Testing of Antimony Selective Media for Treatment of Liquid radwaste

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ABSTRACT

Nuclear power plants have sought radiation source term reduction and reduced discharge of radioactive constituents for many years. In the case of pressurized water reactors (PWR's), the latter efforts have been directed toward capture and immobilization of recalcitrant (ubiquitous radionuclides with long half-lives) species such as Cs-134 and Cs-137 and Co-58 and Co-60. As these plants resolved, or at least mitigated, the problems with radiocesium and radiocobalt, antimony radionuclides (Sb-122, Sb-124, and Sb-125) have become a primary concern in liquid liquid radwaste systems

Graver Technologies developed a granular composite metal oxide media with good selectivity for radioantimony. Initial laboratory data were collected using non-radioactive salts of antimony, cesium, and cobalt to judge efficacy of selective removal of antimony. Based on success of those trials, the media, designated Gravex GX187, was tested in partnership with Energy Solutions (nee Duratek) using actual liquid liquid radwaste in two PWR plants. One of these plants performed extensive slip-stream trials comparing the GX187 with strong base anion resins. With more than 2500 bed volumes of throughput, the GX187 outperformed the other competitors by reducing both Sb-124 and Sb-125 radionuclides below minimum detectable activity (MDA) with average decontamination factors (DF's) of 170, even when subjected to high levels of borate. Based on these favorable results, Energy Solutions installed the GX187 in a layered bed in their ALPS liquid radwaste processing system at this plant in August 2005. After one year of intermittent, batchwise operation including an outage, the GX187 processed more than 2.25 million liters (>600,000 gallons) of liquid liquid radwaste while reducing the Sb-125 activity to 2.9 E-08 Bq/L (DF=111) on average. This evaluation is ongoing and will continue at least until the fall 2006 outage at this plant.

Concurrently, Graver developed a second generation antimony selective media designated Gravex GX194. Again in partnership with Energy Solutions, the GX194 is currently undergoing slip-stream testing in the liquid radwaste system in one PWR and installed and operating in ALPS systems in 3 other PWR's. After 5 months of slip-stream testing, the GX194 media has produced only MDA quality water in terms of Sb-122 and Sb-125 radionuclides, albeit with relatively low levels of influent antimony. The GX194 installed at the first plant has processed more than 1.2 million liters (>325,000 gallons) of liquid radwaste, consistently produced MDA quality water for Sb-125, and typically offered a DF>100. Similarly, the GX194 in the second plant has processed almost 1.3 million liters (~340,000 gallons) of liquid radwaste, produced MDA quality water for Sb-125 for 36% of 42 batches, and typically offered a DF>35. The GX194 was not installed in the third plant until August 2006, so performance data is still pending.

INTRODUCTION

Nuclear power plants have sought radiation source term reduction and reduced discharge of radioactive constituents for many years. In the case of pressurized water reactors (PWR's), the latter efforts have been directed toward capture and immobilization of recalcitrant (ubiquitous radionuclides with relatively long half-lives) species such as Cs-134 and Cs-137 and Co-58 and Co-60. As these plants resolved, or at

least mitigated, the problems with radiocesium and radiocobalt, antimony radionuclides (Sb-122, Sb-124, and particularly Sb-125) became a primary concern in liquid radwaste systems.

Suppliers, processors, and end-users have been searching for media that irreversibly removes antimony from liquid radwaste streams such as floor drains and reactor hold-up tanks. Candidate media must selectively remove remove antimony radionuclides regardless of the physical and chemical form.

BACKGROUND

Chemistry

Antimony chemistry in solution is quite complex. Soluble antimony carries either a +3 or +5 oxidation state. As shown in the Pourbaix diagram in Figure 1, antimony forms relatively stable complexes with hydroxyl ions (OH^-). These complexes include cationic {$Sb(OH)_2^+$}, neutral {$Sb(OH)_3$ (aq) and $Sb(OH)_5$ (aq)}, and anionic {$Sb(OH)_4^-$and $Sb(OH)_6^-$} species depending on the pH and redox conditions. The predominate chemical form of soluble antimony under the oxidative conditions present in PWR liquid radwaste is the anionic $Sb(OH)_6^-$ species.

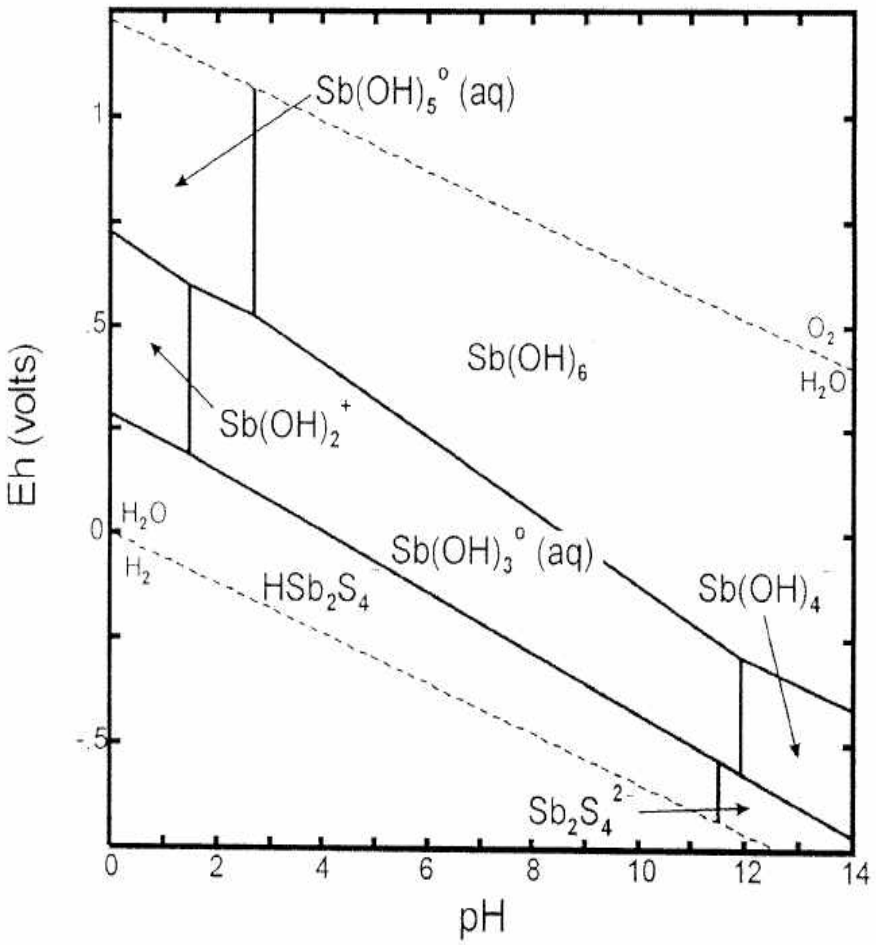

Fig. 1. Pourbaix diagram of aqueous species of antimony. (1)

Similarly to cobalt radionuclides, antimony radionuclides are present in liquid radwaste in both soluble [2] and colloidal forms. Natural antimony (atomic number 51, Group VA) has two isotopes (57.25% Sb-121 and 42.75% Sb-123). In addition, radioisotopes of antimony range from Sb-112 through Sb-134. Most have short half-lives ($t_{1/2}$ <1 day). The most problematic radioisotope of antimony is Sb-125 ($t_{1/2}$ =2.8 years), although Sb-120, Sb-122, Sb-124, Sb-126, and Sb-127 have half-lives between 2.7 and 60 days. Radioisotopes of antimony decay via both beta and gamma radiation. The Sb-125 isotope is a fission product of U-235 that emits both beta and gamma radiation.

Removal Approaches

The most common approach to removal of antimony utilizes strongly basic anion exchange resins. In many cases, the strong base resin works well, at least initially. Since strong base anion resins exchange negatively charged species, the most likely explanation for this reaction involves anionic hydroxyl complexes of antimony, such as the $Sb(OH)_6^-$ moiety. Unfortunately, strong base anion resins are notorious for sloughing antimony suddenly and unexpectedly, especially if high levels of borate are present in the influent. A plausible explanation for this behavior is that the resin has relatively low affinity for the antimony hydroxyl species which is readily displaced by other competing anions such as borate, chloride, and sulfate.

A number of other materials and processes have been tested for removal of radioantimony from liquid radwaste. Standard organic cation exhangers and typical inorganic zeolites have proven generally ineffective for antimony removal [3]. Like the strongly basic anion resins, powdered activated alumina (Al_2O_3) exhibits affinity for antimony, but readily elutes the antimony in borated solutions. The borate elution problem can be mitigated by lowering the feed water pH to 4.0 – 4.5 which converts borates into boric acid that does not displace antimony. Both a mixed bed of strongly basic anion and weakly acidic cation resin and certain metal oxide media utilize the pH adjustment to overcome the borate problem [4]. A recent study [5] claimed that a strong base anion resin loaded with an iron oxide overcame the antimony sloughage problem even at neutral pH.

Other methods that have been investigated for antimony removal from liquid radwaste include chemical complexation with stannous chloride, coagulation / precipitation, ultrafiltration coupled with chemical additives [6], electrodeionization, and hollow fiber filtration [7]. Researchers in Finland have reported success in removing radioantimony from floor drain waters with either titanates or proprietary metal oxides in three recent papers [8, 9, 10].

LABORATORY TRIALS

The initial laboratory trials involved batch equilibrium testing of two metal oxide media using a synthetic, non-radioactive feed water. The feed water was made up in a plastic carboy by adding reagent chemicals (salts) to ultra-pure water made with commercial laboratory equipment utilizing granular carbon, ion exchange resins, and reverse osmosis unit processes. More specifically, the constituents included sufficient cesium chloride, cobalt chloride hexahydrate, antimony chloride, and potassium chloride to achieve the following concentrations:

Cesium	25 mg/L
Cobalt	12.5 mg/L
Antimony	5.0 mg/L
Potassium	2000 mg/L

All of the subsequent batch equilibrium tests utilized a single lot of this synthetic feed water.

For the batch equilibrium tests, 3 liters of feed solution was added to a 4 liter glass beaker containing a magnetic stir bar. The feed solution was agitated slowly by this stirrer and a corresponding magnetic stir plate. One hundred grams (100 g) of metal oxide media was added to the solution to initiate the test. Fifty milliliter (50 mL) samples were extracted from the mixture with disposable pipettes periodically during the 2 hour test.

Each sample was filtered using 0.45 μm filter paper immediately following extraction. The filtered liquids were subsequently stored in plastic bottles for later analysis. Samples were analyzed for cesium, cobalt, and antimony content by Pace Analytical Services (Minneapolis, Minnesota) using an inductively coupled plasma spectrometer equipped with a mass spectrometer (ICP-MS).

Figure 2 illustrates the results of batch equilibrium tests for two metal oxide media. The control media in this trial was a commercial sodium titanate sold by Fortum Nuclear Services, OY (Finland) under the trademark CoTreat. The CoTreat is a granular material (nominal particle size 0.30 to 0.85 mm) primarily designed for selective removal of radicobalt [10, 11, 12]. This media was chosen since it has previously demonstrated capability for removal of radioantimony as well as radiocobalt [2]. The Graver metal oxide media in this comparison was designated as Gravex GX187. This material is also granular with a nominal particle size of 0.25 to 1.20 mm.

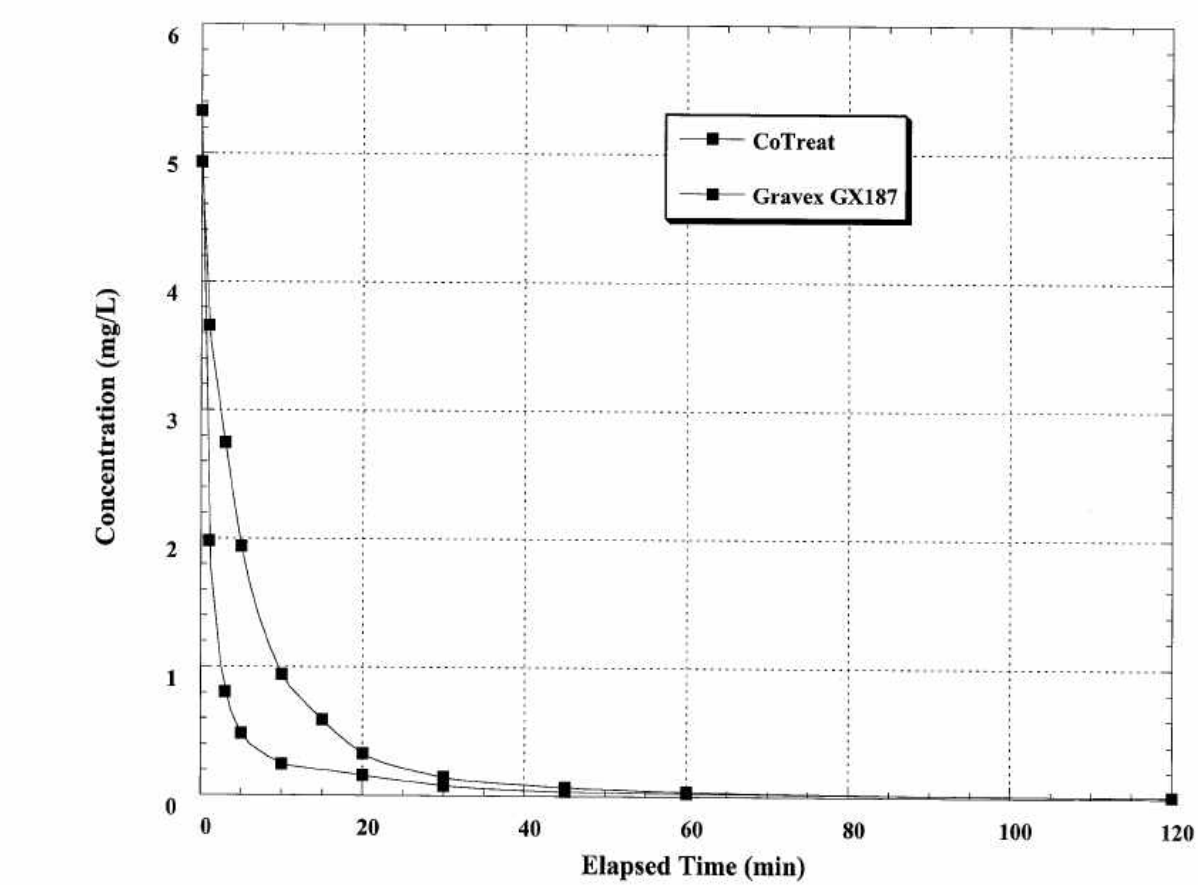

Fig. 2. Antimony removal using stirred batch equilibrium test.

Both the CoTreat and GX187 media removed antimony effectively. Removal was more rapid with GX187 although both media remove virtually all of the antimony after 2 hours of exposure. Table I further illustrates the different rates of removal for the two media. The more rapid removal by the GX187

media may relate to better kinetics (rate of reaction), to better selectivity for antimony versus the competing ions, or both. In any case, GX187 was deemed the superior candidate for further evaluation in a plant setting.

Table I Removal Efficiency Comparison for Batch Equilibrium Trials

	MEDIA →	**CoTreat**	**GX187**
REMOVAL ↓			
>90%		<20 minutes	<5 minutes
>95%		<30 minutes	<10 minutes
>99%		<60 minutes	<45 minutes
At the end of the test		99.75% removal	99.8% removal

PLANT TRIALS

Initial Slip-Stream Testing

Slip-stream testing of the GX187 media on actual liquid radwaste commenced in February 2005 with a brief trial at Plant A under the auspices of Energy Solutions. In this case, Energy Solutions had collected a 2 liter sample in which antimony radioisotopes constituted the bulk of the activity (~85%). The sample was passed through a column containing GX187; activity in both the influent and effluent was measured using gamma spectroscopy.

Table II summarizes the results of this single sample trial. The high decontamination factors[1] (DF's) achieved for both radioantimony isotopes in this trial encouraged further plant testing with the GX187.

Table II Initial Slip-Stream Trial with Gravex GX187 at Plant A

NUCLIDE		**INFLUENT ACTIVITY**	**EFFLUENT ACTIVITY**	**DECONTAMINATION FACTOR**
		(Bq/L)	**(Bq/L)**	
Mn-54		**1.7 E+00**	**MDA**	
Co-58		**1.1 E+02**	**2.2 E+01**	**5.1**
Co-60		**1.8 E+01**	**2.4 E+00**	**7.6**
Sb-124		**3.4 E+02**	**MDA**	**>205**
Sb-125		**5.8 E+02**	**MDA**	**>348**
Cs-137		**3.3 E+01**	**1.2 E+01**	**2.7**

A second, more extensive slip-stream trial of the GX187 media commenced at Plant B in April 2005. In this case, Energy Solutions conducted side-by-side antimony removal trials using three separate columns containing different granular media. These media included a standard gel strongly basic anion (SBA) and a highly macroporous strongly basic anion (MP SBA) resin as well as the GX187 media.

[1] Decontamination factor or "DF" is defined as the ratio of the feed activity divided by the effluent activity as shown in Equation 1.

$$DF = \alpha_{feed} / \alpha_{effluent}, \quad \text{(Eq. 1)}$$

Plant B utilizes an Energy Solutions ALPS system for radwaste processing. The effluent from the initial carbon bed in the ALPS system was used as the influent for the three columns in the slip-stream testing. At the start of the trial the radwaste feed to this system was outage crud burst water that included measurable activities for the following radionuclides: Cr-51, Mn-54, Co-57, Co-58, Fe-59, Co-60, Zn-65, Zr-95, Sb-124, Sb-125, I-131, Cs-134, and Cs-137. Activities ranged from a high of 4.8 E+05 Bq/L (Co-58) to a low of 1.5 E+02 Bq/L (Sb-124) and totaled 5.8 E+05 Bq/L. The liquid radwaste contained detergents, hydrazine, colloids, and dissolved ionic species as well as these outage isotopes. The initial carbon bed in the ALPS system reduced the activities of some of these species. Table III summarizes the results from this comparative trial.

Table III Initial Slip-Stream Trial with Gravex GX187 at Plant B

THROUGHPUT	NUCLIDE		INFLUENT ACTIVITY	EFFLUENT ACTIVITY	EFFLUENT ACTIVITY	EFFLUENT ACTIVITY
				SBA	GX187	MP SBA
(Bed Volumes)			(Bq/L)	(Bq/L)	(Bq/L)	(Bq/L)
14						
	Mn-54		1.7 E+03	1.6 E+03	1.0 E+03	1.8 E+03
	Co-58		5.4 E+04	4.1 E+04	1.6 E+04	4.4 E+04
	Co-60		8.8 E+02	5.8 E+02	8.2 E+02	8.2 E+02
	Sb-124		2.9 E+01	MDA	MDA	MDA
	Sb-125		3.0 E+02	MDA	MDA	MDA
24						
	Mn-54		1.7 E+03	1.8 E+03	1.2 E+02	1.6 E+03
	Co-58		5.4 E+04	4.1 E+04	1.6 E+04	4.4 E+04
	Co-60		8.8 E+02	1.0 E+03	3.3 E+02	8.8 E+02
	Sb-124		2.9 E+01	MDA	MDA	MDA
	Sb-125		3.0 E+02	MDA	MDA	MDA
161						
	Mn-54		1.7 E+03	1.8 E+03	MDA	1.8 E+03
	Co-58		5.4 E+04	6.8 E+04	1.9 E+04	6.8 E+04
	Co-60		8.8 E+02	1.3 E+03	3.0 E+02	1.1 E+03
	Sb-124		2.9 E+01	MDA	MDA	MDA
	Sb-125		3.0 E+02	MDA	MDA	MDA
550						
	Mn-54		6.1 E+02	6.1 E+02	MDA	6.1 E+02
	Co-58		4.1 E+03	7.8 E+03	7.1 E+03	1.4 E+05
	Co-60		1.4 E+02	2.6 E+02	2.0 E+02	3.1 E+02
	Sb-124		4.4 E+00	MDA	MDA	MDA
	Sb-125		1.4 E+02	MDA	MDA	MDA

This slip-stream testing continued through the summer of 2005. Once the boric acid / borate levels in the liquid radwaste increased significantly both anion resin columns showed radioantimony breakthrough (at

~1,500 bed volumes of throughput). Only the GX187 media continued to produce MDA water in terms of antimony and manganese radioisotopes. After 2,345 bed volumes of throughput, this media yielded a DF of >170 for Sb-125 based on an influent activity of 4.4 E+02 Bq/L and an effluent activity of MDA. The GX187 column test continued beyond 5,000 bed volumes of throughput without leaking or sloughing Sb-125 above MDA. Based on these extremely promising results, Energy Solutions and Plant B personnel decided to move forward with a full scale trial.

Full Scale Testing

In late August of 2005, Energy Solutions installed the GX187 media in the ALPS system at Plant B. A later of 0.14 cubic meters (5 ft^3) of the GX187 was added on top of 0.57 cubic meters (20 ft^3) of standard mixed bed in one of the ALPS thirty six inch pressure vessels. Once initial differential pressure problems were resolved, liquid radwaste was processed batchwise through the ALPS system. These batches included radwaste from both floor drain and reactor hold-up tanks. Activity for Sb-125 was monitored on both the inlet and effluent to the vessel containing the GX187.

Table IV summarizes the results of this trial. Initial results for this trial were excellent in terms of Sb-125 removal from both floor drain and reactor hold-up liquid radwaste. Throughout the fall 2005 refueling outage at Plant B the effluent from the GX187 bed contained no measurable Sb-125 (MDA water). Influent activity of the Sb-125 during this time typically ranged from 1.0 to 3.1 E+02 Bq/L while the DF's ranged upwards from 35. The first measurable Sb-125 appeared after more than 1.5 million liters (400,000 gallons) of liquid radwaste throughput. After the outage, liquid radwaste was processed less frequently with the influent Sb-125 activity increasing into the E+02 Bq/L range and the DF's declining somewhat. By early June 2006, more than 2.1 million liters (~550,000 gallons) of liquid radwaste has been processed through the GX187. The media generated MDA quality water for ca. 60% of the 37 batches processed in more than 9 months of operation. The average influent Sb-125 activity was 3.2 E+02 Bq/L while the average effluent Sb-125 activity was 2.9 E+00 Bq/L with an average DF of 111. This bed of GX187 continues to operate well at Plant B.

In response to the initial differential pressure problems experienced at Plant B, Graver developed a second generation antimony selective media designated Gravex GX194. The Gravex GX194 first became available for plant usage in January of 2006. Energy Solutions puchased 0.14 cubic meters (5 ft^3) of the GX194 media for each of three plants (A, C, and D) in mid January 2006.

In late January of 2006, Energy Solutions installed the GX194 media in the ALPS system at Plant A. A layer of 0.14 cubic meters (5 ft^3) of the GX194 was added on top of layers of standard strongly acidic cation and strongly basic anion resins in one of the ALPS thirty six inch pressure vessels at this plant. Liquid radwaste is processed batchwise through the ALPS system. Activity for Sb-125 was monitored on both the influent and effluent to the vessel containing the GX194.

Table V summarizes the results of this trial. Thirty four batches of liquid radwaste have been processed at Plant A since the installation of the GX194 media; 18 of these have contained no measurable (MDA) Sb-125. For the remaining 16 batches, influent Sb-125 activities ranged from 3.2 E+01 to 2.8 E+03 Bq/L. The GX194 media generated MDA quality water (<1.0 E+01 Bq/L) for more than 80% of these. Corresponding DF's ranged from 3 to >240. Including recycling more than 1.5 million liters (>400,000 gallons) of liquid radwaste has passed through the GX194 media.

In early February of 2006, Energy Solutions installed the GX194 media in the ALPS system at Plant C. A layer of 0.14 cubic meters (5 ft^3) of the GX194 was layered added on top of 0.57 cubic meters (20 ft^3) of standard mixed bed in one of the ALPS thirty six inch pressure vessels. The mixed bed itself had already processed 450,000 liters (~120,000 gallons) of radwaste prior to this installation. Liquid radwaste

was processed batchwise through the ALPS system. Activity for Sb-125 was monitored on both the influent and effluent of the vessel containing the GX194. Table VI summarizes the results of this trial.

The initial results for the Plant C trial indicated only modest Sb-125 removal; however, the DF values rose steadily as additional batches were processed during the first week of operation. Since this initial "run-up" DF's through the GX194 bed have remained relatively constant. Moreover, no change in performance was observed when radwaste from the spring refueling outage at Plant C was processed in

Table IV Full Scale Trial with Gravex GX187 at Plant B

DATE	LIQUID RADWASTE SOURCE		INFLUENT ACTIVITY (Sb-125)	EFLLUENT ACTIVITY (Sb-125)	DECONTAMINATION FACTOR
			(Bq/L)	(Bq/L)	
08/25/05	Floor Drain		1.9 E+02	MDA	>69
08/26/05	Floor Drain		1.7 E+02	MDA	>64
08/30/05	Floor Drain		1.4 E+02	MDA	>50
08/31/05	Floor Drain		1.5 E+02	1.0 E+01	14.5
09/02/05	Reactor Hold Up		1.05 E+02	MDA	>39
09/06/05	Reactor Hold Up		1.25 E+02	MDA	>46
09/07/05	Reactor Hold Up		1.3 E+02	MDA	>47
09/08/05	Floor Drain		3.0 E+02	MDA	>108
09/09/05	Reactor Hold Up		1.3 E+02	MDA	>46
09/12/05	Reactor Hold Up		1.4 E+02	MDA	>52
09/16/05	Floor Drain		1.8 E+02	MDA	>65
09/28/05	Floor Drain		1.8 E+02	MDA	>65
10/06/05	Reactor Hold Up		2.0 E+02	MDA	>73
10/07/05	Reactor Hold Up		9.5 E+01	MDA	>35
10/20/05	Floor Drain		1.8 E+02	MDA	>67
11/07/05	Floor Drain		2.9 E+02	MDA	>107
11/10/05	Reactor Hold Up		9.9 E+01	MDA	>37
12/20/05	Floor Drain		1.3 E+02	2.4 E+01	5.4
02/09/06	Reactor Hold Up		7.1 E+02	9.5 E+01	7.5
02/16/06	Floor Drain		4.1 E+02	5.8 E+01	7.1
03/04/06	Floor Drain		2.5 E+02	4.1 E+01	6.1
03/28/06	Floor Drain		1.5 E+03	1.0 E+02	14.3
04/05/06	Floor Drain		5.8 E+02	1.1 E+02	5.2
04/13/06	Floor Drain		1.3 E+03	9.9 E+01	12.8
05/03/06	Floor Drain		5.4 E+01	7.5 E+01	0.7
05/04/06	Reactor Hold Up		1.6 E+02	MDA	>57.5
05/11/06	Floor Drain		1.3 E+02	MDA	>46.3
05/18/06	Floor Drain		3.2 E+02	MDA	>9.5
05/23/06	Reactor Hold Up		3.4 E+02	MDA	>10.8
06/01/06	Reactor Hold Up		2.0 E+02	MDA	>4.6
06/08/06	Floor Drain		3.4 E+02	5.4 E+01	9.4

April 2006. Influent activity of the Sb-125 to the bed during the trial typically has ranged from low E+03 to low E+04 Bq/L while the effluent activity after the initial decline commonly ran from the E+01 Bq/L range down to MDA levels. As of early June 2006, almost 1.3 million liters (~343,000 gallons) of radwaste had been processed through the GX194. The media generated MDA quality water for ca. 36 % of the 42 batches processed in more than 4 months of operation. The average influent Sb-125 activity was 5.8 E+03 Bq/L while the average effluent Sb-125 activity was 1.5 E+02 Bq/L which translates to an average DF of 39. This bed of GX187 at Plant C was removed from service in June 2006 due to

Table V. Full Scale Trial with Gravex GX194 at Plant A

DATE	TOTAL THROUGHPUT		INFLUENT ACTIVITY (Sb-125)	EFLLUENT ACTIVITY (Sb-125)	DECONTAMINATION FACTOR
	(L)		(Bq/L)	(Bq/L)	
01/26/06	7570		MDA	MDA	
03/11/06	54900		MDA	MDA	
03/13/06	78000		MDA	MDA	
03/29/06	101400		MDA	MDA	
03/30/06	150600		MDA	MDA	
04/04/06	200600		MDA	MDA	
04/06/06	243400		MDA	MDA	
04/08/06	293700		MDA	MDA	
04/11/06	337200		MDA	MDA	
04/13/06	386400		MDA	MDA	
04/18/06	422000		3.2 E+01	MDA	>3
04/19/06	471200		1.6 E+03	MDA	>160
04/20/06	481500		7.1 E+02	1.1 E+01	68
04/23/06	507600		6.5 E+02	MDA	>63
04/24/06	537800		2.1 E+03	MDA	>203
04/27/06	591600		5.1 E+02	MDA	>50
05/02/06	626400		3.4 E+02	MDA	>33
05/08/06	659000		7.1 E+02	MDA	>70
05/09/06	695300		1.1 E+03	MDA	>110
05/10/06	707000		2.8 E+03	MDA	>240
05/16/06	734300		1.1 E+03	MDA	>103
09/19/06	760400		MDA	MDA	
09/25/06	811900		MDA	MDA	
10/04/06	894400		MDA	MDA	
10/09/06	931900		1.2 E+02	3.2 E+01	3.9
10/11/06	978400		MDA	MDA	
10/16/06	1007900		MDA	MDA	
10/18/06	1018500		MDA	MDA	
10/19/06	1043500		5.1E+02	8.8 E+01	5.6
10/23/06	1080200		8.2E+01	MDA	8.0
10/24/06	1117000		8.2E+01	MDA	8.0
10/31/06	1174900		MDA	MDA	
11/01/06	1189200		MDA	MDA	
11/06/06	1234700		5.8E+01	MDA	5.8

11/14/06	**1274800**		**MDA**	**MDA**	
11/15/06	**1289600**		**MDA**	**MDA**	
11/22/06	**1322500**		**MDA**	**MDA**	

Table VI. Full Scale Trial with Gravex GX194 at Plant C

DATE	TOTAL THROUGHPUT		INFLUENT ACTIVITY (Sb-125)	EFLLUENT ACTIVITY (Sb-125)	DECONTAMINATION FACTOR
	(L)		(Bq/L)	(Bq/L)	
02/01/06	20900		5.4E+03	7.7E+03	0.7
02/02/06	49200		1.4E+04	8.2E+03	1.7
02/03/06	78800		5.8E+03	1.3E+03	4.6
02/06/06	105900		5.8E+03	1.1E+03	5.3
02/07/06	136800		6.5E+03	5.8E+02	11
02/09/06	167700		1.5E+04	2.5E+02	59
02/10/06	190900		1.5E+04	3.4E+02	43
02/13/06	217900		1.1E+04	2.1E+02	71
02/14/06	248500		1.1E+04	2.1E+02	52
02/17/06	273100		8.2E+03	1.8E+02	45
02/20/06	305800		8.2E+03	2.6E+02	31
02/21/06	333800		7.8E+03	2.7E+02	29
02/22/06	355200		7.5E+03	2.2E+02	34
02/23/06	385200		1.1E+04	3.4E+02	33
02/24/06	406800		3.1E+03	MDA	>107
02/27/06	436600		3.1E+03	1.9E+02	16
02/28/06	458000		2.6E+03	MDA	>90
03/01/06	482500		2.6E+03	MDA	>90
03/02/06	506300		1.8E+03	MDA	>62
03/03/06	531400		2.9E+03	MDA	>102
03/07/06	556800		2.7E+03	MDA	>94
03/09/06	584600		8.7E+02	MDA	>30
03/13/06	601100		2.2E+03	MDA	>76
03/16/06	638300		2.6E+03	7.8E+01	33
03/21/06	675300		3.4E+03	MDA	>107
03/22/06	699600		1.6E+03	MDA	>57
03/24/06	721000		4.3E+03	8.8E+01	48
03/28/06	750300		3.2E+03	1.2E+02	27
03/30/06	795200		3.9E+03	1.5E+02	26
04/11/06	835200		2.8E+03	9.2E+01	30
04/13/06	864900		2.6E+04	MDA	>918
05/01/06	899300		1.6 E+04	8.5 E+02	19
05/02/06	939400		1.5 E+04	5.6 E+02	28
05/04/06	964200		1.1E+04	7.1E+02	15
05/05/06	986700		7.5E+03	5.8E+02	13
05/25/06	1015400		6.5E+03	4.9E+02	13
05/26/06	1048600		6.5E+03	MDA	>223
06/01/06			7.9 E+03	3.19 E+03	2.5
06/02/06	1149500		3.4 E+03	3.7 E+03	9.3
06/06/06			3.4E+03	MDA	>120
06/07/06			1.7E+03	MDA	>474
06/09/06	1299400		1.35 E+03	MDA	>474

administrative limits on the entire ALPS system which had processed a total of more than 2.1 million liters (>550,000 gallons) of radwaste..

In late August, a new bed of GX194 was loaded into the ALPS system in Plant C. In this case, Energy Solutions layered 0.28 cubic meters (10 ft^3) of the GX194 on top of 0.28 cubic meters (20 ft^3) of mixed bed resin. By the beginning of 2007, the new bed of GX194 had processed more than 375,000 L (>100,000 gallons) of radwaste had been processed through the new bed of GX194. The media generated MDA quality water for all 14 batches processed during this time. The average influent Sb-125 activity was 6.3 E+03 Bq/L which translates to an average DF of 97.

Table VII. Second Full Scale Trial with Gravex GX194 at Plant C

DATE	TOTAL THROUGHPUT		INFLUENT ACTIVITY (Sb-125)	EFLLUENT ACTIVITY (Sb-125)	DECONTAMINATION FACTOR
	(L)		(Bq/L)	(Bq/L)	
09/07/06			7.6E+03	MDA	>265
09/08/06			7.6E+03	MDA	>265
09/12/06			8.3E+03	MDA	>290
09/13/06			8.3E+03	MDA	>290
09/14/06			2.8E+04	MDA	>980
09/15/06			1.3E+04	MDA	>471
09/25/06			1.3E+04	MDA	>46
	168500				
10/11/06			1.9E+03	MDA	>65
10/17/06			7.3E+02	MDA	>26
10/18/06			8.0E+02	MDA	>28
10/19/06			6.6E+02	MDA	>23
10/23/06			7.1E+02	MDA	>25
	327300				
01/02/07			1.0E+03	MDA	>35
01/03/07			1.2E+03	MDA	>44
	378800				

In late August of 2006, Plant D installed the GX194 media in their radwaste system. In this case, the GX194 was placed in the polishing position in series with an existing antimony removal media. By year end, no radioantimony had been detected in the feed water to the GX194; consequently, effectively, this trial has yet to begin.

Second Generation Slip-Stream Trials

Slip-stream testing of the GX194 media commenced in early April 2006 in a trial at Plant B with the assistance of Energy Solutions. The protocol involved side-by-side testing of three granular antimony selective media from different vendors. Initially, the testing was similar to the original slip-stream trials on GX187 at Plant B. After several months of testing, the protocol was modified to include deeper beds of the candidate media and to utilize the final effluent from the ALPS system as the feed to the trial columns. Although this change significantly reduced the overall activity in the feed stream, it also lowered the MDA level for radioantimony.

To date, radwaste from both reactor hold up and floor drain tanks have been processed through the reconfigured slip-stream media apparatus. Table VIII summarizes the limited results from this revised comparative trial.

Table VIII. Slip-Stream Trial with Gravex GX194 at Plant B

NUCLIDE	DATE	LIQUID RADWASTE SOURCE		INFLUENT ACTIVITY	*GX194* EFFLUENT ACTIVITY	*MEDIA X* EFFLUENT ACTIVITY	*MEDIA Y* EFFLUENT ACTIVITY
				(Bq/L)	(Bq/L)	(Bq/L)	(Bq/L)
Sb-122							
	06/01/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	06/08/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	06/27/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	06/28/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	07/10/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	07/19/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	07/20/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	07/26/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	08/03/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	08/08/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	08/16/06	**Reactor Hold Up**		**MDA**	**MDA**	**MDA**	**MDA**
	08/18/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	08/22/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
Sb-125							
	06/01/06	**Reactor Hold Up**		**2.0E+02**	**MDA**	**MDA**	**MDA**
	06/08/06	**Floor Drain**		**3.4E+02**	**MDA**	**MDA**	**MDA**
	06/27/06	**Reactor Hold Up**		**3.7E+02**	**MDA**	**MDA**	**MDA**
	06/28/06	**Reactor Hold Up**		**1.2E+02**	**MDA**	**MDA**	**MDA**
	07/10/06	**Floor Drain**		**4.8E+02**	**MDA**	**MDA**	**MDA**
	07/19/06	**Reactor Hold Up**		**3.7E+02**	**MDA**	**MDA**	**4.1 E+02**
	07/20/06	**Floor Drain**		**7.8E+01**	**MDA**	**MDA**	**MDA**
	08/03/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**
	08/08/06	**Floor Drain**		**3.0E+01**	**MDA**	**MDA**	**MDA**
	08/16/06	**Reactor Hold Up**		**1.6E+01**	**MDA**	**MDA**	**MDA**
	08/18/06	**Floor Drain**		**1.3E+02**	**MDA**	**MDA**	**MDA**
	08/22/06	**Floor Drain**		**MDA**	**MDA**	**MDA**	**MDA**

CONCLUSIONS

Both the slip-stream and plant trials of the first generation Gravex GX187 and second generation GX194 granular media yielded significant activity reduction for radioantimony (e.g., Sb-125). In many cases, the activity was reduced below the minimum detectable activity (MDA) level for the Sb-125 radionuclide in that plant. The media proved robust since these trials were conducted with a variety of influents from different liquid radwaste sources.

Throughout these trials radioantimony activity reduction continued even with high total influent activity levels and with significant quantities of competing standard salts and other radionuclides. In particular, introduction of high levels of boric acid did not result in sloughage of radioantimony as frequently occurs with strongly basic anion resins. In part, this behavior reflects the fact the GX187 and GX194 media do not operate as ion exchangers and thereby permanently bind the radioantimony until their operating capacity is exhausted.

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