Escherichia coli O157:H7 Transport in Saturated Porous Media: Role of Solution Chemistry and Surface Macromolecules

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The transport and deposition behavior of Escherichia coli O157:H7 was investigated in saturated packed-bed columns and micromodel systems over a range of ionic strength (IS) (1, 10, and 100 mM) and pH (5.8, 8.4, and 9.2) conditions. At a given IS, elevated solution pH resulted in decreased deposition as a result of the increase in the measured zeta potential of the quartz sand. This deposition trend was consistent with predictions from classic Derjaguin—Landau—Verwey—Overbeek (DLVO) theory. Conversely, the E. coli O157:H7 deposition was inversely proportional to IS (1–100 mM) at high pH conditions (8.4 and 9.2), whereas no effect of IS was observed at pH 5.8. This deposition trend was not consistent with DLVO theory, but could be explained by pH-associated electrostatic stabilization. This phenomenon is driven by the pH-dependent protonated state of functional groups on E. coli O157:H7 surface macromolecules and the corresponding conformational state of the bacterial polymers. Results from this study demonstrate that retention of E. coli O157:H7 cells in porous media is a complex process that depends on the solution chemistry, cell–cell interactions, and pore structure. The findings in this study also imply that previous work conducted at lower pH and IS conditions may underestimate the rate of cell deposition due to conformational changes of polymers that are induced by charge neutralization and ion bridging mechanisms (e.g., refs 12, 15, 22). However, it was observed that the extent of deposition decreased at a high salt concentration (25, 27), implying steric stabilization was induced by IS regardless of the ions’ valence. Additionally, it has been reported that the relative amount of exposed surface functional groups and the corresponding localized charge heterogeneity can influence bacterial deposition behavior (28), suggesting that bacterial acid–base properties, linked to the aquatic condition, may also be important in conformational change of polymers. However, the effect of pH and/or IS on a cell adhesion has typically been assumed to be less significant than ion valence (15, 16, 24, 25). The aim of this study is to investigate the coupled role of solution chemistry and cell polymers on microbe adhesion (15, 16, 24, 25). Studies have revealed that the presence of a divalent cation (Ca2+) can substantially enhance the rate of cell deposition due to conformational changes of polymers that are induced by charge neutralization and ion bridging mechanisms (e.g., refs 25 and 26). However, it was observed that the extent of deposition decreased at a high salt concentration (25, 27), implying steric stabilization was induced by IS regardless of the ions’ valence. Additionally, it has been reported that the relative amount of exposed surface functional groups and the corresponding localized charge heterogeneity can influence bacterial deposition behavior (28), suggesting that bacterial acid–base properties, linked to the aquatic condition, may also be important in conformational change of polymers. However, the effect of pH and/or IS on a cell adhesion has typically been assumed to be less significant than ion valence (15, 16, 24, 25). The aim of this study is to investigate the coupled role of IS and pH on E. coli O157:H7 transport and deposition in porous media. For this purpose, transport experiments were carried out in saturated packed-bed columns over a range of pH (5.8, 8.4, and 9.2) and IS (1, 10, and 100 mM) values were chosen as representative experimental conditions. A carbonate buffer (a mixture of 9 × 10−4 M NaHCO3 and 1 × 10−4 M Na2CO3) was utilized to increase the pH of the

Introduction

Groundwater contamination, caused by the migration of pathogenic bacteria through soil, leads to almost half of the outbreaks of waterborne diseases each year in the United States (U.S.) (1). Among numerous pathogens, Escherichia coli O157:H7, a gram-negative bacterium, is reported to be responsible for severe diarrhea (hemorrhagic colitis) and hemolytic-uremic syndrome (2), leading to approximately 75,000 infections and 61 deaths per year in the U.S. (3). It has been reported that cattle are an important host for this virulent E. coli strain, and that most human infections originate from contact with contaminated water, meat, vegetables, etc. (4). Accordingly, the highly pathogenic nature of this organism demands a clear understanding of its transport and fate in subsurface environments in order to assess and mitigate the potential risk to public health. Although much research has been conducted to investigate physical and chemical factors influencing bacterial transport in well-defined systems (e.g., refs 5 and 6), considerably less is known about complex biological factors that also influence bacteria transport.

Classic Derjaguin—Landau—Verwey—Overbeek (DLVO) (7, 8) and extended DLVO (XDLVO) theories (9) have been extensively used to interpret the deposition mechanism of colloids and bacteria (e.g., refs 10 and 11). Recent research has indicated that the classic DLVO and XDLVO theories are not always valid to predict microbial interactions due to the presence of complex polymer layers extending into the liquid medium (e.g., refs 12–14). Depending on the solution ionic strength (IS), the presence of large macromolecules or other polymeric substances may influence the extent of microbe adhesion due to electrostatic interactions (e.g., refs 12–14) which are not considered in DLVO nor XDLVO theories (14, 15).

Several studies have investigated the role of microbial surface macromolecules in cell adhesion under controlled conditions via atomic force microscopic methods (e.g., refs 16–18), batch adhesion tests (e.g., refs 19–21), radial stagnation point flow systems (e.g., refs 12, 15, 22), and packed-bed columns (e.g., refs 19, 21, 23). These studies indicate that surface macromolecules may enhance (12, 20, 23) or hinder (15, 20–22) adhesion to different types of abiotic surfaces, and that surface macromolecules can play a significant role on microbial deposition.

Some work has been conducted to investigate the coupled role of solution chemistry and cell polymers on microbe adhesion (15, 16, 24, 25). Studies have revealed that the presence of a divalent cation (Ca2+) can substantially enhance the rate of cell deposition due to conformational changes of polymers that are induced by charge neutralization and ion bridging mechanisms (e.g., refs 25 and 26). However, it was observed that the extent of deposition decreased at a high salt concentration (25, 27), implying steric stabilization was induced by IS regardless of the ions’ valence. Additionally, it has been reported that the relative amount of exposed surface functional groups and the corresponding localized charge heterogeneity can influence bacterial deposition behavior (28), suggesting that bacterial acid–base properties, linked to the aquatic condition, may also be important in conformational change of polymers. However, the effect of pH and/or IS on a cell adhesion has typically been assumed to be less significant than ion valence (15, 16, 24, 25). The aim of this study is to investigate the coupled role of IS and pH on E. coli O157:H7 transport and deposition in porous media. For this purpose, transport experiments were carried out in saturated packed-bed columns over a range of pH (5.8, 8.4, and 9.2) and IS (1, 10, and 100 mM) representative of groundwater conditions.

Materials and Methods

Bacterium and Solution Chemistry. E. coli O157:H7/pGFP strain 72 (5) was used in transport experiments conducted over a wide range of pH and IS conditions found in groundwater (29, 30). Information about the bacterial growth and preparation is found in the Supporting Information (SI). Three pH (5.8, 8.4, and 9.2) and IS (1, 10, and 100 mM) values were chosen as representative experimental conditions. A carbonate buffer (a mixture of 9 × 10−4 M NaHCO3 and 1 × 10−4 M Na2CO3) (31) was utilized to increase the pH of the
solution, except for an unadjusted electrolyte solution at pH 5.8. As needed, the final pH was adjusted using either 0.1 N HCl or NaOH solution (changes in the IS due to pH adjustment were negligible). The IS of the solutions was adjusted using KCl as the background electrolyte. The solution pH was measured (φ 350 pH/Temp/mV Meter, Beckman Coulter, Fullerton, CA) and the IS was calculated. All chemicals were reagent grade (Fisher Scientific).

**Cell Characterization.** Cell viability, size, shape, electrophoretic mobility, and hydrophobicity were evaluated as a function of pH and IS at 0 h and compared to similar data measured after 3 h of cell acclimation in the desired electrolyte. The results are presented in Table S1, Figures S1 and S2. This acclimation time was selected based on the length of the time of the column experiments to determine whether there were any physiological changes in the cells.

The experimental details for the cell characterization are provided in the SI. The viability results show that the percentage of viable cells at 0 and 3 h was greater than or equal to 87 and 80%, respectively, with no statistical difference at given solution conditions (P > 0.01) (SI Table S1). The average effective cell diameter was approximately 0.72 ± 0.02 μm over the pH and IS ranges investigated for 0 and 3 h acclimated cells (SI Table S1). Statistical differences were observed at certain conditions (P < 0.01); however, no strong relationship between the change in cell size and the observed transport behavior was found at given conditions. The average aspect ratio also ranged from 2.8 ± 1.6 (minimum) to 3.7 ± 2.2 (maximum) with no significant difference regardless of solution conditions and acclimation time (P > 0.01) (SI Table S1). Additionally, the cells showed similar mobility values and they were relatively hydrophilic over the IS and pH ranges investigated regardless of acclimation time (SI Figures S1 and S2). Hence, cell die-off and changes in cell size, shape, mobility, or hydrophobicity are not significant parameters.

**Porous Media.** Ultrapure quartz sand (Iota quartz, Unimin Corp., NC) was utilized as the porous medium for the transport experiments. Sand preparation procedures are discussed in the SI. The average sand diameter (d₅₀) was approximately 275 μm. The gravimetrically measured bed porosity was determined to be ca. 0.47. In order to characterize the electrokinetic properties of the porous medium, the streaming potential of the quartz sand was measured as a function of pH and IS using an Electro Kinetic Analyzer (EKA) (Anton Paar GmbH, Graz, Austria) equipped with a cylindrical cell (32) and this information is presented in Table 1 and SI Figure S3.

**Column Experiments.** An adjustable chromatography column (Omnisift, Boonton, NJ) with a 1.5 cm inner diameter and a length of 10 cm was used for transport experiments. The superficial velocity was maintained at 0.1 cm min⁻¹ for all experiments. A pulse of bacteria suspension, at a concentration of 5 × 10⁹ cells mL⁻¹, was injected into the column for four pore volumes (PV), followed by a bacteria free electrolyte solution (>20 PV). The profile for E. coli O157:H7 cells retained in the column was obtained after recovery of the breakthrough curve (BTC) (5). The effluent and retained cell concentrations were measured using a spectrophotometer (SP-890, Barnstead International, Dubuque, IA) and fluorometer (FM109545, Barnstead International, Dubuque, IA), respectively. The total percentage of recovered cells (MEff) was calculated from percentages in the effluent (Meff) and within the sand (Msand). Additional information about the column experiments is provided in the SI.

**Micromodel Experiments.** In order to visually examine the deposition of E. coli O157:H7 cells in the quartz sand, a few representative transport experiments were conducted in a specially designed micromodel. The detailed information regarding the micromodel system can be found elsewhere (5,33) and in the SI. The E. coli O157:H7 suspension was injected to the chamber at an approach (Darcy) velocity of 0.1 cm min⁻¹. During and after micromodel experiments, the deposition behavior of the fluorescent E. coli O157:H7 cells was microscopically examined.

**Numerical Modeling.** Some of the cell BTCs were simulated using the HYDRUS 1D computer software (34) in order to better interpret mechanisms of cell retention and transport. The cell transport equations that were solved are provided in the SI and the fitted model parameters are summarized in SI Table S2.

**Results and Discussion**

**Observed Bacterial Transport and Deposition Behavior.**

(a) **BTCs.** Figure 1 shows the BTCs for E. coli O157:H7 cells as a function of IS at pH 5.8, 8.4, and 9.2. Almost no difference in effluent cell concentration with IS was observed at pH 5.8 (i.e., in Figure 1a 0.9 and 1.9% of injected cells eluted for 1 and 100 mM, respectively). In contrast, at the higher pH conditions (i.e., 8.4 and 9.2) cell retention was inversely proportional to IS (Figures 1b and c). Comparison of panels a–c of Figure 1 reveals that the amount of E. coli O157:H7 cells eluting the column increased with pH for a given IS. Mass balance results presented in Table 1 indicate that for a pH of 5.8, 8.4, and 9.2 the cell breakthrough was observed to be 0.9, 2.7, and 32.9% when the IS was 1 mM, and 1.9, 100.3, and 102.9% when the IS was 100 mM, respectively. Similarly, 21.4 and 103.7% of the injected cells came out of the column when the IS was 10 mM and the pH was 8.4 and 9.2, respectively.

(b) **Deposition Profiles.** Figure 2 presents the corresponding deposition profiles of E. coli O157:H7 cells retained in the column as a function of dimensionless depth. Observed

**TABLE 1.** Electrokinetic Properties of Quartz and E. coli O157:H7 Cells as well as Column Mass Balance Information

<table>
<thead>
<tr>
<th>pH</th>
<th>ionic strength (mM)</th>
<th>zeta potential of quartz (mV)</th>
<th>electrophoretic mobility of cells (μm V s⁻¹ cm⁻¹)</th>
<th>MEff (%)</th>
<th>MSand (%)</th>
<th>Mtotal (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.8</td>
<td>1</td>
<td>55.1</td>
<td>0.073</td>
<td>20.4</td>
<td>38.9</td>
<td>59.3</td>
</tr>
<tr>
<td>8.4</td>
<td>100</td>
<td>-16.0</td>
<td>-0.112</td>
<td>1.9</td>
<td>57.4</td>
<td>59.3</td>
</tr>
<tr>
<td>9.2</td>
<td>1</td>
<td>-63.8</td>
<td>-0.168</td>
<td>32.9</td>
<td>54</td>
<td>86.9</td>
</tr>
</tbody>
</table>

*Here, MEff, MSand, and Mtotal refer to the effluent, retained, and total percentage of E. coli O157:H7 cells recovered from column experiments, respectively. All values represent the relative percentage of the cells by number. MEff was determined by integrating beneath the effluent concentration profile in Figure 1. MSand was determined from the experimentally determined deposition profile in Figure 2. Average value obtained from SI Figure S3. Average value obtained from SI Figure S1a. Value including the amount eluted by first and second flow interruption (each 0.4% of cells eluted), and DI water flush (18.7% eluted).
trends with respect to the cell retention as a function of pH and IS were consistent with the BTC data discussed above. The amount of cell deposition did not monotonically decrease with distance from the top of the column as predicted by irreversible attachment (35) or straining (36) models. Specifically, a nonmonotonic decrease in cell concentration with depth was observed. In systems with the greatest amount of cell retention a sharp peak in concentration occurred around a dimensionless depth of 0.1 (i.e., 100 mM at pH 5.8 and 1 mM at pH 8.4). Conversely, in systems with lower amounts of cell retention (i.e., 10 mM at pH 8.4, and 1 mM at pH 9.2) these peaks tended to be broader and flatter.

(c) Micromodel Experiments. Additional transport experiments were conducted using a specially designed micromodel to complement column results with the *E. coli* O157:H7 cells. SI Figure S4 show the images captured at different experimental conditions (1 mM at pH 5.8 and pH 9.2, and 100 mM at pH 9.2). As shown in SI Figure S4, the results obtained from micromodel experiments were qualitatively consistent with those from the column study. Virtually no attachment was observed at 100 mM and pH 9.2 while a substantial amount of bacteria attached to quartz surfaces at 1 mM and pH 5.8, thus, indicating that the conditions tested were relatively unfavorable and favorable for the interaction between the cells and the quartz surfaces, respectively. Additionally, it was observed that a relatively small amount of bacteria were retained in quartz media at 1 mM and pH 9.2 compared with the results at 1 mM and pH 5.8. This was identified by the notably lower levels of cells visualized in the field of view.

**Interpretation of *E. coli* O157:H7 Transport and Retention.** The observed transport and retention behavior for *E. coli* O157:H7 shown in Figures 1, 2, and SI Figure S4 indicate a complex dependence on solution chemistry (pH and IS). Previous research also indicates that *E. coli* O157:H7 retention in porous media will be strongly coupled to the pore space geometry and will depend on cell–cell interactions (5). Each
of these factors will be considered below to provide a meaningful interpretation to the collected data.

(a) Solution Chemistry. Table 1 provides measured zeta potentials for the quartz grains as a function of pH and IS. The zeta potential of the grains was observed to become more negative with decreasing IS and increasing pH due to expansion of the diffuse double layer and dissociation of proton groups, respectively ([39]). In contrast, the electrophoretic mobility of the E. coli O157:H7 cells was relatively insensitive to changes (P > 0.01) in pH and IS over these same ranges, with a value of approximately -0.15 µm V⁻¹ cm⁻¹ (Table 1). As mentioned in the introduction, quantitative application of DLVO theory to predict cell-sand interactions is problematic because of the presence of surface macro-molecules and the fact that the cells are rod shaped. Qualitatively, DLVO theory predicts increasing cell-sand interactions with decreasing pH and increasing IS. Consistent with DLVO theory, Figures 1 and 2 indicate that cell retention at a given IS increased with decreasing pH. In this case, the zeta potential of the sand decreased with decreasing pH (Table 1) and consequently lowered the energy barrier (electrostatic repulsion) to deposition in the primary minimum (Φ₁₉₅₅ₕ) and/or enhanced the depth of the secondary minima (Φ₂₉₅ₕ) that acted on the cells near the grain surfaces.

Mass balance results presented in Table 1 suggest that approximately 40% of the cells were irreversibly retained in a Φ₁₉₅ₕ type interaction at pH 5.8 for both 1 and 100 mM conditions. Figure 1a indicates that approximately 20% of the bacteria were eluted from the column by switching to deionized (DI) water, indicating that only 20% of the cells were reversibly retained in a Φ₂₉₅ₕ type interaction at pH 5.8. Hence, irreversible and reversible attachment played a dominant role in cell retention at pH 5.8 when the IS was 1 and 100 mM. Conversely, retention of the remaining 40% of...
the cells is still unexplained under these conditions, and this finding implies that other cell retention mechanisms were also involved. For higher pH conditions (pH 8.4 and 9.2) mass balance information presented in Table 1 indicates that irreversible cell retention was less likely (relatively good mass balance) and that unfavorable attachment conditions existed. In these cases, reversible attachment of cells is still possible but is expected to be less significant than that observed for the pH 5.8 and 1 mM system (19%).

Figures 1 and 2 indicate that cell retention at pH 8.4 and 9.2 decreased with increasing IS. This trend was not anticipated by classic DLVO theory, which most previous transport studies have reported (e.g., refs 37 and 38). According to DLVO theory, particle deposition in porous media increases with increase in electrostatic force between particles and grain surfaces (39). Our results show the exact opposite trend at pH 8.4 and 9.2, indicating that non-DLVO acid–base interactions were likely involved under these conditions. Possible explanations for these observations include hydrophobic interactions, an anion valence effect, and steric stabilization. Information presented in the SI indicates that hydrophobic interactions and anion valence effect are unlikely to explain this behavior. Below we discuss the potential influence of steric stabilization.

Recent studies indicate that electrostatic force can play a significant role on microbial adhesion to abiotic surfaces due to conformational changes of the polyelectrolyte layers with various functional groups (e.g., refs 15, 22). As discussed by Kuznar and Elimelich (15, 22), the presence of surface macromolecules on Cryptosporidium oocyst walls can considerably hinder the deposition rate on a quartz surface. They attributed this phenomenon to electrostatic repulsion; however, no direct effect of salt concentration on steric stabilization was reported this phenomenon to electrostatic repulsion; however, no direct effect of salt concentration on steric stabilization was observed (15, 22). Li and McLandsborough (27), on the other hand, observed increasing adhesion of E. coli O157:H7 (EC01) cells to beef muscle over a range of IS (i.e., 1.5–150 mM PBS). Chen and Walker (25) observed a similar trend in adhesion experiments using Halomonas pacifica g in the presence of CaCl2 at IS > 300 mM.

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The opposite trend (decrease in effluent concentration with continued addition of cells) was observed for the pH 8.4 and IS 100 mM system and indicates the potential of cell–cell interactions (49, 50) in the porous media. Based on the cell BTCs shown in Figure 1, it is logical to infer that steric stabilization of the surface macromolecules would decrease the strength of cell–cell interactions with increasing pH and IS. Comparison of the cell BTCs at pH 8.4 and 9.2 and IS of 100 mM supports this hypothesis. Specifically, a ripening (i.e., cell–cell interaction) was observed for the BTC at the lower pH condition (pH 8.2), but not at the higher pH condition (pH 9.2). The different degree of electrosteric stabilization with pH could be explained by changes in the separation distance of polymeric layers on the surfaces of cells; i.e., a larger separation at higher pH occurs due to the conformational changes of macromolecules by the changing extent of deprotonation of functional groups (9, 51).

Numerical modeling of the cell BTCs at pH 8.4 and 9.2 was conducted to gain additional insight on the mechanisms of cell retention under unfavorable attachment conditions. Fitted model parameters are shown in SI Table S2. The model provided a reasonable description of the BTCs (\( r^2 > 0.744 \)). At a given IS, the deposition coefficient decreased with pH. This observation is consistent with DLVO predictions and measured changes in the sand zeta potential. Conversely, the deposition coefficient decreased and the release coefficient tended to increase with IS (at a given pH). This trend is attributed to pH-associated electrosteric stabilization of the cell surface macromolecules as discussed above. Fitted values of the maximum solid phase concentration of retained cells demonstrated that only a small fraction of the quartz surface area contributed to cell retention (i.e., 2–23%), and that this fraction decreased with IS and pH. This trend is unlikely due to chemical heterogeneity since thoroughly cleaned ultrapure quartz was used in this study. Instead, this observation supports the potential importance of pore structure in cell retention. A number of studies have recently demonstrated that retention in low velocity regions that occur near grain–grain junctions and/or in small pore spaces may significantly contribute to colloidal/microbial deposition in porous media under unfavorable attachment conditions (e.g., refs 5 and 52–54). Furthermore, the fraction of the solid surface area contributing to cell retention in these locations has been demonstrated to increase with the adhesive force (55, 56); e.g., the solution chemistry. As the small fraction of the surface area that contributes to cell retention fills, the deposition rate decreases with time and theBTCs shown in Figure 1 therefore exhibit a time-dependent shape. When cell retention is confined to small fractions of the surface area the cells may interact with each other within these regions and aggregate. For example, Bradford et al. (5) observed nonmonotonic deposition profiles for E. coli O157:H7 and cell aggregates in aquifer sand under unfavorable attachment conditions. These authors attributed the nonmonotonic deposition profiles to the release and re-entrainment of the cells, which were initially deposited and aggregated near grain–grain contacts.

**Implications.** In this study, we demonstrated that pH-associated electrosteric stabilization can explain the observed trend of decreasing deposition of E. coli O157:H7 cells to
quartz sand with increasing IS (1–100 mM) under higher pH conditions (i.e., 8.4 and 9.2). One important implication of this mechanism is that previous work may have underestimated E. coli O157:H7 travel distances at higher salt and pH conditions in typical groundwater environments (i.e., pH 6–9 and IS 1–50 mM). This study also provides insight into the role of solution chemistry on E. coli O157:H7 fate and the impact of pH-induced electrosteric stabilization. It should be mentioned that the E. coli O157:H7 strain used in this study is not representative for all other pathogenic E. coli strains, and therefore the results may vary between cell strains as observed from other studies (e.g., refs 57–60), indicating that species variation could be important. Nevertheless, the findings in this work demonstrate some potentially important factors that are typically neglected in classic microbial interaction studies. This finding can help guide the design and efficient development of processes for the removal of the E. coli O157:H7 cells during soil passage such as sand/riverbank filtration, and infiltration trenches and basins.

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Supporting Information Available
Additional methods (experiments and numerical modeling), discussion (hydrophobic interactions and anion valence effect), and data pertaining to the viability, size, aspect ratio, electrophoretic mobility, and hydrophobicity of E. coli O157:H7 cells, zeta potentials of quartz sand as a function of pH (effect), and data pertaining to the viability, size, aspect ratio, electrophoretic mobility, and hydrophobicity of E. coli O157:H7 cells during soil passage and basins.

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Environmental Science, University of California—Riverside, CA.


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