

DOWNLOAD PDF IN SITU URANIUM STABILIZATION THROUGH POLYPHOSPHATE REMEDIATION

Chapter 1 : PNNL: EED - Vince Vermeul

In Situ Uranium Stabilization through Polyphosphate Remediation: Development and Demonstration at the Hanford Site Area, Washington State.

Box , Richland, WA ABSTRACT This paper describes the pilot-scale treatability test that was conducted to evaluate the efficacy of using a polyphosphate injection approach to treat uranium-contaminated groundwater in situ within the Area aquifer at the Hanford Site in Richland, Washington. Primary test objectives were to assess 1 direct treatment of available uranium contributing to the groundwater plume through precipitation of the uranyl-phosphate mineral autunite, and 2 emplacement of secondary-treatment capacity via precipitation of the calcium-phosphate mineral apatite, which acts as a long-term sorbent for uranium. Based on an injection design analysis that incorporated results from both bench-scale testing and site-specific characterization activities, a three-phase injection approach was selected for field-scale testing. This approach consisted of 1 an initial polyphosphate injection to facilitate direct treatment of aqueous uranium in the pore space, 2 a second phase consisting of a calcium chloride injection to provide an available calcium source for the creation of apatite, and 3 a subsequent polyphosphate injection to supply a phosphate source for the formation of apatite. The total-solution volume injected during this field test was approximately 3. In addition, data from this test will provide valuable information for designing a full-scale remedial action for uranium in groundwater beneath the Area of the Hanford Site, and a detailed understanding of the fundamental underpinnings necessary to evaluate the efficacy and potential for utilization of the polyphosphate technology at other sites with varying geochemical and hydrodynamic conditions. The remedial action objective of the ROD was reduction of groundwater uranium to the U. Because the remedial action objectives of the ROD were not met, a Phase III Feasibility Study was initiated in to identify and evaluate remedial alternatives that will accelerate monitored natural attenuation of the uranium plume. For fiscal year , the U. After a peer review and selection process, nine projects were selected to meet the objectives of the appropriation. This technology works by forming uranyl- and calcium-phosphate minerals autunite and apatite in the aquifer that directly sequester the existing aqueous uranium in autunite minerals and precipitates apatite minerals for sorption and long-term treatment of uranium migrating into the treatment zone, thus reducing current and future 1 WM Conference, February , , Phoenix, AZ aqueous uranium concentrations. Polyphosphate injection was selected for testing based on technology screening as part of the FF-5 Phase III Feasibility Study for treatment of uranium in the Area. A groundwater plume containing uranium from past-practice discharges of liquid waste associated with nuclear fuel-fabrication activities has persisted beneath the Area for many years. This plume originated from a combination of purposeful discharges of wastewater to cribs, trenches, and ponds, along with some accidental leaks and spills during nuclear fuel-fabrication activities, and is identified as the FF-5 Operable Unit. Elevated uranium concentrations are transported through the high-conductivity sandy gravels of the Hanford formation and enter the nearby Columbia River shoreline and the riparian and river biota through seeps. In the current conceptual site model [1, 2], researchers assume that resupply of the plume is occurring, with continuing release coming primarily from the deep vadose zone beneath waste sites and the zone of water table fluctuations. Comparison of uranium concentrations in groundwater at high- and low-river stage conditions provides some insight as to the distribution of uranium contamination in the deep vadose zone. During high-water conditions, elevated uranium concentrations are observed in localized areas that can be attributed to past-disposal operations. These increases in uranium concentration are most likely associated with contamination remaining in the deep vadose zone and zone of water table fluctuations associated with changes in Columbia River stages. The polyphosphate treatability test site is located near one of the two delineated deep vadose sources. The persistence of this plume is enigmatic for several reasons, including 1 discharges containing uranium-bearing effluent to ground disposal sites ended in the mids; 2 contaminated soil associated with these waste sites was removed during the s, with backfilling complete by early ; and 3 the aquifer is

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comprised of highly transmissive fluvial sediment, suggesting rapid movement of groundwater. This paper provides a preliminary status of pilot-scale treatability test activities for the polyphosphate technology, which is currently ongoing. Topics include brief descriptions of the technology and field test site; associated characterization activities that were performed, including a large-scale tracer injection and drift test; a discussion of the field-scale injection design analysis approach; a description of test operations; and a discussion of performance assessment monitoring data collected to date. Although only a preliminary assessment of the ability of the technology to meet remedial objectives is presented in this paper, available test results do provide valuable insight into implementation challenges at this site and will form the basis for designing additional bench- and field-scale testing as required to develop an injection approach that meets site remedial objectives. Precipitation of phosphate minerals occurs when phosphate compounds degrade in water, due to hydrolysis, to yield the orthophosphate molecule PO_4^{3-} . The rate of the hydrolysis reaction that leads to production of orthophosphate is related to the length of the polyphosphate chain. Release of uranium from the autunite structure may only occur through dissolution of the autunite structure. Extensive testing demonstrates the very low solubility and slow-dissolution kinetics of autunite under conditions relevant to the Hanford Site 2 WM Conference, February 1998, Phoenix, AZ subsurface [3]. In addition to autunite, excess phosphorous can result in apatite mineral formation, providing a long-term source of treatment capacity. Results Previous research [8, 9] demonstrated the efficacy of hydroxyapatite for reducing the aqueous uranium suggested the binding of uranium, irrespective of dissolved carbonate concentration or aqueous uranium concentration, occurred via surface complexation; long-term retention occurred through the transformation of sorbed apatite to chernikovite. Similar evidence for the long-term retention of uranium via initial sorption and subsequent transformation to uranium mineral phases of low solubility has been observed down gradient of the uranium ore deposit at Koongarra, Australia [10]. The location is sufficiently far inland from the Columbia River such that transport of river water during normal high-river stage conditions does not reach the location, an impact that would complicate test result interpretations. Pressure responses associated with fluctuations in river stage are significant at this location and must be considered when analyzing water-level trends and hydraulic test response data. The site was selected due to its proximity to a known uranium source area and because of its contaminant profile; i. Depth-discrete uranium concentrations in groundwater samples collected during drilling of the test site injection well, Fig. A total of 16 wells were installed in support of pilot-scale field test, including one injection well 23 and 15 monitoring wells Fig. One additional monitoring well A that was installed in support of the Hanford Site groundwater monitoring network was also used as a downgradient monitoring location. The uppermost unconfined aquifer beneath the polyphosphate treatability test site is composed of unconsolidated gravel-dominated sediments of the Hanford formation, which consists predominantly of coarse sandy gravel to gravel; where present, the matrix sand is composed of medium to coarse sand. The Ringold Formation that lies beneath the Hanford formation is composed of mostly gravelly silty sand to sand. The Hanford - Ringold contact boundary ranged in depth between approximately 10 to 20 feet. Typical depths to water range from 10 to 20 feet. Results from hydraulic tests conducted at the field test site indicate that hydraulic conductivity in the Hanford formation is up to three orders of magnitude higher than that in the underlying Ringold Formation, which acts as a lower bounding aquitard at this location and effectively isolates treatability test injection operations to the high-permeability Hanford formation. Well location map and generalized construction diagram for the treatability test injection and monitoring well network. During installation of the injection well, depth-discrete slug withdrawal tests were conducted throughout the underlying Ringold Formation, resulting in hydraulic conductivity estimates ranging from 0.01 to 0.1 ft/d. Pressure-buildup data collected during a Tracer Injection and Drift Test A tracer injection test was conducted at the polyphosphate treatability test site on December 13, 1998, [12]. The objective of the tracer test was to evaluate formation heterogeneities, to assess the downgradient transport of the tracer plume i. Results from the tracer test provided information on 4 WM Conference, February 1998, Phoenix, AZ the effective porosity of the aquifer, expected arrival times at the monitoring wells, and polyphosphate solution volume requirements for the

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targeted treatment zone thickness and radial extent. The tracer test was conducted by injecting a solution containing a conservative, nonreactive bromide Br tracer into the central injection well, as shown in Fig. Bromide was measured in the injection stream and the surrounding monitoring wells to determine the arrival times and spatial extent of the tracer plume. The injection stream was maintained at a constant rate of L gal per minute throughout the test duration for. In addition to the collection of aqueous samples, downhole Ion Selective Electrode ISE probes continuously monitored bromide concentrations in the wells during the test. A total of aqueous samples were collected from the injection stream and surrounding monitoring wells and were analyzed in the field laboratory trailer for bromide using an ISE probe. Specific conductance SpC, dissolved oxygen DO, pH, oxidation-reduction potential ORP, and temperature were also measured using an in-line electrode in the sampling manifold. The ion chromatography IC analyses were conducted on each of the archive samples at an offsite laboratory as an additional method of measuring bromide concentration. For wells located within a radial distance of 8. These results indicate a general correlation between tracer arrival time and radial distance from the injection well, with a few notable outliers that provide some insight as to the level of heterogeneity present at this site. Effective porosities were calculated for each of the eight monitoring wells within the targeted injection volume, except for the two extreme outlier wells. This value is consistent with porosity estimates based on physical property analysis of core samples collected from the site [11]. In addition to monitoring wells located within the injection pore volume, two additional monitoring wells were monitored downgradient from the injection well beyond the radial extent of the targeted injection volume; wells Fig. The tracer arrival curve for well Fig. For this reason, hydraulic conductivities were estimated for both the interpreted preferential flow path, resulting in an early tracer arrival and the bulk porous media attributed to transport of the main plume body. Breakthrough curves showing bromide concentrations through time for downgradient wells a and b. The estimated hydraulic conductivity using these parameters is about 4, m 14, ft per day. The fast-path hydraulic conductivity was calculated using the same equation used for the main tracer plume, but with some notable differences in the sources of the parameter values. The breakthrough curve for well, the more distant downgradient monitoring well radial distance of m [ft], shows a more dispersed tracer plume arrival Fig. Although first arrival of the tracer plume at this location is generally consistent with the 15 m 50 ft per day velocity calculated from the well tracer arrival data i. Results from the bromide tracer injection and drift test provide valuable information for design of the polyphosphate treatability test. For example, the, L, gal injected during the tracer test appears to be a suitable volume to create a 9-m ft diameter pore volume. Lastly, the equipment and sampling methods used in the tracer test were successful at capturing arrival responses and should be suitable for operation and monitoring of the polyphosphate injection. This simplified approach for evaluation of tracer injection and drift data provides for a quantitative estimate of treatability test-scale transport properties and variability in those properties, and forms the basis for a more technically rigorous evaluation based on local-scale flow and transport modeling [12]. Results from both of these analyses were used to develop a field-scale injection design. This analysis was based on the results of bench-scale studies, the tracer injection and drift test, and the use of analytic and numerical models [12]. The injection volumes, which are based on the observed arrival of conservative species during the tracer injection test, have been increased to account for increased aquifer thickness during spring high-river stage conditions. These volumes were also increased to account for retardation associated with the reactive species used for the polyphosphate treatability test, as determined based on bench-scale laboratory tests. The injection volume for a conservative species at the polyphosphate treatability test site was estimated based on the results of the bromide tracer test and adjusted volumetrically to account for the higher river stage conditions expected for the polyphosphate injection in June. The tracer arrival data presented in Table I show there is significant variability in the tracer arrival due to heterogeneities at the site. In addition to this tabular comparison, results of an analytic solution for advection and dispersion for wells around an injection well at different radial distances [13] were used to assess tracer arrivals. This analytic solution assumes a homogeneous, isotropic aquifer with constant thickness. Comparisons with the analytic solution were used to show the relative

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differences between the measured tracer arrivals and the expected response assuming homogeneous conditions. This approach allowed for the wells to be categorized as fast or slow relative to this measure. Based on these comparisons, most of the wells southeast of the injection well had slower tracer arrivals than wells in other directions. Well , to the southwest of the injection well, had a very rapid arrival indicating preferential flow in that direction. Wells to the northeast and northwest had similar arrivals to the average expected at the site. This response may be associated with the same higher conductivity feature that results in early arrival at lower zone monitoring well. Chemical requirements for the polyphosphate treatability test were based on 1 bench-scale experiments designed to determine reactive species retardation and rate information, and 2 the determination of the fluid-volume requirements based on the tracer test for treatment of a specified volume in the subsurface. Based on this evaluation, a field-scale injection approach was developed that was composed of three separate injection phases. The first phase consisted of injecting a polyphosphate amendment for sequestration of aqueous uranium in phosphate mineral phases i. This approach relied on bench-scale results that showed both calcium and phosphate species would sorb to formation sediments, helping to facilitate mixing between these two required components for apatite formation. The injection volume for the calcium-chloride solution in the second phase was scaled up to 1, L, gal based on a retardation factor of 4. The injection rate was specified at L gal per minute, the same rate used during the tracer injection test. Table II presents the pilot-scale treatability test amendment formulation. Moreover, the values are not independent solubility values; rather, they are the maximum solubility within the total polyphosphate formulation. The amendment solution was prepared by mixing in order the sodium orthophosphate, sodium pyrophosphate, and sodium tripolyphosphate to achieve a pH of 7 and prevent degradation of polymerized phosphate molecules during preparation of the remedy solution. As indicated above, the injection design specified that the test be conducted in three separate phases. The first phase consisted of a , L , gal injection of polyphosphate solution, followed by a second phase consisting of 2, L , gal of calcium-chloride solution. The third phase consisted of another , L , gal injection of polyphosphate solution. All injection phases were conducted at a constant rate of L gal per minute, for a total test duration of 3. Solute concentrations were held constant during each phase of the injection and analytes of interest were measured in the injection stream and the surrounding monitoring wells to determine arrival times at each location and assess the overall extent of treatment.

Chapter 2 : PNNL: EED - Dawn Wellman

A site specific treatability test was conducted to optimize polyphosphate remediation technology for implementation through a field-scale technology demonstration to accelerate monitored natural.

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