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A review of visualization techniques of biocolloid transport processes at the pore scale under saturated and unsaturated conditions

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Abstract

Field and column studies of biocolloid transport in porous media have yielded a large body of information, used to design treatment systems, protect water supplies and assess the risk of pathogen contamination. However, the inherent "black-box" approach of these larger scales has resulted in generalizations that sometimes prove inaccurate. Over the past 10–15 years, pore scale visualization techniques have improved substantially, allowing the study of biocolloid transport in saturated and unsaturated porous media at a level that provides a very clear understanding of the processes that govern biocolloid movement. For example, it is now understood that the reduction in pathways for biocolloids as a function of their size leads to earlier breakthrough. Interception of biocolloids by the porous media used to be considered independent of fluid flow velocity, but recent work indicates that there is a relationship between them. The existence of almost stagnant pore water regions within a porous medium can lead to storage of biocolloids, but this process is strongly colloid-size dependent, since larger biocolloids are focused along the central streamlines in the flowing fluid. Interfaces, such as the air–water interface, the soil–water interface and the soil–water–air interface, play a major role in attachment and detachment, with significant implications for risk assessment and system design. Important research questions related to the pore-scale factors that control attachment and detachment are key to furthering our understanding of the transport of biocolloids in porous media.

Keywords: Colloid; Sorption; Desorption; Filtration; Straining; Groundwater

1. Introduction

The transport of biocolloids (e.g., viruses, bacteria, spores and other microorganisms) through saturated and unsaturated porous media is of significant interest, from the perspective of protection of groundwater supplies from contamination (e.g., [87,53,128,85,104,99]), assessment of risk from pathogens in groundwater (e.g., [9,40,49,51,54,111,143,1,28,124,14,88,108,95,130]), natural and enhanced bioremediation (e.g., [106,96,141,3,38,33,37,2,68]) and for the design of better water treatment systems to remove biocolloids from drinking water supplies (e.g., [42,81,114]). Microorganisms can also travel attached to abiotic particles (e.g., [83,24,55,61]). In addition, certain microorganisms can also facilitate the transport of metals and other chemicals (e.g., [72,35,132,145]). Thus, it is important to understand the transport of colloids in general, and that of biocolloids in specific.

Biocolloids are affected by many of the physical and chemical processes that influence solute transport, i.e., advection, diffusion, dispersion and adsorption (Fig. 1). Advection is the motion of the biocolloids along the trajectories of the fluid streamlines. This mechanism can create dispersion of the biocolloids because of the heterogeneity of the fluid velocity field and the tortuosity of the paths through the porous media. Dispersion can be more important for colloids than for solutes, since it can lead to earlier breakthrough of the colloids, as presented below. In addition, random interactions among molecules and/or particles result in Brownian movements [117] that diffuse the

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Nomenclature

A	Hamaker constant (J)	N_R	Reynolds number (–)
a_p	particle radius (m)	N_{vdW}	van der Waals number (–)
A_s	soil porosity function (–)	SWA	soil–water–air interface (–)
AWI	air–water interface (–)	SWI	soil–water interface (–)
D_∞	colloid bulk diffusion coefficient (m^2/s)	T	temperature (K)
d_g	effective grain diameter (m)	T/C	pore throat to colloid diameter (m/m)
d_p	particle effective diameter (m)	U	pore water velocity (m/s)
g	gravitational acceleration (m/s^2)	η	collision efficiency (–)
k_B	Boltzmann's constant (J/K)	θ_m	soil matrix porosity (–)
N_A	Hamaker number (–)	μ_w	viscosity of aqueous solution (kg/m s)
N_G	gravity number (–)	ρ_p	particle density (kg/m^3)
N_{Pe}	Peclet number (–)	ρ_w	density of aqueous solution (kg/m^3)

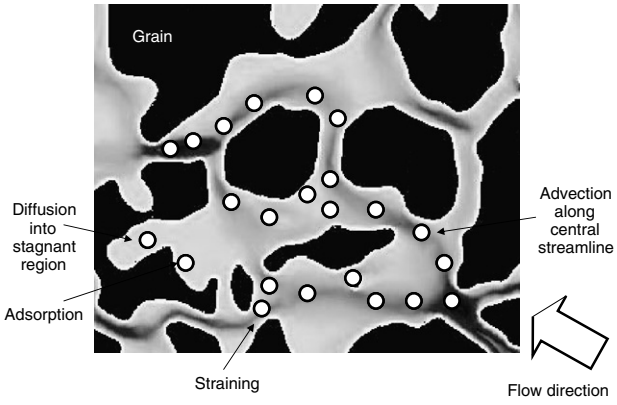

Fig. 1. Schematic of pore scale processes under saturated flow.

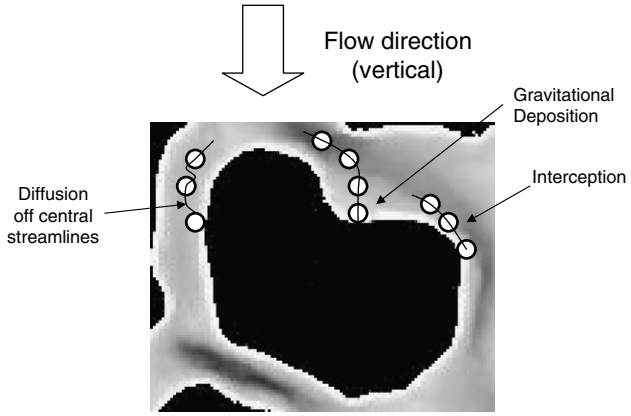

Fig. 2. Schematic of processes that lead to attachment.

biocolloids. The biocolloids can attach to the soil–water interface (SWI), the air–water interface (AWI), or the triple contact of soil–water–air (SWA). Attachment/adsorption to these interfaces can be reversible or essentially irreversible under certain conditions, and is perhaps the most complex process, given the large number of colloid and grain surface characteristics that determine the probability of attachment, and the influence of the dissolved chemical species in the aqueous solution on attachment and detachment. In addition to those four processes, colloids are subject to removal by physical mechanisms, such as straining, interception, diffusion to the wall and gravitational deposition. These physical processes are precursors to attachment (Fig. 2).

At the level of the individual biocolloid, there are processes that can result in the formation of clusters of biocolloids, either attached to an interface or mobile within the aqueous phase. Clusters can also be initiated via biological processes, to form biofilms (e.g., [23,76,129,21,94,126,7]). Individual or clustered biocolloids can break off from the film, releasing them into the flowing aqueous medium (e.g., [18,140,127]).

Biological processes such as growth, death, predation, parasitism and other processes can result in the increase or removal of mobile or attached microorganisms (e.g., [30,36,48,91]). Many of these biological processes are also influenced by physical and chemicals conditions, and the changes in these conditions. Although these processes are extremely important, they are outside the scope of this manuscript, which will focus on the transport of biocolloids through porous media.

Conventional methods to investigate biocolloid transport through saturated and unsaturated porous media often include column and field studies (e.g., [58,67,86,30,42,53,114,130]). These experiments are generally limited to the evaluation of effluent breakthrough curves and destructive sampling at the end of the experimentation that represent some average behavior of biocolloids. Some studies focus on the collection of biogeochemical parameters that can monitor the biological process. Unfortunately, direct observations of the internal processes occurring are not possible, and mechanisms that control biocolloid transport are therefore poorly understood. A useful method to investigate pore scale processes implicates the use of micromodels.

In recent years, micromodels have been increasingly employed to study the fate and transport of colloids and

Table 1
Micromodel and flow cell studies of biocolloid transport

Conditions	Reference	Material	Pattern	Dimensions	Key findings
Saturated porous medium	[75]	Etched glass	Homogeneous periodic network	Pore depth 80 μm, pore width 360 μm	Dispersion of *E. coli* and determination of dispersion coefficient
	[16]	PDMS on glass	Homogeneous network of squares	2 × 2 μm square arrays spaced 1 μm apart	Particle deposition (adsorption)in heterogeneously charged surfaces
	[8]	Etched silicon	Homogenous network of circles, 300 μm diameter	Pore depth 50 μm, pore space 173 μm, pore throat 35 μm	Transport along streamlines and attachment
	[121,122]	Etched silicon	Realistic sand pore network	Pore depth 15 μm, pore diameters: 2.4–30 μm, pore throat 1–10 μm	Pathway a function of colloid size, higher dispersion for small colloids
	[4]	PDMS	Homogeneous network of squares	Pore depth 12 μm, pore throats 10 and 20 μm	Influence of colloidal size on colloidal dispersion
Saturated porous medium with biofilm	[123] Adhesion of	Poly(methyl biocolloids to solid substrata	methacrylate)(PMMA) and glass	Parallel plate flow cell	5.5 × 3.8 × 0.06 cm (l × w × h)
	[31]	Etched silicon	Network of squares; simulation of a fine homogeneous sand; porosity 37%	Pore depth 200 μm, mean channel width 75 μm grain sizes (0.5 mm), pore sizes (50–200 μm)	Rerouting of flow due to biomass growth
	[32]	Etched silicon	Network of squares, channel width randomly distributed	Pore depth 200 μm, channel width 75 and 123 μm	– Conductivity decreases correlated with biofilm growth – Microorganisms strongly attaching to surfaces and to each other are the most effective at reducing permeability – Continuous, rather than periodic, disinfection is recommended
	[73]	Etched glass	Homogeneous triangular lattice	Pore bodies 300 μm, pore throats 30–100 μm	– Biomass accumulation causes permeability reduction – Existence of a critical shear stress
	[125]	Etched glass	Homogeneous triangular lattice	Pore bodies 300 μm, pore throats 30–130 μm	Exopolymer production by bacteria leads to biomass plug and pressure drop increase
	[90]	Etched silicon	Homogenous network of circles	1 cm × 1 cm packed array of 300 μm diameter silicon posts separated by 35 μm pore throats 15 μm deep	Biomass growth changes water flow paths
	[78]	Steel and glass	Flow cells packed with quartz sand	8 × 3 × 54 mm	Colloid–biofilm interactions have implications for colloid transport and remobilization Solution low ionic strength (I) remobilizes attached bacterial biomass Biomass and clay colloids remobilized by deplecting I or increasing flow rate

Columns	Reference	Material	Pattern	Dimensions	Key findings
	[11]	Glass	Sand stone rock	Pore depth 34.9 μm	Biofilm development and accumulation in leading faces of obstructions
Un-saturated porous medium	[138,139]	Etched glass	Hexagonal, quadrilateral and heterogeneous networks	Pore sizes of 20–400 μm 1250 pore bodies each	AWI is an additional sorbent phase for colloids
	[46]	poly(methyl methacrylate) (PMMA)	Parallel plate flow chamber	$76 \times 5 \times 0.6$ mm ($l \times w \times h$)	AWI detaches particles from collector surface
	[121,122]	Etched silicon	Realistic sand pore network	Pore depth 15 μm, pore diameters: 2.4–30 μm, pore throat 1–10 μm	Colloids attached to AWI form a cluster with the dissolution of air bubble
	[25]	Semi-translucent silica sand	Infiltration chamber $26 \times 4.8 \times 0.5$ cm	0.43–0.60 mm grain diameter	Colloidal trapping at SWA Interface
	[5]	PDMS	Realistic sand pore network	Pore diameters: 30–60 μm	Remobilization of biocolloids by intermittent unsaturated flow

specifically biocolloids at the pore scale (Table 1). Micromodels are transparent physical models of porous media, with a pore size in the range of 10–100 μm, etched in glass (e.g., [138,75]), silicon wafers (e.g., [69,121,122,19,8]), or polymer substrates (e.g., [4]) like the ones presented in Fig. 3. Some recent studies have used silica particles as the porous media in three dimensions, visualizing the top surface (e.g., [25]). In addition, flow cells have also been used to study physical processes such as attachment, detachment and mass transfer rates (e.g., [59,123,80,82]). Recent work by Sherwood et al., Olson et al., [118,97] using magnetic resonance imaging has also served to better understand biocolloid transport at small scales. The main purpose of these microscale experiments has been to visualize biocolloid transport processes at the dimensions of a pore or collection of pores, validating or negating hypothesis that have been put forward with regards to processes that had not been actually observed; a secondary objective has been to quantify the importance of these processes. Although the use of micromodels has increased, there are still many questions that need to be answered with regards to attachment and detachment from interfaces, and the role of physical, chemical and biological heterogeneity in such processes.

In this paper, we review the most recent findings on biocolloid migration and immobilization at the pore scale using micromodels. The experimental details can be found in the original papers, so only the most relevant conditions are discussed in this manuscript. We begin by examining the processes that affect biocolloid advection and dispersion under saturated conditions. We then explore the role of interfaces on biocolloid retention in saturated and unsaturated porous media. We conclude with recommendations for future research.

2. Biocolloid transport processes in saturated porous media

2.1. Advection, diffusion, dispersion

Imposing a pressure gradient across a porous medium rapidly generates a stable flow field with defined streamlines. Even fairly significant changes in pressure gradient have minimal influence on the streamlines that define the pathways within the medium, although these changes certainly affect the rate of transport. Colloids and solutes undergo advective transport moving with the pore-water, whose velocity is governed by the hydraulic pressure gradient, porosity, and permeability distribution [44]. Solution of the Navier–Stokes equations at the pore scale (e.g., [121]) indicates that even for fairly complex geometries, the local velocity profile is nearly parabolic [26,8], with the faster streamlines in the center of the pore throats, and slower streamlines along the solid–water interface (Fig. 4). In the complex geometries of natural porous media, there are many regions which are almost stagnant (darker blue regions in Fig. 4), while only a few pathways exhibit significant flow (lighter blue to yellow to red regions

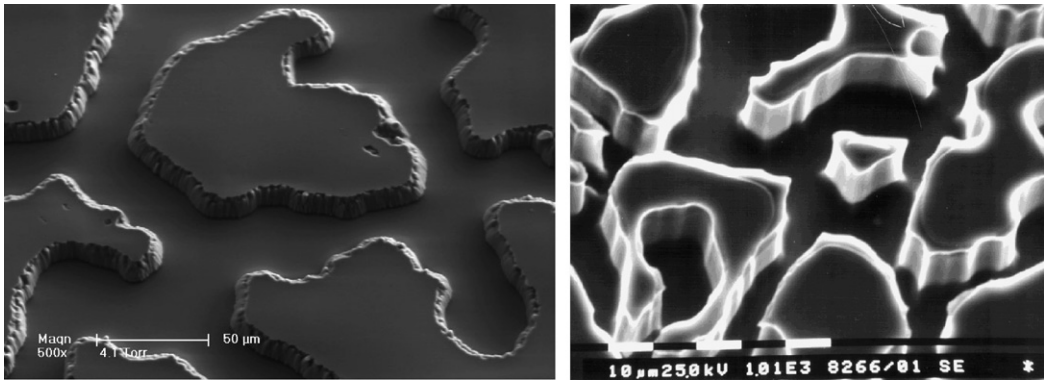

Fig. 3. Scanning electron micrographs of PDMS and silicon wafer micromodels. Typical pore size 10–100 µm, pore throats 3–20 µm.

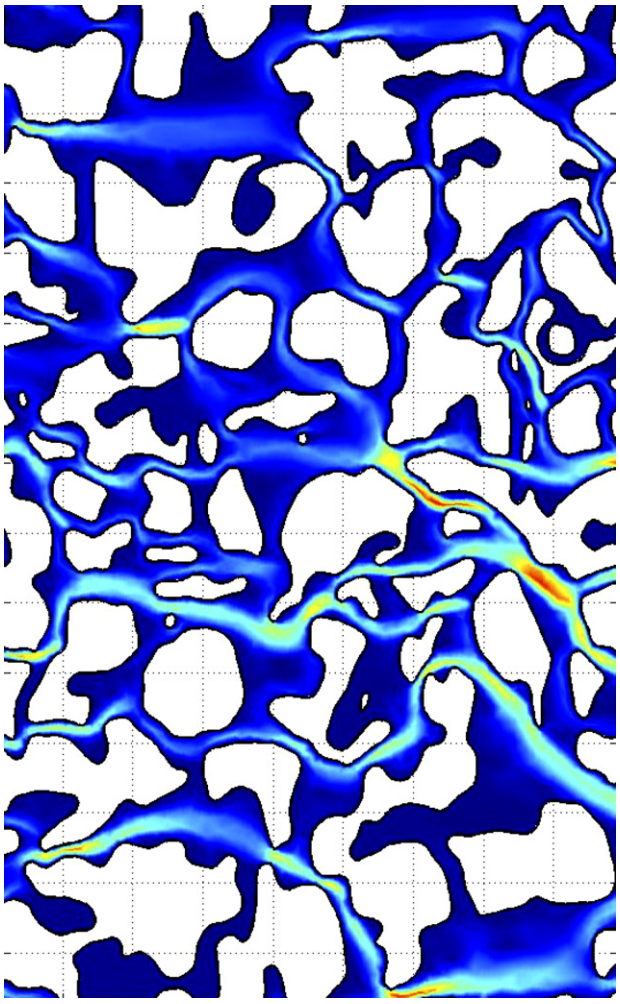

Fig. 4. Solution of Navier–Stokes equation for a complex pore space geometry using FEMLAB. Flow is from right to left, and in the laminar regime.

in Fig. 4). The pressure gradient is from right to left in this simulation that solves the Navier–Stokes equations for a realistic pore space. Thus, solutes or colloids that begin their transport near the central streamlines are advected at a considerably higher rate than those along the SWI.

Since diffusion due to Brownian motion is inversely proportional to the mass of the molecule or particle, solutes have a much higher probability of transferring among streamlines than colloids. Even the smallest colloids observed to date (MS-2 viruses, about 50 nm in diameter) exhibit very low transfer among streamlines within the length of a typical micromodel (a few mm). At larger scales, with increasing transport time, transfer among streamlines will eventually occur, slowing down some of the faster colloids and speeding up some of the slower ones. However, low diffusion tends to focus mobile colloids along the certain streamlines; slower colloids near the SWI have a higher probability of depositing onto the SWI by a number of processes. Larger colloids are forced to remain near the central streamlines, while the smaller colloids can sample a wider range of streamlines (Fig. 5). The schematic shows two sizes of colloids (2 and 7 µm in diameter) in two different pore throats (10 and 20 µm in diameter) and the range of streamlines they can travel through as indicated by the black rectangles. For a smaller pore throat to colloid diameter ratio (T/C ratio), the colloid is severely constrained to the central streamlines.

Under controlled conditions, Auset and Keller [4] showed that some colloids follow these streamlines even

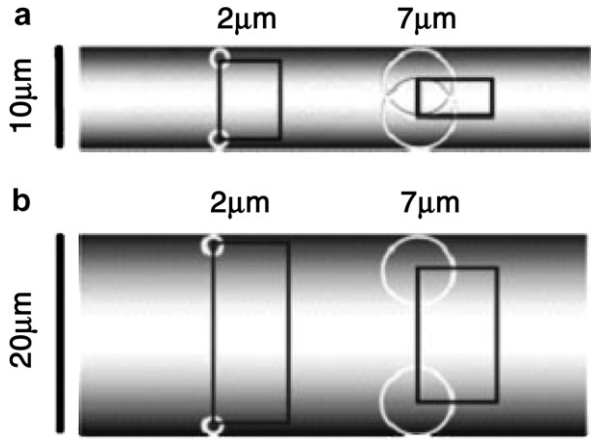

Fig. 5. Schematic of possible distribution of small (2 µm) and large colloids (7 µm) within pore throats of different diameters (10 and 20 µm).

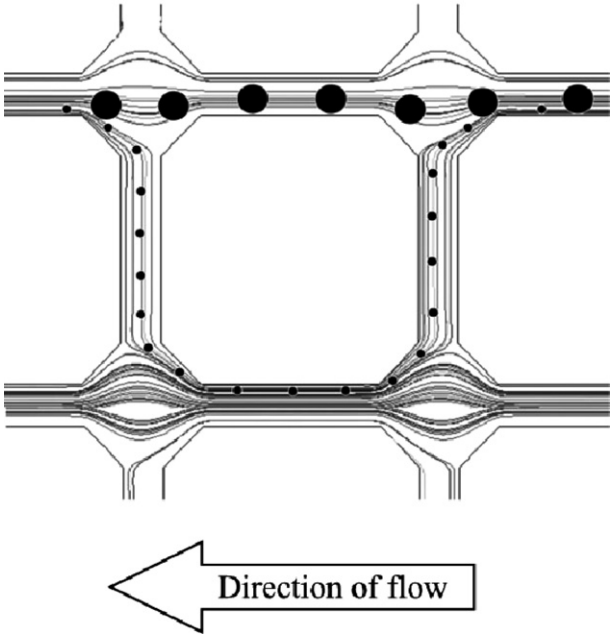

Fig. 6. Schematic of differential colloid transport along streamlines calculated using the solution to the Navier–Stokes equation for a simple pore geometry.

along sharp turns into perpendicular pore throats at the end of pore bodies. Smaller colloids can easily follow along the pore walls, making many detours along their path, while the larger colloids tend to stay on the central streamlines and in general have fewer detours (Fig. 6). For the same size of colloids, travel through narrower pore throats results in shorter average residence time and a narrower distribution of residence times, relative to a wider pore network (Fig. 7a,b). Travel through a more complex network, closer to real pore spaces, results in longer average residence time and a broader distribution than those of simple pore networks (Fig. 7c). Colloid residence time is also a function of the pressure gradient (Fig. 8); large gradients result in wider differences in residence time between colloids of different sizes, while small gradients tend to reduce the differences. Torquato [131] also discusses the effect of heterogeneity on colloid dispersion.

For complex pore geometries such as that shown in Fig. 4, the difference in colloid size has increasing importance. Smaller colloids sample many of the pathways available to them, traveling though both narrow and wide pore throats, and are thus more likely to move into regions where flow is almost stagnant (Fig. 4). Larger colloids are ''excluded'' from many regions and pathways, in part because they remain in the central streamlines, as shown by Sirivithayapakorn and Keller [121]. This differential behavior can have a significant effect on the average residence time of different colloid sizes, since the larger colloids can travel at significant faster velocities through the porous medium compared to the smaller colloids. At the pore scale, this phenomenon can result in colloid velocities that are 1.5–3 times greater than the average water velocity (Fig. 9). This effect has been designated as a ''velocity

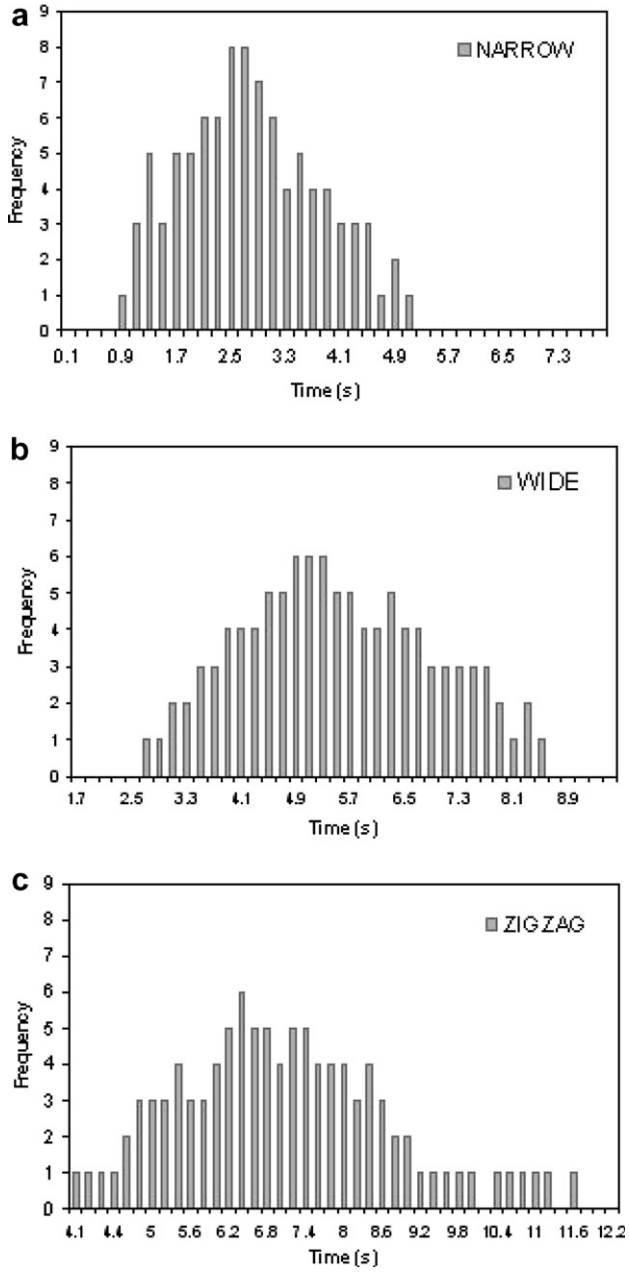

Fig. 7. Experimentally measured residence time distributions for 2 μm colloids in different pore geometries, with a pressure gradient of 500 Pa across the micromodel (visualization method as presented in [4]).

enhancement'' (e.g., [52,4,70]). Mathematically, it has been proposed that this could be handled as a retardation factor less than unity or a lower effective porosity [43]. Due to colloid removal processes the magnitude of this effect decreases with travel distance, as shown by Keller et al. [70], but can nevertheless result in earlier breakthrough of colloids moving through a porous medium, as seen in larger scale studies (Table 2).

An important result from these studies is that dispersivity, which is generally considered an intrinsic property of the porous medium [10], is a function of colloid size [4]; it may be more appropriate to denominate it apparent dispersivity when discussing colloid transport. The effect had

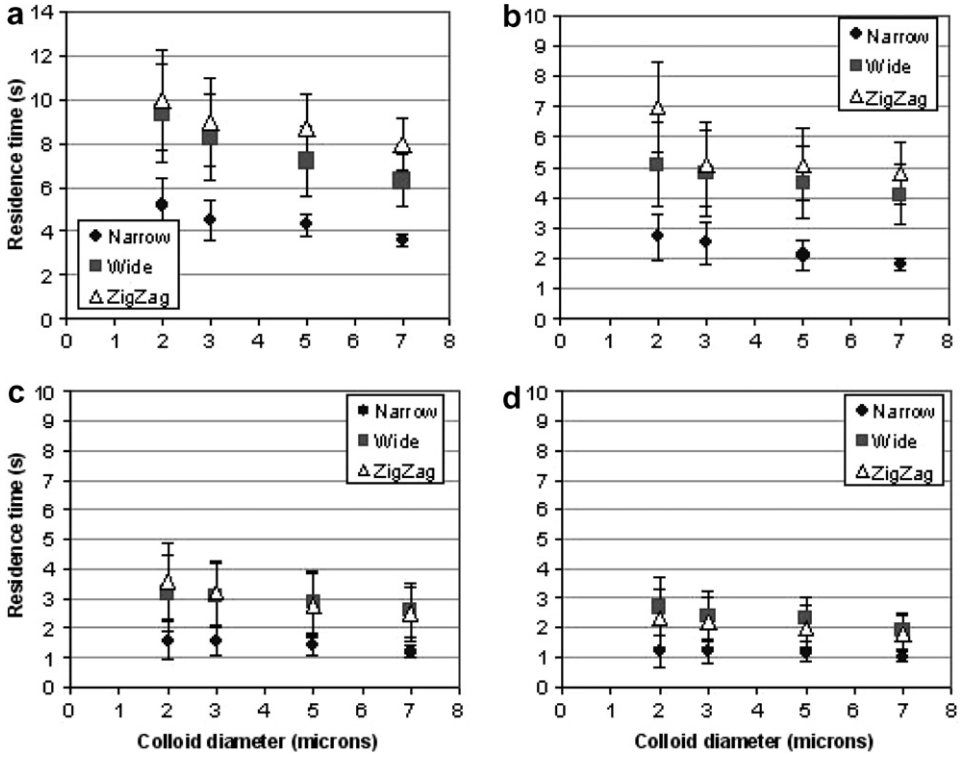

Fig. 8. Comparison between geometries at different pressure gradients. Mean residence time as a function of colloidal diameter. Ten micrometer-channel model (circles), 20 μm-channel model (squares), zig-zag model (triangle). (a) 1500 Pa; (b) 1000 Pa; (c) 500 Pa and (d) 100 Pa.

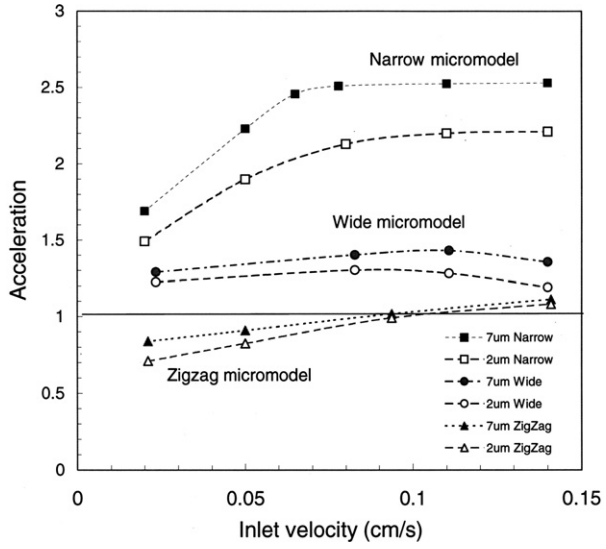

Fig. 9. Ratio of ensemble mean velocity and the "straight path" mean velocity for four colloid sizes, at the highest-pressure gradient (1500 Pa) in each pore geometry.

been observed at larger scales. For example, Shonnard et al.,Pang et al. [119,98], analyzing earlier breakthrough of microbes relative to a tracer, assigned a lower dispersivity for microbes than for solutes. They noted differences in dispersion that led to faster breakthrough, although they were unable to pinpoint the mechanism that caused these differences. Sinton et al. [120] reported reductions in the dispersivity when modeling migration of different sized microorganisms in an alluvial gravel aquifer. Schulze-Makuch et al. [116] also found variable longitudinal dispersivities between bromide and MS2 virus in a model aquifer and showed that vertical dispersion of MS2 is actually less than that of bromide. The micromodel studies have provided the visual explanation for these macroscale observations.

2.2. Exclusion

A number of colloid exclusion processes have been discussed in the literature (e.g., [43,44,13,12]). The most evident exclusion process occurs when the colloid diameter is larger or equal to the pore throat to be entered, resulting in either exclusion (the colloid does not enter the downgradient pore space) or straining, with attachment of the colloid to the SWI. A more subtle exclusion process was observed in the micromodel experiments conducted by Sirivithayapakorn and Keller [121,122] which revealed that the pore T/C ratio threshold for entering a pore throat was about 1.5, due to the hydrodynamics of the system. Since colloids are focused towards the central streamlines, they rarely enter small pore throats. In these studies, more than 100 colloids were tracked through various pores, and the T/C threshold seemed to hold for various sizes and types of colloids, including viruses [121,122]. The pressure gradient was seen not to have a significant effect on the T/C threshold. Although the exact T/C ratio threshold was not determined, one can use this value to consider that biocolloids larger than about 1–5 μm can be excluded from

Table 2
Column and field studies of colloid velocity enhancement

Reference	Travel distance	Medium, particle size	Colloids, size (μm)		Velocity enhancement	Velocity (m/d)
Colloid transport through laboratory columns						
[153]	110 cm	Particles, 18, 40, 58 μm	Microspheres,1, 2, 3, 5, 7, 10 μm		1.03–1.09	3.27
[150]	30 cm	Quartz powder, 30 μm	Microspheres	0.04 μm	1.06	0.144
				0.17 μm	1.11	
				0.31 μm	1.13	
[147]	60 cm	Column sediments, 0.5–1 mm	0.2 μm		1.9	1.4
			0.7 μm		1.7	
			1.3 μm		1.6	
[50]	46 cm	Soil aggregates 1–2 mm	Microspheres 0.11 μm		1.4	10
[52]	10 cm	Coarse sand, 1.4–2.4 mm Medium sand, 0.4–0.5 mm Fine sand, 0.18–0.25 mm	*Cryptosporidium parvum* oocysts, 4.5–5.5 μm		1–1.38	0.7 7
[29]	40 cm 50 cm	Sand sediments	*Comamonas* sp., 0.6 × 1.1 μm		1.1–1.551.8	0.5
[149]	120 cm	Crushed flint gravel,1.5–3 mm	Aeolian quartz silt, 2–60 μm		0.75–1.08	10.4–432
[116]	109 cm	Sieved play sand	Phage MS2 = 0.024 μm		0.88 (pH 6.1) 1.03 (pH 7.5) 1.14 (pH 8.1)	230
[70]	60 cm	Medium sand	Microspheres, 3 and 0.05		1.05–1.09	1.4
			Phage MS2 = 0.025 μm		1.11–1.14	14
Field studies of colloid transport						
[152]		Aquifer	*Escherichia coli*		1.16–1.2	
[148]	0.57 m	Sand aquifer			1.00–1.3	0.43–1.08
	1.62 m		Fulvic acid, 1 nm		1.1–2.3	0.36–0.624
			Polystyrene sulphonate, 20 nm		1.04–1.11	0.6–1.3
					1.0–1.4	0.43–1.08
[146]	6.9 m downgradient	Sandy aquifer, 0.5 mm	Carboxylated microspheres	0.23 μm	1.4	0.33
				0.53 μm	1.4	
				0.91 μm	1.4	
				1.35 μm	1.1	
[151]	385 m	Alluvial gravel aquifer	Fecal coliforms		1.29	160
			F-RNA coliphages		1.88	
			Escherichia coli,J6-2		1.05	
			Phage MS2		1.25	
[98]	61.63 m	Alluvial gravel aquifer	*Bacillus subtilis* endospores		1.16	64
[120]	12–18 m	Alluvial gravel aquifer	*Escherichia coli*, 1.5–6 μm		1.3 and 2	94
			Endospores, 0.8–1.5 μm		1.22	94
			Phage MS2, 0.026 μm		1.21	72
[144]	0.30 m	Colluvial aquifer,silt to gravel size particles	Microsphere, 0.98 μm		1.81	
	0.55 m				1.5	
	1.81 m				1.1	

most small pore throats on the order of a few μm [121,122]. A third exclusion process can occur for higher pressure gradients, since the colloids will tend to by-pass relatively stagnant regions, traveling along the central streamlines. In addition, larger colloids are excluded from some of the streamlines near the pore body and pore throat walls [121,122]. Finally, biocolloids may have surface charges that result in repulsion from the grain surfaces, thus excluding them from certain pore regions (e.g., [113]).

The size of the microbe had previously been observed to be an important factor in bacterial transport in porous media (e.g., [41,39,29]). Variation on the macroscopic transport behavior of different sized biocolloids can be now explained by mechanisms that occur at the scale of pores and pore networks. All four exclusion processes result in selectively faster transport of larger colloids, relative to smaller colloids.

2.3. Collision with SWI

From a theoretical perspective, colloids are thought to reach the SWI based on three mechanisms: interception, diffusion and gravitational deposition. The theoretical framework was put forward by Yao et al. [142] and Rajagopalan and Tien [102], and has since been refined by several authors, in particular by Rajagopalan et al., Ryan and Elimelech [103,109]. Based on this theoretical approach, the probability of a collision can be estimated from:

$$\eta = 0.897\sqrt[3]{A_s}\left(\frac{k_B T}{\mu_w d_p d_g U}\right)^{2/3} + \frac{3}{2}A_s\left(\frac{d_p}{d_g}\right)^2 + \frac{(\rho_p-\rho_w)g}{18\mu_w U}d_p \quad (1)$$

$$A_s = 2(1-p^5)/[2-3p+3p^5-2p^6], \quad p=(1-\theta_m)^{1/3} \quad (2)$$

where ρ_p = particle density (kg/m^3), ρ_w = density of aqueous solution (kg/m^3), g = gravitational acceleration constant (m/s^2), μ_w = viscosity of aqueous solution (kg/m s), U = pore water velocity (m/s), d_p = particle effective diameter (m), d_g = effective grain diameter (m), A_s = soil specific constant related to θ_m = soil matrix porosity (–), k_B = Boltzmann's constant (J/K), and T = temperature (K). Recently, Tufenkji and Elimelech [133] have proposed the following refined correlation based on experimental evidence:

$$\eta = 2.4A_S^{1/3}N_R^{-0.081}N_{Pe}^{-0.715}N_{vdW}^{0.052} + 0.55A_S N_R^{1.675}N_A^{0.125} + 0.22N_R^{-0.24}N_G^{1.11}N_{vdW}^{0.053} \quad (3)$$

where $N_R = \frac{d_p}{d_g}$, $N_{Pe} = \frac{Ud_p}{D_\infty}$, $N_{vdW} = \frac{A}{k_B T}$,

$N_A = \frac{A}{12\pi\mu_w a_p^2 U}$, $N_G = \frac{2}{9}\frac{a_p^2(\rho_p-\rho_f)g}{\mu U}$,

D_∞ = colloid bulk diffusion coefficient (m^2/s), A = Hamaker constant (3×10^{-21}–4×10^{-20} J), and a_p = particle radius (m). The three terms correspond to interception, diffusion and gravitational deposition.

Using general values for biocolloids, such as a particle density of 1050 kg/m^3 (ref) and particle size ranging from 50 nm to 5 μm, the relative importance of these three processes as a function of velocity can be estimated using Eq. (3) (Fig. 10), considering a porosity of 30% and an effective grain diameter of 100 μm (fine sand). The range of flow velocities corresponds to a few cm/d to about 100 m/d, which is the range of interest for transport in porous media. From Eq. (1), interception is considered to be mostly a function of the relative size ratio between the colloid and the grains of the porous medium, as well as the porosity via A_s, independent of U. Interception is expected to be strongly influenced by matrix porosity, particularly as porosity decreases below 10%. However, the empirical evidence used to parameterize Eq. (3) indicates that interception is in fact a function of flow velocity, decreasing with

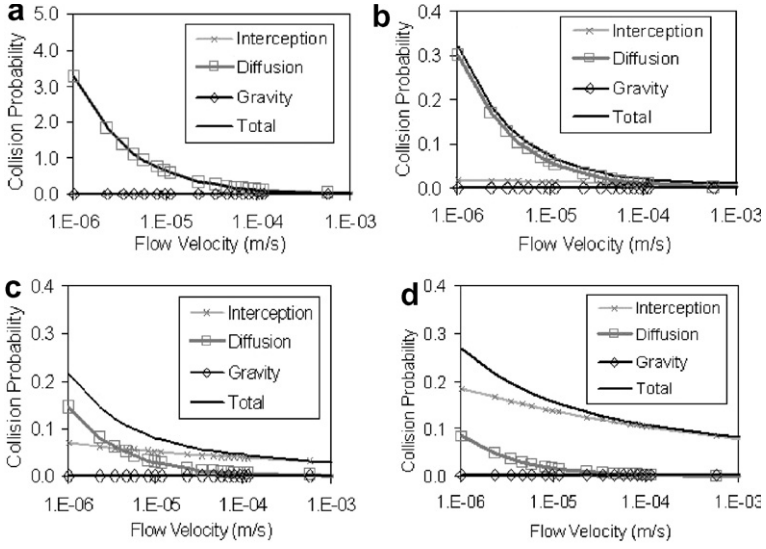

Fig. 10. Estimate of relative importance of interception, diffusion and gravitational deposition, and total collision probability (Eq. (3)) at different flow velocities, for colloids of (a) 50 nm; (b) 1.0 μm; (c) 2.5 μm and (d) 5.0 μm.

increasing velocity. This was recently observed in micromodel studies by Baumann and Werth [8]. These experiments show that at high flow velocities interception is less probable, since the colloids follow along the streamlines and are generally diverted from the grain surfaces.

For small biocolloids such as viruses and microorganisms up to about 1 μm, interception is thought to be negligible, while diffusion dominates over gravitational deposition at all flow velocities of interest (Fig. 10a,b). From the micromodel studies and calculation of the velocity field within a complex pore network, there are regions of stagnant water which are shielded from the main flow direction by the grains, are in crevasses or dead end pores, or along the walls of wide pore bodies (Fig. 11). Small biocolloids are likely to accumulate initially in these regions, since they are more likely to be traveling along these streamlines and can more easily diffuse into stagnant regions. For larger biocolloids, interception should dominate, followed by diffusion (Fig. 10d). Gravitational deposition becomes important only for flow velocities less than 1×10^{-6} m/s, or on the order of mm/day, since biocolloids that are almost buoyancy neutral.

2.4. Attachment

Once the biocolloid collides with the SWI, the probability of attaching to the surface, also denominated the attachment efficiency, α, is thought to be controlled by electrostatic and van der Waals interactions [84]. These interactions have been estimated using Derjaguin–Landau–Verwey–Overbeek (DLVO) theory of biocolloidal stability [62]. Recently, work by Tufenkji and Elimelech, Redman et al. [134,105] suggests that additional aspects need to be considered. Grain surface composition and charge have been shown to be important (e.g., [135,89,56]), as well as the biocolloid surface proteins and other charged chemical species (e.g., [74,136]). Based on theoretical calculations, Baumann and Werth [8] estimated that the probability of attachment for their colloids is in the range of 10^{-4}–10^{-6}. Schijven et al. [115] reported values for a of 0.00027–0.0014 for MS2 viruses in dune sand. Keller et al. [70] reported values of 0.008–0.0026 for MS2 viruses in medium sand at flow velocities of 1.4–14 m/day. For *Cryptosporidium*, Harter et al. [52] reported values from 0.37 to 1.1 in sand. In Sediment cores, Dong et al. [29] measured values of 0.003–0.025 for *Comamonas* sp. Most of the experimental evidence is from column studies, leaving this as an area of open research at the microscale.

A number of studies have addressed the mechanisms of biocolloid attachment to the SWI and/or a growing biofilm. A biofilm may include cells as well as exopolymeric substances that serve as a substrate modifier for a number of reasons (e.g., [22,17,47]). Different bacterial strains may exhibit differential attachment (e.g., [6]). Surface physicochemical properties influence the ability of biocolloids to form biofilms (e.g., [15,79,63,92]). These biofilms can induce changes in hydrodynamic properties that influence the transport of subsequent biocolloids (e.g., [107]). Detachment of biocolloids from these biofilms is a source for downgradient sites, and may be influenced by a variety of processes including flow velocity and associated shear

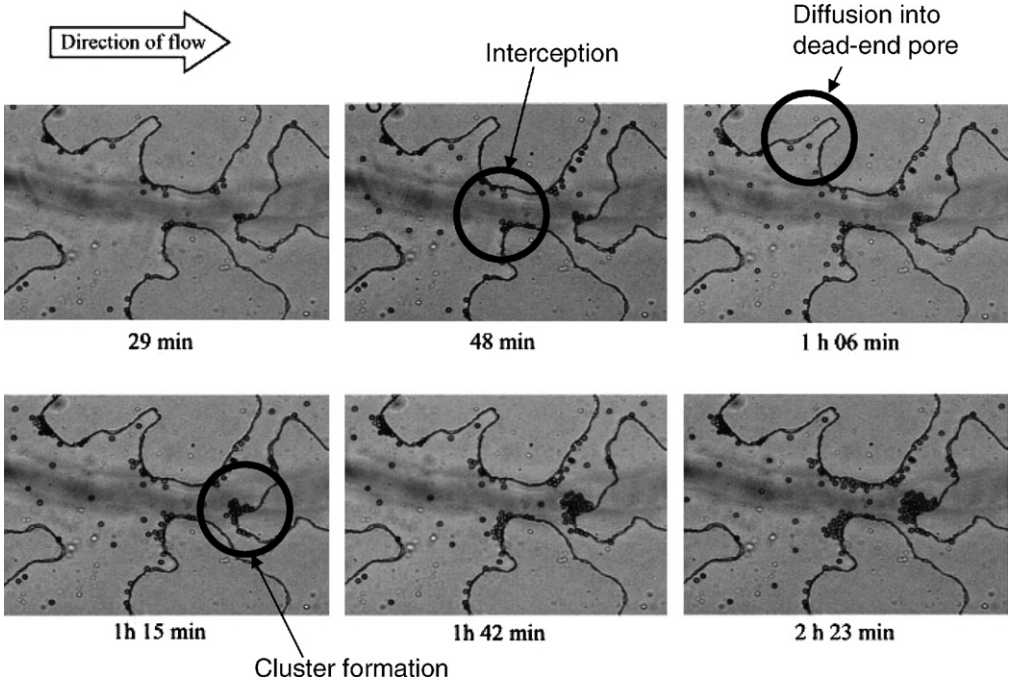

Fig. 11. Images of collision via interception and diffusion into dead-end pores for 5 μm latex microspheres at an average velocity of 143 μm/s = 12 m/day, within a PDMS micromodel. Clusters of colloids form even at very low ionic strength (deionized water) (visualization method as presented in [4]).

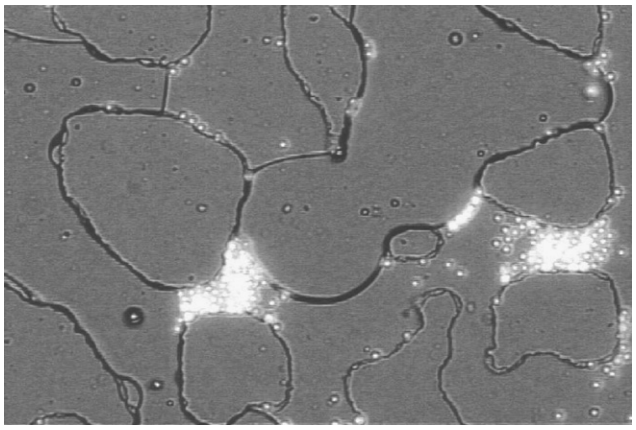

Fig. 12. Clustering of colloids that results in significant modification of permeability and colloid transport pathways (visualization method as presented in [4]).

stress, chemical conditions or biofilm thickness (e.g., [57,45,140,60]).

Particle–particle interactions may lead to attachment and clustering (Fig. 11e,f, Fig. 12). Attached biocolloids can also form large clusters up to a certain thickness (i.e., biofilms, aggregates and filaments, biowebs), until some of the cells in the interior become starved of a particular chemical (e.g., electron acceptor, nutrients), leading to rupture of the biofilm and subsequent release (biosloughing) (e.g., [32]). These clusters can result in significant modification of the permeability of the porous matrix, and will also influence the pathways of subsequent colloids, modifying the dispersivity of the matrix. Clusters of colloids can also form at the AWI (discussed in the next section), which upon release from the interface can lead to a collision with the SWI and subsequent attachment.

3. Biocolloid transport processes in unsaturated porous media

Flow and transport mechanisms in the unsaturated zone become more complicated than those in the saturated zone because of the presence of the AWI, flow discontinuities and wetting history [93,110]. Investigations at larger scales have shown that volumetric moisture content and pore water velocity play a key role in biocolloid transport in the vadose zone (e.g., [101]). Biocolloid sorption at the AWI has been recognized as an important process for several years [138,139,100].

Pore scale studies in unsaturated conditions [139,122] have shown that, like the SWI, the AWI serves as collector of biocolloids (Fig. 13). Some of the colloids might also be trapped at the triple junction, the SWA. These interfaces (AWI and SWA) are therefore important barriers for biocolloid transport. Colloids can interact with the AWI through the same collision processes described before. However, in part due to the hydrophobicity of the AWI and the proliferation of these interfaces as the porous matrix drains, the probability of attachment increases significantly.

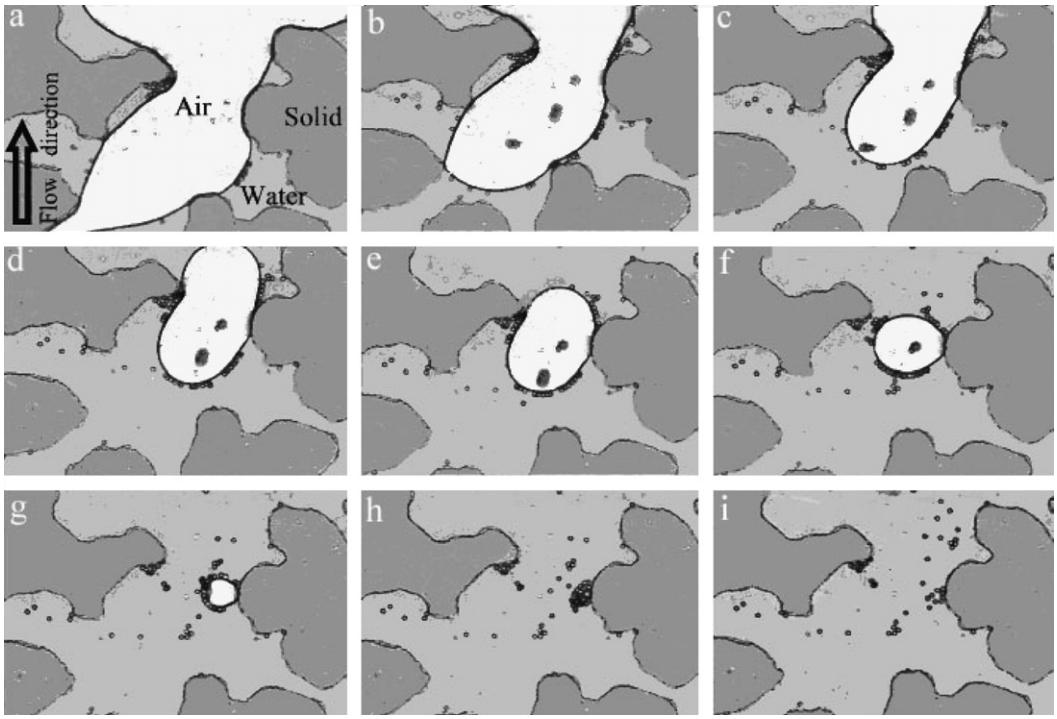

Fig. 13. Sequence showing the imbibition process that displaces the air phase, eventually leading to a detached air bubble with several colloids at the AWI. Eventually the air bubble dissolves, leading to the formation of clusters of colloids. The clusters can then be transported through the pore space, or can break up (visualization method as presented in [4]).

The earlier work visualizing colloid sorption at the AWI was done under steady pore water flow [138], and it led to the conclusion that colloids were sorbed irreversibly at the AWI. Increasing air saturation increased retention at the AWI [139]. These observations were supported by results of experiments on mass balance of breakthrough colloid concentrations in sand columns [112,64–66,77] where more particles were retained at lower water contents. Calculations by Sirivithayapakorn and Keller [122] using DLVO theory and evaluating the electrostatic and capillary forces indicated that colloids, including MS2 viruses, should be held almost irreversible at the AWI, once they cross over the energy barrier for attachment. The energy barrier increases with particle size, but is on the scale of 1–15 nm measured from the AWI into the bulk solution. Thus, colloids can migrate slowly very near the AWI along streamlines perpendicular to the interface and not be captured unless they cross the energy barrier due to some mechanism (diffusion or interception). On the other hand, water flow around entrapped air bubbles decreases substantially, since the dimensions of the water films through which water flow is on the order of a few μm^2 at most [69].

As with attachment to the SWI, biocolloid attachment to the AWI is a function of ionic strength and the surface properties of the biocolloid, such as hydrophobicity and surface charge [138]. Increases in ionic strength will reduce the magnitude of the repulsive energy barrier between the negatively charged air–water interface and the biocolloids, leading to progressively more favorable conditions for attachment and faster rates of air–water interface capture.

Wan and Tokunaga [137] introduced an additional mechanism of colloid immobilization in partially saturated porous media. They used the concept of film straining to suggest that the transport of suspended colloids could be retarded due to physical restrictions imposed when the thickness of water films is smaller than the diameter of the colloids. Wan and Tokunaga [137] estimated that these films should be on the order of 20–40 nm, which is considerably thinner than a 1 μm *Escherichia coli*, but may not completely immobilize a 25 nm virus. Chu et al. [20] estimated a film thickness in the range of 15–21 nm for different soils, at a water content of 0.17–0.29 $cm^3\ cm^{-3}$. In their column studies, Keller et al. [70] estimated a thickness for the water films of 30–60 nm in a medium sand and average water content of 0.11–0.18 $cm^3\ cm^{-3}$.

According to Wan and Tokunaga [137], colloid retention by film straining depends on the existence of pendular ring discontinuity, on the ratio of biocolloid size to film width and on flow velocity. A pendular ring is defined as water retained by capillarity around the adjacent grains. The possibility of pendular ring discontinuity augments as the capillary pressure decreases [27]. When the biocolloid diameter is smaller than film thickness, straining remains ineffective. When the biocolloid diameter is similar or bigger than film thickness than surface tension forces retain biocolloids against grain surfaces. Crist et al. [25] provided visual evidence that biocolloid retention can also occur via trapping at the solid–water–air (SWA) interface. These thin water films serve as storage locations for biocolloids under unsaturated conditions, but may also serve to place the biocolloid in direct contact with the SWI if the water film thickness decreases even more.

Transient flow, generated by rainfall and snowmelt events interspersed between dry periods or due to artificial aquifer recharge or other anthropogenic actions, can promote very rapid biocolloid mobilization (e.g., [34]). Under transient conditions it has been observed that the movement of biocolloids is affected by the movement of air bubbles and AWI (e.g., [46,45,71]). Sirivithayapakorn and Keller, Auset et al. [122,5] observed in micromodels how infiltration events can mobilize the AWI, thicken the water films where colloids are immobilized, dissolve some of the gas phase and mobilize air bubbles (Fig. 13). First, the AWI is displaced as the water re-imbibes into the porous media. Colloids trapped in stagnant water regions are able to remobilize. At some point, an air bubble breaks off from the main air phase. Colloids which were attached to the AWI remain attached until the AWI disappears. Eventually, these rewetting processes lead to the remobilization of all colloids trapped at the AWI or in thin water films.

Depending on colloid surface properties, the colloids may tend to cluster even in solution. However, in many cases surface charges are similar, creating an electrostatic barrier that reduces the likelihood of clustering, as calculated by Sirivithayapakorn and Keller [122] for MS2 viruses and latex microspheres in a weak ionic solution. Nevertheless, colloid clusters can form at the AWI as the size of the AWI shrinks, as observed in Fig. 13g–h. These clusters might be stable enough to travel as a single body, or they might break up while traveling (Fig. 13i). These observations suggest that coagulation at the AWI may increase the overall filtration for biocolloids transported through the vadose zone.

Whether all colloids on the AWI are actually at the SWA interface is an open question. In most micromodel visualizations, the pore space being observed reflects a thin (10–50 μm) wedge between the top and bottom surfaces (see for example the diagram in [25]). Colloids which appear to be at the AWI could in fact be at the SWA. Certainly, some of the colloids observed in these experiments are at the SWA, as suggested by Crist et al. [25]. Even as the imbibing water front displaces the air phase in Figs. 13b–g, some of the colloids remain in place, suggesting attachment to the SWI at the same time that the colloids were in contact with the AWI. However, in other sequences (e.g., [5,122]; and Fig. 13), some colloids are seen to rapidly travel through the porous media as soon as the AWI disappears, suggesting no attachment to the SWI.

Rewetting processes and intermittent wetting and drying events thus can result in significant mobilization of biocolloids that had been considered irreversibly retained at the AWI. Colloid remobilization appears to be a strong function of particle size [71]. Although intermittent filtration provides significant pathogen removal capacity, it is important to

take into consideration the potential for biocolloid remobilization over time.

These pore-scale mechanisms in the unsaturated zone play a significant role in the macroscopic transport of biocolloids; biocolloids can be significantly retarded in their transport through the porous media due to the interaction with the AWI and SWA interface, but they can also be released from these interfaces to continue their path. In addition, sorption of biocolloids at the AWI, SWA interface and thin water films can result in increased probability of sorption onto the SWI.

4. Future research directions

Although significant advances have been made in our understanding of biocolloid fate and transport in saturated and unsaturated porous media with pore scale visualizations, there are a number of important questions still left unanswered. The conditions that result in attachment to the SWI need to be better understood at this scale. Surface heterogeneity needs to be characterized, so that we can make better predictions of the probability of attachment to a particular surface given information on the biocolloid, the attachment surface and the aqueous solution composition. Detachment from the surface has also not been studied in any detail at this scale, whether it is directly from the surface or from a growing biofilm. We need to understand the effect of changes in physical, chemical and/or biological conditions that result in detachment of biocolloids. Biological processes that influence surface characteristics and heterogeneity are also targets for future research at this scale. Pore scale observations can help to validate our understanding of the mechanisms that govern the macroscopic equations used to simulate the fate and transport of biocolloids in porous media.

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