A New Paradigm for Pathogen Transport and Deposition in Porous Media: The Role of Pore Structure and Colloid-Colloid Interactions

USDA-CSREES Research (2006-02541)

Existing colloidal filtration theory (CFT) fails to predict deposition when colloid-collector repulsion exists (i.e., presence of energy barriers);

Experiments and simulations indicate dominant colloid deposition mechanisms: wedging/straining, retention at secondary energy minima;

Underlying concept for retention in the presence of energy barriers involves translation of colloids across collector surfaces (outboard of energy barriers) until they become retained by above mechanisms;

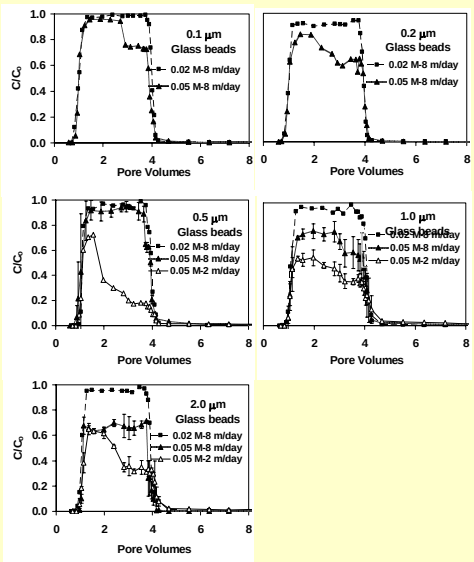
Near-surface accumulation of colloids renders colloid-colloid interactions important.

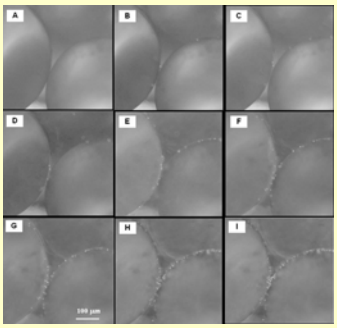
Objectives

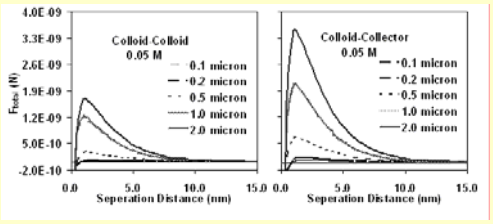
Develop models for pathogen & colloid transport and retention in porous media from pore scale to continuum scale and predict collector efficiency;

Investigate experimentally pathogen & colloid deposition in porous media across a spectrum of environmental conditions and scales.

Observation of colloid aggregation at grain-to-grain contacts





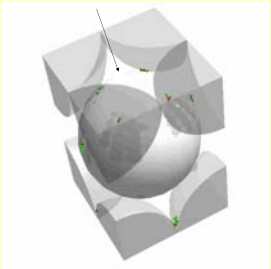


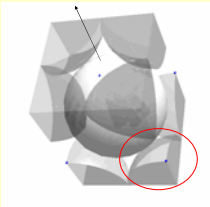
M. Tong, H. Ma, W.P. Johnson, Funneling of fluid into grain to grain contacts drives colloid-colloid aggregation in the presence of an energy barrier. *Environ. Sci. Technol.,* 2008, accepted.

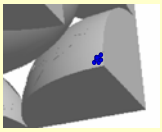
Mechanistic modeling: Lagrangian trajectory analysis

$$(m_p + m_{vm})\frac{du}{dt} = F_G + F_{VDW} + F_{EDL} + F_B + F_L + F_D$$

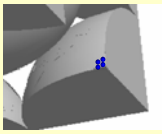
Representative unit cell with specified porosity:







without colloid-colloid interaction

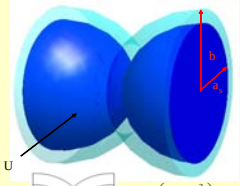


with colloid-colloid interaction

Red: wedged at grain-to-grain contacts
Green: attached to wedged colloids

Blue: retained at secondary energy minima

Representative unit cell with varied porosity:



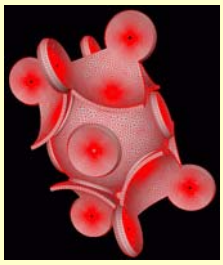
$$\theta = \frac{3\left(1 - \frac{a_s^2}{b^2}\right)}{3 - \frac{a_s^2}{b^2}}$$

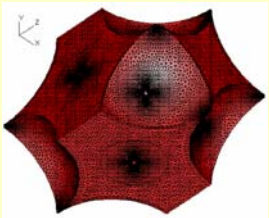
Computational mesh for quarter unit cell (due to symmetry) is built;

Fluid field within the mesh is solved using Star-CD;

Fluid field thus generated has been merged into particle trajectory model to simulate transport & retention.

Assemblage of unit cells (allow to investigate deposition along transport distance:



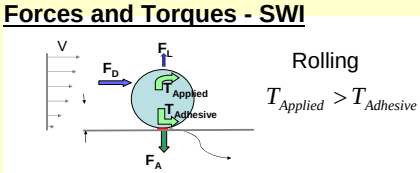


University of Utah and Pacific Northwest National Lab

THE UNIVERSITY OF UTAH

H. Ma[1], M. Tong[1], V. Gupta[1], W.P. Johnson[1]
S. Torkzaban[3], S.A. Bradford[2], H. Kim[3], S.L. Walker[3]
[1]University of Utah, Salt Lake City, UT
[2]US Salinity Laboratory, USDA-ARS, Riverside, CA
[3]University of California Riverside, CA

Forces and Torques - SWI

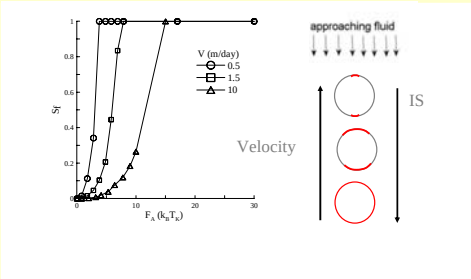


$$T_{Adhesive} = l_A F_A \qquad T_{Applied} = 1.4 r_c F_D$$

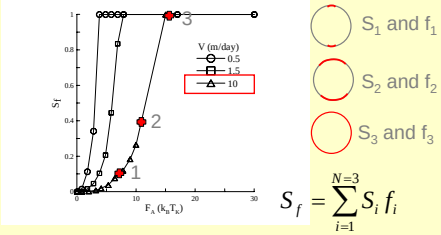
$$l_A = \left(\frac{F_A r_c}{4K}\right)^{1/3} \qquad F_D = 10.205 \pi \mu \tau_w r_c^2$$

(Johnson et al., 1971) (Goldman et al., 1967)

Surface area that is hydrodynamically and chemically "favorable" for retention

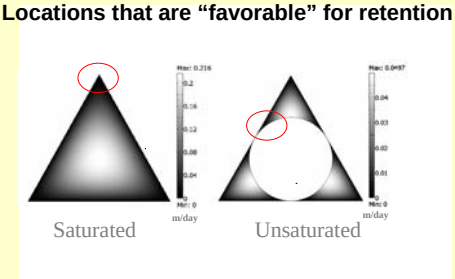


Solid or Colloid Chemical Heterogeneity



$$S_f = \sum_{i=1}^{N=3} S_i f_i$$

Locations that are "favorable" for retention



- Torkzaban, S., S. A. Bradford, and S. L. Walker. 2007. Resolving the coupled effects of hydrodynamics and DLVO forces on colloid attachment to porous media. Langmuir, 23, 9652-9660.
- **Bradford, S. A., and S. Torkzaban. 2008. Colloid transport and retention in unsaturated porous media: A review of interface, collector, and pore scale processes and models. Vadose Zone Journal, In Press.**

Continuum Scale Modeling

- Advective-dispersion equation

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial z^2} - v\frac{\partial C}{\partial z} - k_d C + \frac{\rho_b k_r}{\theta_w} S$$

$$\frac{\rho_b}{\theta_w}\frac{\partial S}{\partial t} = k_d C - \frac{\rho_b k_r}{\theta_w} S$$

- Filtration theory can be used when k_r=0.

$$k_d = \frac{3(1-\theta_w)}{2d_{50}}\eta\alpha v$$

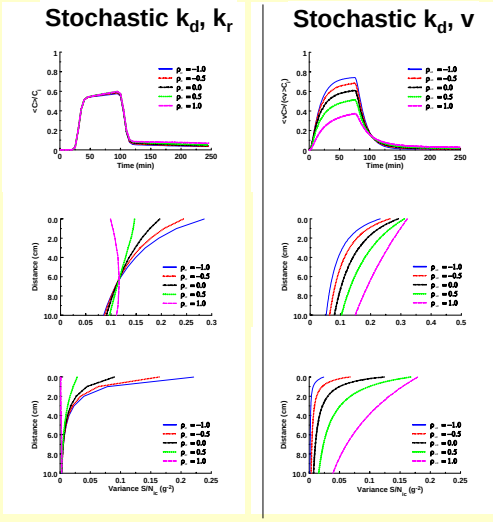
- 2 Stochastic parameters.

$$\langle C(x,t)\rangle = \int_0^\infty\int_0^\infty C(x,t;k_d,k_r)P(k_d,k_r)dk_d dk_r$$

$$\langle S(x,t)\rangle = \int_0^\infty\int_0^\infty S(x,t;k_d,k_r)P(k_d,k_r)dk_d dk_r$$

$$P(k_d,k_r) = \frac{1}{2\pi\sigma_d\sigma_r k_d k_r\sqrt{1-\rho_{dr}^2}}\exp\left[-\frac{Y_r^2 - 2\rho_{dr}Y_r Y_d + Y_d^2}{2(1-\rho_{dr}^2)}\right]$$

$$\rho_{dr} = \int_0^\infty\int_0^\infty Y_r Y_d P(k_d,k_r)dk_d dk_r$$



- **Bradford, S. A., and N. Toride. 2007. A stochastic model for colloid transport and deposition. Journal of Environmental Quality, 36, 1346-1356.**

Role of Growth Phase and Media on E. coli O157:H7

Research Objectives

- ✓ To investigate the sensitivity of bacterial surface properties in same nutrient condition (trypsic soy), but in different type of media (broth vs. agar) .
- ✓ To select optimum growth media for transport study.
- ✓ To investigate the influence of growth phase on bacterial surface properties.

Role of Growth Media

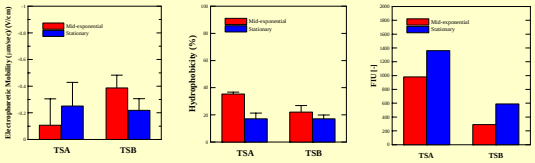


Figure 1. Variation of electrophoretic mobility, hydrophobicity, and fluorescence intensity of *E. coli* O157:H7 prepared in tryptic soy broth (TSB) and agar (TSA). Red and blue bars represent the cells in mid-exponential (2.5 h) and stationary phases (18 h), respectively. Experiments were conducted [illegible]).

Role of Growth Phase

Table 1. Surface characteristics of *E. coli* O157:H7 in different growth phase (mid-exponential vs. stationary). Experiments were conducted at pH 9.

Growth Phase	Ionic Strength (mM)	Live (%)[a]	Radius (μm)[b]	Aspect Ratio[c]	Mobility (μm/sec)/(V/cm)	MATH (%)[d]	Acidity (meq/10[8] cell)[e]	Surface Charge density (μC/Cm²)[f]
Mid-exponential (2.5h)	1	92.7 ± 0.8	0.47±0.06 (n=136)	2.9±1.5	-0.18 ± 0.28	26.7 ± 9.3	8.48×10^{-5} ± 3.75×10^{-5}	2840 ± 1260
	25	99.5 ± 0.8			-0.31 ± 0.20	34.3 ± 5.9		
Stationary (18h)	1	88.0 ± 4.2[g]	0.47±0.06 (n=212)	1.8±0.7	-0.30 ± 0.13	32.3 ± 6.0	1.67×10^{-5} ± 3.34×10^{-6}	574 ± 115
	25	90.9 ± 0.3[g]			-0.13 ± 0.22	45.4 ± 4.0		

[a] Percent of cell population determined to be viable based on the Live/Dead BacLight® kit at 10 mM electrolyte. [b] Value for equivalent spherical radius calculated from experimentally measured length and breadth of individual cells. [c] Aspect ratio is defined as the ratio of major and minor axis. [d] Microbial adhesion to hydrocarbon (MATH) indicates the relative hydrophobicity of the cell as the percent of cells partitioned in dodecane versus electrolyte. [e] Acidity determined from the amount of NaOH consumed during a titration between pH 4 and 10 for cells suspended in 10 mM electrolyte. [f] Indicates the density of charged functional groups across the cell surface. Value determined from the experimentally measured acidity, and accounting for the exposed surface area of the cells (calculated for a spherical cell) and Faraday's constant of 96,485 C/mol. [g] Values measured at pH 9.2.

- ✓ Regardless of growth media and phase, electrokinetic mobility was near neutral. (Figure 1 and Table 1).
- ✓ There was no significant different between the cells grown in the two different medias (TSA and TSB) in terms of electrokinetic and hydrophobic properties; however, the cells grown in TSA showed greater fluorescence intensity than those in TSB (Figure 1).
- ✓ The cells in mid-exponential stage were relatively more hydrophilic than those in stationary phase, indicating the cells have more functional groups on the surface of membrane and extracellular polymeric substances (EPS) as shown in surface charge density (Table 1).

Go
UC Riverside
University Advancement
Search for:

Role of Solution Chemistry [illegible] and pH)

Research Objectives

- ✓ To investigate the influence of solution chemistry on *E. coli* O157:H7 transport through column studies and complimentary cell characterization
- ✓ To determine the impact of solution chemistry on bacterial surface polymer (EPS) presence and the subsequent influence of EPS in transport.

Characterization Study

Table 2. Viability and size of *E. coli* O157:H7. Standard deviation is reported in parentheses.

IS (mM)	Live (%)			Radius (μm)			Aspect ratio[c]		
	pH								
	5.8	8.4	9.2	5.8	8.4	9.2	5.8	8.4	9.2
1	92.8 (2.1)	89.5 (0.9)	88.0 (4.2)	0.39 (0.06)	0.36 (0.05)	0.35 (0.06)	3.7 (2.2)	3.3 (1.9)	3.1 (2.0)
10	92.6 (1.4)	87.3 (0.5)	88.0 (1.1)	0.34 (0.05)	0.36 (0.05)	0.37 (0.05)	2.9 (1.9)	3.4 (2.0)	3.1 (1.9)
25	92.8 (1.1)	90.5 (2.0)	90.9 (0.3)	0.38 (0.05)	0.36 (0.04)	0.37 (0.04)	3.7 (1.9)	3.6 (2.0)	3.1 (1.9)
100	95.9 (2.4)	94.1 (1.4)	93.2 (1.7)	0.37 (0.04)	0.36 (0.05)	0.34 (0.04)	2.9 (1.6)	3.0 (1.6)	2.9 (1.6)

Table 3. *E. coli* O157:H7 EPS Content and Composition. (Ethanol extraction method used for EPS)

EPS[a] (mg/10[10] cell)	Sugar (μg/10[10] cell)[b]	Protein (μg/10[10] cell)[c]	Sugar / Protein ratio
79.1 ± 31.2	175 ± 16	13961 ± 1271	1.3×10^{-2} ± 4.6×10^{-4}

[a] 10 mM KCl was used as background solution. For the cells which were 91.5±1.4% alive. [b] Based on Xanthan gum as the standard. [c] Based on Bovine Serum Albumin(BSA) as the standard.

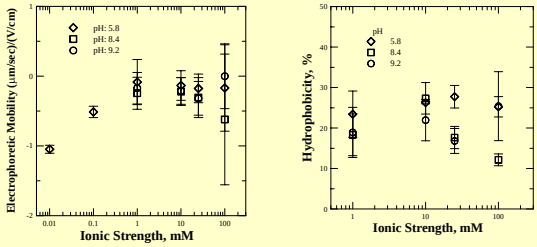


Figure 2. Electrophoretic mobility and hydrophobicity of *E. coli* O157:H7 as a function of IS at different pH.

Transport Study

➢ Experimental conditions

- Sand type: pure quartz (d_{avg}: 275 μm)
- Superficial velocity: 0.1 cm/min
- Porosity: 0.44 – 0.47
- 1 pore volume: ~ 47 min
- Column length: 10 cm
- Cell injection concentration: $5*10^8$ cell/mL
- Temperature : 25 °C
- pH : 8.4 and 9.2 (carbonate buffer)
- Ionic strength (KCl) : 1, 10, and 100 mM

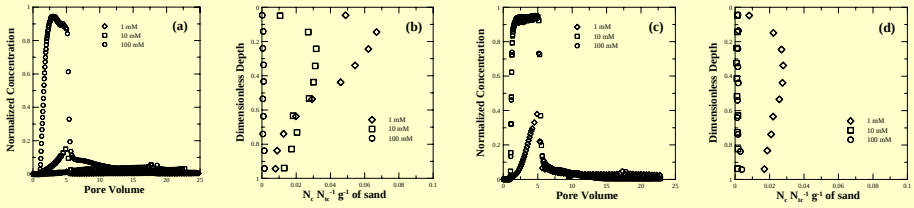


Figure 3. Representative breakthrough curves and deposition profiles for *E. coli* O157:H7 as a function of ionic strength and pH. Data obtained at conditions of pH of 8.4 (a and b) and 9.2 (c and d).

- ✓ Bacterial transport behavior was very sensitive to solution chemistry (IS and pH) (Figure 3), which cannot be explained by macroscopic surface properties (such as viability, size, shape, electrophoretic mobility, and hydrophobicity) (Table 2).
- ✓ It is hypothesized that pH sensitivity may be due to protonated state of surface functional groups of the cells. Additionally, compositional change of bacterial surface polymer (i.e., folding and unfolding) may occur with changing IS since the cells show substantially low sugar/protein ratio of EPS.

Ongoing Research

I. Role of Surface Macromolecules

Research Objectives

- ✓ To investigate the role of surface macromolecules on cell surface properties and transport behavior of *E. coli* O157:H7.
- ✓ To be conducted through the removal of polymers with proteinase K and the study of cell properties/transport with and without surface polymers.

Preliminary Experimental Results

Table 4. Viability and size of *E. coli* O157:H7 before and after proteinase K-treatment (pH 8.4)

IS (mM)	Live (%)		Radius (μm)		Aspect ratio	
	before	after	before	after	before	after
1	89.5 (0.9)	90.6 (1.9)	0.36 (0.05)	0.33 (0.04)	3.3 (1.9)	3.5 (1.5)
10	87.3 (0.5)	92.9 (1.4)	0.36 (0.05)	0.33 (0.04)	3.4 (2.0)	2.4 (1.3)
25	90.5 (2.0)	93.5 (2.8)	0.36 (0.04)	0.34 (0.03)	3.6 (2.0)	2.5 (1.5)
100	94.1 (1.4)	95.8 (1.9)	0.36 (0.05)	0.34 (0.04)	3.0 (1.6)	2.6 (1.6)

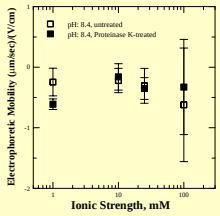


Figure 4. Electrophoretic mobility of *E. coli* O157:H7 before and after proteinase K-treatment.

- ✓ Results shown in Table 4 suggest that the cells were intact after enzyme (proteinase K) treatment.
- ✓ There was no significant difference in cell mobility before and after proteinase K-treatment. The results suggest that the potential contributed by surface polymers are relatively negligible as compared to that of the cell outer membrane.
- ✓ In future experiments, the relative composition of surface functional groups of the cells will be examined using FT-IR. Other surface properties (hydrophobicity, EPS, and surface charge density) will be evaluated, followed by column studies.

II. Role of Geometry (Aspect Ratio and Size)

Research Objectives

- ✓ To prepare ellipsoidal colloids of varying aspect ratio (1, 3, and 5) to study as bacterial surrogates.
- ✓ To investigate the role of particle geometry on transport and adhesion processes.

Preliminary Experimental Results

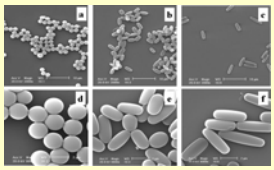


Figure 5. SEM images of spherical and ellipsoidal colloids; original colloids (a and d), aspect ratio of 3 (b and e), and aspect ratio of 5 (c and f).

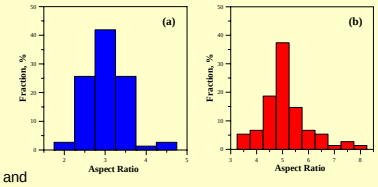


Figure 6. Aspect ratio distribution of ellipsoidal colloids. (a) 3 and (b) 5.

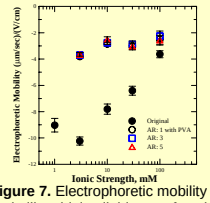


Figure 7. Electrophoretic mobility of spherical and ellipsoidal colloids as a function of ionic strength.

- ✓ A modified film stretching method (FSA) was developed and the ellipsoidal colloids prepared were shown in Figure 5. Furthermore, the ellipsoidal colloids exhibit narrow aspect ratio distribution (Figure 6).
- ✓ There was no different in electrophoretic mobility of colloids as a function of aspect ratio although the magnitude was smaller than original colloids (Figure 7).
- ✓ As future experiments, particle aggregation, hydrophobicity, and surface charge density will be evaluated, followed by micro-model and column studies.

UNIVERSITY OF CALIFORNIA UC RIVERSIDE