier fluid was a dispersion of silica nanoparticles, the ratio of PS particles in the column outlet would change from 1:1 large-to-small particles to slightly over 2:1, implying that more of the smaller, 0.82 μm particles were being trapped on the surface of the collector beads due to depletion forces. Experiments with a single particle size (either 1.5 or 0.82 μm PS particles only) were also performed and the results were consistent with those obtained with the binary mixture. Equilibrium potential energy profiles between a PS particle and a flat silica plate were calculated using the equilibrium force-balance model of Walz and Sharma [J. Colloid Interface Sci. 168 (1994) 485]. The long-range, secondary energy barrier (arising from structuring of the nanoparticles in the gap region) for the 1.5 μm particles was two times greater than that for the 0.82 μm particles, meaning that the 0.82 μm particles were far more likely to overcome the energy barrier and get trapped on the surface of the collector beads. Although not explored, separation of particles based on other properties, such as surface charge density, should also be possible with this approach, as the depletion and structural forces are sensitive to such properties.

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1. Introduction

The efficient separation of colloidal particles remains a topic of great interest and new techniques or advances to existing techniques continue to be made [1–3]. While batch separation processes techniques, such as elutriation and sieving, have been used for over a hundred years, most modern technique are flow-based

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and involve capitalizing on differences in transport rates of particles flowing through either open or packed capillaries [4]. In many of these techniques, the variation in transport rates arises from differences in average radial position between particles of different properties being carried along by fluid with a parabolic velocity profile, such that particles that have an average position closer to an immobile surface (e.g., the capillary wall) will transport slower than those with an average position farther in the bulk.

For example, in hydrodynamic chromatography, in which the fluid is pumped through a column packed with a nonporous material, separation based on size arises from the simple fact that smaller particles can move closer to the immobile substrate and thus have a slower average transport rate than larger particles [5–7]. Ruckenstein and Prieve showed that the nature of the interaction between the particle and immobile surface also plays a critical role in the transport rate and utilized this fact to develop the technique of potential barrier chromatography [8–10]. Here the magnitude of the potential energy barrier that particles must overcome in order to deposit on the surface of the immobile collector is manipulated such that the barrier is lower (favoring deposition) for a specific type of particle.

Perhaps the most common commercial technique that is that of field flow fractionation (FFF), which was first proposed by Giddings in 1966 [11]. Here the average transverse position of a particle in a parabolic velocity profile is altered by an external field acting perpendicular to the flow direction. A wide variety of applied fields have been utilized, including gravity, magnetic, electrical, thermal, and cross-flow [12–14].

In this paper, we propose a new particle separation technique that utilizes a completely different approach for manipulating the transverse position of particles in a parabolic velocity field. The approach capitalizes on the change in the potential profile between particles and an immobile surface that is produced by the addition of nonadsorbing nanoparticles to the carrier fluid. Specifically, these nanoparticles create long-range depletion and structural forces that are very sensitive to the size and charge of the particles to be separated. In addition, the magnitude and range of these forces can be carefully controlled via the size and concentration of the nanoparticles. As such, this approach provides a tool for efficiently separating particles without the need for external fields.

This paper presents the results of a preliminary experimental study demonstrating the viability of the concept. Specifically, a bidisperse solution of polystyrene latex particles is effectively separated in a simple packed bed using highly charged silica nanoparticles as the depletant. The separation mechanism is then explained based on the predicted changes in the potential energy profile between the particles and collected beads in the bed.

2. Background—separation of particles using depletion forces

While the separation scheme proposed in this work is novel, the concept of separating particles using depletion effects is certainly not a new one. Bibette used depletion interactions to purify a polydisperse emulsion via a batch fractionating crystallization scheme [15]. In their work, a silicone oil–water emulsion was purified using sodium dodecylsulfate (SDS), which was added to the oil–water emulsion to a concentration of 0.02 M, just above the critical micelle concentration (cmc). After 24 h, a top layer would form, which was removed and placed in a different container. SDS at a slightly higher concentration was again added to the two different mixtures and the process repeated until a narrow droplet size distribution was reached.

Bonard et al. used a similar scheme and showed that a dispersion of carbon nanotubes could, in practice, be purified using depletion forces arising from micelles [16]. Khripin et al. were able to separate a mixture of different length carbon nanotubes by adding polyethylene glycol (PEG) in stepwise increases [17]. As the nanotubes precipitated, either the supernatant or sediment would be removed and the addition of PEG repeated until the size distribution of the nanotubes was sufficient for the desired application. Park et al. separated a dispersion of gold nanorods and nanospheres using a micelle-induced depletion interaction [18]. When cetyltrimethylammonium bromide (CTAB) surfactant was added to the dispersion, the gold nanorods would aggregate and sediment to the bottom of the container, leaving the nanospheres in the supernatant. After only one sedimentation step, the number fraction of rods could be increased from 77% to ~99%.

While effective, such batch processes are rather cumbersome, as achieving a thorough separation of a dispersion of particles can involve multiple separation steps. Clearly the ability to produce such separations in a continuous device would be more efficient and more attractive for commercial application.

The approach described in this manuscript for accomplishing this is based on earlier work by Weronki et al. who showed that polystyrene microparticles flowing through a packed column of glass beads could be trapped within the column by attractive depletion forces when the carrier fluid was a dispersion of silica nanoparticles [19]. In the work of Weronki et al., a pulse of polystyrene microparticles was injected into a carrier fluid at the column inlet. When the carrier fluid was a dispersion of nanoparticles, the recovery of the polystyrene particles was as low as 34%, whereas non-nanoparticle experiments had recoveries near 100%. When the carrier fluid was switched from a dispersion of nanoparticles to either an electrolyte solution or water, some of the trapped particles were released, suggesting that a fraction of the trapped particles were held in reversible energy wells that disappeared once the carrier fluid was changed. The authors showed that the particle recovery was a decreasing function of both the silica nanoparticle concentration and the carrier fluid residence time, and an increasing function of the fluid velocity in the bed.

In addition to involving a continuous flow of material, another advantage of the approach described here is that rather than having multiple separation steps, the extent of separation can be readily controlled by changing the residence time of the colloidal particle dispersion within the packed bed, which can be easily done by either changing the flow rate of the carrier fluid or by changing the length of the packed bed.

3. Theory

The experimental system in this work consists of a binary, negatively charged dispersion of colloidal particles in an electrolyte solution that is flowing through a stationary bed of negatively charged, spherical collector beads. The dispersion can also contain negatively charged spherical nanoparticles that induce depletion and structural forces between the colloidal particles and the collector beads. A simple schematic of the system is shown in Fig. 1.

Our objective here is to calculate the potential energy profile between the colloidal particles and the collector beads, which we assume can be written as the sum of electrostatic, van der Waals and depletion/structural interactions. Thus

$$E_{\text{Tot}}(h) = E_{\text{elec}}(h) + E_{\text{vdw}}(h) + E_{\text{dep}}(h) \quad (1)$$

where $E_{\text{elec}}(h)$ the electrostatic repulsion, $E_{\text{vdw}}(h)$ is the van der Waals attraction, and $E_{\text{dep}}(h)$ is the depletion/structural interaction.

The electrostatic interaction potential is calculated using the linear superposition approximation (LSA) developed by Bell et al. [20].

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