We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists



186,000

200M



Our authors are among the

TOP 1% most cited scientists





WEB OF SCIENCE

Selection of our books indexed in the Book Citation Index in Web of Science™ Core Collection (BKCI)

Interested in publishing with us? Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected. For more information visit www.intechopen.com



Geological Disposal of Nuclear Waste: Fate and Transport of Radioactive Materials

Additional information is available at the end of the chapter

http://dx.doi.org/10.5772/50391

1. Introduction

Prabhakar Sharma

Nuclear power plants use nuclear fission for generating tremendous amount of heat for the production of electrical energy. Currently, there are many nuclear power plants in operation worldwide, which produces high-level nuclear wastes at the same time. Nuclear wastes are being produced as by-product of nuclear processes, like nuclear fission (spent fuel) in nuclear power plants, the radioactive elements left over from nuclear research projects and nuclear bomb production. The management and disposal of these previously stored and continuously generated nuclear wastes is a key issue worldwide. A huge amount of radioactive wastes have been stored in liquid and solid form from nuclear electricity/bomb production plants from several decades at different locations in the world. For example, the Hanford Site is a most decommissioned nuclear production complex on the Columbia River in the U.S. state of Washington, operated by the United States federal government as shown in fig 1 [21, 28, 50]. Hanford was the first large-scale plutonium production reactor in the world. The Hanford site represents approximately two-thirds of the nation's high-level radioactive waste by volume [28].

Radioactive/nuclear wastes are specific or mixture of wastes which contain radioactive chemical elements that can not be used for further power production and need to be stored permanently/long term in environmentally safe manner [63]. The ultimate disposal of these vitrified radioactive wastes or spent fuel elements requires their complete isolation from the environment. One of the most favorite method is disposal in dry and stable geological formations approximately 500 meters deep. Recently, several countries in Europe, America and Asia are investigating sites that would be technically and publicly acceptable for deep geological storage of nuclear wastes. For example, a well designed geological storage of nuclear wastes from hospital and research station is in operation at relatively shallow level in Sweden and a permanent nuclear repository site is planning to be built at deep subsurface system for nuclear spent fuel in Sweden in order to accommodate the stored and running nuclear waste from ten operating nuclear reactors which produce about 40 percent of Sweden's electricity (In Sweden, the responsibility for nuclear waste management has been



©2012 Sharma, licensee InTech. This is an open access chapter distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/3.0), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

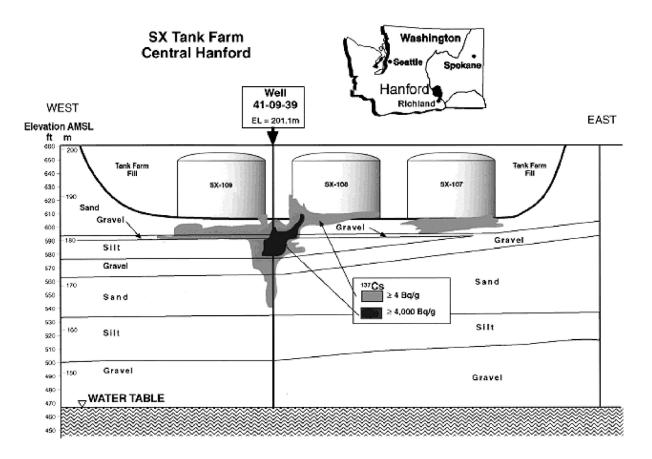


Figure 1. An example of underground storage of radioactive waste and leakage into subsurface system at Hanford site, Richland, WA, USA (from McKinley et al. 2001) [50].

transferred in 1977 from the government to the nuclear industry, requiring reactor operators to present an acceptable plan for waste management with a so called absolute safety to obtain an operating license. The conceptual design of a permanent repository was determined by 1983, calling for a placement of copper-clad iron canisters in a granite bedrock about 500 m underground, below the water table known as the KBS-3 method, an abbreviation of kärnbränslesäkerhet, nuclear fuel safety. Space around the canisters will be filled with bentonite clay. On June 3rd 2009, Swedish government choose a location for deep level waste site at Östhammar, near Forsmark nuclear power plant.).

The recent accident in 2011 in nuclear power plant in Fukushima, Japan due to Tsunami has caused release of underground stored radioactive elements/wastes into the subsurface system. This is a big concern for clean-up operation as they can migrate to farther locations with pore water flow of subsurface system and can create big environmental disaster. It has led to re-thinking of researcher and responsible organizations for protecting their underground stored radioactive wastes and implementing multi-protection mechanisms for deep geological storage of the hazardous radioactive wastes. In the event of accidental release/leakage of radioactive materials into the subsurface system, there is a possibility of its migration with the soil-pore water flow and to be transported to the surface and groundwater bodies as shown in fig 1 [21, 50, 76]. Furthermore, some radioactive contaminants do not move through soil pores in dissolved form but rather attach strongly to fine soil particles (1 nm to 1 μ m size, commonly called "colloid"). These contaminant-attached soil

Geological Disposal of Nuclear Waste: Fate and Transport of Radioactive Materials 61

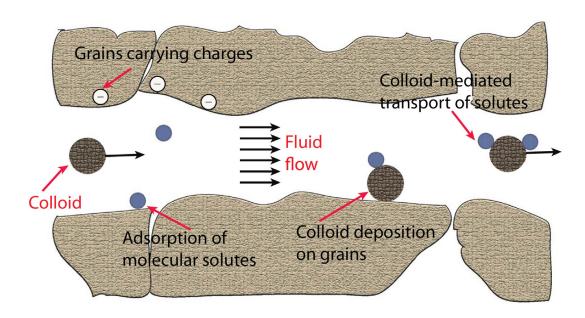


Figure 2. Flow and transport of colloidal particles attached with possible radioactive contaminants.

particles may themselves become mobile and move through the soil and eventually reach the water bodies (fig 2). This is commonly known as colloid-facilitated contaminant transport [9, 11, 14, 16, 20, 23, 41, 52, 56]. Colloids are a ubiquitous component of subsurface systems and play an important role in radioactive contaminant fate and transport. Mobile colloidal particles in the subsurface can enhance the movement of otherwise immobile radioactive contaminants attached with colloidal particles or colloids can be the radioactive elements themselves [11, 20, 23, 33, 34]. The colloidal particles can be transported in the groundwater through soil solution as affected by physico-chemical condition of the surrounding medium and colloids (fig 2). The understanding of colloid transport is of significant interest for the protection of subsurface environment from contamination by intentional or unintentional release of nuclear wastes.

In the infiltration/rainfall events, the colloidal size radioactive particles or radioactive elements attached with mobile colloids would be transported to the groundwater through unsaturated porous media, where gaseous phase can play a critical role in association with the liquid and solid phases [13, 68, 82]. Several mechanisms are responsible for colloid transport in unsaturated zone in addition to that of saturated zone, such as, liquid-gas interface capture, solid-liquid-gas interface capture, liquid-film straining, and storage in immobile liquid zones [13, 18, 27, 44, 51, 68, 70, 79, 85, 87]. The strong force (capillary force) associated with the moving liquid-gas interfaces led to particle mobilization in the natural subsurface environment. As the water content decreases, a thin film of liquid forms over the grain surfaces and in the pendular ring created between the pore spaces had different opinion in different literature [73, 81]. This chapter will review all the possible mechanisms responsible for attachment of colloids in the partially saturated system. The discrepancies in literature about colloid removal and deposition mechanisms at different locations in three phase system

will also be discussed to guide the researcher and decision making bodies for designing deep geological storage for storing nuclear wastes (to ensure uninterrupted and cheap nuclear power generation) and to combat the extreme situation of their release into subsurface systems through unsaturated zone and protecting the natural water bodies and environment from radioactive contamination.

2. Mechanism of colloid attachment

The colloid retention in saturated porous media is primarily controlled by attachment at the solid-liquid interface in relation to the surface properties of the solid and background solution, which has been well documented in literature [37, 42, 46, 48, 49, 60, 65, 69]. Whereas the presence of gaseous phase in the unsaturated subsurface system introduces an additional mechanism for colloid retention. Although several steps has been taken to enhance the understanding of mechanisms responsible for colloid transport and retention through unsaturated porous media, there is a need to put extra effort in this area for better understanding [4, 24, 42, 47, 55]. In the unsaturated porous media, the additional mechanisms (compared to saturated system) for colloid transport were reported as: colloid captured at the liquid-gas interface [1, 12, 43, 44, 54, 66–68, 70, 72, 80, 83], colloid captured due to straining [4, 7, 74, 78], the colloid captured at solid-liquid-gas interface [10, 17, 18, 27, 51, 87, 88], and colloid storage in immobile zone [15, 25, 26, 61]. The flow chart lists the above four retention mechanisms (fig 3). The colloids trapped due to different mechanisms, as mentioned in the flow chart, govern the movement of colloidal/nano-size particles in a porous media (Fig. 4). The figure 4 shows the example of the colloid captured by liquid-gas interface, solid-liquid-gas triple point, straining, immobile zone, and solid-liquid interface. Many of the colloid retention mechanisms are still poorly understood and debating [73, 81]. To improve our knowledge and understanding about the fate of radioactive particles (alone or attached with colloidal particle) in unsaturated porous media, the colloid capture mechanisms are discussed in detail below.

2.1. Attachment at the liquid-gas interfaces

It has been stated in the past that the moving liquid-gas interface plays an important role in colloid mobilization in unsaturated porous medium [1, 12, 43, 44, 54, 66–68, 70, 72, 80, 83]. A considerable amount of colloids were captured at the liquid-gas interfaces and moved with the infiltration front depending on flow velocity and the solution ionic strength [68]. This has been verified by numerical solution of the Young-Laplace equation that expanding water film can lift the subsurface colloids from the mineral surfaces [66]. The detachment of sub-micron sized particles from initially wet solid surfaces had been investigated by air-bubble experiments to understand the strength of moving liquid-gas interfaces [30–32, 45, 53]. In a direct visualization experiments, it had been found that a significant number of colloids were detached from initially dried solid surfaces by the moving liquid-gas interface and remain attached to the liquid-gas interfaces (Fig 5) [70]. The irreversible nature of colloid attachment from the liquid-gas interface has been observed earlier, which validate the strength of moving liquid-gas interface [1, 80].

Three consequent steps might occur in the colloid detachment from a solid surface and its attachment to the liquid-gas interface. These are interception of the particle, attachment or thinning of the liquid film in between the particle and the liquid-gas interface, and

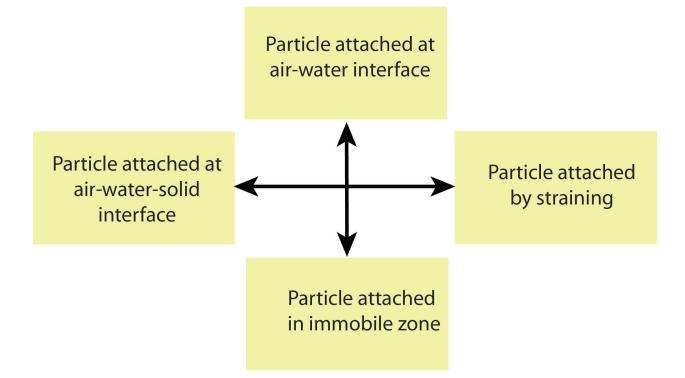


Figure 3. Different possible colloid retention mechanisms in unsaturated porous media.

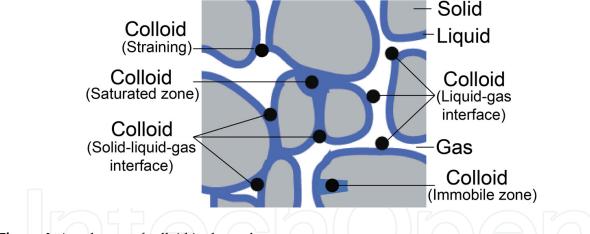


Figure 4. Attachment of colloid in three-phase system.

stabilization of the particle on the liquid-gas interface [22, 30, 67, 70]. The total detachment probability (P_{det}) is defined as [22]:

$$P_{det} = P_{int} \times P_{att} \times P_{sta} \tag{1}$$

where P_{int} is the interception/colloision probability, P_{att} is the attachment probability, and P_{sta} is the stability probability. P_{int} depends on the actual number of colloids intercepted by the moving liquid-gas interface. It depends on the velocity and direction of the liquid-gas interface. It could be zero, means no colloid detachment from solid surface, if interface contact time will be less than induction time. The induction time is time to form a three-phase contact line by thinning the liquid film between the particle and the liquid-gas interface. P_{sta} can be assumed

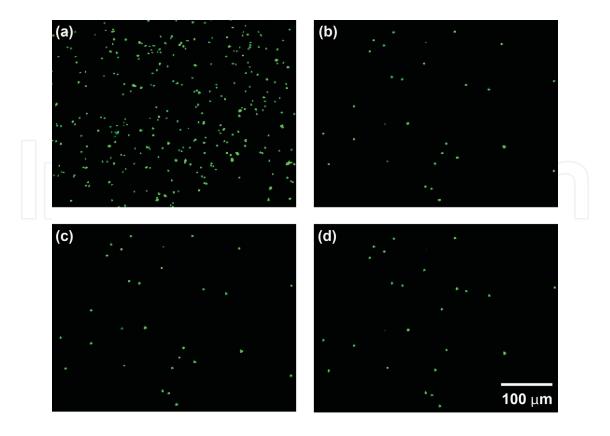


Figure 5. Detachment of amine-modified microspheres from glass slide after moving the liquid-gas interface: (a) no interface movement, (b) 1 interface movements, (c) 2 interface movements, and (d) 3 interface movements.

as one because of irreversible nature of colloids attached on the liquid-gas interface, i.e., particles remain attached to the interface. Thus, the above equation (1) shows the importance of velocity (specially in the range of porous media velocity) on detachment of colloid by the moving liquid-gas interface. Sharma et al [70] tested the velocity effect in the range of 0.4 to 400 cm h⁻¹ on colloid detachment from the solid surface. They found that colloid detachment from the solid surface was more at lower interface velocity. A similar observation has been made for particle detachment by air-bubble moving at higher speed (> 8 cm h⁻¹) [30, 31].

For the transport and mobilization of radioactive materials in colloidal size or its attachment with colloidal particle, the balance among electrostatic, hydrodynamic, and capillary forces are responsible for attraction of particle towards the liquid-gas interface [29, 67, 68, 70, 72, 79]. If capillary force dominates then the colloidal particles attracted towards the liquid-gas interface and if electrostatic dominates then the colloidal particles remain stay over the grain surface. The hydrodynamic forces may be neglected for the colloidal size particles [57, 58, 64]. Figure 6 shows the force balance between electrostatic force and the capillary force for hydrophilic and hydrophobic particle attached with the solid surface when liquid-gas interface moved in the upward direction. The attachment force (F_{att}) is the sum of electrostatic force and van der Waals force acting toward the solid surface whereas the detachment force (F_{det}) is the horizontal component of the capillary force, which is responsible for detachment of colloids from the solid surface and attachment into the liquid-gas interface as shown below in detail. Colloids detach from the solid surface only if $F_{det} > F_{att}$. The detail of force balance calculation are shown below:

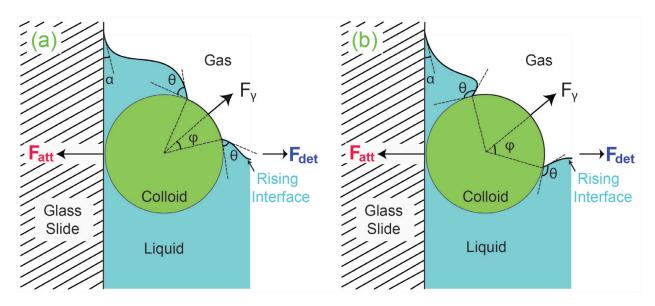


Figure 6. Schematic of forces exerted on an adhered particle: (a) hydrophilic and (b) hydrophobic particle, in contact with a liquid-gas interface. Gravity and buoyancy forces are neglected. (F_{det} : detachment force, F_{att} : attachment force, F_{fl} : surface tension force, θ : contact angle for colloids, α : contact angle for glass surface, ϕ : filling angle).

2.1.1. DLVO forces

The DLVO profiles for the colloids and their interaction with the glass surface were calculated according to [35]:

$$\Delta G_{\rm el} = 64\pi\epsilon R \left(\frac{kT}{ze}\right)^2 \left[\tanh\left(\frac{ze\psi_{0,1}}{4kT}\right)\right] \left[\tanh\left(\frac{ze\psi_{0,2}}{4kT}\right)\right] \exp(-\kappa h) \tag{2}$$

where ΔG_{el} is the electrostatic interaction energy, ϵ is the dielectric permittivity of the medium, R is the radius of the colloids, k is the Boltzmann constant, T is the absolute temperature; z is the ion valence, e is the electron charge, $\psi_{0,1}$ and $\psi_{0,2}$ are surface potential of the colloids and the glass slide respectively, which are taken as the colloid and the glass ζ -potentials, h is

the separation distance, κ is the inverse Debye-Hückel length, $\kappa = \sqrt{\frac{e^2 \sum n_j z_j^2}{\epsilon kT}}$, where n_j is the number concentration of the ions in solution, and z_j is the ion valence.

The van der Waals interaction energy was calculated by [36]:

$$\Delta G_{\rm vdw} = -\frac{AR}{6h} \left[1 - \frac{5.32h}{\lambda_0} \ln \left(1 + \frac{\lambda_0}{5.32h} \right) \right] \tag{3}$$

where *A* is the effective Hamaker constant of colloid-water-glass system, and λ_0 is a characteristic length of 100 nm. The effective Hamaker constant ($A = A_{123}$) was calculated using individual Hamaker constant of colloid, water, and glass [38].

$$A_{123} = (\sqrt{A_{11}} - \sqrt{A_{22}})(\sqrt{A_{33}} - \sqrt{A_{22}}) \tag{4}$$

where A_{11} is the Hamaker constant of the colloids, A_{22} is the Hamaker constant of the fluid, and A_{33} is the Hamaker constant of the glass.

				Experimental Conditions				
Polystyrene	Diameter ^a	Contact	Surface	CaCl ₂	pН	Electrophoretic	ζ-	Colloid
colloids		$angle^b$	charge ^a	conc.	-	mobility ^c	potential ^d	conc.
	(µm)	(deg)	(meq/g)	(mM)	(-)	$(\mu m/s)/(V/cm)$	(mV)	(particles/L)
Amino-modified	$1.0 {\pm} 0.02$	20.3±1.9	0.1047	6	5.9	$0.15 {\pm} 0.02$	$1.9{\pm}0.2$	7.2×10^8

^{*a*}Values provided by manufacturer. ^{*b*}Measured with a goniometer (DSA 100, Krüss, Hamburg, Germany). ^{*c*}Measured with a ZetaSizer 3000HSa (Malvern Instruments Ltd., Malvern, UK) at the electrolyte concentration and pH indicated in the table. ^{*d*}Obtained from measured electrophoretic mobilities using the von Smoluchowski equation [38].

Table 1. Selected properties of polystyrene colloids and suspension chemistry used in the experiments.

Finally, the total DLVO forces were calculated as:

$$F_{\text{DLVO}} = \frac{d}{dh} \left(\Delta G_{\text{tot}} \right) = \frac{d}{dh} \left(\Delta G_{\text{el}} + \Delta G_{\text{vdw}} \right)$$
(5)

To see an example of particle detachment from initially dried glass surfaces, [70] performed experiments by selecting different types of colloids with their modified surface properties. Parameters for the DLVO calculations for one of the colloids are shown in Table 1, and the Hamaker constant was chosen as that for a polystyrene-water-glass system (polystyrene: $A_{11} = 6.6 \times 10^{-20}$ J, water: $A_{22} = 3.7 \times 10^{-20}$ J, glass: $A_{33} = 6.34 \times 10^{-20}$ J; all data taken from Israelachvili [39]; the combined Hamaker constant calculated with equation 4 is $A_{123} = 3.84 \times 10^{-21}$ J).

2.1.2. Surface tension forces

The total force exerted by a moving liquid-gas interface on a colloidal particle is the sum of gravity, buoyancy, and interfacial forces. However, the gravity and buoyancy forces can be neglected for small particles with radii < 500 μ m [57, 58, 64, 70]. In experimental setup, when the liquid-gas interface moves in upward direction over the vertically mounted glass slide, the horizontal component of surface tension force (F_{γ}) is the detachment force (F_{det}) which is opposed by the DLVO force (F_{att}) (Figure 6). The detachment force (the maximum horizontal surface tension force) can be calculated by [45, 57, 58, 64]:

$$F_{\rm det} = 2\pi R \gamma \sin^2\left(\frac{\theta}{2}\right) \cos\alpha \tag{6}$$

where *R* is the radius of the particle, γ is the surface tension of liquid, and θ and α are the advancing contact angles for colloids and the glass slide, respectively.

The experiments were conducted using hydrophilic and hydrophobic modified surface and positively and negatively charged colloids attached over the negatively charged glass slide to estimate the number of colloids removed by moving liquid-gas interface [70]. Colloids over the glass slide were visualized using laser scanning confocal microscopy. Figure 5 shows an example of confocal images before and after moving the liquid-gas interfaces over the glass slide. The figure shows that a considerable amount of colloids were removed by the passage of the first liquid-gas interface (Fig 5a,b), however more number of passages of liquid-gas interface did not affect the colloid left after the first interface movement (Fig 5c,d). This was caused because some of the particles might have attached in the primary energy minimum from the glass slide, so F_{att} would be much larger than F_{det} for those particles [70]. From the DLVO calculations using eq 5, there is a favorable attachment for amino-modified

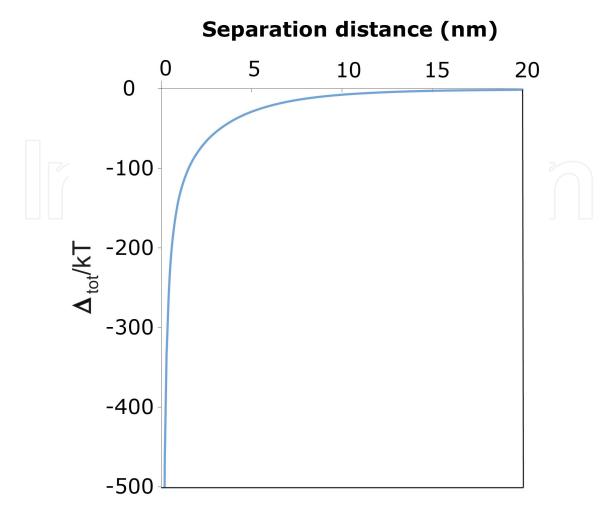


Figure 7. DLVO profile of amino-modified colloid (at given condition in Table 1).

microspheres i.e., a strong attractive force between colloids and the glass surface (fig 7). Shang et al [67] had recently studied the total force balance exerted on a particle passing through a liquid-gas interface. They considered different shape and size (1 to 6 μ m) of particles of different surface properties to pass through a liquid-gas interface and measured the forces exerted on the particle over time using a tensiometer and compared with their theocratical force balance calculations. It has been observed that the liquid-gas interface due to capillary force generates a strong repulsion of particles from the stationary surfaces when water film expands or move through the solid surfaces [67]. In order to detach a particle from the solid surfaces, a liquid film larger than the particle diameter must build up around the particle so that the repulsive capillary force can dominate and a lift can occur.

The above discussion based on force balance complimented with visualization experiments imply that moving liquid-gas interface tends to dominate colloid movement during water infiltration into soils and sediments. The strong force associated at the liquid-gas interface can overcome colloid aggregation and settling, which otherwise dominate colloid dispersion and mobility in porous media. The strong affinity of colloidal particle towards the liquid-gas interface may also be applied in remediation technology, as the inert gases in the form of gas-bubbles can be injected in soils or aquifers to preferentially mobilize colloidal size radioactive contaminants.

2.2. Attachment of colloid by straining

The infiltration and drainage scenarios are quite common in the event of rainfall and drying on the unsaturated zone of the subsurface system. This processes can complex the mobilization of radioactive particles in the upper layers if there is any spill or leakage of those material. In the unsaturated zone as the water drain, sorb or evaporate, the water thickness over the solid surface becomes thinner and thinner; and once the water film becomes thinner than colloid diameters that mechanism is called water film staining. In this case, a strong force exerted on the colloid towards the solid surface which is called capillary force [70, 77, 78, 90]. Other possibility of film straining was explained by the colloid trapped in the pendular rings (smaller pores) region separated by thin water films from the remaining fluids [4, 78], which can be remobilized after expanding the water films [26, 61]. The straining of colloids also happen if the pore sizes are smaller than colloid size. This phenomenon commonly occurs in the saturated zone which can also happen in the unsaturated zone.

Different types of straining mechanism for colloid attachment were studied by Bradford group and others [2–8, 19, 40, 59, 71, 74, 84–86]. Figure 8 shows the different types of straining locations for colloids in the saturated and unsaturated media. Colloids trapped at the intersection point of two solid grains in the saturated systems at location 1 by single and 2 by multiple colloidal particles are also called wedging [40] and bridging [59] respectively. The straining of single particle (location 1) occurs if the pore spaces in a porous medium are smaller than the colloid diameter, which is a common phenomenon applied in mechanical filtration [49]. However, straining of multiple particles (location 2) occur as a result of aggregation of colloidal particles in the solution, although the pore space is larger than the single colloid diameter.

In addition, straining of colloids in the saturated system also depends on solution properties, colloid size, colloid shape, colloid size distribution as well as grain size and heterogeneity [2, 3, 5, 6, 71, 85, 86]. The straining of colloids were more dominant for large, irregular shape, and multi-disperse colloids [84-86]. Straining of colloids in the unsaturated porous media become very complex due to the presence of gaseous phase. The capillary force controls the distribution of liquid and gas phases in the pores. As the amount of liquid decreased from the porous medium, the liquid form a film over the solid surface or retain the smaller pores due to strong capillary forces and the larger pores are filled with gases [75]. Straining behavior of colloids due to pore sizes in the unsaturated systems were not studied yet, however few efforts had been taken on straining of colloid by liquid film [62, 78] and the colloid attachment at the solid-liquid-gas triple point [17, 18, 27, 51, 87, 88]. The example of colloids retained at the solid-liquid-gas triple point are shown in Fig 8 at location 3, which has been discussed in detail in the next section. Straining of colloids in the unsaturated porous media due to liquid film occurred if the liquid thickness is smaller than colloid diameters (location 4 in Fig 8). [78] concluded, using different size of colloids and by changing flow velocity, that colloids with smaller diameter than water film thickness passed easily but colloids bigger than film thickness were trapped on the water film.

2.3. Attachment at the solid-liquid-gas interfaces

The contact point of solid-liquid and liquid-gas is called solid-liquid-gas interface. Steenhuis and coworkers used infiltration chambers, light source, and imaging system (camera setup or confocal microscope) to study the colloids attached at so called air/water-meniscus/solid

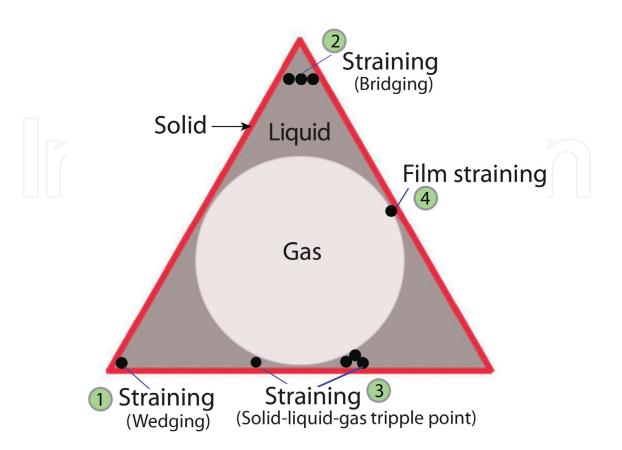


Figure 8. Attachment of colloids in porous media due to straining.

(AW_mS) interface (inside the narrow portion of the pendular ring) in the unsaturated porous media [17, 18, 27, 51, 87, 88]. They established from their visualization experiments that colloids tend to accumulate at AW_mS interface in the unsaturated porous media. They reiterated that the hydrophilic colloids were deposited at AW_mS interface whereas hydrophobic colloids deposited at the solid-liquid interface, but none of them were present at the liquid-gas interface [17, 18]. The capillary force calculation had given the theoretical explanation why the colloids attracted towards AW_mS interface [27]. Their force calculation showed that the colloid retention at AW_mS interface is only possible for hydrophilic colloids with contact angle less than 45° for sand grain medium (Fig 9). So the colloid were not attached at the AW_mS interfaces in the friction coefficient were bellow the tangent of contact angle.

Contrarily, the deposition of colloids were found at the liquid film (liquid-gas interface) from glass micromodel experiments and modeling studies [78, 80]. In another visualization studies, colloids accumulation were found at thin films outside the pendular ring, which was air-water interface not connected with the solid grains [26]. A column and micromodel experiments and thermodynamic calculations showed that colloids were most likely to be retained near the sediments of liquid-gas interface i.e., solid-liquid-gas interface attachment [10]. These discrepancies in the literature between the colloid attachment mechanisms due to the presence of the solid-liquid-gas interface in the unsaturated porous media had been debated [73, 81]. [81] argued that the possible cause of colloid attachment at AW_mS interface was evaporation

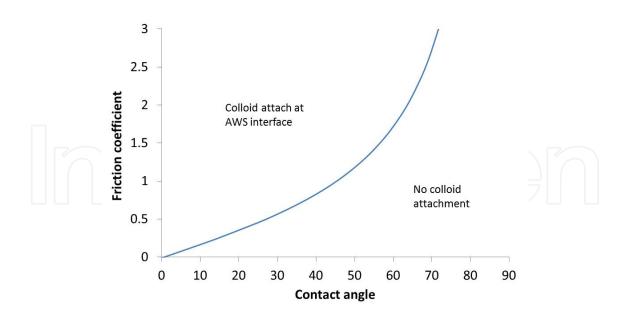


Figure 9. Relationship between grain contact angle and friction coefficient for colloid retention at the AW_mS interface.

in the chamber, drying of thin water film over the grain surface, and advaction of colloids to AW_mS contact line. But it had been refuted by Steenhuis et al [73].

2.4. Attachment at the immobile zones

In the partially saturated systems, colloids were found to be captured into stagnant/immobile zone. There was evidence of exchange of colloids between immobile and mobile zone due to long breakthrough curve tailing on colloid transport through unsaturated porous media [15, 25, 61, 68]. In a visualization study, it was found that colloids present in the immobile zone at the liquid-gas interface were not moved to mobile zone in steady flow, but the exchange of colloids between immobile and mobile zone occurred in varied flow rate [26]. The exchange of colloids between mobile and immobile zones were likely controlled by slow advection in addition to diffusion. The occurrence of larger quantity of colloids from unsaturated column studies were found in transient flow condition due to movement of colloids present in immobile zone [61, 66, 89]. All these studies indicated that the colloid can be attached in the immobile zone created by heterogeneity of the medium and by the presence of gaseous phase, which could be remobilized in the large rainfall and infiltration events.

3. Conclusions and future directions

The study of colloid fate and transport in important as there is strong affinity of radioactive contaminants to attach with the moving colloidal particles or radioactive elements can fall under colloidal size range. In subsurface systems (like soils and sediments) moving air-water interfaces are common, e.g., during infiltration and drainage of water, air and water displace each other in continuous cycles. Such moving air-water interfaces have a profound effect on detachment of colloids from surfaces. Several research efforts had been made to understand the mechanism of colloid retention and mobilization in unsaturated porous media. The

possible cause for colloid attachment in the presence of gaseous phase are discussed in this chapter. As discussed in this chapter, it is difficult to draw firm conclusions about the colloid capture locations in unsaturated porous media. The column experiments, modeling techniques, and visualization studies reveal a number of possible mechanisms of colloid retention and deposition in the partially saturated systems. It is likely that the colloidal particles attached with the solid grain can be removed by moving liquid-gas interface and then colloids can be either deposited and restrained from further moving due to different types of straining, solid-liquid-gas interface capture, and the presence of immobile zone of heterogeneous medium or remain attached at the liquid-gas interfaces.

The strong attachment of radioactive particles to liquid-gas interfaces leading to removal of stationary surfaces offers opportunities for management of subsurface systems in terms of flow and transport. Infiltration fronts in soils can be readily generated by flooding, for instance, and radioactive particle can be effectively "washed" out of a soil profile. Air-bubbles in the form of N₂ or other inert gases may be injected in soils or aquifers to preferentially mobilize and remove radioactive contaminants. Such techniques offer ways to enhance the mobility of otherwise immobile particles in the vadose zone and in groundwater. The results from this study point to the relevance of moving air-water interfaces for nuclear waste mobilization and transport in the vadose zone. Such moving air-water interfaces are common in soils and near-surface sediments, where rainfall, snow melt, or irrigation cause infiltration and drainage. Current theory for colloid transport in unsaturated porous media does not consider the effect of moving air-water interfaces for release of contaminants. Evidently, the colloid removal, transport, and deposition mechanisms remain a fertile area of research with much still left to investigate and opportunities for progress in both theory and experiments that are likely to have significant practical impact in vadose zone fate and transport of colloid attached contaminants for better understanding of any radioactive contamination transport from the release point to farther location.

4. Abbreviations

AW_mS: Air/water-meniscus/solid DLVO: Derjaguin, Landau, Verwey and Overbeek KBS: Kärnbränslesäkerhet

Author details

Prabhakar Sharma Department of Earth Sciences, Uppsala University, Uppsala, Sweden

5. References

- [1] Abdel-Fattah, A. I. & El-Genk, M. S. [1998]. On colloidal particle sorption onto a stagnant air-water interface, *Adv. Colloid. Interface Sci.* 78: 237–266.
- [2] Bradford, S. A. & Bettahar, M. [2005]. Straining, attachment, and detachment of *Cryptosporidium* oocysts in saturated porous media, *J. Environ. Qual.* 34: 469–478.
- [3] Bradford, S. A., Bettahar, M., Šimůnek, J. & van Genuchten, M. T. [2004]. Straining and attachment of colloids in physically heterogeneous porous media, *Vadose Zone J.* 3: 384–394.

- [4] Bradford, S. A. & Torkzaban, S. [2008]. Colloid transport and retention in unsaturated porous media: A review of interface-, collector-, and pore-scale processes and models, *Vadose Zone J.* 7: 667–681.
- [5] Bradford, S. A., Šimůnek, J., Bettahar, M., Tadassa, Y. F., van Genuchten, M. T. & Yates, S. R. [2005]. Straining of colloids at textural interfaces, *Water Resour. Res.* 41: W10404, doi:10.1029/2004WR003675.
- [6] Bradford, S. A., Šimůnek, J., Bettahar, M., van Genuchten, M. T. & Yates, S. R. [2006]. Significance of straining in colloid deposition: Evidence and implications, *Water Resour. Res.* 42: W12S15, doi:10.1029/2005WR004791.
- [7] Bradford, S. A., Šimůnek, J. & Walker, S. L. [2006]. Transport and straining of *E. coli* O157:H7 in saturated porous media, *Water Resour. Res.* 42: W12S12, doi:10.1029/2005WR004805.
- [8] Bradford, S. A., Yates, S. R., Bettahar, M. & Šimůnek, J. [2002]. Physical factors affecting the transport and fate of colloids in saturated porous media, *Water Resour. Res.* 38: 1327, doi:10.1029/2002WR001340.
- [9] Chawla, F., Steinmann, P., Loizeau, J. L., Hossouna, M. & Froidevaux, P. [2010]. Binding of 239pu and 90sr to organic colloids in soil solutions: Evidence from a field experiment, *Environ. Sci. Technol.* 44: 8509–8514.
- [10] Chen, G. & Flury, M. [2005]. Retention of mineral colloids in unsaturated porous media as related to their surface properties, *Colloids Surf. Physicochem. Eng. Aspects* 256: 207–216.
- [11] Chen, G., Flury, M. & Harsh, J. B. [2005]. Colloid-facilitated transport of cesium in variable-saturated Hanford sediments, *Environ. Sci. Technol.* 39: 3435–3442.
- [12] Chen, L., Sabatini, D. A. & Kibbey, T. C. G. [2008]. Role of the air-water interface in the retention of *TiO*₂ nanoparticles in porous media during primary drainage, *Environ. Sci. Technol.* 42: 1916–1921.
- [13] Cheng, T. & Saiers, J. E. [2009]. Mobilization and transport of in situ colloids during drainage and imbibition of partially saturated sediments, *Water Resour. Res.* 45: W08414, doi:10.1029/2008WR007494.
- [14] Cheng, T. & Saiers, J. E. [2010]. Colloid-facilitated transport of cesium in vadose-zone sediments: The importance of flow transients, *Environ. Sci. Technol.* 44: 7443–7449.
- [15] Cherrey, K. D., Flury, M. & Harsh, J. B. [2003]. Nitrate and colloid transport through coarse hanford sediments under steady state, variably saturated flow, *Water Resour. Res.* 39: 1165, doi:10.1029/2002WR001944.
- [16] Crancon, P., Pili, E. & Charlet, L. [2010]. Uranium facilitated transport by water-dispersible colloids in field and soil columns, *Sci. Total Environ.* 408: 2118–2128.
- [17] Crist, J. T., McCarthy, J. F., Zevi, Y., Baveye, P., Throop, J. A. & Steenhuis, T. S. [2004]. Pore-scale visualization of colloid transport and retention in party saturated porous media, *Vadose Zone J.* 3: 444–450.
- [18] Crist, J. T., Zevi, Y., McCarthy, J. F., Troop, J. A. & Steenhuis, T. S. [2005]. Transport and retention mechanisms of colloids in partially saturated porous media, *Vadose Zone J*. 4: 184–195.
- [19] Cushing, R. S. & Lawler, D. F. [1998]. Depth filtration: Fundamental investigation through three-dimensional trajectory analysis, *Environ. Sci. Technol.* 32: 3793–3801.
- [20] Czigany, S., Flury, M., Harsh, J. B., Williams, B. C. & Shira, J. M. [2005]. Suitability of fiberglass wicks to sample colloids from vadose zone pore water, *Vadose Zone J*. 4: 175–183.

- [21] Dai, M., Buesseler, K. O. & Pike, S. M. [2005]. Plutonium in groundwater at the 100k-area of the U.S. DOE Hanford site, *J. Contam. Hydrol.* 76: 167–189.
- [22] Dai, Z., Fornasiero, D. & Ralston, J. [1999]. Particle-bubble attachment in mineral flotation, J. Colloid Interface Sci. 217: 70–76.
- [23] Flury, M., Mathison, J. B. & Harsh, J. B. [2002]. *In situ* mobilization of colloids and transport of cesium in Hanford sediments, *Environ. Sci. Technol.* 36: 5335–5341.
- [24] Flury, M. & Qiu, H. [2008]. Modeling colloid-facilitated contaminant transport in the vadose zone, *Vadose Zone J.* 7: 682–697.
- [25] Gamerdinger, A. P. & Kaplan, D. I. [2001]. Physical and chemical determinants of colloid transport and deposition in water-unsaturated sand and Yucca Mountain tuff material, *Environ. Sci. Technol.* 35: 2497–2504.
- [26] Gao, B., Saiers, J. E. & Ryan, J. N. [2006]. Pore-scale mechanisms of colloid deposition and mobilization during steady and transient flow through unsaturated granular media, *Water Resour. Res.* 42: W01410, doi:10.1029/2005WR004233.
- [27] Gao, B., Steenhuis, T. S., Zevi, Y., Morales, V. L., Nieber, J. L., Richards, B. K., McCarthy, J. F. & Parlange, J. Y. [2008]. Capillary retention of colloids in unsaturated porous media, *Water Resour. Res.* 44: W04504, doi:10.1029/2006WR005332.
- [28] Gephart, R. E. [2010]. A short history of waste management at the hanford site, *Physics Chem. Earth* 35: 298–306.
- [29] Gillies, G., Kappl, M. & Butt, H. [2005]. Direct measurements of particle-bubble interactions, *Adv. Colloid. Interface Sci.* 114–115: 165–172.
- [30] Gomez-Suarez, C., Noordmans, J., van der Mei, H. C. & Busscher, H. J. [1999a]. Detachment of colloidal particles from collector surfaces with different electrostatic charge and hydrophobicity by attachment to air bubbles in a parallel plate flow chamber, *Phys. Chem. Chem. Phys.* 1: 4423–4427.
- [31] Gomez-Suarez, C., Noordmans, J., van der Mei, H. C. & Busscher, H. J. [1999b]. Removal of colloidal particles from quartz collector surfaces as simulated by the passage of liquid-air interfaces, *Langmuir* 15: 5123–5127.
- [32] Gomez-Suarez, C., Noordmans, J., van der Mei, H. C. & Busscher, H. J. [2001]. Air bubble-induced detachment of polystyrene particles with different sizes from collector surfaces in a parallel plate flow chamber, *Colloids Surf*. 186: 211–219.
- [33] Graham, M. C., Oliver, I. W., MacKenzie, A. B., Ellam, R. M. & Farmer, J. G. [2008]. An integrated colloid fractionation approach applied to the characterisation of porewater uranium/Uhumic interactions at a depleted uranium contaminated site, *Sci. Total Environ.* 404: 207–217.
- [34] Graham, M. C., Oliver, I. W., MacKenzie, A. B., Ellam, R. M. & Farmer, J. G. [2011]. Mechanisms controlling lateral and vertical porewater migration of depleted uranium (DU) at two UK weapons testing sites, *Sci. Total Environ.* 409: 1854–1866.
- [35] Gregory, J. [1975]. Interaction of unequal double layers at constant charge, J. Colloid Interface Sci. 51(1): 44–51.
- [36] Gregory, J. [1981]. Approximate expressions for retarded van der Walls interaction, J. *Colloid Interface Sci.* 83(1): 138–145.
- [37] Grolimund, D., Elimelech, M., Borkovec, M., Barmettler, K., Kretzschmar, R. & Sticher, H. [1998]. Transport of in situ mobilized colloidal particles in packed soil columns, *Environ. Sci. Technol.* 32: 3562–3569.
- [38] Hiemenz, P. C. & Rajagopalan, R. [1997]. *Principles of Colloid and Surface Chemistry*, Marcel Dekker Inc., New York.

- 74 Nuclear Power Practical Aspects
 - [39] Israelachvili, J. [1992]. Intermolecular and Surface Forces, Academic Press, London.
 - [40] Johnson, W. P., Li, X. & Yal, G. [2007]. Colloid retention in porous media: Mechanistic confirmation of wedging and retention in zones of flow stagnation, *Environ. Sci. Technol.* 41: 1279–1287.
 - [41] Kersting, A. B., Efurd, D. W., Finnegan, D. L., Rokop, D. J., Smith, D. K. & Thompson, J. L. [1999]. Migration of plutonium in groundwater at thenevada test site, *Nature* 397: 56–59.
 - [42] Kretzschmar, R., Borkovec, M., Grolimund, D. & Elimelech, M. [1999]. Mobile subsurface colloids and their role in contaminant transport, *Adv. Agron.* 66: 121–193.
 - [43] Lazouskaya, V. & Jin, Y. [2008]. Colloid retention at air-water interface in a capillary channel, Colloids Surf. Physicochem. Eng. Aspects 325: 141–151.
 - [44] Lazouskaya, V., Jin, Y. & Or, D. [2006]. Interfacial interactions and colloid retention under steady flows in a capillary channel, *J. Colloid Interface Sci.* 303: 171–184.
 - [45] Leenaars, A. F. M. & O'Brien, S. B. G. [1989]. Particle removal from silicon substrates using surface tension forces, *Philips J. Res.* 44: 183–209.
 - [46] Lenhart, J. J. & Saiers, J. E. [2003]. Colloid mobilization in water-saturated porous media under transient chemical conditions, *Environ. Sci. Technol.* 37: 2780–2787.
 - [47] McCarthy, J. F. & McKay, L. D. [2004]. Colloid transport in the subsurface: Past, present, and future challenges, *Vadose Zone J.* 3: 326–337.
 - [48] McCarthy, J. & Zachara, J. [1989]. Subsurface transport of contaminants, *Environ. Sci. Technol.* 23: 496–502.
 - [49] McDowell-Boyer, L. M., Hunt, J. R. & Sitar, N. [1986]. Particle transport through porous media, *Water Resour. Res.* 22: 1901–1921.
 - [50] McKinley, J. P., Zeissler, C. J., Zachara, J. M., Serne, R. J., Lindstrom, R. M., Schaef, H. T. & Orr, R. D. [2001]. Distribution and retention of Cs-137 in sediments at the Hanford Site, Washington, *Environ. Sci. Technol.* 35: 3433–3441.
 - [51] Morales, V. L., Gao, B. & Steenhuis, T. [2009]. Grain surface-roughness effects on colloidal retention in the vadose zone, *Vadose Zone J.* 8: 11–20.
 - [52] Moridis, G. J., Hu, Q., Wu, Y. S. & Bodvarsson, G. S. [2003]. Preliminary 3-D site-scale studies of radioactive colloid transport in the unsaturated zone at Yucca Mountain, Nevada, J. Contam. Hydrol. 60: 251–286.
 - [53] Noordmans, J., Wit, P. J., van der Mei, H. C. & Busscher, H. J. [1997]. Detachment of polystyrene particles from collector surfaces by surface tension forces induced by air-bubble passage through a parallel plate flow chamber, *J. Adhesion Sci. Technol.* 11: 957–969.
 - [54] Oettel, M. & Dietrich, S. [2008]. Colloidal interactions at fluid interfaces, *Langmuir* 24: 1425–1441.
 - [55] Ouyang, Y., Shinde, D., Mansell, R. S. & Harris, W. [1996]. Colloid enhanced transport of chemicals in subsurface environments: A review, *Crit. Rev. Environ. Sci. Technol.* 26: 189–204.
 - [56] Pedrot, M., Dia, A., Davranche, M., Coz, M. B., Henin, O. & Gruau, G. [2008]. Insights into colloid-mediated trace element release at the soil/water interface, *J. Colloid Interface Sci.* 325: 187–197.
 - [57] Pitois, O. & Chateau, X. [2002]. Small particles at a fluid interface: effect of contact angle hysteresis on force and work of detachment, *Langmuir* 18: 9751–9756.
 - [58] Preuss, M. & Butt, H. [1998]. Measuring the contact angle of individual colloidal particles, J. Colloid Interface Sci. 208: 468–477.

- [59] Ramachandran, V. & Fogler, H. S. [1999]. Plugging by hydrodynamic bridging during flow of stable colloidal particles within cylindrical pores, *J. Fluid Mech.* 385: 129–156.
- [60] Ryan, J. N. & Elimelech, M. [1996]. Colloid mobilization and transport in groundwater, Colloids Surf. Physicochem. Eng. Aspects 107: 1–56.
- [61] Saiers, J. E. & Lenhart, J. J. [2003a]. Colloid mobilization and transport within unsaturated porous media under transient-flow conditions, *Water Resour. Res.* 39: 1019, doi:10.1029/2002WR001370.
- [62] Saiers, J. E. & Lenhart, J. J. [2003b]. Ionic-strength effects on colloid transport and interfacial reactions in partially saturated porous media, *Water Resour. Res.* 39: 1256, doi:10.1029/2002WR001887.
- [63] Salbu, B., Krekling, T. & Oughton, D. H. [1998]. Characterisation of radioactive particles in the environment, *Analyst* 123: 843–849.
- [64] Scheludko, A., Toshev, B. V. & Bojadjiev, D. T. [1976]. Attachment of particles to a liquid surface (Capillary theory of flotation), *J. Chem. Soc. Faraday Trans. I* 72: 2815–2828.
- [65] Sen, T. K. & Khilar, K. C. [2006]. Review on subsurface colloids and colloid-associated contaminant transport in saturated porous media, *Adv. Colloid. Interface Sci.* 119: 71–96.
- [66] Shang, J., Flury, M., Chen, G. & Zhuang, J. [2008]. Impact of flow rate, water content, and capillary forces on in situ colloid mobilization during infiltration in unsaturated sediments, *Water Resour. Res.* 44: W06411, doi:10.1029/2007WR006516.
- [67] Shang, J., Flury, M. & Deng, Y. [2009]. Force measurements between particles and the air-water interface: Implications for particle mobilization in unsaturated porous media, *Water Resour. Res.* 45: W06420, doi:10.1029/2008WR007384.
- [68] Sharma, P., Abdou, H. & Flury, M. [2008]. Effect of the lower boundary condition and flotation on colloid mobilization in unsaturated sandy sediments, *Vadose Zone J*. 7(3): 930–940.
- [69] Sharma, P., Flury, M. & Mattson, E. [2008]. Studying colloid transport in porous media using a geocentrifuge, *Water Resour. Res.* 44: W07407, doi:10.1029/2007WR006456.
- [70] Sharma, P., Flury, M. & Zhou, J. [2008]. Detachment of colloids from a solid surface by a moving air-water interface, *J. Colloid Interface Sci.* 326: 143–150.
- [71] Shen, C., Huang, Y., Li, B. & Jin, Y. [2008]. Effects of solution chemistry on straining of colloids in porous media under unfavorable condition, *Water Resour. Res.* 44: W05419, doi:10.1029/2007WR006580.
- [72] Sirivithayapakorn, S. & Keller, A. [2003]. Transport of colloids in unsaturated porous media: a pore-scale observation of processes during the dissolution of air-water interface, *Water Resour. Res.* 39: 1346, doi:10.1029/2003WR002487.
- [73] Steenhuis, T. S., McCarthy, J. F., Crist, J. T., Zevi, Y., Baveye, P. C., Throop, J. A., Fehrman, R. L., Dathe, A. & Richards, B. K. [2005]. Reply to "comments on 'pore-scale visualization of colloid transport and retention in partly saturated porous media", *Vadose Zone J.* 4: 957–958.
- [74] Torkzaban, S., Bradford, S. A., van Genuchten, M. T. & Walker, S. L. [2008]. Colloid transport in unsaturated porous media: The role of water content and ionic strength on particle straining, *J. Contam. Hydrol.* 96: 113–127.
- [75] Tuller, M. & Or, D. [2001]. Hydraulic conductivity of variably saturated porous media: Film and corner flow in angular pores, *Water Resour. Res.* 37: 1257–1276.
- [76] Utsunomiya, S., Kersting, A. B. & Ewing, R. C. [2009]. Groundwater nanoparticles in the far-field at the Nevada test site: Mechanism for radionuclide transport, *Environ. Sci. Technol.* 43: 1293–1298.

- [77] Veerapaneni, S., Wan, J. & Tokunaga, T. [2000]. Motion of particles in film flow, *Environ*. *Sci. Technol.* 34: 2465–2471.
- [78] Wan, J. M. & Tokunaga, T. K. [1997]. Film straining of colloids in unsaturated porous media: conceptual model and experimental testing, *Environ. Sci. Technol.* 31: 2413–2420.
- [79] Wan, J. M. & Tokunaga, T. K. [2002]. Partitioning of clay colloids at air-water interfaces, J. Colloid Interface Sci. 247: 54–61.
- [80] Wan, J. M. & Wilson, J. L. [1994]. Visualization of the role of the gas-water interface on the fate and transport of colloids in porous media, *Water Resour. Res.* 30(1): 11–23.
- [81] Wan, J. & Tokunaga, T. K. [2005]. Comments on "pore-scale visualization of colloid transport and retention in partly saturated porous media", *Vadose Zone J.* 4: 954–956.
- [82] Wan, J., Tokunaga, T. K., Kim, Y., Wang, Z., Lanzirotti, A., Saiz, E. & Serne, R. J. [2008]. Effect of saline waste solution infiltration rates on uranium retention and spatial distribution in Hanford sediments, *Environ. Sci. Technol.* 42: 1973–1978.
- [83] Williams, D. F. & Berg, J. C. [1992]. The aggregation of colloidal particles at the air-water interface, *J. Colloid Interface Sci.* 152: 218–229.
- [84] Xu, S., Gao, B. & Saiers, J. E. [2006]. Straining of colloidal particles in saturated porous media, *Water Resour. Res.* 42: W12S16, doi:10.1029/2006WR004948.
- [85] Xu, S., Liao, Q. & Saiers, J. E. [2008]. Straining of nonspherical colloids in saturated porous media, *Environ. Sci. Technol.* 42: 771–778.
- [86] Xu, S. & Saiers, J. E. [2009]. Colloid straining within water-saturated porous media: Effects of colloid size nonuniformity, *Water Resour. Res.* 45: W05501, doi:10.1029/2008WR007258.
- [87] Zevi, Y., Dathe, A., Gao, B., Richards, B. & Steenhuis, T. [2006]. Quantifying colloid retention in partially saturated porous media, *Water Resour. Res.* 42: W12S03, doi:10.1029/2006WR004929.
- [88] Zevi, Y., Dathe, A., McCarthy, J. F., Richards, B. K. & Steenhuis, T. S. [2005]. Distribution of colloid particles onto interfaces in partially saturated sand, *Environ. Sci. Technol.* 39: 7055–7064.
- [89] Zhuang, J., McCarthy, J. F., Tyner, J. S., Perfect, E. & Flury, M. [2007]. In situ colloid mobilization in Hanford sediments under unsaturated transient flow conditions: Effect of irrigation pattern, *Environ. Sci. Technol.* 41: 3199–3204.
- [90] Zimon, A. D. [1969]. Adhesion of Dust and Powder, Plenum Press, New York, NY.

