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Tetraphenylborate Decomposition Testing using Savannah River Site High Level Waste

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SUMMARY

This report describes batch testing of six different tank wastes for catalytic NaTPB decomposition. The waste samples came from Tanks 7F, 13H, 26F, 30H, 35H, and 46F. Tests with the six wastes occurred at both ambient (22-26 °C) and elevated (45 °C) temperature. Testing lasted six months. Samples obtained from the tests yielded the following observations.

- Under the conditions tested (45 °C), the maximum reaction rate observed – for samples from Tank 35H -- equates to a theoretical benzene generation rate of 0.2 mg/(L-h).
- The waste sample from Tank 35H contained the highest concentration of soluble mercury, an element known to participate in the reaction sequence. This finding suggests the value of continuing the efforts to explore the influence of soluble mercury concentration on the catalytic reaction.
- The maximum rate (0.08 mg/(L-h)) observed at 25 °C fell well within the design basis limit for the proposed facility.
- Palladium concentrations in the six waste samples, when diluted to a target concentration of 4.7 M sodium, ranged from 0.05 to 0.16 mg/L. Analyses did not detect the presence of platinum in the waste. The total concentration of noble metals (i.e., palladium, rhodium, and ruthenium) ranged from 1.96 to 6.49 mg/L
- The observed reaction rate appeared to correlate directly with the measured soluble sodium content of the waste. This finding suggests that lack of slow mass transfer and dissolution of tetraphenylborate likely limited the rate of reaction.
- Review of the data suggests four variables influenced the observed reactivity: temperature, concentration of noble metals, presence of soluble mercury, and presence of soluble tetraphenylborate.
- Post-mortem analysis of residual test materials, 11 months after initiating the tests, showed that 9 of the tests exhibited evidence of decomposition.

INTRODUCTION

The Department of Energy discontinued operation of the In-Tank Precipitation facility due to the potential for catalytic decomposition of sodium tetraphenylborate.¹ The Salt Disposition Systems Engineering Team identified Small Tank Tetraphenylborate Precipitation (STTP) as an alternative to replace the In-Tank Precipitation Facility at the Savannah River Site. The STTP process applies the same process chemistry for removal of cesium from the radioactive wastes but at a controlled lower temperature and in a smaller facility that offers engineering features to mitigate potential for a catalytic reaction. However, additional understanding of the catalytic reaction, through further experimental investigation, is needed to better define the potential for a reaction to occur in the proposed facility. Simulant testing indicates that the minimum species required to catalyze sodium tetraphenylborate (NaTPB) decomposition are either copper or a supported, reduced noble metal (e.g., Pd(0) on alumina).² The most reactive simulant system includes diphenylmercury (or Hg(II) salt) along with the alumina supported Pd(0) and diphenylborinic acid (2PB).³ The most reactive high level waste system observed was from batch testing in 1997-98.⁴ High Level Waste Engineering (HLWE) requested that the Savannah River Technology Center (SRTC) further evaluate the potential catalytic properties of additional high level waste tank samples.^{5,6,7} This report provides the details of those experiments and partially addresses Item 2.0 (Cesium Removal Kinetics and Equilibrium) of the HLW Applied Technology Scope of Work Matrix for Small Tank Tetraphenylborate (Demonstration Phase).⁸

EXPERIMENTAL

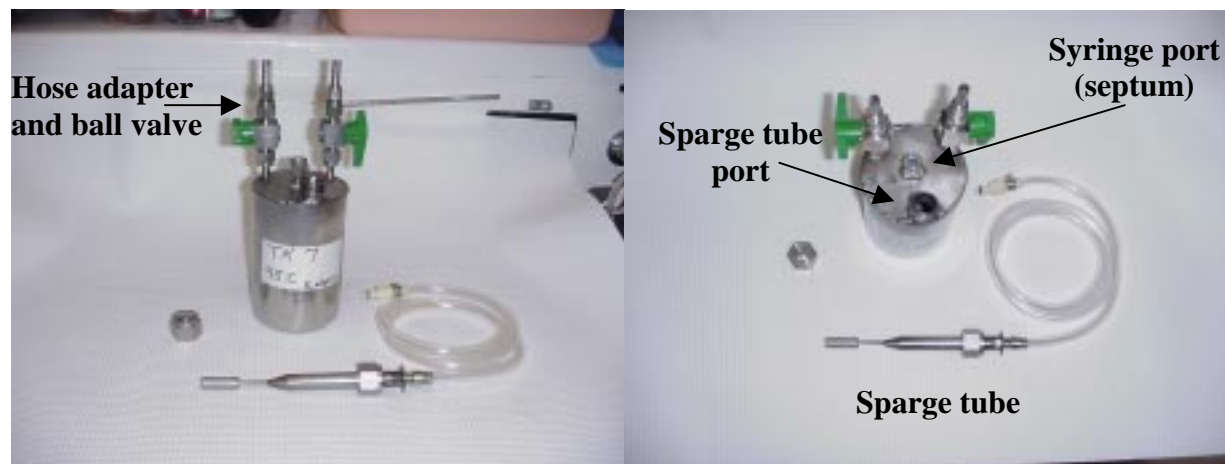
Experiments tested the catalytic activity of six high level waste tank samples. The experiments occurred in remote cells using manipulators to perform test operations. The tank wastes used in the program provided varying process histories and composition. The samples were as follows:

- Tank 7F: F-Area Waste Removal Tank
- Tank 13H: H-Area Waste Removal Tank
- Tank 26F: F-Area Evaporator Feed Tank
- Tank 30H: H-Area Evaporator Receipt Tank
- Tank 35H: H-Area Canyon Receipt Tank
- Tank 46F: F-Area Concentrate Receipt Tank

Analysis of filtered and unfiltered aliquots of the samples by inductively coupled plasma mass spectroscopy (ICP-MS), inductively coupled plasma emission spectroscopy (ICP-ES), plutonium scintillation with thenoyl trifluoroacetone (PuTTA), ion chromatography (IC), titration, atomic absorption (AA), scintillation, and gamma scan yielded both soluble and insoluble compositions of the waste. Appendix 1 provides the results of the characterizations.

Each waste solution was prepared for testing by diluting its entire sample volume to a target concentration of 5.05 M Na^+ . The actual volume of each solution, after dilution, was unique due to their varying starting volumes and sodium concentrations. The experiments used custom designed 250-mL stainless steel vessels that permitted oxygen depletion (see Figure 1). Each vessel was fitted with two gas purge ports (hose adapters and ball valves) as well as a septum sample port and sparge tube assembly port. Each sample was split in half. The twelve resulting fractions were placed in individual specially built vessels.

Figure 1. Custom designed 250-mL stainless steel reaction vessels with sparge tubes.



As a prerequisite to catalytic testing, the authors evaluated the conditions required to produce a deaerated solution. We performed a sparge test in one of the unique vessels with a stainless steel frit sparge tube. An Ocean Optics FOXY fiber optic oxygen sensor was used to monitor oxygen depletion during nitrogen sparge at a fixed flow rate. The sensor was initially calibrated in a large (1 L) stainless steel baffled vessel with impeller and air or nitrogen sparge at room temperature. The sparge rates were kept reasonable so as not to cool the liquid substantially. The sensor was then placed in the small stainless vessel, with no agitation, containing 150 mL

deionized water. Gas flow was controlled via a rotameter and monitored with a Humonics flow meter (0-500 standard cubic centimeters per minutes (ccm) flow range). Six experiments were performed over several days in order to assess the variability of the measurement scheme. The target flow rate was 200 ccm, and this varied during the course of the development testing with a standard deviation of 34 ccm (several runs contained a large drift component due to the instability in the rotameter). Initial conditions were reached by flowing air through the vessel until the sensor reached steady state at approximately 22 vol % oxygen. At this point, the system was switched to nitrogen and adjusted to 200 ccm flow. The data were acquired at 30 s and 5 s increments, but the FOXY software recorded the hour and minute values, leaving the second values as zeros. Therefore, the time axis had to be interpolated, and this was accomplished by applying a smoothing filter to the time axis, which essentially generated increments smaller than a minute. The data were then analyzed via Microsoft Excel calculations to find the least squares fit of the depletion curve to an exponential function. From this fit, time constants were obtained for each run. The data are shown in Table 1 and Figure 2.

Table 1. Sparge test results for 150 mL liquid volume, nominal 200 ccm flow rate, sparge time constants determined using FOXY sensor to monitor oxygen depletion.

Experiment <u>number</u>	Flow rate at end of <u>run, ccm</u>	exponential time constant, <u>seconds</u>
1	223.0	259.9
2	200.0	187.1
3	288.0	205.7
4	201.0	260.9
5	200.0	258.0
6	208.0	493.4
Average	220.0	277.5
std. dev.	34.5	110.4

The data show a time constant for the depletion of oxygen from the system of 277 seconds. Three times the time constant yielded an approximate sparge time for the conditions of 832 seconds, or 14 minutes. The standard deviation on this number was 330 seconds (3 times the standard deviation of the runs), or approximately 6 minutes. Therefore, a minimum recommended sparge time for the experiment was 20 minutes to accomplish oxygen depletion by nitrogen sparging at 200 ccm.

Based upon the simulant sparge test findings, each test solution was conservatively sparged for 180 minutes. The sparge tubes were removed immediately upon completion of the deaeration step and the port capped. The vapor space of the vessels was then purged with nitrogen to complete the vessel inerting process.

Each vessel was charged with 0.55 M NaTPB solution via syringe through the septum port. The quantity of NaTPB added was sufficient to react stoichiometrically with the measured potassium and provide a 0.03 M residual excess of NaTPB. The percent excess added for each tank is shown in Table 2. Additionally, each vessel was charged with 500 mg/L of phenylboronic acid (1PB) to promote reactivity and alleviate the lengthy induction periods observed in the previous batch tests⁴ with high level waste. The resulting target sodium concentration for each test after the additions was 4.7 molar. Six test vessels, representing the six waste samples, were placed in a heater-shaker

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Figure 2. Sparge test results for 150 mL liquid volume, nominal 200 ccm flow rate, sparge time constants determined using FOXY sensor to monitor oxygen depletion.

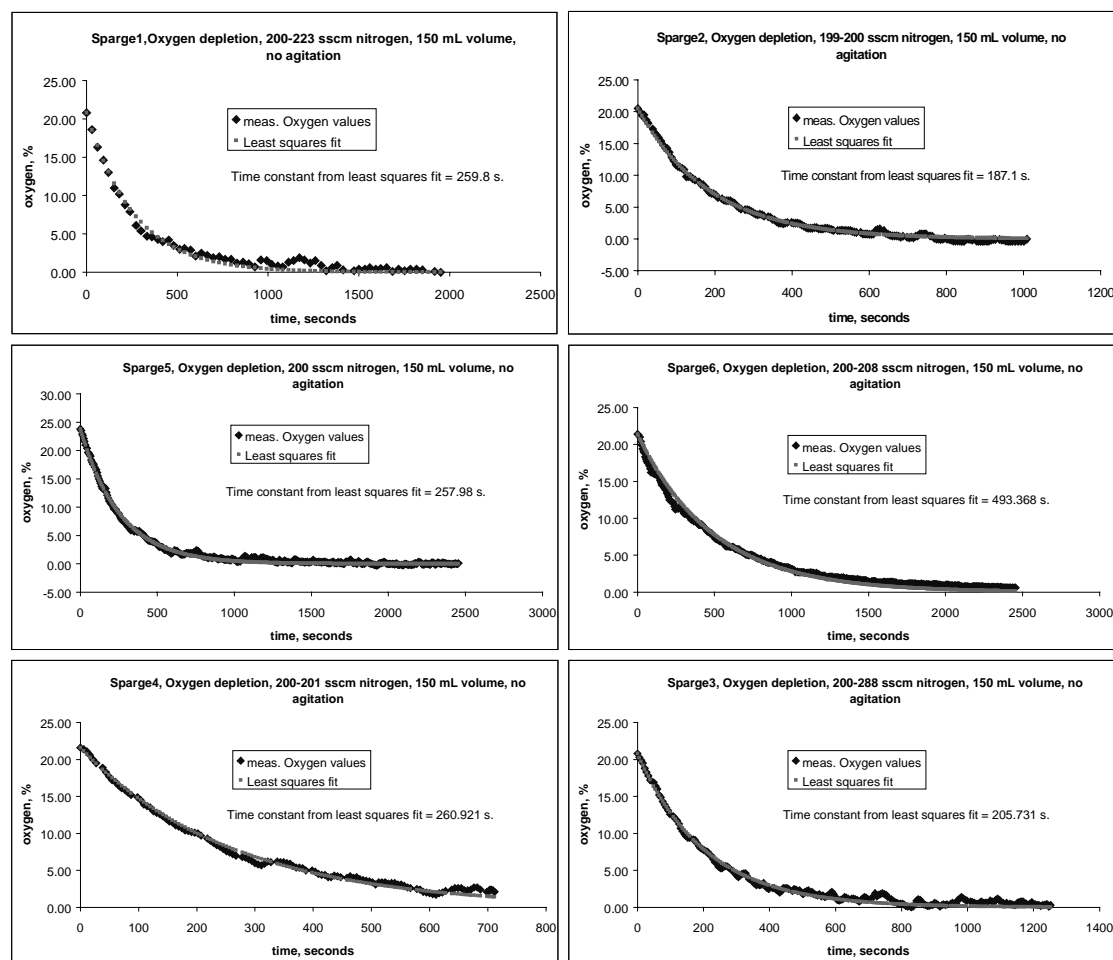


Table 2. Percent Excess NaTPB added to each real waste batch test.

Tank	% Excess
7F	65
13H	86 (174)*
26F	106
30H	112 (224)*
35H	224
46F	99

*The value in parentheses for Tanks 13H and 30H represents the total percent excess NaTPB added to Tests 13H-L and 30H-L after a second addition of NaTPB solution to each test.

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where they were shaken continuously at ~190 rpm and heated at 45 ± 3 °C. The six test vessels were designated with the letter H to indicate they were the higher temperature of the two sets (e.g., Test 7F-H, 13H-H, etc.). The remaining six test vessels were labeled with the letter L (e.g., Test 7F-L, 13H-L, etc.) to indicate they were the lower temperature set. These six were also shaken continuously but no temperature control was provided. Ambient temperature in the shielded cell facility ranged from 28 °C (start of testing) to 22 °C (end of testing). Figure 3 is a photograph of the two shakers and test vessels in the shielded cell. Testing lasted a period of 6 months. Periodically, samples were obtained via syringe, filtered immediately using 0.45 micron nylon disposable filter cups, and the filtrate typically submitted for Cs-137 (gamma scan), phenylborates (High Performance Liquid Chromatography, HPLC), and soluble boron (microwave digestion followed by ICP-ES) analysis. The available test volume and previous sample data dictated the sample frequency and required analysis. Data for the twelve tests are contained in Appendix 2.

Figure 3. Photograph of both the heater-shaker and ambient temperature shaker in operation in the shielded cell.



RESULTS AND DISCUSSION

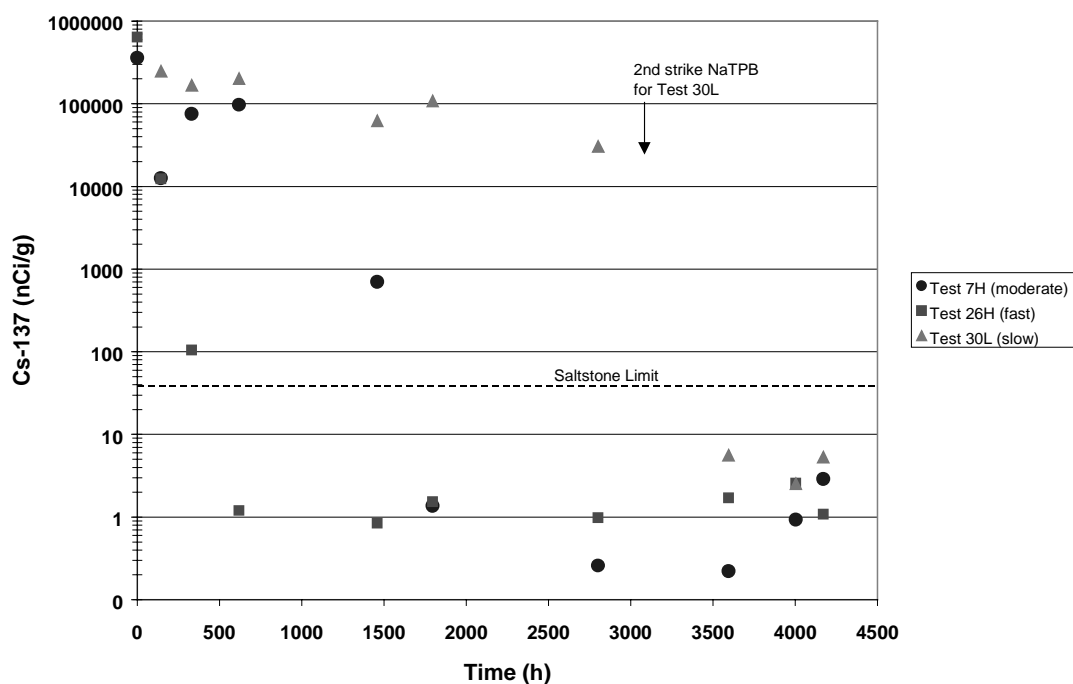
Cesium Precipitation

Cesium decontamination of the test solutions proved erratic between the twelve tests. Five of the tests (i.e., 7F-L, 26F-H, 35H-L, 35H-H, and 46F-H) exhibited rapid cesium precipitation, reaching the Saltstone limit of 45 nCi/g in a few hundred hours. Three tests (7F-H, 26F-L, and 30H-H) precipitated cesium at a slower rate, reaching the Saltstone limit around a thousand hours of testing. The remaining four tests (13H-L, 13H-H, 30H-L, and 46F-L) took considerably longer. Figure 4 shows the three distinct types of cesium precipitation behavior. The lack of any pattern between tank number, temperature, or even percent excess NaTPB of the affected tests indicates that another property affected the rate of decontamination. The most probable cause is the extremely low solubility of NaTPB in high ionic strength salt solutions such as these tested. One could expect approximately an order of magnitude change (~300 mg/L to ~30 mg/L) in NaTPB solubility in going from 5.0 M sodium to 7.0 M sodium. (A post-mortem analysis, see section on p. 13, of the test residues showed all had sodium concentrations in excess of 5.3 M, one as high as 8.5 M.) Once added, the majority of the NaTPB precipitated and its

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dissolution was very slow under the mild degree of agitation. An attempt was made to increase the shaker speeds but this resulted in breaking of the clamps holding the vessels in place. The decision was made to add additional NaTPB to the tests. However, the available material from Test 13H-H was depleted earlier than expected. As a result, no additional NaTPB was added to the test and it never obtained decontamination. Additional NaTPB was not added to Test 46F-L since it exhibited signs of increasing decontamination. Test 30H-L achieved decontamination soon after the second addition of NaTPB. Test 13H-L showed only a moderate level of decontamination and failed to reach the Saltstone limit. Interestingly, the cesium data from Test 13H-L does not rule out the possibility that reaction of solid tetraphenylborate was occurring at the end of the test (however, the soluble boron data does not verify this observation). It should be noted that the presence of soluble cesium indicates the amount of soluble NaTPB is very low and hence reaction of soluble NaTPB in the system is mass transfer limited. However, the long test duration magnifies detection of the even the slowest of reactions and offers ample time for overcoming induction periods or long catalyst activation times. Of the twelve tests, seven showed measurable levels of soluble NaTPB. These were Tests 7F-L, 7F-H, 26F-L, 26F-H, 35H-L, 35H-H, and 46F-H.

Figure 4. Graph exhibiting the three distinct types of cesium precipitation behavior exhibited by the twelve tests.



Evaluation of High-Level Waste Test Data for Catalyzed Decomposition of NaTPB

Eleven of the twelve tests (Test 13H-H material depleted too quickly to perform HPLC analysis) showed decomposition of the added 1PB and the resulting formation of nearly equivalent amounts of phenol. This demonstrates that the solutions are capable of producing phenylborate decomposition. However, the formation of triphenylborane (3PB) and to some extent 2PB provides the most direct evidence of catalytic NaTPB decomposition. Only tests 35H-L and

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35H-H showed detectable quantities of 3PB. It is feasible that a reaction might occur which produced no detectable 3PB, 2PB, or 1PB (i.e., 3PB would form at a slow rate and then be consumed along with any 2PB and 1PB at a sufficiently fast rate so that no 3PB, 2PB, or 1PB would be detected). Test 35H-H also produced detectable quantities of 2PB.

Figure 5 provides the reaction profile for Test 35H-H. For comparative purposes, the reaction profile of Test 26F-H, a non-reactive case, is shown in Figure 6. The rate of increase in soluble boron from Test 35H-H conservatively indicates the benzene generation rate from NaTPB decomposition to be less than 0.2 mg/(L•h). A similar analysis of Test 35H-L yields a conservative rate of benzene generation of less than 0.08 mg/(L•h). Note that the calculated rates are based upon small changes in soluble boron concentration and are conservative. The actual values are likely less than indicated. In consideration, such small changes in soluble boron are evident in other tests (e.g., Test 30H-H). However, Tests 35H-L and Test 35H-H differ from the others in that the boron data shows a more consistent trend, rather than scatter, and reaction did occur as evidenced by formation of 3PB and 2PB (not observed elsewhere). Regression of the data from Test 35H-H yields rate constants of 3.5 E-5, 3.3 E-4, 2.9 E-3, and 5.8 E-3 h⁻¹ for NaTPB, 3PB, 2PB, and 1PB, respectively. These rates are of similar magnitude to those obtained from the decomposition of the same species in Tank 49H in January 2000. Rate constants were not obtainable from the Test 35H-L data. The low benzene generation rate observed in Test 35H-H is more than 2 orders of magnitude less than observed (~ 40 mg/(L•h)) in the previous demonstration with high level waste.⁴ Note, that the latter rate serves to define the baseline reaction rate for the facility design.⁹

A comparison of the composition of Tank 35H waste (post-dilution) versus the other five tank wastes yields some insight into its greater (relative) degree of reactivity. Most notably, the soluble mercury level in the Tank 35H waste is greater than in any other tank waste sample (see Table 3). Interestingly, Tank 46F waste sample contains more total mercury, but it is largely insoluble (most likely insoluble HgO). Examination of the Pd, Rh, and Ru concentrations

Figure 5. Reaction profile of Test 35H-H, a reactive test case.

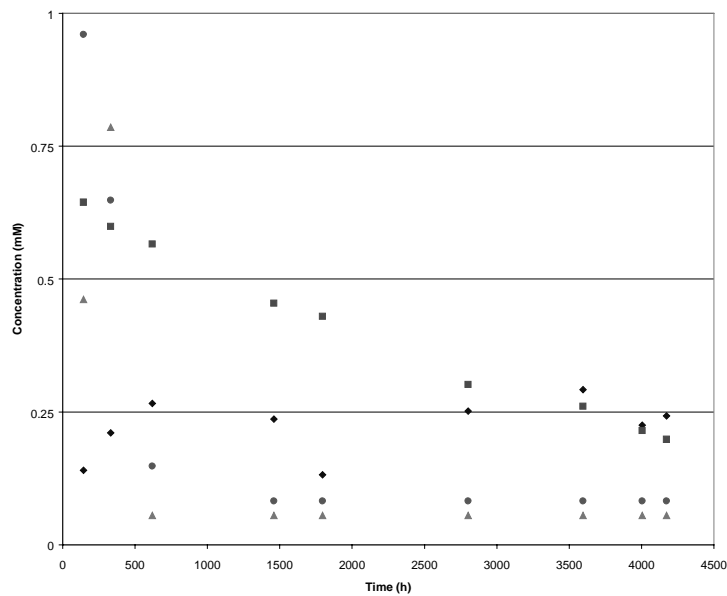
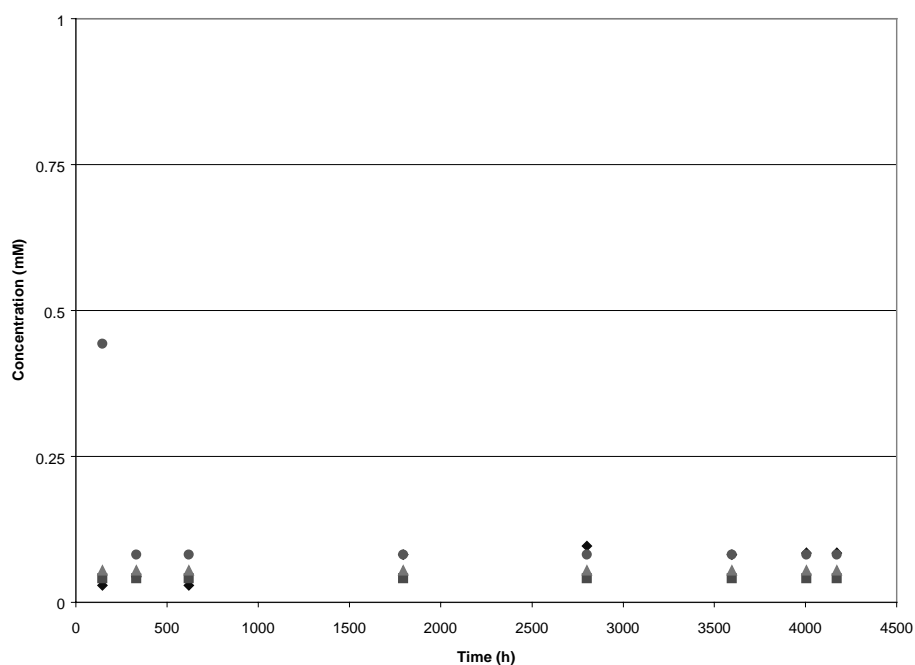


Figure 6. Reaction profile of Test 26F-H, a non-reactive test case.

(also post-dilution –see Table 4) indicates Tank 35H contained the largest concentrations of each of the three metals. Although all tests had measurable concentrations of each species. Palladium has been shown to be ~ 4X more active than Ru and Rh. The concentration of Pd in the tests was remarkably close and ranged from 0.05 to 0.16 mg/L. In almost all cases the majority of Pd was soluble in the as received samples. Therefore, all should have approximately the same level of reactivity if all of the Pd in each case were able to activate. A Pd concentration of 0.2 mg/L approximates the most observed in previous real waste tests with significant reactivity.⁴ The lack of reactivity suggests that another variable (e.g., mercury, or Rh, or Ru) or combination of variables also participates in and influences the reaction process.

Table 3. Mercury concentrations of the six waste tank samples used in testing (after dilution to 4.7 M).

Tank	Soluble Hg (mg/L)	Insoluble Hg (mg/L)
7F	3.4	0
13H	2.9	8.2
26F	3.1	4.3
30H	7.2	4.7
35H	47.5	0
46F	4.0	47.6

Table 4. Palladium, rhodium, and ruthenium concentrations of the six waste tank samples used in testing (after dilution to 4.7 M).

Tank	Soluble Pd (mg/L)	Insoluble Pd (mg/L)	Soluble Rh (mg/L)	Insoluble Rh (mg/L)	Soluble Ru (mg/L)	Insoluble Ru (mg/L)	Total Pd, Rh, Ru (mg/L)
7F	0.16	0.00	1.16	0.00	0.64	0.00	1.96
13H	0.05	0.00	1.91	0.00	1.99	0.00	3.95
26F	0.06	0.04	0.75	1.33	1.14	1.04	4.36
30H	0.12	0.00	1.68	0.00	0.98	0.73	3.51
35H	0.14	0.00	0.42	2.09	0.33	3.51	6.49
46F	0.04	0.04	0.83	0.00	1.20	0.00	2.11

Post-Mortem Analysis

Researchers analyzed the residual slurries from each remaining test for both sodium content and low concentrations of phenylborates. The post-mortem analyses arose from questions received several months after the tests ended. The actual time between the start of testing and post-mortem analyses was approximately 11 months. During that time, the residual materials were stored in capped, polyethylene bottles at ambient temperature (unmonitored). The vapor space in the bottles contained air.

Table 5 and Table 6 contain the post-mortem sodium and phenylborate data, respectively. The sodium data proved significantly higher than the value (i.e., 4.8 M) originally targeted or measured. The cause may be attributed to either poor analytical results, underdilution (resulting from error in the initial characterization of the concentrated wastes prior to use), evaporation, or more likely a combination of all three. The latter would explain the gross deviation from the target concentration (4.7 M Na) as well as the slight deviations between high and low tests with

Table 5. Post-mortem sodium data.

Tank ^a	Na (M)	Relative Rate of DF
7F-L	5.5	rapid
7F-H	5.8	slow
13H-L	8.5	very slow
26F-L	5.7	slow
26F-H	5.5	rapid
30H-L	6.2	very slow
30H-H	6.8	slow
35H-L	5.5	rapid
35H-H	5.3	rapid
46F-H	5.5	rapid

^aTank samples 13H-H and 46F-L were previously depleted (i.e., no residue remained for analysis).

Table 6. Post-mortem phenylborate data.

Tank^a	NaTPB (mg/L)^b	3PB (mg/L)^b	2PB (mg/L)^b	1PB (mg/L)^b	Phenol (mg/L)^b
7F-L	20 (<10)	5 (<10)	<1 (<10)	<10 (13)	423 (473)
7F-H	<1 (21)	<1 (<10)	<1 (<10)	<10 (<10)	26 (194)
13H-L	<1 (<10)	2 (<10)	4 (<10)	21 (<10)	191 (<10)
26F-L	19 (16)	3 (<10)	10 (<10)	15 (<10)	270 (253)
26F-H	57 (29)	23 (<10)	2 (<10)	<10 (<10)	177 (114)
30H-L	2 (<10)	4 (<10)	2 (<10)	<10 (<10)	110 (150)
30H-H	20 (<10)	2 (<10)	<1 (<10)	<10 (<10)	<10 (726)
35H-L	1 (69)	<1 (30)	8 (<10)	34 (<10)	938 (191)
35H-H	53 (83)	58 (48)	2 (<10)	16 (<10)	1054 (1084)
46F-H	3 (11)	9 (<10)	5 (<10)	<10 (<10)	36 (178)

^aTank samples 13H-H and 46F-L were previously depleted (i.e., no residue was available).

^bValue shown in parentheses represents the prior analysis (February 2001).

the same waste tank material (e.g., Tank 7F waste was diluted and then split to produce Tank 7F-L and 7F-H samples). The most confusing observation is the difference in obtaining decontamination in Tank 7F-L versus 7F-H. The solutions were equivalent at the outset of testing and one would expect the higher temperature to drive Test 7F-H to obtain decontamination faster than 7F-L. The cause of this discrepancy is unknown. Even though the vessels were sealed, evaporation during and after testing may have been brought on by heating, repeated gas purging, and high ventilation flow through the Shielded Cells. The action of the shakers was shown in at least one instance to have loosened the plugs on a vessel. The observed high sodium values would reduce the solubility of NaTPB. The data appears to correlate well with the observed relative rates of decontamination.

In the case of the phenylborate data, researchers increased the analytical sensitivity by approximately 10X from that used during the previous measurements. Installation and use of a new HPLC instrument and detector improved the signal to noise ratio and therefore provided a lower detection limit for NaTPB, 3PB and 2 PB. For this reason, it was possible to detect NaTPB, 3PB, and 2PB concentrations greater than 1 mg/L. The data shows that all but two of the tests now contained measurable concentrations of NaTPB. Furthermore, all but two (Tank samples 7F-H, and 35H-L) contained 3PB and all but three (Tank samples 7F-H, 7F-H, and 30H-H) showed the presence of 2PB, indicating some degree of reaction occurred in the other tests. The phenylborate and phenol data also suggest a substantial reaction occurred in Test 35H-L after the testing ceased. One puzzling observation is noted with the phenol data. In many instances (e.g., Test 30H-H), the phenol concentration decreased significantly. There is no explanation for this phenomena (i.e., it doesn't react or decompose or precipitate out).

Table 7 contains a summary of relative reactivity for the twelve tests as well as the most likely variables affecting reactivity. Clearly, Tank 35H waste contained the most favorable of each factor (i.e., lowest Na - rapid DF, highest metals, and highest soluble Hg). No other waste tests contained the same degree of favorable conditions. The testing serves to demonstrate the complexity and dependence of the reaction system on multiple factors.

Table 7. A comparison of relative reactivity and potential factors.

Tank^a Sample	Na (M)	Relative Rate of DF	Total Pd, Rh, Ru (mg/L)	Total Soluble Hg (mg/L)	Relative Rate of Reaction^b
7F-L	5.5	rapid	1.96	3.4	no indication of reaction
7F-H	5.8	slow	1.96	3.4	slow w/ intermed. formed
13H-L	8.5	very slow	3.95	2.9	insufficient data
13H-H	unknown ^a	very slow	3.95	2.9	insufficient data
26F-L	5.7	slow	4.36	3.1	no indication of reaction
26F-H	5.5	rapid	4.36	3.1	slow w/ intermed. formed
30H-L	6.2	very slow	3.51	7.2	no indication of reaction
30H-H	6.8	slow	3.51	7.2	slow w/ intermed. formed
35H-L	5.5	rapid	6.49	47.5	measureable
35H-H	5.3	rapid	6.49	47.5	fastest
46F-L	unknown ^a	very slow	2.11	4.0	no indication of reaction
46F-H	5.8	rapid	2.11	4.0	slow w/ intermed. formed

^aTank samples 13H-H and 46F-L were previously depleted (i.e., no residue remained for analysis).

^bRelative reactivity based upon production of 3PB, 2PB, and an increase in soluble boron during actual test time.

CONCLUSIONS

This report describes batch testing of six different tank wastes for catalytic NaTPB decomposition. The waste samples came from Tanks 7F, 13H, 26F, 30H, 35H, and 46F. Tests with the six wastes occurred at both ambient (22-26 °C) and elevated (45 °C) temperature. Testing lasted six months. Samples obtained from the tests yielded the following observations.

- Under the conditions tested (45 °C), the maximum reaction rate observed – for samples from Tank 35H -- equates to a theoretical benzene generation rate of 0.2 mg/(L-h).
- The waste sample from Tank 35H contained the highest concentration of soluble mercury, an element known to participate in the reaction sequence. This finding suggests the value of continuing the efforts to explore the influence of soluble mercury concentration on the catalytic reaction.
- The maximum rate (0.08 mg/(L-h)) observed at 25 °C fell well within the design basis limit for the proposed facility.
- Palladium concentrations in the six waste samples, when diluted to a target concentration of 4.7 M sodium, ranged from 0.05 to 0.16 mg/L. Analyses did not detect the presence of platinum in the waste. The total concentration of noble metals (i.e., palladium, rhodium, and ruthenium) ranged from 1.96 to 6.49 mg/L
- The observed reaction rate appeared to correlate directly with the measured soluble sodium content of the waste. This finding suggests that lack of slow mass transfer and dissolution of tetraphenylborate likely limited the rate of reaction.
- Review of the data suggests four variables influenced the observed reactivity: temperature, concentration of noble metals, presence of soluble mercury, and presence of soluble tetraphenylborate.
- Post-mortem analysis of residual test materials, 11 months after starting the tests, showed that 9 of the tests exhibited evidence of decomposition.

Future Program Elements

Additional real waste batch tests are warranted if Small Tank Tetraphenylborate Precipitation is selected as the technology for deployment in the Salt Processing Facility. Specifically, additional testing with Tank 35H waste using increased agitation would likely provide more reliable reaction rates. Furthermore, continued batch testing with additional tank waste samples and increased agitation rates would serve to increase the available data sets investigating the reactivity of high level waste.

QUALITY ASSURANCE

Personnel utilized reagent grade chemicals and prepared solutions with calibrated balances checked daily before use.¹⁰ The weights used for balance checks received calibration by the SRTC Standards Laboratory. The accuracy of glassware used to measure volumes was verified by gravimetric methods using water as a standard.¹¹ All M&TE used in this task received calibration or verification for accuracy prior to their use. The Analytical Development Section performed all chemical analyses. Laboratory Notebook WSRC-NB-2000-00125 (M. Barnes) contains the experimental log for these experiments. Anderson recorded the data from examination of the nitrogen sparging efficiency in Laboratory Notebook WSRC-NB-2000-00214. This report, in part, finalizes the work identified by Item 2.0 of "Applied Technology Integration Scope of Work Matrix for Small Tank TPB Precipitation (Demonstration Phase)," HLW-SDT-99-0353, Rev. 5, November 21, 2000.

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APPENDIX 1: WASTE SAMPLE CHARACTERIZATION

SUPERNATE COMPOSITION*

Analysis	Tank 7F	Tank 13H	Tank 26F	Tank 30H	Tank 35H	Tank 46F
Na (M)-ICPES	6.94	5.99	9.30	7.55	6.69	12.15
Na (M)-AA	6.32	5.57	8.50	6.96	6.16	11.66
K (M)-AA	0.0667	0.0434	0.0534	0.0413	0.0177	0.0790
OH (M)	4.4	7.2	8.0	8.0	5.9	4.4
Al (M)-ICPES	0.53	0.37	0.57	0.47	0.36	0.73
Cs-137 (uCi/mL)	623.5	1944	1451.5	2097.5	948.5	2103.5
Sr-90 (dpm/mL)	103000	890000	334000	1770000	3225000	675000
F (M)	0.012	< 0.001	0.009	< 0.001	< 0.001	< 0.001
Cl (M)	0.0056	0.0054	0.0113	0.0037	< 0.006	0.0141
NO2 (M)	0.96	1.64	1.56	1.16	1.01	1.76
NO3 (M)	1.30	2.18	2.04	3.05	2.35	1.30
PO4 (M)	0.026	0.028	0.015	0.022	< 0.011	0.017
SO4 (M)	0.049	0.019	0.013	0.048	0.031	0.006
C2O4 (M)	0.0031	< 0.001	< 0.001	0.0015	< 0.011	< 0.001
HCO2 (M)	< 0.002	< 0.002	0.0211	< 0.002	0.0071	0.0200
Pu-239/40 (dpm/mL)	< 1070	< 1650	< 1095	< 9650	< 7700	226.5
Pu-238 (dpm/mL)	6150	51000	8250	630000	425500	5550
Ag (mg/L)-ICPES	< 3.000	< 3.000	< 3.000	< 3.000	< 3.000	< 3.000
B (mg/L)-ICPES	29.15	89.7	213.3	104.75	122.2	317.1
Ba (mg/L)-ICPES	< 1.000	4.3	< 1.000	< 1.000	< 1.000	1.3
Ca (mg/L)-ICPES	86.6	79.65	79.8	79.75	79.25	82.5
Cd (mg/L)-ICPES	1.9	< 1.500	1.75	2.45	< 1.500	2.05
Co (mg/L)-ICPES	3.05	3.2	2.75	< 2.500	< 2.500	3.8
Cr (mg/L)-ICPES	197.05	222.2	248.05	232.05	105	375.2
Cu (mg/L)-ICPES	< 1.500	2.25	1.6	2.15	1.7	2.95
Fe (mg/L)-ICPES	13.75	10.85	15.25	16.85	13.6	22.05
Hg (mg/L)-CVAA	2.365	0.77	0.66	5.5	26.18	0.99
Hg (mg/L)-ICPMS	5.00	3.65	5.91	11.0	62.72	10.49
La (mg/L)-ICPES	< 5.500	< 5.500	< 5.500	< 5.500	< 5.500	< 5.500
Li (mg/L)-ICPES	< 1.000	1.6	< 1.000	1.2	6.4	1.45
Mg (mg/L)-ICPES	1.9	1.9	2.05	2	1.9	2.4
Mn (mg/L)-ICPES	0.65	0.8	1.4	0.75	0.6	1.9
Mo (mg/L)-ICPES	51.9	179.85	90.5	173.15	46.95	129.95
Ni (mg/L)-ICPES	< 3.500	3.9	< 3.500	< 3.500	< 3.500	< 3.500
P (mg/L)-ICPES	371.35	206.8	308.9	227.35	203.5	460.6
Pb (mg/L)-ICPES	25.3	20.8	27.3	38.05	30.1	32.2
Si (mg/L)-ICPES	16.55	18.65	36.05	22	44.8	49.05
Sn (mg/L)-ICPES	26.1	23.2	20.95	19.8	16.4	25.5
Sr (mg/L)-ICPES	< 0.500	< 0.500	< 0.500	< 0.500	< 0.500	< 0.500
Ti (mg/L)-ICPES	< 1.000	< 1.000	< 1.000	1.2	< 1.000	1.45
V (mg/L)-ICPES	3.85	4.45	4.2	4.2	2.45	7.6
Zn (mg/L)-ICPES	12.3	9.85	20.3	10.25	17.15	28.2
Zr (mg/L)-ICPES	2.35	2.8	< 2.000	2.6	2.2	3.15

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SUPERNATE COMPOSITION (continued)*

Analysis	Tank 7F	Tank 13H	Tank 26F	Tank 30H	Tank 35H	Tank 46F
Ag (mg/L)-ICPMS	0.23	0.27	0.25	0.24	0.22	0.22
Pd (mg/L)-ICPMS	0.24	0.06	0.11	0.18	0.19	0.11
Rh (mg/L)-ICPMS	1.71	2.44	1.48	2.70	0.60	2.14
Ru (mg/L)-ICPMS	0.95	2.53	2.25	1.57	0.47	3.10
Pt (mg/L)-ICPMS	< 2.49E-03	< 2.49E-03	< 2.49E-03	< 2.49E-03	< 2.49E-03	< 2.49E-03
Tc-99 (mg/L)-ICPMS	7.10	14.9	13.0	19.0	9.80	18.3

*Data obtained from analysis of filtered samples.

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SOLIDS COMPOSITION*

Analysis	Tank 7F	Tank 13H	Tank 26F	Tank 30H	Tank 35H	Tank 46F
Pu-239/40 (dpm/mL)	0.00E+00	1.80E+02	0.00E+00	2.15E+04	1.09E+04	4.84E+02
Pu-238 (dpm/mL)	6.50E+02	6.00E+04	3.55E+03	1.40E+06	5.15E+05	9.35E+03
Sr-90 (dpm/mL)	1.65E+04	1.53E+06	1.29E+05	3.47E+07	2.28E+06	0.00E+00
Ag (mg/L)-ICPES	0.65-3.65	< 3.0	< 3.0	< 3.0	< 3.0	< 3.0
Al (mg/L)-ICPES	0	5065	800	3185	805	2450
B (mg/L)-ICPES	0.0	44.1	9.5	26.4	9.4	13.2
Ba (mg/L)-ICPES	1.0-1.2	2.40	< 1.0	21.2-22.2	< 1.0	0.00
Ca (mg/L)-ICPES	13.90	0.00	1.85	0.45	0.00	0.00
Cd (mg/L)-ICPES	0.00	0.75-2.25	0.20	0.00	<1.5	0.20
Co (mg/L)-ICPES	0.10	0.00	0.00	0.25-2.75	0.45-2.95	0.00
Cr (mg/L)-ICPES	0.00	105.55	9.80	49.35	7.50	17.95
Cu (mg/L)-ICPES	1.15-2.65	2.90	1.95	4.20	2.70	0.75
Fe (mg/L)-ICPES	0.00	8.55	15.60	60.55	16.90	34.80
Hg (mg/L)-ICPMS	0.00	10.3	8.14	7.24	0.00	124.3
La (mg/L)-ICPES	1.35-6.85	< 5.5	< 5.5	< 5.5	< 5.5	< 5.5
Li (mg/L)-ICPES	0.75-1.75	0.35	<1.0	1.55	0.50	0.00
Mg (mg/L)-ICPES	1.90	0.00	0.00	1.95	0.35	0.00
Mn (mg/L)-ICPES	0.85	0.45	0.60	124.15	4.65	0.40
Mo (mg/L)-ICPES	0.00	84.25	3.65	37.65	3.00	3.90
Na (M)-ICPES	0.00	2.90	0.44	1.85	0.56	0.93
Ni (mg/L)-ICPES	1.35-4.85	0.00	<3.5	1.95-5.45	<3.5	<3.5
P (mg/L)-ICPES	0.00	91.55	0.00	35.05	0.00	6.80
Pb (mg/L)-ICPES	8.65	18.20	0.00	0.00	0.00	0.00
Si (mg/L)-ICPES	5.05	2.85	2.15	2.10	1.80	0.00
Sn (mg/L)-ICPES	0.00	1.65	1.75	8.60	7.20	0.00
Sr (mg/L)-ICPES	0.05-0.55	<0.5	<0.5	0.1-0.6	<0.5	<0.5
Ti (mg/L)-ICPES	1.05-2.05	<1.0	<1.0	0.00	<1.0	0.00
V (mg/L)-ICPES	0.55	0.85	0.50	0.40	0.00	0.00
Zn (mg/L)-ICPES	0.00	3.25	0.45	1.15	0.00	1.10
Zr (mg/L)-ICPES	2.25	0.95	0.15-2.15	1.40	0.00	0.00
Ag (mg/L)-ICPMS	1.42	0.46	1.35	1.84	2.01	1.67
Pd (mg/L)-ICPMS	0.00	0.00	0.07	0.00	0.00	0.11
Rh (mg/L)-ICPMS	0.00	0.00	2.64	0.00	2.97	0.00
Ru (mg/L)-ICPMS	0.00	0.00	2.05	1.17	4.99	0.00
Pt (mg/L)-ICPMS	0.00	0.00	0.00	0.00	0.00	0.00
Tc-99 (mg/L)-ICPMS	0.00	0.00	13.11	0.00	16.6	0.00

*The listed concentrations are based on a total solution volume basis and not a dry weight basis since solids were not actually isolated and analyzed alone. Concentrations were calculated from the difference in concentration of filtered and unfiltered samples. Where no difference in concentration was observed, the insoluble concentration is reported as 0.00 mg/L.

APPENDIX 2: TEST DATA

Test 7F-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	353333	NA*	NA	NA	NA	NA	NA
145	65000	NA	NA	NA	NA	NA	NA
332	NA	8.2	NA	NA	NA	NA	NA
619	1.2	8.1	0.09	< 0.04	< 0.05	0.38	4.7
1459	2.1	8.5	0.09	< 0.04	< 0.05	< 0.08	4.8
1795	0.4	8.4	< 0.03	< 0.04	< 0.05	< 0.08	4.9
2802	0.4	9.0	0.06	< 0.04	< 0.05	< 0.08	5.0
3595	1.0	7.9	0.09	< 0.04	< 0.05	< 0.08	5.0
4004	1.1	10.4	< 0.03	< 0.04	< 0.05	0.10	5.1
4171	1.4	8.6	< 0.03	< 0.04	< 0.05	0.11	5.0

*Indicates analysis was not performed.

Test 13H-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	1291667	NA*	NA	NA	NA	NA	NA
145	908333	NA	NA	NA	NA	NA	NA
332	791667	NA	NA	NA	NA	NA	NA
619	640833	NA	NA	NA	NA	NA	NA
1459	480583	NA	NA	NA	NA	NA	NA
1795	505000	NA	NA	NA	NA	NA	NA
2802	439133	NA	NA	NA	NA	NA	NA
3595	39750	NA	NA	NA	NA	NA	NA
4004	37000	16.3	< 0.03	< 0.04	< 0.05	< 0.08	0.11
4171	109967	15.7	< 0.03	< 0.04	< 0.05	< 0.08	0.11

*Indicates analysis was not performed.

Test 26F-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	640833	NA*	NA	NA	NA	NA	NA
145	99833	16.4662	< 0.03	< 0.04	< 0.05	4.0	1.2
332	54025	22.5717	< 0.03	< 0.04	< 0.05	3.3	1.8
619	87667	17.2988	< 0.03	< 0.04	< 0.05	2.2	2.5
1459	4.9	NA	NA	NA	NA	NA	NA
1795	2.2	17.4	0.04	< 0.04	< 0.05	0.52	3.4
2802	1.2	17.0	0.05	< 0.04	< 0.05	0.25	3.4
3595	1.4	17.8	< 0.03	< 0.04	< 0.05	< 0.08	3.2
4004	9.2	17.1	< 0.03	< 0.04	< 0.05	0.11	3.0
4171	1.5	16.2	0.05	< 0.04	< 0.05	< 0.08	2.7

*Indicates analysis was not performed.

Test 30H-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	1136667	NA*	NA	NA	NA	NA	NA
145	247667	17.2	< 0.03	< 0.04	< 0.05	3.2	1.6
332	167750	13.9	< 0.03	< 0.04	< 0.05	2.5	2.3
619	201583	13.4	< 0.03	< 0.04	< 0.05	1.2	3.1
1459	62165	NA	NA	NA	NA	NA	NA
1795	108308	13.4	< 0.03	< 0.04	< 0.05	< 0.08	3.2
2802	30499	14.2	< 0.03	< 0.04	< 0.05	< 0.08	2.2
3595	5.6	13.4	< 0.03	< 0.04	< 0.05	< 0.08	1.6
4004	2.5	13.1	< 0.03	< 0.04	< 0.05	< 0.08	1.6
4171	5.3	12.9	< 0.03	< 0.04	< 0.05	< 0.08	1.6

*Indicates analysis was not performed.

Test 35H-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	598333	NA*	NA	NA	NA	NA	NA
145	4.1	12.1	0.16	0.04	< 0.05	1.2	2.5
332	7.2	12.1	0.19	0.10	< 0.05	0.40	3.2
619	2.7	13.0	0.24	0.15	0.06	< 0.08	3.4
1459	7.6	13.3	0.19	0.14	< 0.05	< 0.08	3.0
1795	3.0	13.3	0.16	0.17	< 0.05	0.17	3.0
2802	0.4	14.1	0.22	0.11	< 0.05	< 0.08	2.5
3595	1.3	13.9	0.20	0.13	< 0.05	0.09	2.3
4004	1.1	13.7	0.24	0.12	< 0.05	< 0.08	2.0
4171	2.7	13.1	0.20	0.12	< 0.05	< 0.08	2.0

*Indicates analysis was not performed.

Test 46F-L							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	670000	NA*	NA	NA	NA	NA	NA
145	47950	23.4	< 0.03	< 0.04	< 0.05	3.7	1.2
332	36033	18.8	< 0.03	< 0.04	< 0.05	3.1	1.6
619	34608	18.8	< 0.03	< 0.04	< 0.05	2.3	2.4
1459	7171	NA	NA	NA	NA	NA	NA
1795	4311	19.1	< 0.03	< 0.04	< 0.05	0.58	3.2
2802	1.4	14.4	< 0.03	< 0.04	< 0.05	0.21	3.2
3595	0.8	19.1	< 0.03	< 0.04	< 0.05	0.14	2.9
4004	0.8	18.4	< 0.04	< 0.04	< 0.05	0.10	2.7
4171	11	17.9	< 0.03	< 0.04	< 0.05	0.11	2.7

*Indicates analysis was not performed.

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Test 7F-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	358333	NA*	NA	NA	NA	NA	NA
145	12533	10.0	< 0.03	< 0.04	< 0.05	2.2	3.0
332	75392	6.2	< 0.03	< 0.04	< 0.05	1.0	2.3
619	97417	6.0	< 0.03	< 0.04	< 0.05	< 0.08	2.7
1459	701	NA	NA	NA	NA	NA	NA
1795	1.4	6.7	0.05	< 0.04	< 0.05	< 0.08	2.8
2802	0.3	6.8	0.04	< 0.04	< 0.05	< 0.08	2.5
3595	0.2	7.0	0.07	< 0.04	< 0.05	< 0.08	2.3
4004	0.9	7.0	0.04	< 0.04	< 0.05	< 0.08	2.1
4171	2.9	6.8	0.06	< 0.04	< 0.05	< 0.08	2.1

*Indicates analysis was not performed.

Test 13H-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	1292500	NA*	NA	NA	NA	NA	NA
145	941667	NA	NA	NA	NA	NA	NA
332	786667	NA	NA	NA	NA	NA	NA
619	763333	NA	NA	NA	NA	NA	NA
1459	342558	NA	NA	NA	NA	NA	NA

*Indicates analysis was not performed.

Test 26F-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	640000	NA*	NA	NA	NA	NA	NA
145	12275	14.5	< 0.03	< 0.04	< 0.05	0.44	2.6
332	105	15.9	0.04	< 0.04	< 0.05	< 0.08	2.7
619	1.2	16.1	< 0.03	< 0.04	< 0.05	< 0.08	2.8
1459	0.8	15.4	NA	NA	NA	NA	NA
1795	1.5	15.1	0.08	< 0.04	< 0.05	< 0.08	2.0
2802	1.0	16.2	0.10	< 0.04	< 0.05	< 0.08	1.8
3595	1.7	16.1	0.08	< 0.04	< 0.05	< 0.08	1.5
4004	2.6	16.1	0.08	< 0.04	< 0.05	< 0.08	1.3
4171	1.1	15.3	0.08	< 0.04	< 0.05	< 0.08	1.2

*Indicates analysis was not performed.

Test 30H-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	1133333	NA*	NA	NA	NA	NA	NA
145	102750	15.5	< 0.03	< 0.04	< 0.05	0.56	2.9
332	355667	14.1	< 0.03	< 0.04	< 0.05	< 0.08	3.9
619	182917	14.0	< 0.03	< 0.04	< 0.05	< 0.08	3.6
1459	25	NA	NA	NA	NA	NA	NA
1795	1.6	15.1	< 0.03	< 0.04	< 0.05	< 0.08	5.0
2802	1.6	17.0	< 0.03	< 0.04	< 0.05	< 0.08	6.2
3595	5.4	18.0	< 0.03	< 0.04	< 0.05	< 0.08	7.5
4004	5.4	17.8	< 0.03	< 0.04	< 0.05	< 0.08	7.6
4171	41	17.8	< 0.03	< 0.04	< 0.05	< 0.08	7.7

*Indicates analysis was not performed.

Test 35H-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	596667	NA*	NA	NA	NA	NA	NA
145	6.2	17.4	0.14	0.64	0.46	0.96	3.2
332	5.1	16.5	0.21	0.60	0.79	0.65	8.0
619	13	17.7	0.27	0.57	0.05	0.15	10.2
1459	26	16.9	0.24	0.45	< 0.05	< 0.08	10.1
1795	2.4	17.6	0.13	0.43	< 0.05	< 0.08	10.3
2802	3.4	18.6	0.25	0.30	< 0.05	< 0.08	10.9
3595	9.3	19.3	0.29	0.26	< 0.05	< 0.08	11.6
4004	1.2	19.4	0.23	0.21	< 0.05	< 0.08	11.3
4171	2.0	19.2	0.24	0.20	< 0.05	< 0.08	11.5

*Indicates analysis was not performed.

Test 46F-H							
Reaction Time (h)	Cs-137 (nCi/g)	Boron (mM)	NaTPB (mM)	3PB (mM)	2PB (mM)	1PB (mM)	Phenol (mM)
0	672500	NA*	NA	NA	NA	NA	NA
145	49417	17.1	< 0.03	< 0.04	< 0.05	2.08	2.8
332	35967	18.9	0.13	0.16	0.24	0.89	3.7
619	91	19.0	0.04	< 0.04	< 0.05	< 0.08	4.0
1459	12	18.5	0.05	< 0.04	< 0.05	< 0.08	3.5
1795	1023	19.0	0.04	< 0.04	< 0.05	< 0.08	3.2
2802	1.2	19.4	< 0.03	< 0.04	< 0.05	< 0.08	2.9
3595	7.3	19.4	< 0.03	< 0.04	< 0.05	< 0.08	2.3
4004	157	19.1	0.09	< 0.04	< 0.05	< 0.08	2.1
4171	2.3	18.5	0.03	< 0.04	< 0.05	< 0.08	1.9

*Indicates analysis was not performed.