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On colloid retention in saturated porous media in the presence of energy barriers: The failure of α, and opportunities to predict η

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[1] This contribution reviews recent findings that illuminate the processes governing colloid retention in porous media under environmentally relevant conditions. In the environment, colloids act as conveyors of contaminants, or even as contaminants themselves; however, despite decades of research, we are unable to accurately predict the retention of colloids in granular aquifer media under environmental conditions, where repulsion exists between colloids and surfaces. This failure cannot be blamed solely on the complexities of the subsurface, since colloid filtration theory (CFT) works well in the absence of colloid-collector repulsion despite its idealization of porous media as consisting of spherical grains completely surrounded by fluid envelopes. Rather, the failure of CFT stems from failure to incorporate the correct mechanisms of retention when repulsion exists. Recent observations implicate wedging in grain-to-grain contacts and retention in secondary energy minima as dominant mechanisms of colloid retention in the presence of an energy barrier. Mechanistic simulations in unit cells containing grain-to-grain contacts corroborate these mechanisms of colloid retention. The resulting concept for colloid retention in the presence of an energy barrier involves translation of colloids across the collector surfaces until they become wedged within grain-to-grain contacts, or are retained via secondary energy minima (without attachment) in zones where the balance of fluid drag, diffusion, gravitational, and colloid-collector interaction forces allow retention. The above findings highlight the pore domain geometry as a dominant governor of colloid retention in so far as the geometry gives rise to grain-to-grain contacts and zones of relatively low fluid drag.

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1. Introduction

1.1. Similarities of Transport Among Biological and Nonbiological Colloids

[2] The long-term mobility of contaminants in the subsurface is entwined with the transport of colloidal phases [e.g., *Kersting et al.*, 1999; *Novikov et al.*, 2006], to which they are bound through sorption, complexation, and precipitation processes. Colloids themselves may act as contaminants in the form of pathogens [*Tufenkji et al.*, 2002; *Jiang*, 2006; *Abulreesh et al.*, 2006; *Muniesa et al.*, 2006] and potentially toxic novel nano-materials [*Dhawan et al.*, 2006; *Templeton et al.*, 2006; *Lyon et al.*, 2006]. Although colloid transport in the subsurface is an important challenge to the protection of water resources, existing readily implemented colloid transport models do not capture the fundamental processes governing colloid transport in the environment, where repulsion exists between colloids and porous media grain surfaces. In this paper, the term colloid is used to refer to “particles” in the size range between 10 nm and 10 μm regardless of origin (biological or non-biological). Biological colloids (e.g., viruses, bacteria, and protozoa) and non-biological colloids (e.g., microspheres and mineral colloids) differ in terms of potential physiological influences on the transport of biological colloids in porous media. Despite these important differences between biological and abiotic colloids, their transport behaviors in environmental porous media share important similarities; the most fundamental among them being that they undergo deposition in porous media even when existing theory suggests otherwise; that is, retention occurs despite the presence of a formidable energy barrier that should prevent direct attachment to the surface. Additional important similarities are apparent in the dynamics of their deposition and reentrainment, as reflected in their distribution in porous media with distance from a source, and the influences of fluid velocity on their deposition and reentrainment rate coefficients, as reviewed recently by *Johnson et al.* [2007a].

1.2. Classic Filtration Theory

[3] The challenge in simulating the subsurface transport of constituents is to develop an accurate representation of

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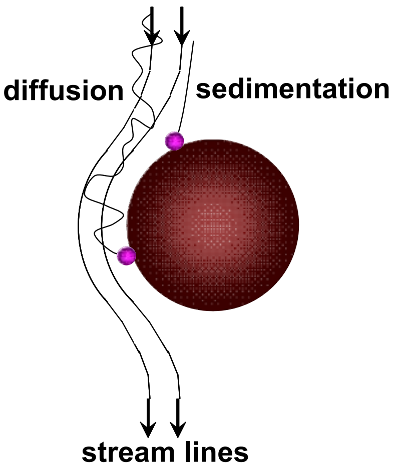

Figure 1. Schematic of Happel sphere-in-cell unit collector. The hatched circle represents the fluid shell completely surrounding the collector. Streamlines denote fluid flow, whereas the schematic shows that colloid trajectories are influenced by sedimentation and diffusion, in addition to fluid drag.

mass transfer, for example, by use of rate coefficients in the advection-dispersion-deposition-reentrainment equation,

$$\frac{\partial C}{\partial t}\theta = -v\frac{\partial C}{\partial x}\theta + D\frac{\partial^2 C}{\partial x^2}\theta - k_f C\theta + k_r C_s \rho_b \quad , \tag{1}$$

where C is the aqueous concentration of the constituent, and C_s is the reversibly retained stationary phase concentration of the constituent; v is the pore fluid velocity; D is the dispersion coefficient of the constituent under the conditions considered; k_f and k_r are the forward (removal from the aqueous phase) and reverse (addition to the aqueous phase) rate coefficients, respectively; θ is the volumetric water content and ρ_b is the bulk density of the stationary phase.

[4] Relating D mechanistically to the observed properties of the porous media is a major challenge taken up in the subfield of stochastic hydrology. The difficulty in mechanistic determination of k_f and k_r varies with the process considered. For radioactive decay, these rate coefficients are not dependent on porous media properties or solution chemistry; hence values can be adapted from the literature for a wide range of contexts. In the case of colloid deposition, the porous media, solution, and colloid properties each influence the process, yielding complex mechanistic relationships of k_f and k_r to attributes of the system.

[5] Complex transport behaviors in groundwater are often attributed to heterogeneous physical properties of the subsurface, which motivates the characterization of increasingly realistic pore domains [e.g., *Y. Li et al.*, 2005; *Yang et al.*, 2006]. However, the benefit of increased realism must be balanced against the cost of decreased simplicity. Approaches that incorporate rigorous mechanisms of mass transfer within representative (nonrealistic) pore domains are valuable if they are effective for a range of porous media. Such an approach that is widely used despite its well-noted limitations [*Elimelech and O'Melia*, 1990; *Nelson and Ginn*, 2005] is colloid filtration theory (CFT) [*Yao et al.*, 1971; *Rajagopalan and Tien*, 1976; *Tufenkji and Elimelech*, 2004a; *Nelson and Ginn*, 2005], which utilizes the results of numerical simulations of pore-scale processes to develop phenomenological functions to scale up to the continuum. The approach of CFT is sometimes misunderstood, as exemplified by a recent review article [*Rockhold et al.*, 2004] wherein existing filtration theory was referred to as "essentially an empirical correlation function that contains a number of dimensionless terms…". CFT is based on mechanistic simulation of colloid transport; wherein the mechanistic results are encompassed by a phenomenological expression to allow prediction of colloid transport without developing the complex numerical simulation.

[6] CFT is based upon the Happel sphere-in-cell model [*Happel*, 1958], which is represented by an isolated solid sphere immersed in a concentric fluid shell (Figure 1), where the thickness of the fluid shell is chosen such that its porosity is equal to the actual porosity of a packed bed. The flow field in this unit cell is solved under the conditions that "no slip" occurs on the solid-fluid interface, and that the undisturbed (uniform) flow field is reestablished at the outer boundary of the fluid envelope [*Happel*, 1958]. Colloid trajectory models were developed within the flow field of the Happel sphere-in-cell model to examine the probability of colloid interception with the collector surface based on a mechanistic force balance [*Rajagopalan and Tien*, 1976; *Rajagopalan et al.*, 1982]. The forces examined were fluid drag with hydrodynamic retardation near the collector surface, gravity, and colloid-surface van der Waals and electrostatic interactions. Diffusion was recently added into the force balance by *Tufenkji and Elimelech* [2004a] and *Nelson and Ginn* [2005]. The numbers of colloids that intercept the surface relative to those released into the unit cell constitute the collector efficiency (η), which is easily transformed to a deposition rate coefficient according to [*Logan et al.*, 1995; *Nelson and Ginn*, 2005]

$$k_f = \frac{3(1-\theta)^{1/3}}{2d_c} v\eta, \tag{2}$$

where d_c is the collector diameter, and η is defined as by *Nelson and Ginn* [2005]. The values of η from mechanistic particle trajectory simulations (in the absence of electrostatic repulsion) have been regressed to dimensionless parameters to provide phenomenological expressions for estimation of η under the spectrum of conditions expected in the environment [*Rajagopalan and Tien*, 1976; *Tufenkji and Elimelech*, 2004a]. These expressions are described in detail in the associated references, and so are not reproduced here.

[7] By assuming steady state transport, negligible diffusion relative to deposition, and negligible detachment, the following equation can be derived, which predicts a log linear decrease in the concentration of suspended (and retained) colloids as a function of distance (x) from the source.

$$Ln\frac{C}{C_0} = -\frac{k_f}{v}x, \tag{3}$$

where C_0 is the concentration at the source.

1.3. Interaction Forces

[8] The correlation equations for η [*Rajagopalan and Tien*, 1976; *Tufenkji and Elimelech*, 2004a] are quite accu-

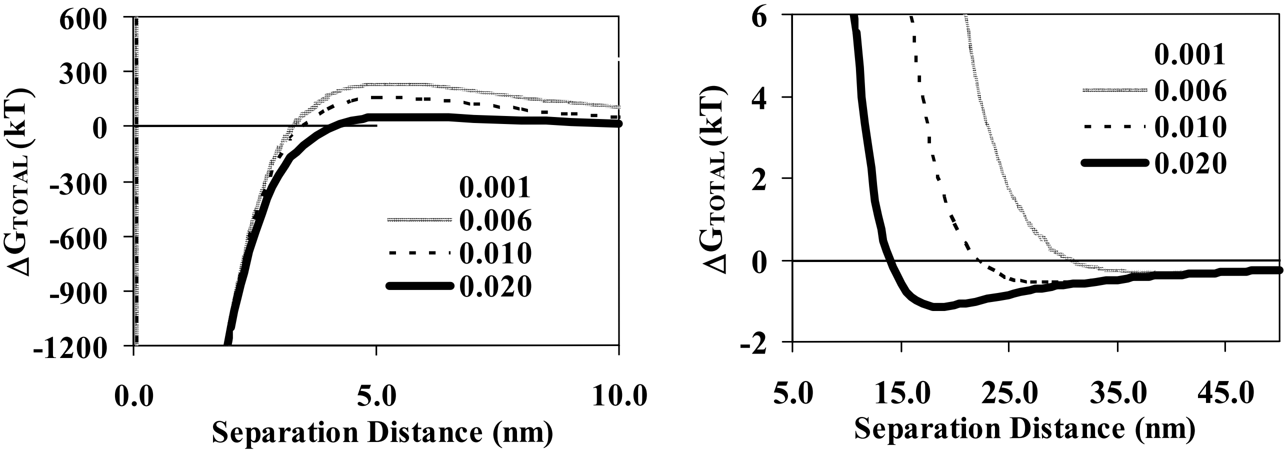

Figure 2. Example DLVO interaction energy profiles at various ionic strengths for 1.1-μm carboxylate modified latex microspheres, for separation distances (left) within 5 nm and (right) between 5 and 50 nm from the collector surface. The zeta potentials of the colloid and the collector were -20 mV and -60 mV, respectively. The DLVO calculations were performed as described by *Tong et al.* [2005b].

rate in granular aquifer media when no electric double layer repulsion exists between the colloid and collector surfaces (energy barrier is absent). The energy barrier is absent when the surfaces are oppositely charged, or when the ionic strength is sufficiently high to compress the electric double layer repulsion to short separation distances [*Derjaguin and Landau*, 1941; *Verwey and Overbeek*, 1948]. The profile of net interaction energy (ΔG_{total}) is strongly negative (attractive) at colloid-surface separation distances less than a few nm (the so-called primary energy minimum) owing to the dominance of van der Waals attraction (Figure 2). At intermediate separation distances, ranging from a few to tens of nm, ΔG_{total} may be strongly positive (repulsive) for like-charged surfaces, yielding an energy barrier to deposition with magnitude inversely proportional to solution ionic strength. At even greater separation distances, weak van der Waals attraction may exceed weak electric double layer repulsion, yielding an attractive secondary energy minimum (Figure 2, right).

[9] Under conditions absent an energy barrier (e.g., oppositely charged colloid and collector surfaces), the concentrations of mobile and retained colloids are observed to decrease exponentially with distance from the source, as expected from equation (3) above [*Li et al.*, 2004; *Tufenkji and Elimelech*, 2004b; *X. Li et al.*, 2005]. As well, the reentrainment of colloids from porous media is negligible in the absence of an energy barrier, consistent with existing filtration theory [*Li et al.*, 2004; *X. Li et al.*, 2005], and the estimated value of η predicts well the magnitude of colloid deposition during transport through porous media [e.g., *Rajagopalan and Tien*, 1976; *Elimelech and O'Melia*, 1990; *Tong and Johnson*, 2006].

1.4. Presence of Energy Barriers

[10] In the presence of even relatively small energy barriers to deposition (e.g., >10 kT), the explicit numerical models of colloid deposition based on the Happel sphere-in-cell model predict that no colloid deposition will occur [e.g., *Elimelech and O'Melia*, 1990]. This is shown in Figure 3 by

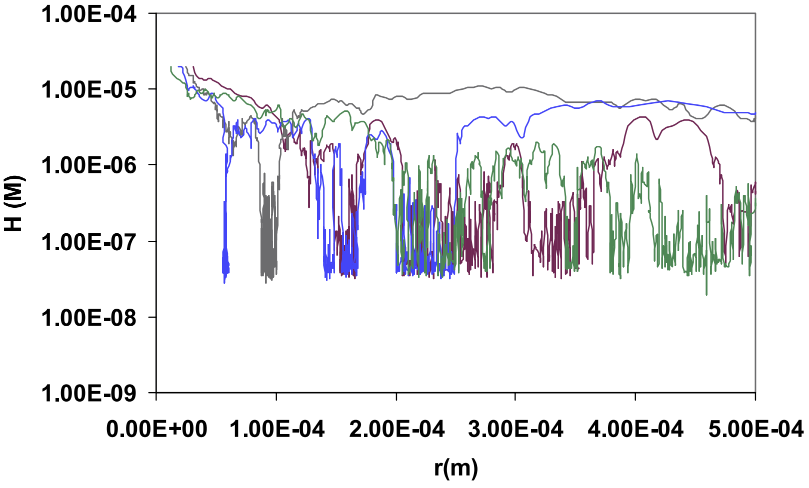

Figure 3. Example of simulated trajectories for colloids approaching an oppositely charged surface in an impinging jet system. Colloids enter the system from the top left portion of the diagram. H is the separation distance between the colloid and the flat surface, and r is the distance from the axis of the impinging jet. The simulations were performed as described by *Johnson and Tong* [2006].

a simulated trajectory of colloid approaching an oppositely charged surface (using the model of *Johnson and Tong* [2006]), where the energy barrier prevents the colloid from approaching closer than 20 nm separation distance between the two surfaces. Surfaces of environmental colloids (biological and nonbiological) and porous media tend to display overall like charge (negative), yielding overall repulsive electric double layer interactions between them. The overall negative surface charge emanates from acidic functional groups, which dominate the surfaces of biological colloids and the surfaces of silicate mineral grains under most environmentally relevant groundwater conditions (e.g., pH 6–9) [*Davis*, 1982; *Tipping and Cooke*, 1982]. Despite the resulting formidable energy barriers to deposition, the prevalence of colloid deposition in environmental systems is well demonstrated; for example, by the general success of filtration as a water treatment technology and by the generally higher quality of groundwater relative to surface water, in addition to hundreds (if not thousands) of published experiments in the colloid transport literature.

[11] The failure of classic filtration theory when repulsion exists has been traditionally addressed by assuming that interception of the surface does not always yield attachment in the presence of an energy barrier, and is accounted for by multiplying η by another term (α) to represent the probability of attachment upon interception. The term α may represent attachment at locations where the repulsive energy barrier is reduced or eliminated. Some of the minor mineral phases in porous media, such as iron oxides, may display positive surface charge at environmental pH conditions. One can characterize the bulk of subsurface media as being negatively charged, but with varying extents of surface charge heterogeneity arising from mineral phases such as iron oxides [*Song and Elimelech*, 1993, 1994; *Coston et al.*, 1995; *Johnson et al.*, 1996]. Iron oxyhydroxide coatings have been shown to increase bacterial attachment in natural [*Scholl and Harvey*, 1992] and model sediments [*Johnson and Logan*, 1996], owing to electrostatic attraction between negatively charged bacteria and the positively charged iron oxyhydroxides (lack of an energy barrier between the colloid and the surface). The same has been demonstrated for bacteriophage [*Powelson et al.*, 1991; *Pieper et al.*, 1997; *Foppen et al.*, 2006]. This conclusion is also based on the observation that bacterial attachment in iron oxyhydroxide coated sand is sensitive to increases in pH that result in charge reversal of the iron oxyhydroxide coatings [*Scholl and Harvey*, 1992]. Even in the absence of discreet metal oxide phases, nano-scale defects in mineral structures can confer charge heterogeneity or otherwise influence the extent of repulsion from otherwise negatively charged surfaces. Hence colloid deposition onto overall like-charged surfaces (relative to the colloid) can result from localized nanoscale patches of attractive surface charge [*Elimelech and O'Melia*, 1990; *Song and Elimelech*, 1993, 1994; *Johnson et al.*, 1996] and surface roughness [*Bhattacharjee et al.*, 1998; *Shellenberger and Logan*, 2002] which act to locally reduce or eliminate the energy barrier to deposition. Values of α have been successfully predicted under particular conditions for media with significant (and known) percentages of attractive surface area [*Song and Elimelech*, 1993, 1994; *Johnson et al.*, 1996]. Hydrophobicity is another surface attribute that may locally influence the extent of repulsion or attraction; however, this attribute is only tentatively characterized relative to surface charge, and so is not discussed here.

[12] Some researchers have attempted to correlate α (expressed as the ratio of η observed in the presence of an energy barrier relative to η observed or calculated in the absence of an energy barrier) to other properties of porous media (e.g., hydraulic conductivity) with some success [*Rehmann et al.*, 1999; *Ren et al.*, 2000]; however, such an approach cannot be transferred across different sites, or likely even different locations at the same site. Others have empirically correlated observed values of α to dimensionless parameters representing the energy barrier [*Elimelech*, 1992; *Bai and Tien*, 1996, 1999; *Vaidyanathan and Tien*, 1989; *Chang and Huang*, 1998]. Unfortunately, these types of correlations are not mechanistically based, and so can be applied only to the limited set of experimental conditions to which the empirical correlation was developed.

2. New Observations Calling for New Models of Colloid Filtration

2.1. Failure of α and New Insights

[13] Recently, major flaws in the strategy utilizing α have become apparent. First, the approach is predicated on the assumption that η is correctly predicted in the presence of an energy barrier, such that that α depends solely on colloid-collector interaction forces. Accumulating results demonstrate this assumption to be false, and that α decreases with increasing fluid velocity [*X. Li et al.*, 2005; *Johnson et al.*, 2007a]. Furthermore, the approach using α does not account for important differences in the mechanisms of colloid retention in porous media versus unbounded surfaces (lacking crevices or grain-to-grain contacts). Experiments comparing colloid deposition on unbounded versus porous media surfaces under conditions where the surfaces in the two systems display equivalent surface characteristics, and where the near-surface tangential fluid velocities in the two systems are similar, yield insight into deposition mechanisms in the presence of an energy barrier. The most commonly used system for examining colloid deposition onto unbounded surfaces is the impinging jet system, in which the solution is directed normal to the flat surface, where upon impinging the surface it spreads radially [*Adamczyk et al.*, 1995; *Weiss et al.*, 1998; *Yang et al.*, 1998]. Colloid deposition efficiencies (ratio of colloid deposition rate in presence versus absence of an energy barrier) have been demonstrated to be much greater in porous media relative to impinging jet systems (factors of 2 to 50) under equivalent conditions [*Redman et al.*, 2004; *Walker et al.*, 2004; *Brow et al.*, 2005; *Tong and Johnson*, 2006], suggesting that the vast majority of colloid retention in porous media is not due to surface heterogeneity, which presumably operates in both the impinging jet and the porous media.

[14] A mechanism driving the observed excess colloid retention in porous media relative to flat surfaces is indicated by the fact that the majority of the colloids retained in porous media in the presence of an energy barrier are released in response to elution of the packed porous media with low ionic strength solution [*Franchi and O'Melia*, 2003; *Hahn and O'Melia*, 2004; *Hahn et al.*, 2004; *Tong*

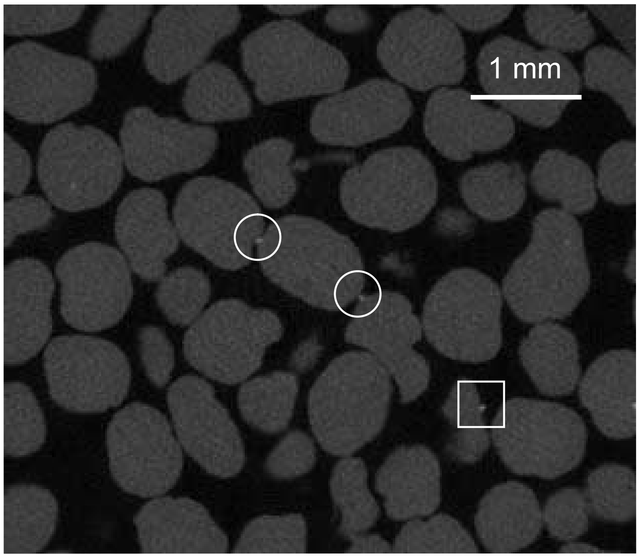

Figure 4. Representative cross-sectional XMT image of quartz sand (gray areas) and gold-coated hollow microspheres (white spots). Black areas represent pore water. The microsphere in the white square demonstrates single contact retention. Microspheres in the white circles are retained at grain-grain contacts. After *Li et al.* [2006b].

and Johnson, 2006]. In contrast, elution of the equivalently loaded impinging jet system with low ionic strength solution (and increases in fluid velocity up to a factor of 200) yields negligible colloid release [*Johnson and Tong*, 2006; *Tong and Johnson*, 2006]. These observations indicate that a significant fraction of the colloids retained in porous media in the presence of an energy barrier are associated with surfaces via secondary energy minima [*Hahn and O'Melia*, 2004; *Hahn et al.*, 2004]; whereas those retained on unbounded surfaces are associated with surfaces via primary energy minima. Release from the porous media would not occur if the colloids were deposited in the primary energy minimum, since the barrier to detachment from the primary energy minimum is negligibly changed as ionic strength is decreased (see the interaction energy profiles, Figure 2) [*Hahn and O'Melia*, 2004; *Hahn et al.*, 2004]. Hence the colloids reentrained from porous media in response to decreased ionic strength appear to have been associated with surfaces via the secondary energy minimum, since the secondary energy minimum is reduced or eliminated via decreased ionic strength (Figure 2). Colloids associated with secondary energy minima would be expected to translate across the grain surface owing to tangential hydrodynamic drag, unless they are retained in zones where fluid drag forces are insufficient to overcome other forces resisting their down-gradient translation (e.g., rear stagnation points or leeward sides of protrusions) [*Johnson and Tong*, 2006; *Tong and Johnson*, 2006]. A portion of the greater deposition efficiency in porous media relative to impinging jets (excess retention) is therefore thought to result from retention of secondary minimum-associated colloids in loose association with the grain surfaces [*Redman et al.*, 2004; *Walker et al.*, 2004; *Brow et al.*, 2005; *Tong and Johnson*, 2006], whereas impinging jets (flat surfaces) lack zones to "protect" colloids from fluid drag and translation out of the system.

[15] Observations demonstrate that colloid retention via secondary energy minima depends on the geometry (or bounding) of the surface; whereas CFT is based on an unbounded spherical surface. This limitation of CFT is most apparent in the retention of colloids via straining, which has recently been inferred from column experiments to be an important colloid retention mechanism in porous media even for colloid:collector ratios as low as 0.005 [*Bradford et al.*, 2002, 2003, 2004, 2006; *Tufenkji et al.*, 2004; *Yoon et al.*, 2006; *Xu et al.*, 2006]. In the absence of an energy barrier to deposition, straining (in the form of wedging in grain-to-grain contacts) is not a dominant colloid retention mechanism, since in the absence of an energy barrier, colloids attach wherever they intercept the surface. This was demonstrated experimentally by *Li et al.* [2006a, 2006b] for colloid:collector ratios near 0.05, where the profiles of retained near-neutrally buoyant gold-and-surfactant-coated 18-μm radius hollow glass microspheres (Figure 4) were (in the absence of an energy barrier) evenly distributed across the 390-μm radius collector grain surfaces (were not associated with grain-to-grain contacts or other porous media features), and their concentrations decreased log linearly with distance from the source, in accordance with expectations from CFT (Figure 5, left). In contrast, in the presence of an energy barrier, the deposited colloids were dominantly located in grain-to-grain contacts, and their concentrations did not decrease with distance from the source according to filtration theory (Figure 5, right), but rather varied nonmonotonically. These contrasting profiles in the presence versus the absence of an energy barrier reflect those for much smaller colloid:collector ratios ranging from 0.0004 to 0.008 [*Tong et al.*, 2005; *Li and Johnson*, 2005; *Tong and Johnson*, 2006; *Bradford et al.*, 2007]. These ratios correspond to a large range of colloid sizes (0.1 to 10 μm) in medium grained (250 $\sim$ 500 μm) sand, and so are of broad relevance.

2.2. Mechanistic Confirmation of Wedging and Retention in Secondary Energy Minima

[16] The experimental findings reviewed above demonstrate that particular retention mechanisms become important in the presence (but not the absence) of an energy barrier, i.e., wedging/straining and retention in secondary energy minima; hence the relationship between η and the mass transfer mechanisms (interception, sedimentation, diffusion) can be expected to differ in the presence versus the absence of an energy barrier. Hence α (as a modification to the Happel-based collector efficiency in the presence of an energy barrier) is not a robust construct for prediction of colloid deposition. The problem of prediction of colloid deposition in the presence of an energy barrier therefore lies in the correct estimation of the probability of colloid-collector encounter (η). This determination suggests the need to revisit the original mechanistic model underlying CFT, and to explore changes to the unit cell (traditionally the Happel sphere in cell), to incorporate grain-to-grain contacts and influences of secondary energy minima, in order to achieve accurate prediction of the collector efficiency in the presence of an energy barrier. Unit collectors that have been examined other than the Happel sphere include two-dimensional [*Payatakes et al.*, 1974a, 1974b]

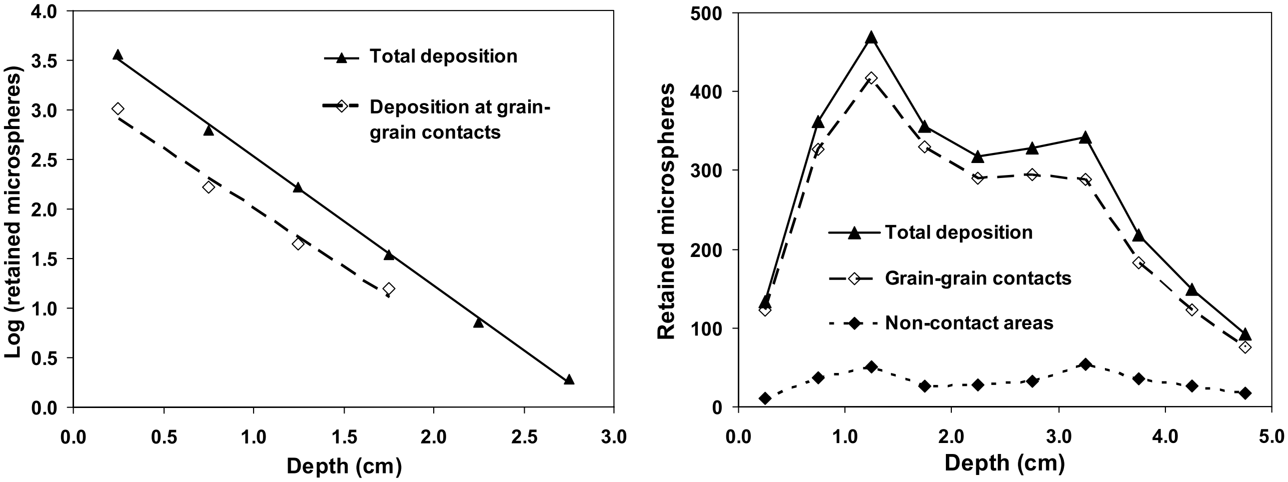

Figure 5. Profiles of total deposition and deposition at grain-to-grain contacts in quartz sand at a pore water velocity of 0.25 cm s^{-1}, in the (left) absence and (right) presence of an energy barrier to deposition. In the absence of an energy barrier to deposition, deposition at grain-to-grain contacts accounted for 27.6% of total deposition, whereas in the presence of an energy barrier to deposition, deposition at grain-to-grain contacts accounted for 84.6% of total deposition. After *Li et al.* [2006a, 2006b].

and three-dimensional [*Paraskeva et al.*, 1991; *Burganos et al.*, 1992, 1994] constricted tube models; however, the smooth parabolic constriction involved in these models does not represent grain-to-grain contacts, which are demonstrated below to be a critical feature in colloid deposition in the presence of an energy barrier.

[17] *Cushing and Lawler* [1998] were, to the knowledge of the authors, the first to explore unit cells of idealized packed porous media composed of spherical grains for prediction of collector efficiencies for aqueous colloids. Their model demonstrated that use of a packed porous media unit cell resulted in colloid deposition in the presence of an energy barrier. However, for unexplained reasons, their collector efficiency was completely insensitive to the presence or absence of the energy barrier, in direct contradiction to experimental results and theoretical expectations. *Johnson et al.* [2007b] continued the exploration of unit cells of representative packed porous media comprising spherical grains, using simple (θ = 0.47) and dense (θ = 0.26) packing structures (Figure 6). This Lagrangian approach integrated velocities from accelerations determined from the same set of forces and torques considered in CFT, but with diffusion added into the force balance. The approach incorporated approximate closed form fluid flow fields for the dense-packed [*Snyder and Stewart*, 1966] and simple-packed [*Sorensen and Stewart*, 1974] unit cells. A variable time step was used, and was conditioned to changes in interaction force and fluid velocity. The mechanistic simulations of *Johnson et al.* [2007b] found that two mechanisms of colloid deposition arose in packed porous media in the presence of an energy barrier: (1) wedging, and (2) retention in secondary energy minima.

[18] Deposition via wedging and retention in secondary energy minima are best illustrated by contrast to deposition in the absence of an energy barrier. In the absence of an energy barrier to deposition, the distribution of colloids that do attach is random across the upstream zones of the collector surfaces, as shown by superposition of the simulated locations of attachment onto the unit collectors (Figure 7). The relatively uniform distribution of attached colloids in the absence of an energy barrier to deposition is observed for both unit collectors, and conforms to the distribution of deposition observed in the Happel sphere-in-cell model. As is the case in the Happel model, the fraction of colloids that attach is dependent on colloid size, fluid velocity, and other characteristics that govern the forces acting on the colloids [*Johnson et al.*, 2007b]. In contrast to the absence of an energy barrier, colloid deposition in the presence of an energy barrier yields a distribution of retained colloids that is highly dependent on the pore domain geometry. All colloid trajectories that resulted in retention showed initial association with the collector surface via the secondary energy minimum [*Johnson et al.*, 2007b]. Upon secondary minimum-association with the surface, the colloids translated along the collector surface in response to fluid drag forces (as in Figure 3). Those colloids that were retained had trajectories that demonstrated either one of two developments: (1) the colloid became bounded by two repulsive force barriers (two surfaces), one of which was overcome owing to fluid drag forces that pushed the colloid into these

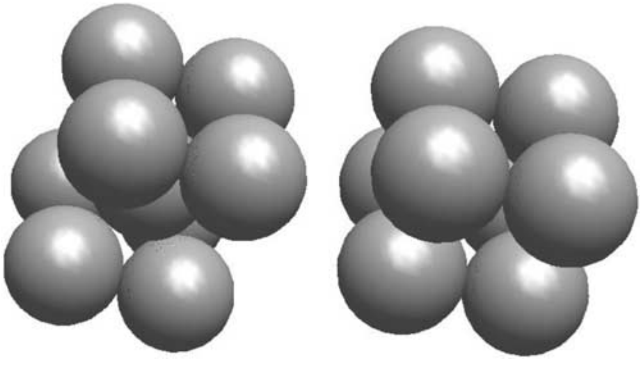

Figure 6. Packing arrangement for (left) dense cubic packing and (right) loose (simple) cubic packing. After *Johnson et al.* [2007b].

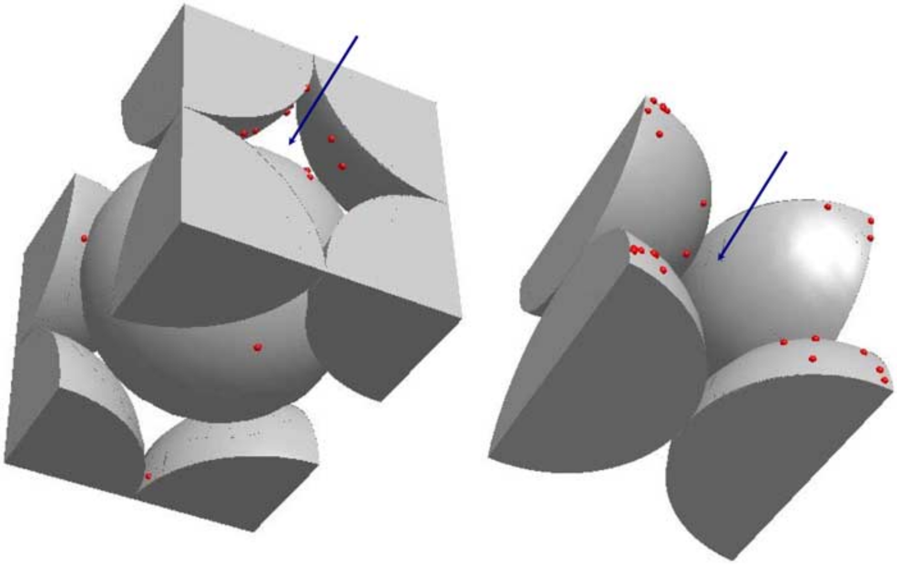

Figure 7. Distribution of attached colloids in the absence of an energy barrier to deposition in (right) the dense-packed unit collector and (left) the simple-packed (loose–packed) unit collector. The distribution was developed by superimposing locations of attachment determined in the simulations onto the unit collectors. Arrows denote directions of flow in z dimension. Colloid and collectors are not proportionally scaled. After *Johnson et al.* [2007b].

bounding barriers, and into direct contact with the collector surface; or (2) the colloid intercepted a zone where fluid drag was insufficient to translate the colloid, where it was retained without attachment but in ‘‘loose’’ secondary minimum-association with the collector surface.

[19] By superimposing the coordinates of colloids that were retained by the first mechanism (attachment via confinement between two bounding surfaces) it was observed that these colloids were retained at grain-to-grain contacts (Figure 8, left), and so represented colloids that were wedged in grain-to-grain contacts. Note that attached colloids located on the outside of the unit collector were wedged between the center spheres of the unit collector shown, and the adjacent unit collector. The simulations demonstrate that wedging (and straining) constitutes attachment that occurs despite the presence of an energy barrier. Fluid drag pushes the colloid toward increased confinement between two (wedging) or more (straining) bounding energy barriers (repulsive surfaces), thereby forcing the colloid to overcome one of the bounding energy barriers and to make

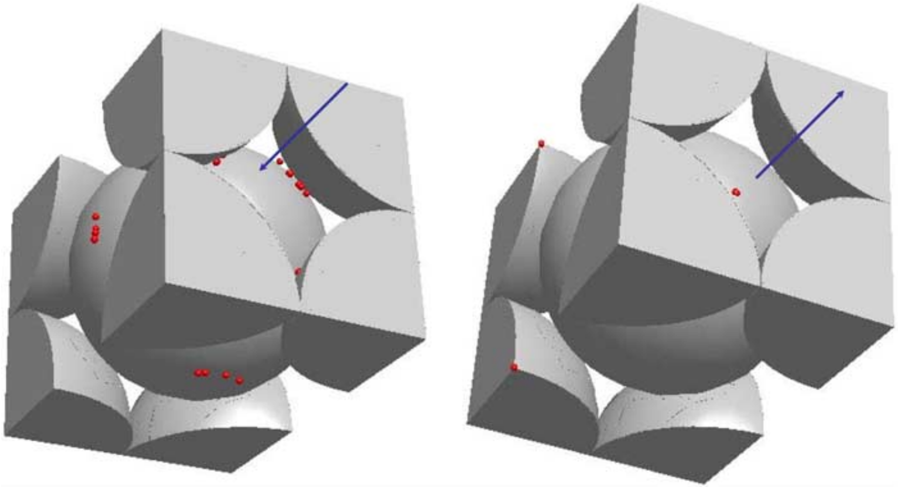

Figure 8. Distribution of (left) wedged colloids and (right) colloids retained in flow stagnation zones in the presence of an energy barrier to deposition in the dense cubic unit collector. The distribution was developed by superimposing onto the unit collectors the locations of wedged colloids determined in the simulations. Note that wedged colloids located on the outside of the unit collector (left) were wedged between the center spheres of the unit collector shown, and the adjacent unit collector. Arrow denotes direction of flow in z dimension. Colloid and collectors are not proportionally scaled. After *Johnson et al.* [2007b].

contact with the corresponding surface. The term wedging was originally used by *Herzig et al.* [1970]. Another group refers to the process by the useful term "contact filtration" [*Yoon et al.*, 2006]. We use the term wedging to distinguish the process from straining; which is traditionally defined as retention in pore throats to small to pass [e.g., *McDowell-Boyer et al.*, 1986; *Bradford et al.*, 2004; *Tufenkji et al.*, 2004, 2006]. As a result of straining, the vast majority of colloids are retained near the entry surface of the porous media [*Bradford et al.*, 2002, 2003, 2004; *Auset and Keller*, 2006]. Straining in pore throats too small to pass would be expected to occur regardless of the presence versus the absence of an energy barrier between the colloid and the grain surface, and so would be expected to yield similar profiles of retained colloids in the presence and absence of an energy barrier, except possibly under conditions where colloid-colloid interaction forces (and hence, aggregation) differ under the two conditions. In contrast, wedging can occur for colloids much smaller than the pore throat, and so it is not an inevitable process, but is promoted by the translation of colloids along the energy barrier at the grain surfaces. The distinction between attachment and straining made in several recent papers is misleading, since straining (and wedging) constitute attachment in the presence of an energy barrier.

[20] By superimposing coordinates of colloids that were retained without attachment onto the unit collector, it was found that all such colloids were retained at zones of low fluid drag (Figure 8, right). A distinction between attachment and retention in flow stagnation zones is useful, since attachment can be considered for the most part irreversible in the absence of charge reversal [e.g., *Johnson and Tong*, 2006], whereas retention in flow stagnation zones is easily reversed, for example, by decreased ionic strength (reduction of secondary energy minimum depth), or by colloid diffusion out of secondary energy minima.

[21] The simulations indicated that wedging was predominant for colloid:collector ratios greater than about 0.005 (colloid sizes greater than 1 μm, where the collector radius was 255 μm), with this threshold ratio increasing with decreasing fluid velocity [*Johnson et al.*, 2007b]. Retention in fluid drag zones was demonstrated to be prevalent for colloid:collector ratios less than about 0.005, with this threshold decreasing with increasing fluid velocity [*Johnson et al.*, 2007b]. Both wedging and retention in flow stagnation zones were sensitive to colloid-surface interaction forces (energy barrier height and secondary energy minimum depth) [*Johnson et al.*, 2007b].

[22] In the simulations performed in the presence of an energy barrier, increased secondary energy minimum depth increased colloid translation along the collector surfaces and led to increased wedging and retention in flow stagnation zones. This result provides an explanation for the observed (in experiments) sensitivity of colloid deposition to colloid-interaction forces despite the presence of a formidable energy barrier. The colloids that are retained do not pass over the energy barrier, but rather translate along the surfaces in secondary-minimum association with the surface until they become wedged or retained in zones of relatively low fluid drag.

2.3. Implications

[23] The above simulations demonstrate that incorporation of grain-to-grain contacts in representative spherical porous media yields colloid deposition by mechanisms previously inferred from experiments (discussed above), but which are not accounted for in CFT. The simulated trends in η as a function of solution chemistry were similar to those observed in experiments [*Johnson et al.*, 2007b] indicating that incorporation of the correct mechanisms of retention in the presence of an energy barrier may yield good predictions of η in actual granular porous media. Although increased realism in the pore domain will lead to improved representation of processes [e.g., *Xu et al.*, 2006], the above simulations demonstrate that the pursuit of realistic pore domains must be balanced with examination of representative pore domains in order to understand the actual value of increased realism. If a representative pore domain is found to provide good predictions for simple granular media, then this model will be useful for its transferability and parsimony. Furthermore, the value of increasingly realistic pore domains can be assessed relative to the representative pore domain.

[24] The retention of secondary minimum-associated colloids in zones of flow stagnation in porous media offers a dynamic mechanism of colloid retention in the presence of an energy barrier that explains the observed release of colloids in response to perturbation with low ionic strength solution, and reentrainment of colloids even in the absence of ionic strength or velocity perturbations (e.g., via diffusion out of secondary energy minima) [e.g., *Cortis et al.*, 2006; *Johnson et al.*, 2007a]. These two mechanisms of deposition (wedging and retention in secondary energy minima) provide a new framework to evaluate the observed distributions of colloids as a function of distance from their sources, which deviate from the log linear expectations of classic filtration theory [*Albinger et al.*, 1994; *Baygents et al.*, 1998; *Simoni et al.*, 1998; *Schijven et al.*, 1999; *Bolster et al.*, 2000; *Redman et al.*, 2001; *Li et al.*, 2004; *Tufenkji and Elimelech*, 2004b; *Li and Johnson*, 2005; *Tong et al.*, 2005; *Johnson et al.*, 2005].

[25] The recent progress made in identifying the mechanisms responsible for colloid retention in porous media in the presence of an energy barrier leads to the possibility that correlation equations will soon be available for easy prediction of η in saturated porous media in the presence of an energy barrier. The variability of the profiles of retained colloids in the presence of an energy barrier promises to complicate prediction of transport distances; however, identification of mechanisms of retention, and development of correlation equations for η in the presence of an energy barrier, constitute important steps toward meeting this challenge.

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