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Unraveling the complexities of the velocity dependency of *E. coli* retention and release parameters in saturated porous media



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HIGHLIGHTS

GRAPHICAL ABSTRACT

- The velocity dependency of cell retention and release parameters were investigated under different IS conditions.
- The sticking efficiency, retention capacity, and irreversible cell retention increased with ionic strength and decreasing water velocity.
- The probability for cells to diffuse over the energy barrier depends on the residence time on heterogeneous surfaces.
- The adhesive strength increased with the residence time and decreasing water velocity.
- Torque balance consideration explained the negligible cell removal with large increases in velocity, and large amounts of cell release following sand excavation.

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ABSTRACT

Escherichia coli transport and release experiments were conducted to investigate the pore-water velocity (v) dependency of the sticking efficiency (α), the fraction of the solid surface area that contributed to retention (S_f), the percentage of injected cells that were irreversibly retained (M_{irr}), and cell release under different (10–300 mM) ionic strength (IS) conditions. Values of α , S_f , and M_{irr} increased with increasing IS and decreasing v, but the dependency on v was greatest at intermediate IS (30 and 50 mM). Following the retention phase, successive increases in v up to 100 or 150 m day⁻¹ and flow interruption of 24 h produced negligible amounts of cell release. However, excavation of the sand from the columns in excess electrolyte solution resulted in the release of >80% of the retained bacteria. These observations were explained by: (i) extended interaction energy calculations on a heterogeneous sand collector; (ii) an increase in adhesive strength with the residence time; and (iii) torque balance consideration on rough surfaces. In particular, α , S_f , and M_{irr} also increased with IS due to lower energy barriers and stronger primary minima. The values of α , S_f , and M_{irr} also increased with decreasing v because the

Abbreviations: IS, ionic strength; BTC, breakthrough curves.

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Retention Surface roughness adhesive strength increased with the residence time (e.g., an increased probability to diffuse over the energy barrier) and lower hydrodynamic forces diminished cell removal. The controlling influence of lever arms at microscopic roughness locations and grain-grain contacts were used to explain negligible cell removal with large increases in *v* and large amounts of cell recovery following sand excavation. Results reveal the underlying causes (interaction energy, torque balance, and residence time) of the velocity dependency of *E. coli* retention and release parameters (k_{sw} , α , and S_f) that are not accounted for in colloid filtration theory.

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1. Introduction

An improved understanding of processes that control the transport, retention, and release of bacteria in porous media is needed for many environmental and industrial applications such as bioremediation of contaminated soils and aquifers, and filtration of pathogenic microorganisms in groundwater or engineered water treatment systems (Liu et al., 2007; Redman et al., 2004). A number of physical, chemical, and biological factors control the retention of bacteria in porous media, including: soil texture (Morales et al., 2015), temperature (McCaulou et al., 1995; Morales et al., 2015), water velocity (Hendry et al., 1999; Meinders et al., 1995), bacterial surface properties (Bolster et al., 2000; Liu et al., 2007; Vandevivere and Baveye, 1992), bacterial motility (de Kerchove and Elimelech, 2008; Huysman and Verstraete, 1993; Massoudieh et al., 2013), solution chemistry (Gannon et al., 1991; Massoudieh et al., 2013; Redman et al., 2004; Simoni et al., 2000; Tong et al., 2005), solid surface roughness (Shellenberger and Logan, 2002), and chemical heterogeneities (Schulze-Makuch et al., 2003; Yee et al., 2000). Although the influence of water velocity on bacteria retention has been studied for many years (Hendry et al., 1999; Meinders et al., 1995), there still are important gaps in knowledge.

Colloid filtration theory (Tufenkji and Elimelech, 2004a; Yao et al., 1971) has been commonly used to explain the transport and retention of bacteria in porous media under saturated conditions. According to colloid filtration theory, the retention rate coefficient (k_{sw}) is a function of the single-collector efficiency (η), the sticking efficiency (α), and the pore-water velocity (v). In particular, k_{sw} is dependent on the mass transfer of colloids from the bulk solution to the collector (sand grain) surface via Brownian diffusion, interception, and sedimentation which is guantified by η (Schijven and Hassanizadeh, 2000; Tufenkji and Elimelech, 2005). Correlation equations have been established from pore-scale simulations of colloid transport to predict η as a function of water velocity, diffusion coefficient, colloid size and density, and collector (grain) diameter and porosity (Ma et al., 2013; Messina et al., 2015; Nelson and Ginn, 2011; Rajagopalan and Tien, 1976; Tufenkji and Elimelech, 2004a; Yao et al., 1971). Colloid filtration theory predicts that differences in the rates of colloid mass transfer cause a nonlinear increase in k_{sw} with v for a given value of α (Tufenkji and Elimelech, 2004b). The fraction of colloid collisions with the collector surface that produces retention (e.g., immobilization) is quantified by the parameter α which changes with the solution and solid phase chemistries (Schijven and Hassanizadeh, 2000; Tufenkji and Elimelech, 2004a). Colloid filtration theory assumed that α only depends on the irreversible adhesive interaction between the colloid and collector surface, but was independent of the water velocity (Elimelech et al., 1998; Elimelech and O'Melia, 1990). In contrast, measured and theoretical values of α and the maximum concentration of colloids on the solid phase (S_{max}) have been shown to depend on solution chemistry and water velocity (Bradford et al., 2012; Johnson et al., 2007; Li and Johnson, 2005; Sasidharan et al., 2014; Shen et al., 2010; Tong et al., 2005; Torkzaban et al., 2007). The value of S_{max} is proportional to the fraction of the solid surface area that contributed to the retention (S_f) (Bradford et al., 2011a; Bradford et al., 2011b; Kim et al., 2009a; Sasidharan et al., 2014). In addition, only a fraction of immobilized colloids is irreversibly retained (Torkzaban and Bradford, 2016; Torkzaban et al., 2010). However, the coupled role of solution chemistry and water velocity on α , *S*_{*f*}, and the reversibility of colloid retention have not yet been completely studied and explained.

Bacteria retention and release depend on the balance of forces and/ or torques that act on cells adjacent to the solid-water interface (Bergendahl and Grasso, 2000; Bradford et al., 2009; Bradford et al., 2011b; Li et al., 2005; Torkzaban et al., 2007; Torkzaban et al., 2008). For neutrally buoyant bacteria, these forces and/or torgues arise from adhesive interactions, random Brownian diffusion, and system hydrodynamics (Ahmadi et al., 2007; Bergendahl and Grasso, 2000; Bradford et al., 2009; Cushing and Lawler, 1998; Goldman et al., 1967; Johnson et al., 1971; O'Neill, 1968; Sharma et al., 1992; Soltani and Ahmadi, 1994; Torkzaban et al., 2007; Torkzaban et al., 2008). Consequently, colloid filtration theory assumes that adhesive interactions always dominant over Brownian diffusion and hydrodynamic forces and torques. In reality, the strength of the adhesive force and torque depends on the solution and solid phase chemistries, colloid size and shape, nanoscale roughness and chemical heterogeneity, deformation, and grain topography (Bayoudh et al., 2009; Bolster et al., 2001; Bradford and Torkzaban, 2012; Bradford and Torkzaban, 2013; Bradford and Torkzaban, 2015; Bradford et al., 2011b; Elimelech, 1994; Kim et al., 2009b; Shen et al., 2013; Shen et al., 2012; Suresh and Walz, 1996; Torkzaban and Bradford, 2016; Torkzaban et al., 2007; Torkzaban et al., 2008; Walker et al., 2005). The hydrodynamic force and torque depend on the average pore-water velocity, the grain size distribution, microscopic roughness, porosity, and colloid size (Bradford et al., 2011b; Burdick et al., 2001; Kuznar and Elimelech, 2007; O'Neill, 1968; Saffman, 2006; Torkzaban et al., 2007; Torkzaban et al., 2008). In many instances, the strength of the adhesive interaction is weak, and a fraction of retained colloids are susceptible to diffusive and/or hydrodynamic removal (Torkzaban et al., 2007; Wang et al., 2016). This result has been used to explain the velocity dependency of α and S_f (Torkzaban et al., 2007).

Not all factors that influence adhesive and hydrodynamic forces and torgues have been considered in previous studies that examined the velocity dependency of α and S_f. For example, some researchers have also reported that the adhesive interaction increases with the residence time (Hemmerle et al., 1999; Meinders et al., 1994; Meinders et al., 1995; Mondon et al., 2003; Stuart and Hlady, 1995; Torkzaban et al., 2013; Vadillo-Rodriguez et al., 2004; Xu and Logan, 2006; Xu et al., 2005). This finding has typically been related to the formation of chemical bonds (Vadillo-Rodriguez et al., 2004; Xu and Logan, 2006), but may also be explained by an increase in the probability that random Brownian motion will produce a kinetic energy of sufficient strength to overcome an energy barrier to achieve a deeper minimum in the interaction energy (Bradford and Torkzaban, 2015; Sasidharan et al., 2017; Torkzaban and Bradford, 2016). An increase in the adhesive interaction with the residence time has important implications for the velocity dependency of α and S_f that have not yet been explained. In particular, the residence time is inversely related to the advective transport velocity. This implies that lower water velocities and flow interruptions with larger residence times may produce greater bacteria retention and less release because of the greater adhesive strength. Furthermore, initially weakly associated colloids that are temporally immobilized in low velocity regions, microscopic roughness locations, and/or grain-grain contacts can experience less diffusive and/or hydrodynamic release with increasing residence time because of an increase

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