Remediation of Toxic Particles from Groundwater

H. E. Nuttall and Rahul Kale

Department of Chemical/Nuclear Engineering, University of New Mexico, Albuquerque, NM 87131 USA

ABSTRACT

The presence of radioactive colloids (radiocolloids) in groundwater has been documented in several studies. There is significant evidence to indicate that these colloids may accelerate the transport of radioactive species in groundwaters. Because field experiments are often fraught with uncertainties, colloid migration in groundwater is an area of active research and the role and existence of radiocolloids is being investigated. This paper describes an ongoing study to characterize groundwater colloids, to understand the geochemical factors affecting colloid transport in groundwater, and to develop an in-situ colloid remediation process. The colloids and suspended particulate matter used in this study were collected from a perched aquifer site (located at Los Alamos National Laboratory's Mortandad Canyon in Northern New Mexico, USA) that has radiation levels several hundred times the natural background and where previous researchers have measured and reported the presence of radiocolloids containing plutonium and americium. At this site, radionuclides have spread over several kilometers. Inorganic colloids collected from water samples are characterized with respect to concentration, mineralogy, size distribution, and electrophoretic mobility (zeta potential), and radioactivity levels. The methods used to investigate the physiochemical factors affecting colloid transport and some of the preliminary results are presented. These include a description of a colloid transport model and the corresponding computational code, water analyses, characterization of the inorganic colloids, and a conceptual description of a process for in-situ colloid remediation using the phenomenon of polyelectrolyte capture. An overview of this project is presented and preliminary test results are described.

INTRODUCTION

The presence of radioactive colloids (radiocolloids) in groundwater has been documented in several studies (Penrose et al., 1990; McCarthy and Zachara, 1989; Travis and Nuttall, 1985) and the facilitate transport of toxic materials in soils and aquifers by is well documented in the literature (Haque and Freed, 1975; Jury et al., 1986) Hence accelerated colloid transport of radioactive species in groundwater is a serious problem leading to pollution of valuable groundwater. The mechanisms of colloid transport in porous media are not well understood and no colloid remediation technologies exist (Hiemenz, 1986; Jain, 1991; Nuttall, Jain, and Fertelli, 1991; Nuttall and Long, 1992). This paper describes an ongoing study designed to characterize groundwater colloids, to understand the geochemical factors affecting colloid transport in groundwater, and to develop an in-situ colloid remediation process. The colloids and suspended particulate matter used in this study were from a perched aquifer site (located at Los Alamos National Laboratory's Mortandad Canyon in Northern New Mexico, USA). This site has radiation levels several hundred times the natural background and researchers have reported the presence

pump system is illustrated schematically in Figure 3. And as shown in Table I the following matrix of column experiments is in progress.

(with/without polyelectrolyte treatment of the packing)				
Tests	Packing	Dispersed	Continuous	
1)	Quartz	Silica spheres	D.I.water	
2)	Quartz	Mortandad	Well water	
	Canyon Colloids			

Table I. Colloid Remediation Experiments vith/without polyelectrolyte treatment of the packing)

Preliminary results showed that Mortandad colloids passed through columns of packed glass beads with little or no capture on the surface of the glass beads. This finding further corroborates the Zeta potential measurements which showed that the colloids are negatively charged. Column tests using commercial latex microspheres have shown that the polyelectrolyte induced flocculation can be adversely affected by the surfactants introduced during manufacturing. These surfactants effectively inhibited flocculation and rendered the latex colloid unusable for our studies.

Treatment of a site with a dilute polymer solution to create polymer-induced capture on the rock matrix is the most promising colloid remediation strategy we have to date. The theory of polymer-induced flocculation is a common waste water clarification technology (Schwoyer, 1981); however, its application to in-situ colloid transport remediation is new and required development. Polymers in ppm concentrations can adsorb on the surface of particles and then form a polymer-bridge type colloid clusters as illustrated in Figure 1. In practice, a polymer type flood could coagulate and remove colloids from the groundwater thus immobilizing the radioactive nuclides. (Rao, 1992; Kale, 1993; Nuttall et al. 1992; Nuttall and Triay, 1992)

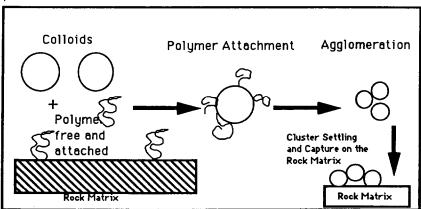


Figure 1. Polymer-Induced Flocculation and Colloid Capture

and migration of radiocolloids containing plutonium and americium (Penrose et al., 1990).

DEVELOPMENT AND TESTING OF COLLOID TRANSPORT REMEDIATION STRATEGIES

An in-situ colloid remediation concept is being developed and tested. This concept treatments a contaminated subsurface zone with dilute polymer solution to create polymer-induced capture on the rock matrix. A schematic diagram of the process on an individual colloid size scale is illustrated in Figure 1. And the process as envisioned for application to a field site is outlined schematically in Figure 2.

Polyelectrolyte induced flocculation has been used in municipal water treatment, and the theory is well developed (Schwoyer, 1981). In the proposed colloid remediation process, a polyelectrolyte solution is injected into the subsurface contaminated zone and the polymer, in very low concentrations, adsorbs onto the rock matrix. Polyelectrolytes are characterized by two properties (i) charge density and (ii) molecular weight. The charge density of the polymer molecule determines the extent of charge neutralization that occurs on the colloid surface when the polymer attaches. The chemical formula for the proposed polyelectrolyte, CATFLOC is poly diallyl dimethyl ammonium chloride. For CATFLOC the monomers have a positive while the colloid surface is negatively charged. The higher the molecular weight and thus the greater the physical length of the polymer the more likely it is to form a bridge between the colloids and the rock matrix. The combination of these two properties of the polyelectrolyte lead to the formation flocs and attachment of colloids to the rock matrix. Ideally the addition of the polymer to a contaminated groundwater site would remove the radiocolloids. Addition of a natural clay type colloid (adsorber) would strongly adsorb the dissolved radionuclides and then polymer bridging (Nuttall, Rao, Jain, Long. and Triay, 1992) could attach these radioactive clay colloids along with natural radiocolloids onto the rock matrix. Flocculation of colloids in the groundwater would also occur as well as attachment of the colloids to the rock matrix.

Both batch and column type laboratory tests, are used in developing of the in-situ colloid remediation process. Tasks include:

• determine efficiency of various polyelectrolytes for removing groundwater colloids.

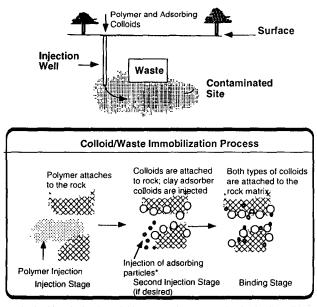
• estimate polyelectrolyte dosage for given water and subsurface media characteristics.

study effects of the following parameters,

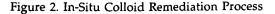
- (i) water chemistry,
- (ii) colloid size and concentration,
- (iii) permeability of the medium, and
- (iv) aging and biodegradation.

• study mechanisms and kinetics of both the polymer attachment to the rock and colloid/rock interactions.

The column studies are performed in small vertical polycarbonate tubes (0.8 cm x 17 cm) with selected packing, colloids, and water composition. The column and syringe



 adsorbing clay/zeolite colloids are added to remove dissolved radionuclides from solution



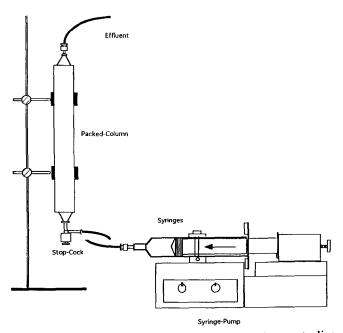


Figure 3. Experimental setup for column studies

Sample	Size (µM)	Zeta Potential (in Ground Water) (mV)	Zeta Potential (in DI water) (mV)
Colloidal Silica	0.300	-42.25	-66.18
Quartz sand	75-300	-25.89	-33.97
Ground Water (well MCO-6B)	1.778	-18.56	
Polystyrene Latex	0.746	-34.72	-41.83
Core Sample	0.1-50	-14.53	-30.19

Table 2. Size, Mobility and Zeta Potential of Samples

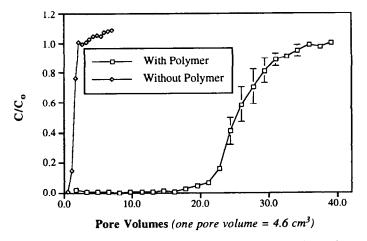


Figure 4. Break-through curves for colloidal silica through quartz sand columns with and without polymer (long runs)

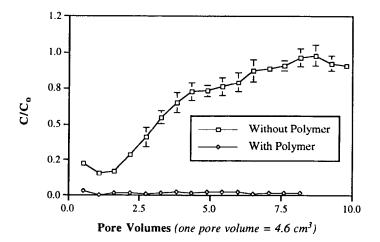


Figure 5. Break-through curves for Mortandad Canyon groundwater colloids with and without polymer (column packing is quartz sand)

The figure shows that the ground water colloids are captured equally well by the polyelectrolyte. The column pre-treated with polymer totally adsorbs all the colloids in 35 cc of suspension passed through it. However, it is observed that there is some adsorption of ground water colloids in the column not treated with the polymer.

CONCLUSIONS

Facilitated transport of radioactive waste in groundwater presents an environmental threat and a challenging restoration problem. The purpose of this study was to investigate radioactive groundwater colloids and to develop a viable in-situ colloid immobilization process. Mortandad Canyon was the chosen investigation site because plutonium and americium have migrated over 2 km in a perch aquifer at this location.

Groundwater colloids from the Mortandad Canyon site within Los Alamos National Laboratory were characterized and studied in a series of packed column experiments under conditions that simulated the natural perched aquifer. Also spherical silica colloids were used as a simulant to represent the natural colloids and provide a better controlled system in which to study colloid transport. Mortandad colloids were composed of feldspar, quartz, and clay and contained trace concentrations of plutonium and americium. The source of colloids is from the surrounding rock which has the same mineral composition. They have a zeta potential of -18 mV indicating that these colloids are electrostatically stabilized. There average size was approximately one micron in diameter. The batch flocculation and column experiments also confirmed the relative stability of the Mortandad colloids. Because of electrostatic stabilization, both Mortandad and silica colloids transported relatively unretarded through the untreated 20 cm quartz packed columns. Treatment of the quartz packing with the polyelectrolyte CATFLOC effectively stopped colloid migration within these laboratory columns. Post analysis of the packing material using an ESEM showed a monolayer absorption coating of colloids on the quartz packing. The mass concentration of the loaded column was x gm of colloids per gram of quartz packing.

This study showed the existence of Mortandad colloids, their chemistry, and charge stabilization properties as well as their ability to transport through a porous media. A polyelectrolyte polymer treatment was successful in stopping colloid migration in laboratory column tests. Further field work is required to demonstrate this technology at an actual site.

ACKNOWLEDGEMENT

The research on which this publication is based was partially financed by the U.S. Department of Energy through the New Mexico Waste Management Education Consortium

REFERENCES

- Haque, Rizwanul and V. H. Freed. Environmental Dynamics of Pesticides. Plenum Press; 1975.
- Hiemenz, Paul C. (1986). Principles of Colloid and Surface Chemistry. Marcel Dekker, Inc.
- Jury, William A. Hesham Elabd, and Margaret Resketo. Field Study of Napropamide Movement Through Unsaturated Soil. Water Resources Research. 1986 May; 22(22): 749-755.
- Jain, Rohit (; 1992) A Model and Computer Code for Particulate Systems, M.S. Thesis, Albuquerque, New Mexico: University of New Mexico.
- Kale, Rahul (1993 in progress) Develop of a Colloid Remediation Process, M.S. Thesis, University of New Mexico.
- McCarthy, John F. and Zachara. John M. (1989) Subsurface Transport of Contaminants (Mobile colloids in the subsurface environment may alter the transport of contaminants. Environmental Science and Technology 23(5), pp. 496-502.
- Nuttall, H. E., Rao, S., Jain, R., Long, R. and Triay, Ines R. (1992) Colloid Remediation in Groundwater by Polyelectrolyte Capture, American Chemical Society Symposium Series. Chapter 6: pp. 71-82.
- Nuttall, H. E. and Long, Richard L.(1992) Mobility of Radioactive Colloidal Particles in Groundwater. Submitted for publication in the Radioactive Waste Management and the Nuclear Fuel Cycle.
- Nuttall, H. E., Jain, Rohit and Fertelli, Yasemin (1991) Radiocolloid Transport in Saturated and Unsaturated Fractures. High Level

Radioactive Waste Management Conference; 1991 Apr 28; Las Vegas, NV. : American Nuclear Society, Inc.; pp. 189-196.

- Nuttall, H. E. and Triay. Ines (1992) Removal of Actinides from Waste Water by Flocculation. Fourth World Congress of Chemical Engineering; 1991 Jun 16; Karlsruhe, Germany. Frankfurt, Federal Republic of Germany; pp. 468-472.
- Penrose, William R., Polzer, Wilfred L, Essington, Edward H., Nelson, Donald M. and Orlandini, Kent A. (1990) Mobility of Plutonium and Americium through a Shallow Aquifer in a Semiarid Region, Environ. Sci. Technol., Vol. 24, No. 2, pgs 228-234.
- Rao, Sudeep (1992) Groundwater Colloids: Characterization, Analysis and the Development of a Remediation Strategy, M.S. Thesis, University of New Mexico.
- Schwoyer, William L. K. (1981) Polyelectrolytes for Water and WasteWater Treatment. CRC Press, Inc.
- Travis, B.J. and Nuttall, H.E. (1985) A Transport Code for Radiocolloid Migration: With an Assessment of an Actual Low-Level Waste Site. Scientific Basis for Nuclear Waste Management VIII 44, pp. 969-976.