HNF-42335-FP
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Potential Use of Activated Carbon to Recover Tc-99 from 200 West Area Groundwater as an Alternative to More Expensive Resins, Hanford Site, Richland, Washington

Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Contractor for the U.S. Department of Energy
under Contract DE-AC06-08RL14788

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Date Published
November 2009

To Be Presented at
Waste Management 2010 Conference

Waste Management Symposia, Inc.
Phoenix, Arizona

March 7-11, 2010

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Printed in the United States of America

Potential Use of Activated Carbon to Recover Tc-99 from 200 West Area Groundwater as an Alternative to More Expensive Resins, Hanford Site, Richland, Washington – 10230

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ABSTRACT

Recent treatability testing performed on groundwater at the 200-ZP-1 Operable Unit at the Hanford Site in Richland, Washington, has shown that Purolite®[1] A530E resin very effectively removes Tc-99 from groundwater. However, this resin is expensive and cannot be regenerated. In an effort to find a less expensive method for removing Tc-99 from the groundwater, a literature search was performed. The results indicated that activated carbon may be used to recover technetium (as pertechnetate, TcO_4^-) from groundwater. Oak Ridge National Laboratory [1] used activated carbon in both batch adsorption and column leaching studies. The adsorption study concluded that activated carbon absorbs TcO_4^- selectively and effectively over a wide range of pH values and from various dilute electrolyte solutions (<0.01 molarity). The column leaching studies confirmed a high adsorption capacity and selectivity of activated carbon for TcO_4^-. Since activated carbon is much less expensive than Purolite A530E resin, it has been determined that a more extensive literature search is warranted to determine if recent studies have reached similar conclusions, and, if so, pilot testing of 200-ZP-1 groundwater will likely be implemented. It is possible that less expensive, activated carbon canisters could be used as pre-filters to remove Tc-99, followed by the use of the more expensive Purolite A530E resin as a polishing step.

INTRODUCTION

The 200-ZP-1 Groundwater Operable Unit is one of two groundwater operable units located in the 200 West Groundwater aggregate area of the Hanford Site near Richland, Washington. An interim pump-and-treat system for the operable unit was implemented in 1994 to control the 2,000 µg/L contour of a 13-square-kilometer (5-square-mile) carbon tetrachloride plume associated with the Plutonium Finishing Plant. In addition to carbon tetrachloride, other groundwater contaminants found in the 200 West Area include Tc-99, trichloroethylene, chromium, hexavalent chromium, I-129, nitrate, tritium, and uranium.

A final Record of Decision for the 200-ZP-1 Operable Unit was issued September 30, 2008. The selected remedy is a robust groundwater pump-and-treat system combined with flow-path control, monitored natural attenuation, and institutional controls. The final pump-and-treat system is currently being designed to treat groundwater at a rate of 9464 L/min (2500 gal/min) and is expandable to 14005 L/min (3700 gal/min). The well network may include as many as 20 extraction wells and 16 injection wells, as shown in Figure 1. The selected treatment process includes the use of ion-exchange resins (e.g., Purolite® A530E and Dowex®[2] 21K) to remove Tc-99, I-129, and uranium from the groundwater. The groundwater then enters fluidized bed reactors for treatment of nitrate, carbon tetrachloride, and trichloroethylene, as well as to reduce hexavalent chromium to insoluble trivalent chromium. Air stripping is used as a final polishing step to remove any volatile organics that may have remained after passing through the fluidized bed reactors. Figure 2 shows the general layout of the 200 West Area groundwater treatment facility.

[1] Purolite® is a registered trademark of The Purolite Company, Bala Cynwyd, Pennsylvania.
[2] Dowex® is a registered trademark of The Dow Chemical Company, Midland, Michigan.

Fig. 1. Final groundwater extraction well and injection well network.

While recent treatability testing performed on 200-ZP-1 groundwater has shown that Purolite A530E resin very effectively removes Tc-99 from treated groundwater, this resin is expensive. In an effort to find a less expensive method for removing Tc-99 from the groundwater, a preliminary literature search was performed. The results of this search found that activated carbon may be used to recover technetium (as pertechnetate, TcO_4^-) from contaminated groundwater (Figure 3). Oak Ridge National Laboratory [1] used activated carbon in both batch adsorption and column leaching studies. The adsorption study concluded that activated carbon absorbs TcO_4^- selectively and effectively over a wide range of pH values and from various dilute electrolyte solutions (<0.01 molarity). The column leaching studies confirmed a high adsorption capacity and selectivity of activated carbon for TcO_4^-.

Since activated carbon is much less expensive than Purolite A530E resin, it has been determined that a more extensive literature search is warranted to determine if recent studies have reached conclusions similar to the 1996 Oak Ridge National Laboratory study [1], and, if so, pilot testing on 200-ZP-1 groundwater will likely be implemented. It is possible that less expensive, activated carbon canisters could be used as pre-filters to remove Tc-99, followed by the use of the more expensive Purolite A530E resin as a polishing step.

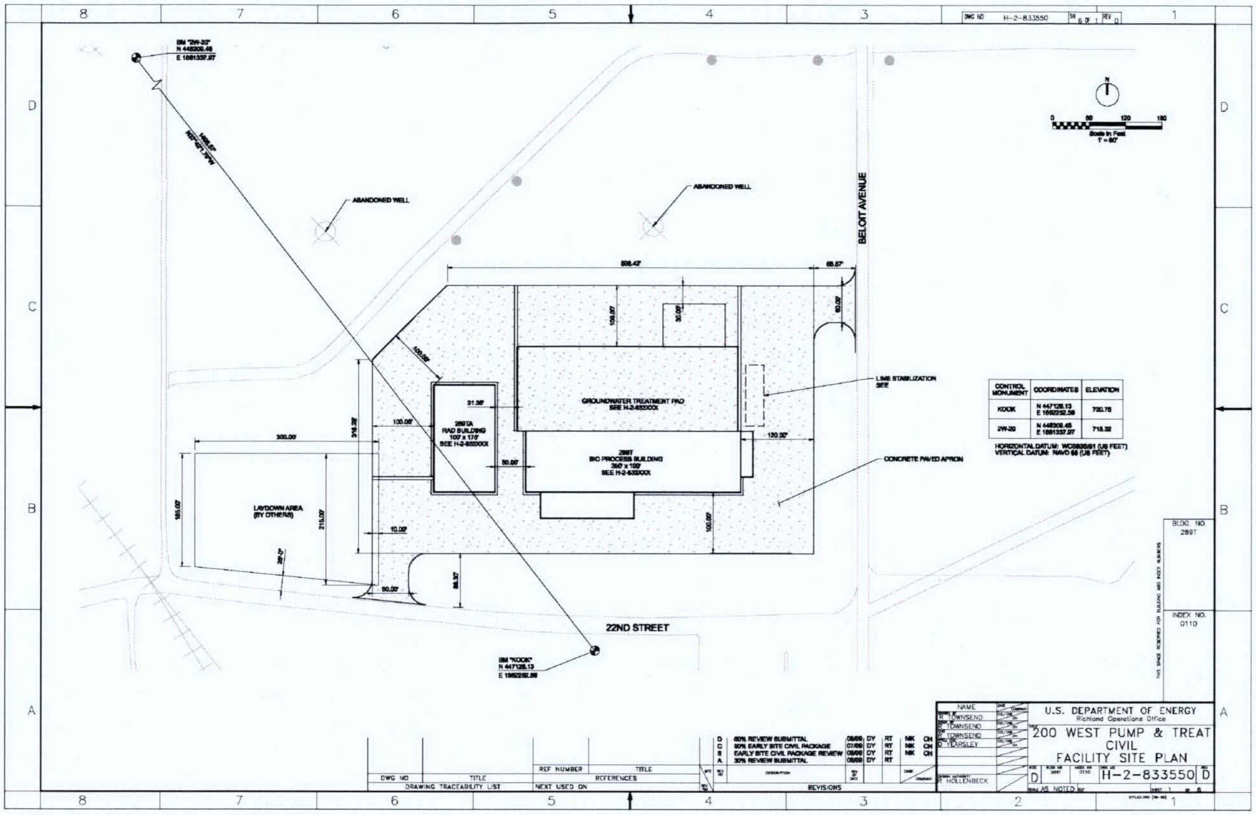

Fig. 2. General layout of the 200 West Area groundwater treatment facility.

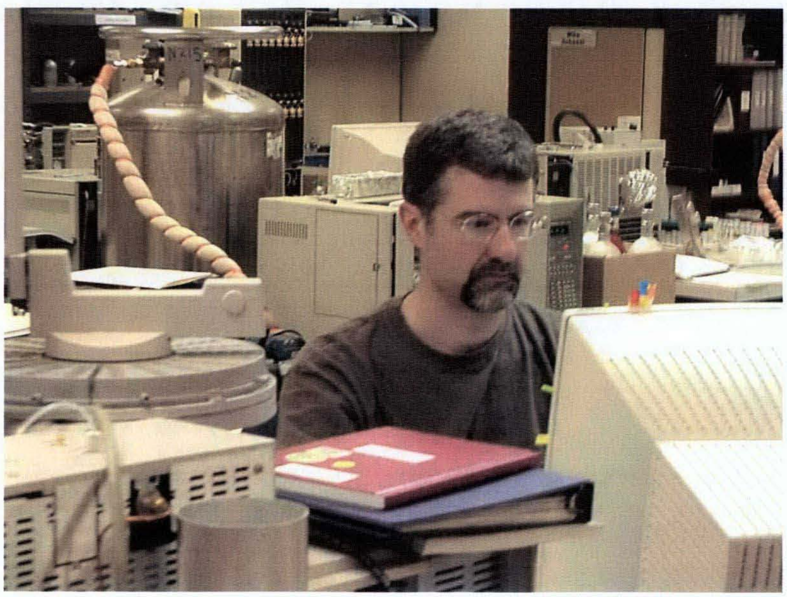

Fig. 3. Laboratory studies have shown that activated carbon may be used to recover technetium.

PROPOSED TESTING PROGRAM

The proposed testing program has been divided into four separate parts. The objectives of testing several commercial sources of carbon are to determine their effectiveness at recovering Tc-99 and their sensitivity to a variety of physical parameters.

Part I

Initial testing will characterize carbon from several commercial sources based on acid-base titration curves. This information will help identify carbon types with various Tc-99 uptake characteristics, such as sensitivity to pH, potential range of distribution coefficient values, and possibly, the approximate Tc-99 loading capacity (based on distribution of functional sites resulting from activation).

Work at Sandia National Laboratories [2] showed that the binding strength of Tc-99 on activated carbon depended on the population of certain carboxylic acid functional groups on the carbon. The concentrations of these functional groups were measured by constructing titration curves of acid dosage as a function of solution pH for slurries of activated carbon.

Carbon titration curves will be used to select a representative group of commercially available carbon types for batch and continuous-flow tests of Tc-99 uptake. Efforts will be made to correlate titration curves with activation methods and approximate distribution coefficients (i.e., distribution coefficient values from published reports).

Parts II and III

Parts II and II of testing will evaluate (a) the basic characteristics of Tc-99 loading, loading rate, and total capacity from batch testing; and (b) process performance under continuous-flow operation (granular or powdered forms may be tested).

In Part II of testing, batch uptake of Tc-99 by representative types of activated carbon will be used to confirm published correlations of carbon functional groups with Tc-99 uptake characteristics. These tests will be used to evaluate Tc-99 uptake kinetics and to calculate partitioning coefficients of Tc-99 between aqueous and solid (carbon) phases, which in turn will serve as guidelines for choosing carbon types continuous-flow column tests.

The Tc-99 loading information will also provide insight about waste disposal. High Tc-99 concentrations may be prohibited from land disposal at the Hanford Site, while low loading may make activated carbon unattractive as a pre-treatment method ahead of the more expensive ion-exchange resin. Potential points for using activated carbon and reducing the use of ion-exchange resin would be evaluated after Tc-99 uptake characteristics are established to help identify a water source for continuous-flow tests.

Part III – Continuous-Flow Column Tests

In Part III of testing, continuous-flow column tests using granular activated carbon will be performed with the type(s) of carbon that best suit the treatment objectives and that fall within known constraints of Tc-99 uptake and disposal. Packed columns will be operated either individually, in series, or with sample taps along the flow path, depending on contact-time requirements and expected removal efficiencies determined by batch tests. Continuous-flow testing will provide information regarding the sharpness of the contaminant interface along the column, the Tc-99 loading capacity, and concentration distribution along the column.

Part IV

Finally, in Part IV of the testing, technetium-containing carbon would be tested for stability against Tc-99 desorption by establishing a baseline, by evaluating "conventional" pozzolanic (high-pH cement or fly ash) treatment, and by near-neutral chemical stabilization by Ceramicrete®[3]. This testing is necessary to understand whether Tc-99 captured by the carbon can be disposed at the Hanford Site's Environmental Restoration Disposal Facility and whether the Tc-99 would remain immobilized.

The Tc-99 adsorption by carbon is either weakly or strongly affected by the pH of the solution in which it is in contact, but the general trend is toward weaker binding at higher pH.

Pozzolanic stabilization with cement, lime fly ash, or combinations of these constituents is most common because it is the most economical approach. However, pozzolanic materials are alkaline, increasing the risk of Tc-99 release through the porous structure. Thus, the stabilized, activated carbon containing Tc-99 will be characterized for structural stability and leaching properties. A near-neutral stabilizing material known commercially as "Ceramicrete" will also be tested for structure and Tc-99 leaching characteristics.

The structural and chemical stabilities of these two stabilization methods will be evaluated and compared for economic and technical performance.

CONCLUSIONS

The results from testing several commercial sources of carbon will be used to determine the effectiveness of using activated carbon to recover technetium (as pertechnetate, TcO_4^-) from groundwater. This testing will identify which commercial source of activated carbon provides the best uptake of Tc-99 and will identify the sensitivity to pH, potential range of distribution coefficient values, and possibly the approximate Tc-99 loading capacity (based on distribution of functional sites resulting from activation). The testing will also provide general information on Tc-99 loading rates and total capacity from batch testing. Granular or powdered forms of activated carbon will likely be evaluated. Finally, carbon containing Tc-99 would be tested for structural integrity and chemical stability against Tc-99 desorption by establishing a baseline, by evaluating "conventional" pozzolanic (high-pH cement or fly ash) treatment, and by near-neutral chemical stabilization by Ceramicrete. This testing is necessary to understand whether technetium captured by the carbon might provide a practical and economical pre-treatment process to reduce the operating cost of ion-exchange resin.

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[3] Ceramicrete is a registered trademark of UChicago Argonne, LLC Corporation Illinois, Chicago, Illinois.