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Uranium Adsorption on Granular Activated Carbon – Batch Testing

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September 2013



Pacific Northwest
NATIONAL LABORATORY

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Richland, Washington 99352

Summary

The uranium adsorption performance of two activated carbon samples (Tusaar Lot B-64, Tusaar ER2-189A) was tested using unadjusted source water from well 299-W19-36. These batch tests support ongoing performance optimization efforts to use the best material for uranium treatment in the Hanford Site 200 West Area groundwater pump-and-treat system. A linear response of uranium loading as a function of the solution-to-solid ratio was observed for both materials. K_d values ranged from ~380,000 to >1,900,000 ml/g for the B-64 material and ~200,000 to >1,900,000 ml/g for the ER2-189A material. Uranium loading values ranged from 10.4 to 41.6 $\mu\text{g/g}$ for the two Tusaar materials.

Acronyms and Abbreviations

μg	microgram(s)
$\mu\text{g/g}$	microgram(s) per gram
μm	micrometer(s) or micron(s)
DOE	U.S. Department of Energy
EPA	U.S. Environmental Protection Agency
GAC	granular activated carbon
ICP-MS	inductively coupled plasma-mass spectrometer
OU	operable unit
K_d	distribution coefficient(s)
mg/g	milligram(s) per gram
mg/L	milligram(s) per liter
ml/g	milliliters per gram
PNNL	Pacific Northwest National Laboratory

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1.0 Introduction

CH2M HILL Plateau Remediation Company is currently operating the Hanford Site 200 West Area groundwater pump-and-treat system as the remedial action selected under the Comprehensive Environmental Response, Compensation, and Liability Act Record of Decision for Operable Unit (OU) 200-ZP-1. This report documents the results of treatability tests Pacific Northwest National Laboratory (PNNL) researchers conducted to quantify the ability of selected granular activated carbon (GAC) products to adsorb uranium from 200 West Area groundwater.

Laboratory sorption experiments included batch tests from which the sorption characteristics of each of the GAC materials were quantified. PNNL researchers tested two GAC materials from Tusaar using batch techniques. The goal of the batch testing was to determine the uranium-loading capacity of these materials.

2.0 Experiment

2.1 Material Preparation

2.1.1 Carbon Material

Carbon material was used as received from Tusaar. The two materials received were B-64 and ER2-189A. No additional preparation was required.

2.1.2 Source Water

Water from well 299-W19-36 was used for testing. The water was sparged for five days in the original received container to remove any volatile organics. Sparging also lowered the pH of the water to 7. Water was filtered through a 0.45- μm filter before use in the batch tests.

2.2 Batch Isotherm Tests

The following resin-to-solution ratios were used for the batch isotherm tests:

- Ratio #1 – 1.00 g resin/50 ml solution
- Ratio #2 – 0.75 g resin/50 ml solution
- Ratio #3 – 0.50 g resin/50 ml solution
- Ratio #4 – 0.25 g resin/50 ml solution.

All batch adsorption tests were conducted for 18 hours. During that time, the centrifuge tubes were agitated continuously at room temperature (22 °C) so the carbon and groundwater remained well mixed. After approximately 18 hours of contact time, the tubes were allowed to stand for five minutes while the carbon settled. A 10-mL sample was obtained and filtered through a 0.45- μm filter. The first 5-mL aliquot of filtered sample went back into the sample tube. The second 5-mL aliquot was analyzed for uranium. See Appendix A for a description of analysis procedure. The batch-testing procedure was repeated for each carbon sample.

3.0 Results

3.1 Source Water

ICP-MS analyses were conducted on the source water upon receipt to quantify the concentrations of iodine-127, technetium-99, uranium, and nitrate. Results are given in Table 3.1. The analytical error associated with ICP-MS is $\pm 15\%$.

Table 3.1. Concentrations of Specified Constituents in 299-W19-36 Source Water

Constituent	Units	As Received
Iodine-127	$\mu\text{g/L}$	12.2
Technetium-99	$\mu\text{g/L}$	1.57
Uranium	$\mu\text{g/L}$	197
Nitrate	$\mu\text{g/mL}$	325

3.2 Batch Tests

The results of the batch isotherm tests for the Tusaar B-64 carbon material are shown in Table 3.2 and Figure 3.1. Batch isotherm results for the Tusaar ER2-189A carbon material are shown in Table 3.3 and Figure 3.2. For both materials, some of the final uranium concentrations were below the instrument detection limit. These samples are marked with an asterisk in Table 3.2 and Table 3.3. A linear response of uranium loading as a function of the solution-to-solid ratio was observed for both materials as shown in Figure 3.1 for B-64 and Figure 3.2 for ER2-189A. K_d values ranged from $\sim 380,000$ to $>1,900,000$ ml/g for the B-64 material and $\sim 200,000$ to $>1,900,000$ ml/g for the ER2-189A material. Uranium loading values ranged from 10.4 to 41.6 $\mu\text{g/g}$ for the two Tusaar materials.

Table 3.2. Uranium-Adsorption Data for Tusaar B-64 GAC

Sample #	U Initial Conc. ($\mu\text{g/L}$)	U Final Conc. ($\mu\text{g/L}$)	GAC Mass (g)	Soln. Vol. (ml)	U Ads. ($\mu\text{g/g}$)	K_d (ml/g)
B64-uranium-pH7-1	207.9	0.027	0.9991	50.007	10.40	381,395
B64-uranium-pH7-2	207.9	0.027	1.0009	50.092	10.40	382,689
B64-uranium-pH7-3	207.9	0.014	0.7491	50.015	13.88	962,942
B64-uranium-pH7-4*	207.9	<0.011	0.7512	50.146	13.88	>1,309,204
B64-uranium-pH7-5*	207.9	<0.011	0.5002	50.013	20.79	>1,960,946
B64-uranium-pH7-6*	207.9	<0.011	0.4993	49.959	20.80	>1,962,360
B64-uranium-pH7-7	207.9	0.058	0.2503	50.068	41.58	717,665
B64-uranium-pH7-8	207.9	0.042	0.2507	50.150	41.58	987,223

* The final uranium concentrations for these samples were below detection limits; therefore, the quantification limit of the instrument was used as the “final” concentration of uranium for calculations.

The “U initial conc.” was measured on a duplicate sample of source water and is within analytical error of the previous measurement.

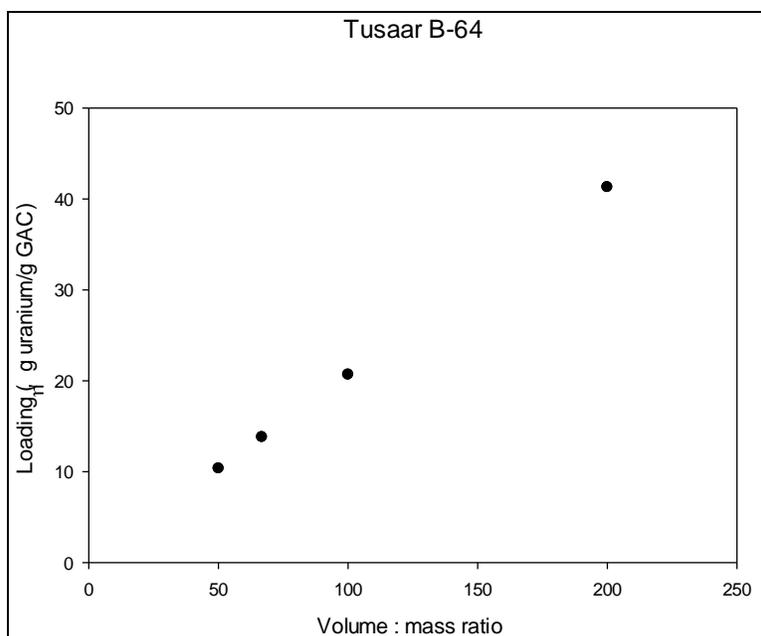


Figure 3.1. Uranium Loading (mg uranium/g GAC) on Tusaar B-64 GAC as a Function of the Volume-to-Mass Ratio

Table 3.3. Uranium-Adsorption Data for Tusaar ER2-189A GAC

Sample #	U Initial Conc. (µg/L)	U Final Conc. (µg/L)	GAC Mass (g)	Soln. Vol. (ml)	U Ads. (µg/g)	K _d (ml/g)
ER2-189A-uranium-pH7-31	207.9	0.039	0.9998	50.011	10.40	265,314
ER2-189A-uranium-pH7-32	207.9	0.047	1.0005	50.021	10.39	219,486
ER2-189A-uranium-pH7-33	207.9	0.036	0.7503	50.020	13.86	386,501
ER2-189A-uranium-pH7-34	207.9	0.040	0.7502	50.017	13.86	348,972
ER2-189A-uranium-pH7-35*	207.9	<0.011	0.5001	50.021	20.79	>1,961,652
ER2-189A-uranium-pH7-36*	207.9	<0.021	0.5000	50.006	20.79	>980,678
ER2-189A-uranium-pH7-37	207.9	0.024	0.2505	50.109	41.58	1,744,826
ER2-189A-uranium-pH7-38	207.9	0.036	0.2503	50.061	41.57	1,145,120

* The final uranium concentrations for these samples were below detection limits, so the estimated quantification limit of the instrument is given.

The “U initial conc.” was measured on a duplicate sample of source water and is within analytical error of the previous measurement.

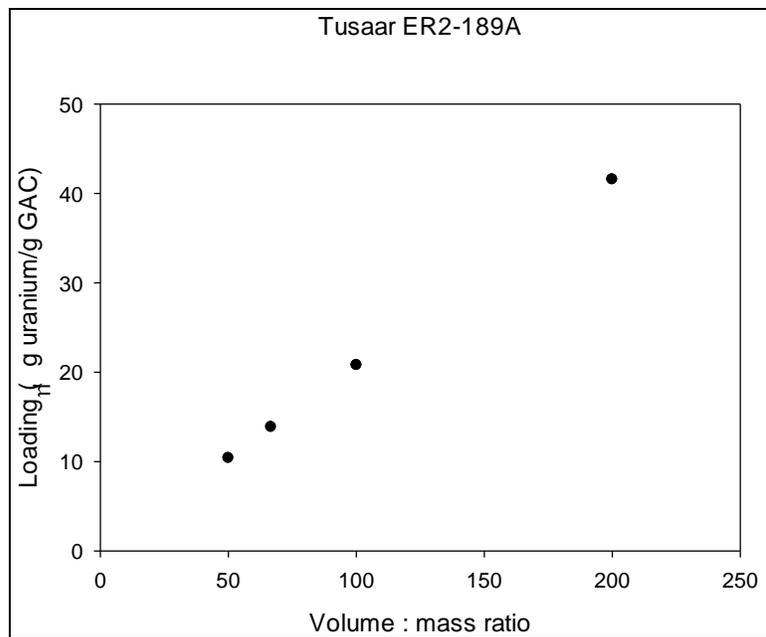


Figure 3.2. Uranium Loading (mg uranium/g GAC) on Tusaar ER2-189A GAC as a Function of the Volume-to-Mass Ratio

4.0 Conclusions

A linear response of uranium loading as a function of the solution-to-solid ratio was observed for both materials. K_d values ranged from ~380,000 to >1,900,000 ml/g for the B-64 material and ~200,000 to >1,900,000 ml/g for the ER2-189A material. Uranium loading values ranged from 10.4 to 41.6 $\mu\text{g/g}$ for the two Tusaar materials. These results are comparable to previous testing conducted in fiscal year 2011. In these tests, uranium adsorption batch tests were completed on five resin materials (Dowex 1, Dowex 21K 16-30 [fresh], Dowex 21K 16-30 [regenerated], Purofine PFA600/4740, and ResinTech SIR-1200).¹ The low-nitrate tests from that work had similar test conditions to the current experiments. The K_d values for Dowex 1 and Dowex 21K 16-30 (fresh) ranged from approximately 800,000 to 1,500,000 ml/g. Purofine PFA600/4740 and ResinTech SIR-1200 resins had K_d values that ranged from ~800,000 to 3,500,000 ml/g. The regenerated Dowex 21K 16-30 resin exhibited attenuated uranium-adsorption performance with K_d values ranging from 15,000 to 34,000 ml/g. Uranium loading values ranged from 8 to 37 $\mu\text{g/g}$ for the resins.

¹ Mattigod SV, EC Golovich, DM Wellman, EA Cordova, and RM Smith. 2010. *Uranium Adsorption on Ion-Exchange Resins - Batch Testing*. PNNL-20135, Pacific Northwest National Laboratory, Richland, WA.

Appendix A

Analytes and Analytical Methods

Appendix A

Analytes and Analytical Methods

The analytes and analytical methods for the testing are described below.

A.1 Analyte List

Influent water was analyzed for uranium, technetium-99, and nitrate. Batch contact solutions were analyzed for uranium only.

A.2 Trace Metals Analysis

Iodine-127, uranium, and technetium-99 analyses of the groundwater/test solution were performed using an inductively coupled plasma-mass spectrometer (ICP-MS) following procedure PNNL-AGG-415, “Inductively Coupled Plasma Mass Spectrometry (ICP-MS) Analysis,”¹ which is similar to SW-846, Method 6020A (EPA 1996). High-purity traceable single element standards traceable to the National Institute of Standards and Technology (Ultra Scientific [(Kingston, RI) and Inorganic Ventures [Lakewood, New Jersey]) were used to generate calibration curves and to verify continuing calibration during the analytical run. A serial dilution was made of select samples to investigate and correct for matrix interferences. Typical instrument detection limits for the ICP-MS are in the range of parts per trillion.

A.3 Anion Analysis

Anion analyses of the groundwater was performed using PNNL’s procedure AGG-IC-001, “Determinations by Ion Chromatography (IC),”² which is similar to EPA SW-846, Method 9056A (EPA 2007). Nitrate was separated on a Dionex AS18 column with a sodium hydroxide gradient elution and measured using a conductivity detector. The only modification to the EPA ion-chromatography method was the use of sodium hydroxide for gradient elution. High-purity calibration standards were used to generate calibration curves and to verify continuing calibration during the analytical run.

¹ Clayton ET. 2008. “Inductively Coupled Plasma Mass Spectrophotometry (ICP-MS),” PNNL-AGG-415, unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

² Lindberg MJ. 2004. “Determinations by Ion Chromatography (IC),” AGG-IC-001, unpublished PNNL Technical Procedure, Pacific Northwest National Laboratory, Richland, Washington.

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U.S. Environmental Protection Agency (EPA). 2007. "Method 9056A, Determination of Inorganic Anions by Ion Chromatography," Rev. 1. In *Test Methods for Evaluating Solid Waste: Physical/Chemical Methods*. EPA SW 846, EPA Office of Solid Waste and Emergency Response, Washington, D.C. Accessed January 11, 2011 at <http://www.epa.gov/wastes/hazard/testmethods/sw846/pdfs/9056a.pdf>.

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