

## Examination of the Potential for Formation of Energetic Compounds in Dry Sludge

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## SUMMARY

This report details initial results from an investigation of the potential formation and fate of energetic compounds in Savannah River Site sludge. The initial studies included examination of archived sludge samples (from Tanks 8F, 12H, and 15H) as well as calorimetry and impact testing of simulated sludge samples. The testing provide the following conclusions.

- Analysis of an archived Tank 12H and Tank 15H samples showed the material experienced prior treatment that makes it non-representative of the current contents of that vessel. In contrast, the composition of the archived Tank 8F sample suggests no prior chemical treatment.
- The Differential Scanning Calorimetry (DSC) analysis of the Tank 8F, Tank 12H and Tank 15H samples showed no exothermic behavior to temperatures as high as 400 °C.
- Impact testing (with a pressure of 5.38 N/m<sup>2</sup>, or 780 psi) of simulated sludge samples showed no visible evidence of exothermic reaction (i.e., smoke) at conservatively high concentrations of mercury and silver and after drying at a temperature of 50 °C. A test on a single sample dried at 350 °C showed no indication of reaction.
- Calorimetric measurements of the same samples showed no indications of exothermic reaction. The lack of exothermic behavior suggests that unstable energetic compounds of mercury or silver do not readily form under the expected conditions for the alkaline sludge.
- The tests examined the influence of organic materials (i.e., sodium oxalate, Ionac 604 resin – either as received or after permanganate digestion, or a mixture of tri-n-butyl phosphate and n-paraffin) on these results. Adding these components at conservatively high concentrations did not result in any exothermic reactions as determined by both impact tests and calorimetry. The organic compounds do not appear to facilitate formation of energetic compounds in dried sludge.

The experiments also studied the behavior of mixtures of simulated sludge with mercury oxalate and with mercury fulminate. These two compounds decompose with moderately high release of energy when impacted or heated. These tests provide the following results.

- Impact testing with mixtures of simulated sludge containing as much as 14.9 wt % mercury oxalate or 21.5 wt % mercury fulminate showed no visible signs of reaction. The nominal mercury fulminate mass in 25 of 59 samples tested either equaled or exceeded the minimal amount of the pure mercury fulminate needed to produce a visible indication (i.e., smoke) for reaction with the impact test equipment. Similarly, 7 of 47 tests using a mixture of simulated sludge and mercury oxalate contained enough oxalate to exceed the detection as determined for the pure compound.
- Calorimetry measurements for mixtures of simulated dry sludge containing as much as 14.9 wt % of dry mercury oxalate showed no net exothermic behavior since the oxalate decomposition occurs at a temperature overlapping a region of endothermic behavior for the pure sludge. Review of the data suggests that the presence of the selected organic compounds did not alter the measurements.



- Calorimetry of mixtures of wet sludge combined with mercury oxalate and then dried suggests negligible destruction of the oxalate in the alkaline media during the short contact and drying period.
- Calorimetry measurements for mixtures of simulated dry sludge containing as much as 21 wt % of mercury fulminate showed an exothermic reaction of magnitude roughly equivalent to that for the pure fulminate. Review of the data suggests that the presence of the selected organic compounds did not alter the measurements.
- Calorimetry of mixtures of wet sludge with approximately 1.68 wt % added mercury fulminate and then dried showed no detectable exothermic reactions. Increasing the fulminate concentration to 6.81 wt % resulted in a small exothermic reaction. Note that 6.81 wt % mercury fulminate equates to 120% of the mercury in Tank 15H converted entirely to the fulminate.

These experimental findings suggest the following conclusions of importance to storage and sampling efforts for dried sludge.

- Experimental evidence does not preclude the formation of energetic compounds in the waste tanks during dry storage. However, simulated sludge with conservatively high concentrations of mercury and silver failed to show any exothermic behavior without the deliberate addition of such compounds. Presence of conservatively high concentrations of several organic species did not alter this result.
- Both mercury oxalate and mercury fulminate survived contact with moist sludge (0.22 M hydroxide) for periods as long as one day. Nevertheless, the mixtures of sludge with mercury oxalate showed no net exothermic behavior under the conservative conditions tested. Testing with mercury fulminate required addition at relatively high concentrations in simulated sludge to provide a relatively low energy exothermic reaction.
- Calorimetry data suggests the presence of the principal organic compounds of interest within simulated sludge does not enhance the energetic behavior of the material, even when both the organic and the energetic standard (i.e., mercury oxalate or mercury fulminate) exist in conservatively high concentrations.

The results of this study indicate that planned sampling of Tank 12H may proceed with minimal risk of experiencing an exothermic reaction during the sample collection.

Previous discussions suggested adding liquid to Tank 12H prior to sampling. Such an addition will reduce the concentration of dust – with associated radionuclides – in the tank vapor space during the sampling evolution. The liquid would also provide a means to dissipate any energy released during sampling. However, an excess addition of liquid could jeopardize the integrity of the collected sample from the perspective of determining whether energetic compounds exist during dry storage.

Future efforts should extend the studies to large masses of simulated sludge. Similarly, testing should examine the influence of extended contact to caustic on the energetic behavior of selected compounds such as mercury oxalate, mercury fulminate, and silver nitride. Additional calorimetry studies should further examine the behavior of simulated sludge with mixtures of

energetic standards, for instance, to include the combined presence of silver nitride and mercury oxalate.

## INTRODUCTION

The Savannah River Site stores High Level Waste in several forms in large carbon steel tanks. One of those forms consists primarily of insoluble iron and aluminum oxides with smaller amounts of uranium, manganese, mercury and nickel and traces of various radionuclides. Trace amounts of silver compounds also exist in the tank farm from transfers in the late 1960s of silver Berl saddles used as part of the canyon process. Ordinarily an aqueous supernate layer remains over the top of the sludge. A previous study indicated that when sludge remains in a wet state, little concern exists for any sort of energetic decomposition.<sup>1</sup> However, recent operations allowed several of the sludge tanks to dry, at least on the surface. A concern exists that certain mercury and silver compounds and mixtures of organic compounds and nitrates capable of energetic decomposition may form in the dry sludge. Also, such materials generally become more reactive when dry. High Level Waste Division requested the assistance of the Savannah River Technology Center (SRTC) in determining the likelihood for the energetic decomposition during sampling of the dry sludge.<sup>2,3</sup> This report documents initial studies to determine whether energetic compounds exist in the dry sludge in sufficient quantities to pose a safety concern.

## EXPERIMENTAL

### Radioactive Sludge Samples

For Tanks 8F, 12H, and 15H each containing dry sludge, archived samples existed in the Shielded Cells of Building 773-A, Figure 1. Original collection of the samples occurred on September 26, June 16, and May 11 of 1977, respectively. However, review of existing records and interviews with personnel failed to provide a definitive understanding of the history for these samples. Researchers could not determine from this review whether the samples received treatment in previous years. Even if previously treated, these samples, dried over an extensive period of time, might still provide insight regarding formation of any energetic compounds during drying. If previously treated and left with a less alkaline supernate, the conditions might prove more favorable to generating these unstable species.<sup>4</sup> Therefore, personnel analyzed these samples to determine how well they represent the current content of the tanks.

Personnel dissolved portions of the dried samples in triplicate by contacting with aqua regia or by fusion with sodium peroxide. Contacting the sodium peroxide fusion with nitric acid allows determination of the composition of the total sample. For both radiological protection and analytical limitations, the analyses required dilution of the digested sludge samples with deionized, distilled water before analysis.

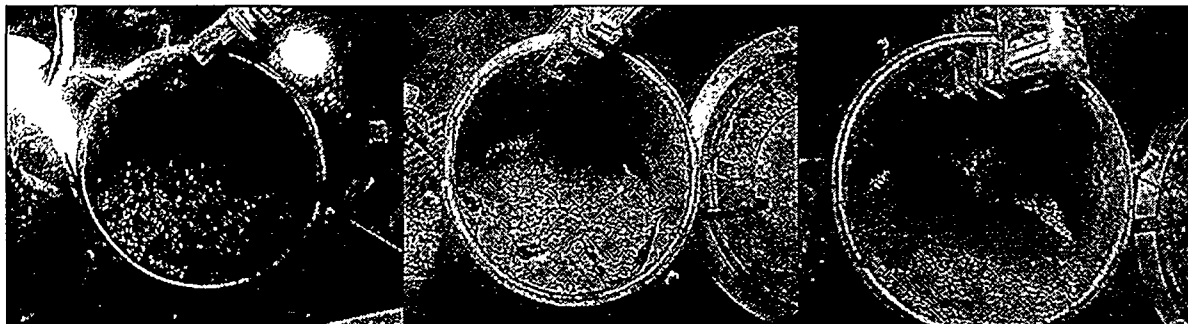


Figure 1. Archived sludge samples. From left to right, samples from Tanks 8F, 12H and 15H.

The Analytical Development Section uses the following analytical methods for determination of specific species. Measurement of sodium, aluminum, iron, and other metallic elements used inductively coupled plasma-emission spectroscopy (ICP-ES). Potassium and mercury determinations used atomic adsorption spectroscopy (AA) with the mercury measurement using the cold-vapor technique (CV). Noble metals (i.e., Ag, Rh, Ru, and Pd) concentrations came from inductively-coupled plasma mass spectrometry (ICP-MS). Gamma spectroscopy provided the concentrations of gamma-emitting fission products. Actinide concentrations derive from a combination of ICP-MS and alpha counting spectroscopy. Personnel determined the  $\text{Sr}^{90}$  content from the beta liquid scintillation counting with strontium separation on selected samples as necessary.

Researchers performed differential scanning calorimetry and thermal gravimetric analysis (TGA) on portions of the Tank 15H sample. (Calibration of the TA Instruments 910 Differential Scanning Calorimeter used the melting point of zinc as a standard. The TGA experiments used calcium oxalate decomposition as a standard.) For samples from Tanks 8F and 12H, researchers only performed the calorimetry measurement. These thermal methods allow identification of any exothermic reactions and associated sample weight loss that occurs when heating a sample. Most energetic compounds exhibit thermal instability, often at relatively low temperatures.

### Simulated Sludge

Table 1 shows the chemical composition of the Tank 15H sludge as previously determined by Coleman et al.<sup>5</sup> Researchers used this composition in developing a simulated sludge to serve as a reference material. Testing added mercury, silver, digested resin, undigested resin, sodium oxalate and a mixture of tri-n-butyl phosphate and n-paraffin to this simulated sludge (see Table 2 for range of each variable). Appendix 1 provides a listing of the individual experiments. The synthesis of the sludge also used a supernate containing 0.22 M hydroxide, 1.3 M nitrate, 0.86 M nitrite, and 0.08 M aluminate. This supernate composition approximates that measured for Tank 15H in September of 1983.<sup>6</sup> Personnel dried these various sludge samples to a constant weight using a range of drying temperatures (i.e.,  $50 \pm 3$ ,  $150 \pm 3$ ,  $250 \pm 3$ , and  $350 \pm 5$  °C). Drying times ranged from 16 to 89 hours. (Researchers checked the samples infrequently for dryness and hence recorded drying time does not correlate with completion of drying.) The highest temperature matches the maximum temperature ever experienced in a waste tank (i.e., Tank 1F during November, 1964).<sup>7</sup> See Figure 1 for views of the resulting sludge samples.

**Table 1. Major elemental composition of Tank 15H radioactive sludge.**

<u>Element</u>	<u>Concentration (elemental wt %)</u>	<u>Element</u>	<u>Concentration (elemental wt %)</u>
Fe	4.9	P	0.30
Al	30.8	Ni	0.46
Na	2.4	Cu	0.05
K	0.0	Ti	0.02
Mn	2.5	Cr	0.02
U	0.045	Th	0.23
Ca	0.21	Ru	0.064
Mg	0.15	Rh	0.013
Si	0.19	Pd	0.002
B	0.01		

**Table 2. Additives and their range of concentration for addition to simulated sludge.**

<u>Additive</u>	<u>Nominal Tank 15H Concentration</u>	<u>Range</u>	<u>Basis/Comment</u>
Mercury	4 wt %	0 - 6 wt %	Upper limit of 1.5 times maximum reported in Ref. 8
Silver	0.0003 wt %	0 - 0.0015 wt %	Upper limit of 5 times maximum reported in Ref. 8
Digested resin	Undetermined	0 - 3 wt %	Selected to exceed highest expected content based on process knowledge
Undigested resin	Undetermined	0 - 3 wt %	Selected to exceed highest expected content based on process knowledge
Tri-n-butyl Phosphate (30 vol %) and n-paraffin mixture	Undetermined	0 - 3 wt %	Selected to exceed highest expected content based on process knowledge
Sodium oxalate	Undetermined	0 - 3 wt %	Selected to exceed highest expected content based on process knowledge

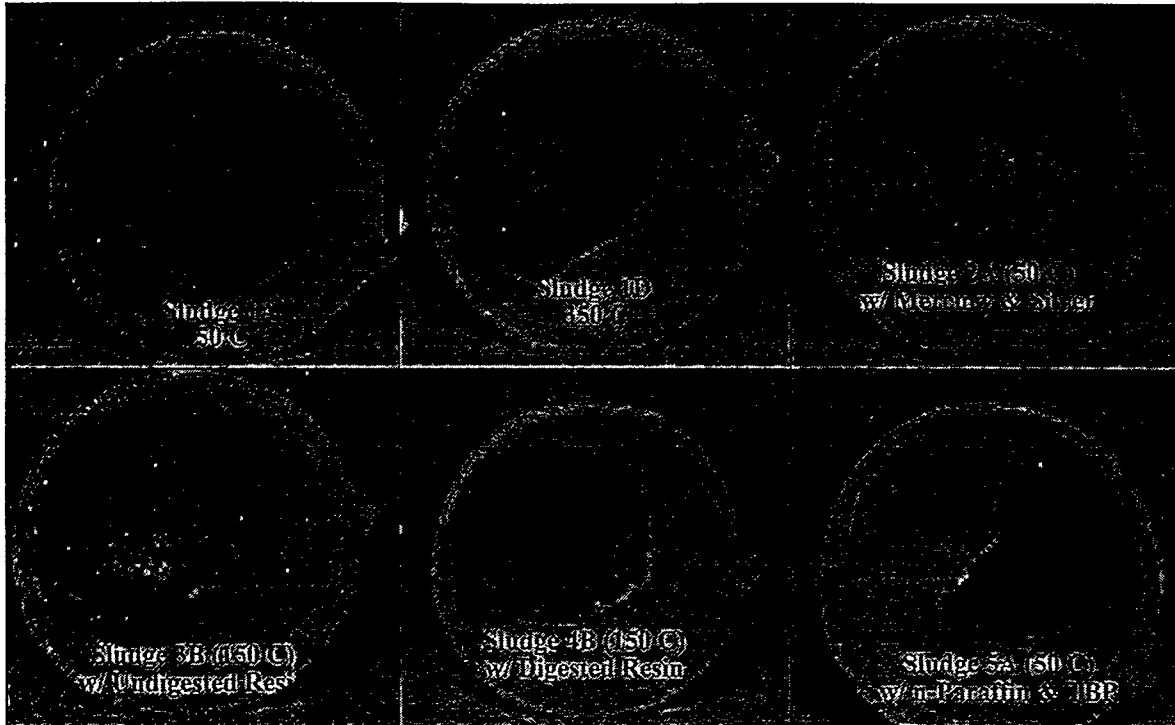


Figure 2. Photographs of various simulated-sludge samples.

After preparation, personnel examined the simulated sludge using standard impact tests.<sup>9,10</sup> Figure 3 and shows the equipment used for these tests. These tests resemble similar studies of Hanford High Level Waste material reported in the past from Los Alamos National Laboratory.<sup>11</sup> Personnel also examined the dried samples for energetic reactions at temperatures to 400 °C using DSC and TGA methods.

### Energetic Standards

Hobbs<sup>4</sup> evaluated known energetic compounds for use as standards in these experiments and recommended any of the following: silver nitride ( $\text{Ag}_3\text{N}$ ), mercury (II) fulminate ( $\text{Hg}(\text{ONC})_2$ ), mercury oxalate, and Millon's base ( $\text{Hg}_2\text{NOH}\cdot 2\text{H}_2\text{O}$ ) or a derivative. Table 3 gives the energies associated with the decomposition of several of these compounds. Given that the mercury concentration far exceeds than of silver in the waste, personnel synthesized two of these compounds for use as standards: mercury oxalate and mercury fulminate.<sup>12</sup> Oxalate preparation involved precipitation from a dilute nitric acid solution saturated in mercuric nitrate. The fulminate synthesis involved direct reaction of a strongly acidic mercuric nitrate solution with ethyl alcohol. Personnel verified the synthesis by performing x-ray diffraction and Raman spectroscopy on the solids.

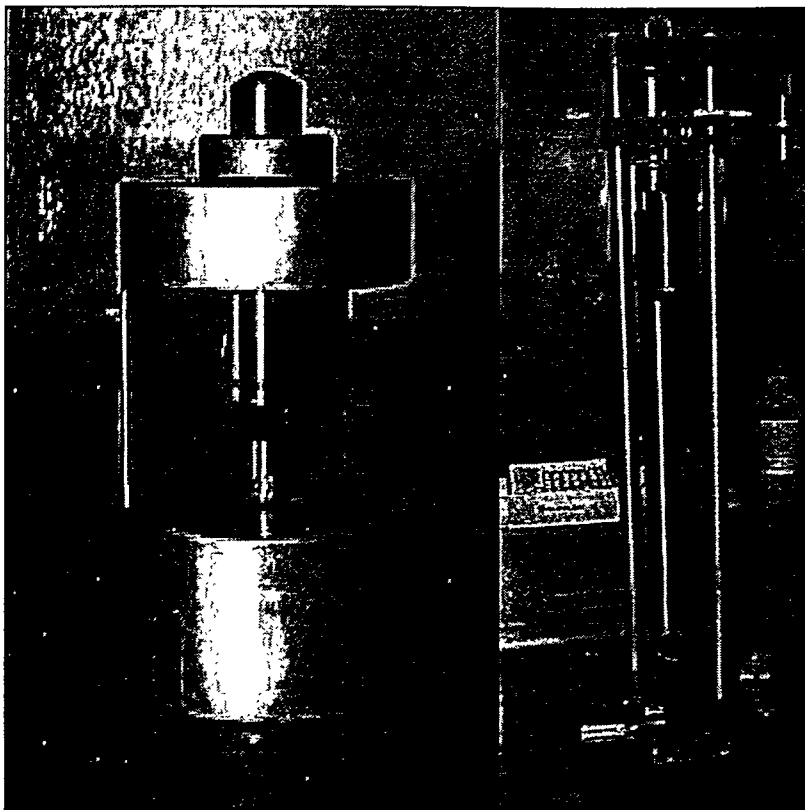


Figure 3. Detailed view of impact test chamber and impact (i.e., drop weight) test equipment.

Table 3. Exothermic properties of potential waste tank energetic compounds.

Mercury fulminate<sup>13</sup>

Heat of explosion	1486 J/g
Decomposition temperature	165 °C

Mercury oxalate<sup>14</sup>

Decomposition energy	377 J/g
Decomposition temperature	172 °C

Silver oxalate<sup>14</sup>

Decomposition energy	372 J/g
Decomposition temperature	136 °C

Silver nitride<sup>15</sup>

Decomposition energy	1860 J/g
Decomposition temperature	270 °C (in presence of impurities) 340 °C (pure)

Trinitrotoluene (TNT - for comparative purposes<sup>13</sup>)

Decomposition energy	4560 J/g
Decomposition temperature	300 °C

## RESULTS AND DISCUSSION

### Radioactive Samples: Characterization and DSC

Table 4 and Table 5 provide the currently available results of the chemical analyses. Sodium and aluminum concentrations provide the primary insight into process history for the samples. The sample from Tank 8F shows a high sodium and low aluminum concentration such as expected based on process history. However, the samples from Tanks 12H and 15H contain much less sodium and more aluminum by comparison, suggesting previous sample handling included caustic leaching and washing. Hence, the Tank 8F appears to more closely resemble current vessel contents.

Figure 4 provides the DSC and TGA results for the archived Tank 15H material. Figure 5 provides the DSC results for the archived Tank 8F and Tank 12H samples. The DSC spectra provide the specific energy released as the material heats to a defined temperature. (For the simulated sludge samples that follow, the line continues through the cooling of the sample.) The experiments start with the material at a low temperature – typically 40 °C for these experiments – and raise the temperature at a pre-selected rate, generally 10 °C/min in this work. Most testing held the samples in air although a few used an argon cover gas. A smooth horizontal line represents the lack of a reaction or heat flow from the material. A line deviating to negative energy flow represents either an endothermic reaction or a phase transition. For instance, near 100 °C most samples evaporate water requiring a heat input to remove the water. When solids undergo a phase transition – such as melting to changing crystal structure – an endothermic peak occurs. On cooling, such reversible phase transitions show an exothermic (i.e., positive energy flow) peak: see Figure 7 above 200 °C. An explosion shows a very strong exothermic peak, narrow in width. The strongest such reactions will often exceed the ability of the equipment to track the energy flow.

These samples showed no evidence of exothermic reactions. Several broader endothermic peaks occur on the calorimetry scans (i.e., centered near temperatures of 55, 130-150, 230, 240, 280, and beyond 300 °C). During sample preparation, personnel did not observe any sparks or evidence of shock-sensitive compounds (although the remote operations make such observations difficult except for the most energetic of reactions).

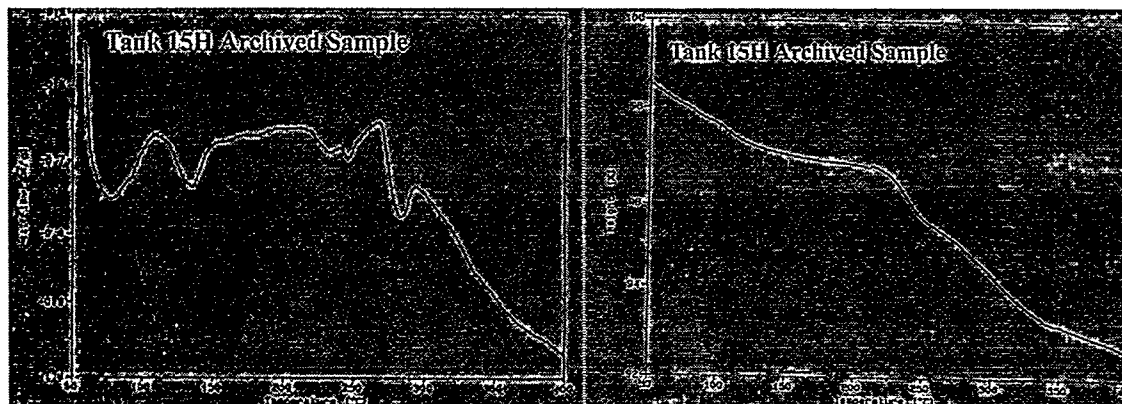


Figure 4. Calorimetry measurements -- DSC spectra, left, and TGA right -- of archived Tank 15H sample.

**Table 4. Results of the Analysis of the Total Dried Solids from Archived Sludge Samples from Tanks 8F, 12H, and 15H.**

Species		Archived Tank 8F Sludge <sup>a</sup>	Archived Tank 12H Sludge <sup>a</sup>	Archived Tank 15H Sludge <sup>a</sup>
Fe	wt %	19.3 (5)	4.12 (9)	4.03 (8)
Na	wt %	11.8 (7)	1.57 (7)	2.51 (7)
Al	wt %	3.01 (7)	33.1 (4)	30.1 (2)
U	wt %	11.5 (2)	0.044 (5)	0.013 (2)
Mn	wt %	3.75 (4)	1.95 (2)	2.43 (4)
Ca	wt %	1.52 (2)	1.12 (1)	0.23 (15)
Mg	wt %	0.32 (7)	0.18 (14)	0.15 (6)
Si	wt %	<1	<1	0.18 (2)
P	wt %	<0.3	<0.3	0.27 (15)
Ni	wt %	3.78 (6)	0.52 (9)	0.42 (5)
Cr	wt %	0.085 (13)	<0.07	0.015 (7)
Hg	wt %	0.46 (2)	0.92 (6)	3.32 (3)
Cu	wt %	0.080 (7)	<0.07	0.042 (14)
Ti	wt %	<0.03	<0.03	0.014 (18)
Pu	wt %	0.013 (3)	0.031 (7)	0.0019 (6)
Ag	wt %	*	*	Not detected
Pd	wt %	*	*	0.00047 (13)
Ru	wt %	*	*	0.059 (3)
Rh	wt %	*	*	0.014 (3)
K		<0.2	<0.2	<0.2
Sr <sup>90</sup>	μCi/g	1.70E+04 (6)	1.51E+04 (4)	1.47E+04 (3)
Cs <sup>137</sup>	μCi/g	101 (3)	88.2 (3)	85.6 (6)
Am <sup>241</sup>	μCi/g	85.3 (11)	32.9 (3)	6.51 (13)
Eu <sup>154</sup>	μCi/g	36.1 (7)	67.9 (4)	78.6 (12)
Co <sup>60</sup>	μCi/g	32.0 (7)	1.54 (5)	1.30 (4)
Eu <sup>155</sup>	μCi/g	30.6 (7)	10.2 (6)	9.34 (14)

<sup>a</sup>Value in parenthesis indicates percent relative standard deviation of three or more determinations on aliquots of the same sample. This deviation provides a measure of the analytical precision and does not account for the sampling uncertainty.

\*Analysis pending.

**Table 5. Uranium and Plutonium Isotopics of the Total Dried Solids from Archived Sludge Samples from Tanks 8F, 12H, and 15H.**



Species		Archived Tank 8F Sludge	Archived Tank 12H Sludge	Archived Tank 15H Sludge
U <sup>233</sup>	wt %	Not detected	1.5E-03 (7)	3.1E-04 (22)
U <sup>234</sup>	wt %	3.3E-04 (15)	1.0E-03 (6)	2.9E-04 (42)
U <sup>235</sup>	wt %	3.6E-02 (3)	5.3E-03 (1)	4.4E-03 (2)
U <sup>236</sup>	wt %	2.0E-03 (5)	1.7E-03 (4)	1.9E-03 (8)
U <sup>238</sup>	wt %	1.14E+01 (2)	3.5E-02 (6)	5.7E-03 (6)
U <sup>total</sup>	wt %	1.15E+01 (2)	4.4E-02 (5)	1.3E-02 (2)
Pu <sup>238</sup>	wt %	1.9E-04 (50)	5.0E-03 (10)	6.1E-04 (8)
Pu <sup>239</sup>	wt %	9.8E-03 (3)	2.0E-02 (4)	1.2E-03 (9)
Pu <sup>240</sup>	wt %	1.4E-03 (10)	4.2E-03 (5)	2.1E-04 (16)
Pu <sup>241</sup>	wt %	1.5E-03 (17)	9.0E-04 (69)	Not detected
Pu <sup>242</sup>	wt %	Not detected	3.5E-04 (19)	Not detected
Pu <sup>total</sup>	wt %	1.3E-02 (3)	3.1E-02 (7)	2.0E-03 (6)

Values in parenthesis indicate percent relative standard deviation of three or more determinations on aliquots of the same sample. This is a measure of the analytical precision and does not account for the sampling uncertainty.

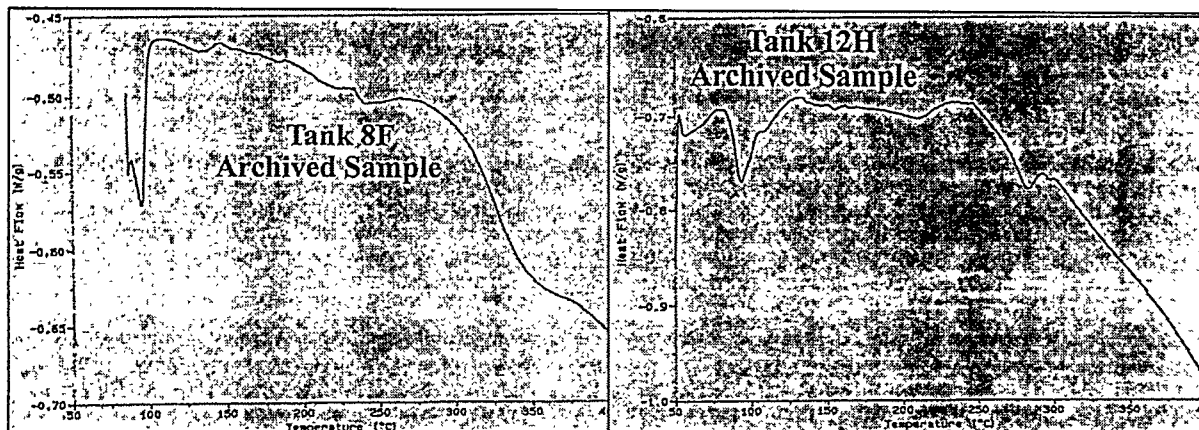


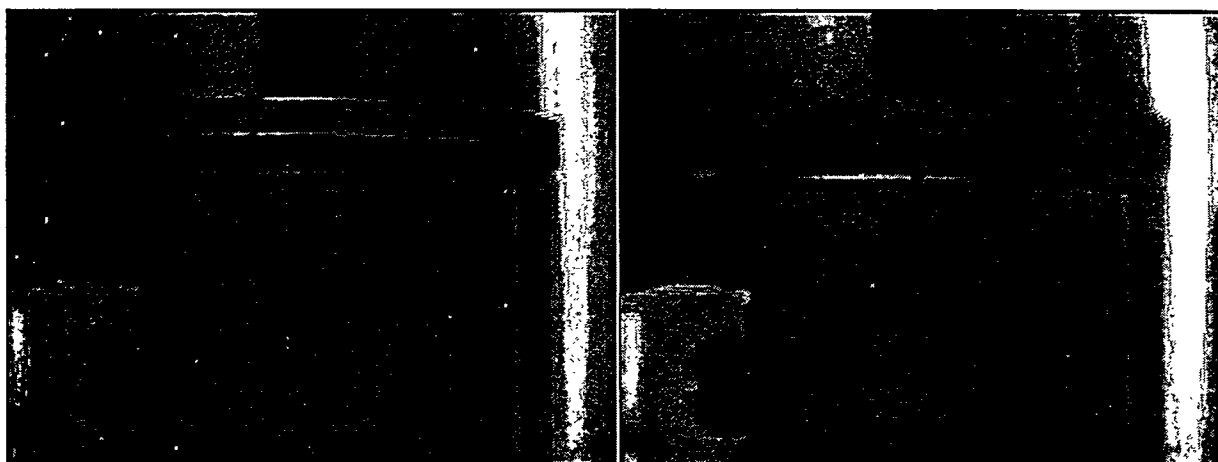
Figure 5. Calorimetry measurements on archived Tank 8F and Tank 12H samples.

### Simulated Sludge: Impact Testing

Impact testing of all sludge samples provided no indication of exothermic reaction or shock sensitive compounds. Such testing generally included multiple – typically five or six – attempts with the same materials. Testing also examined the response with the addition of either mercury

oxalate or mercury fulminate in the sludge materials. With oxalate concentrations as high as 15 wt % (nominal), the tests showed no signs of an exothermic reaction. Similarly, with mercury fulminate added at concentrations as high as 15 wt % (nominal), the experiments did not indicate a reaction upon impact.

Observation of a reaction using the impact test protocol proves subjective with relatively high detection limits. Researchers also tested the pure standards and attempted to define the minimum amount required to provide a positive indication of reaction. Figure 6 shows the results of such a test yielding a reaction. The smoke observed in the figure provided the only reliable means of detection. For mercury oxalate, testing required 3 mg to achieve a visible result. For mercury fulminate, personnel noted a reaction for masses as small as 2 mg. (When using small total masses of the compounds, researchers occasionally added a layer of an inert material, sodium nitrate, underneath the standard to improve contact with the falling piston.) These detection limits for pure materials imply a minimal detection limit for samples containing sludge mixed with the mercury compounds. For sludge containing mercury oxalate, the method could detect an explosion in a sludge mixture containing at least 15 wt % mercury oxalate. Similarly, the method could detect a reaction for a sludge mixture containing at least 10 wt % of mercury fulminate.



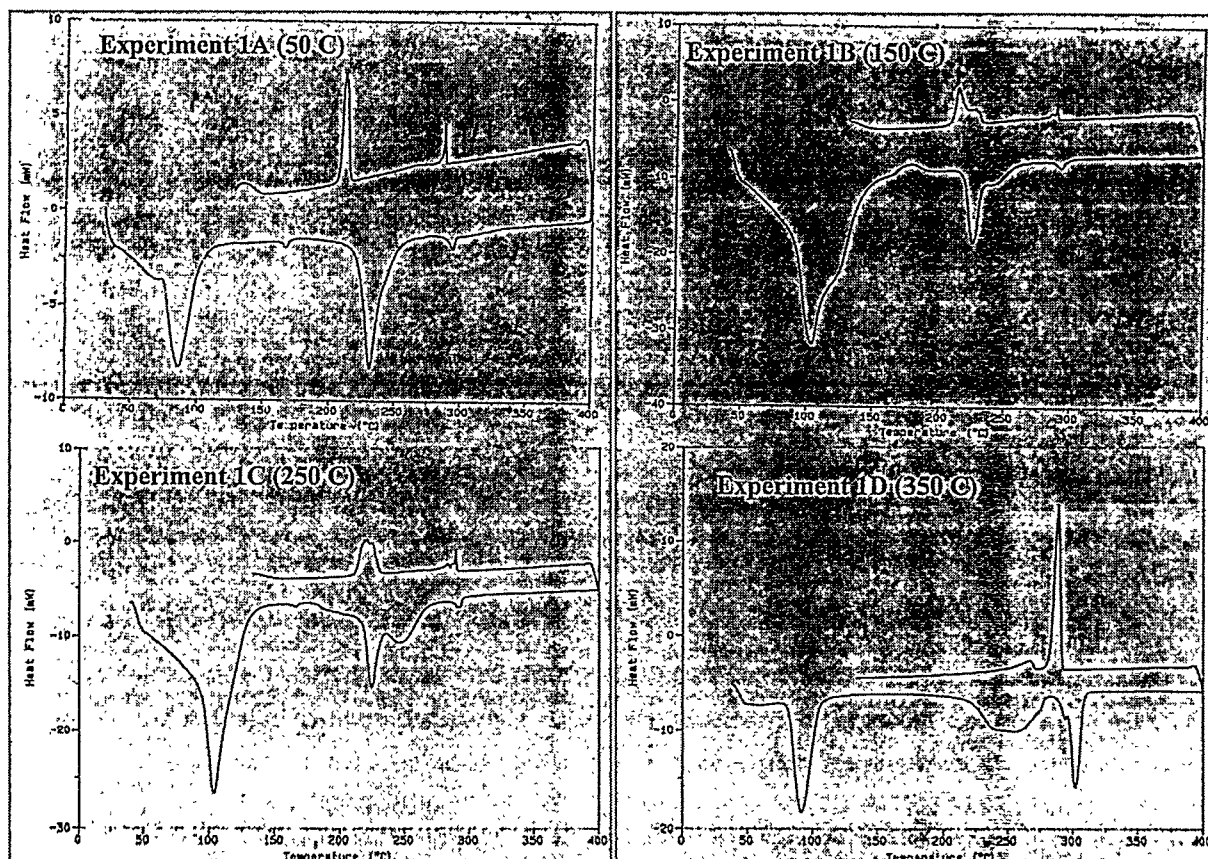
**Figure 6. Typical photos of impact test experiment. Left figure shows a test with no visible indication of reaction. Right figure depicts positive visible indication (i.e., smoke) of reaction.**

Researchers investigated various means to improve detection beyond that available by visible observation. In one case, personnel used a mercury specific detector (Jerome Model 411 Mercury Vapor Analyzer). This detector senses the presence of mercury vapor by means of a gold detection plate that amalgamates with the mercury. Detection limits measure as low as 0.001  $\mu\text{g/L}$  of air. Personnel attempted to direct the gases from the impact test unit into the detector by a simple funnel. The use of the equipment inside a forced flow hood limited the ability to provide reproducible results. For the standards, even at amounts as low as 3 mg mercury oxalate, the concentrations detected exceed the maximum range (i.e., 1.99  $\mu\text{g/L}$ ) of the instrument. For impact tests of sludge mixtures containing no standards but with 6 wt % mercury, the instrument did not detect mercury vapor above the background for the equipment when using no material (i.e., 0.030  $\mu\text{g/L}$ ). For sludge samples with added mercury oxalate or mercury fulminate, the method gave intermediate and non-reproducible results.

Personnel also performed visual microscopy on samples following testing. For the pure standards, the microscopy detected droplets of mercury formed during the decomposition reaction. For the sludge samples, personnel could not visually detect any such droplets. However, the detection limit for this microscopic method remains unknown.

### Simulated Sludge: DSC

Figure 7 (experimental Series 1) shows the influence of drying temperature on the energetic characteristics of the sludge samples that did not contain organic compounds, silver or mercury. As the drying temperature changed from 50 to 350 °C for the sludge, the thermal characteristics remained largely unchanged. Addition of mercury and silver in Series 2 (not shown), at conservatively high concentrations, in the sludge sample prior to drying did not alter this observation. None of these tests showed, during heating, any indication of an exothermic reaction by DSC at temperatures as high as 400 °C. Between 300 and 220 °C, exothermic peaks

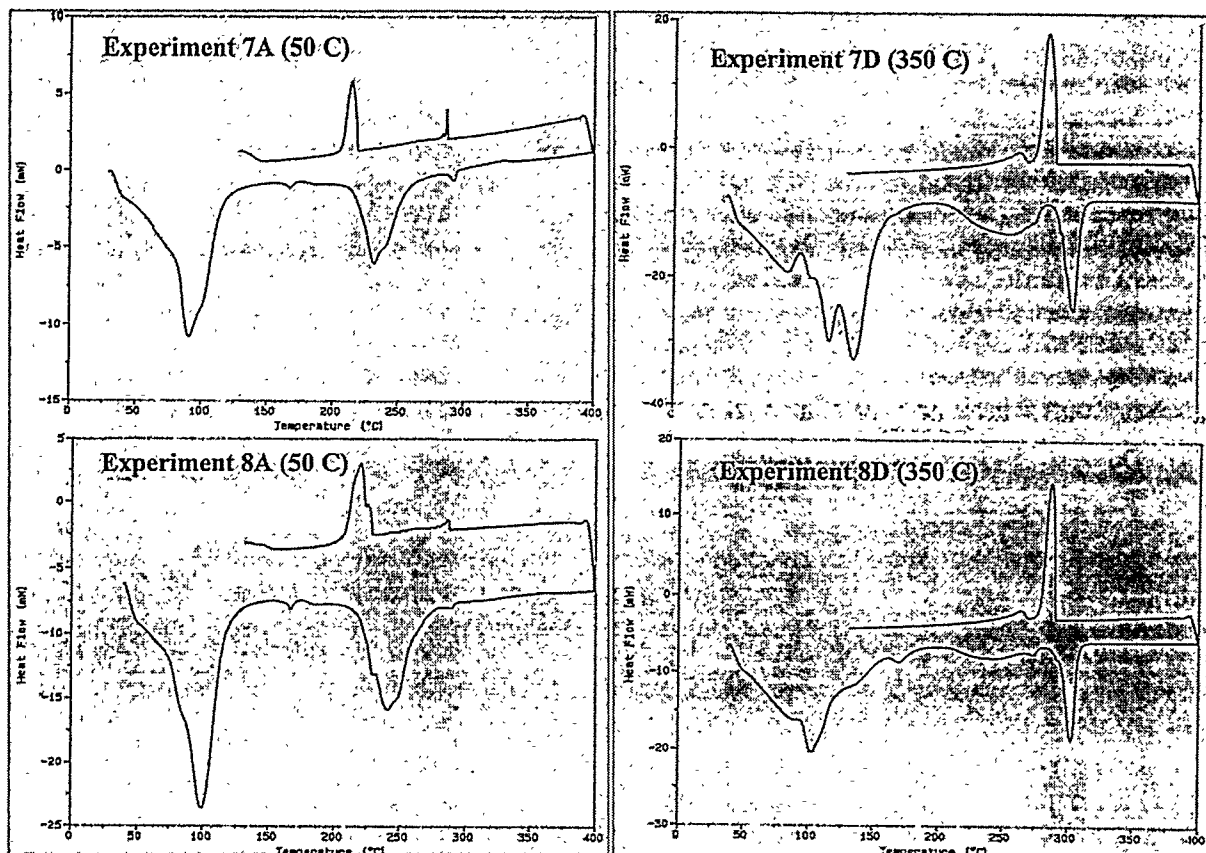


**Figure 7. Influence of temperature on calorimetry measurements for simulated sludge.**

occur during cooling at approximately similar temperatures for endothermic peaks during heating. These pairs likely represent changes in crystalline phases of alumina compounds.<sup>16</sup>

Figure 8 provides insight into the combined influence of organic compounds and drying temperature. Experimental Series 7 did not include mercury or silver in the sludge recipes. Thermal analyses of these samples again showed no exothermic reactions. The endothermic behavior at temperatures below 100 °C and near 250 °C broadened relative to observations in the

absence of the organic compounds. As the drying temperature of the sludge sample increased, additional endothermic peaks occur in the figures, presumably from partial thermal decomposition of the organic compounds during drying. Although not shown, experiments also examined the influence of each individual organic compound (Series 3 through Series 6). Again, tests with the individual organic compounds added did not result in any indication of exothermic behavior.



**Figure 8. Combined influence of organic compounds and temperature on calorimetry measurements for simulated sludge.**

In contrast, Series 8 samples – see Figure 8--contained mercury and silver at conservatively high concentrations. These measurements did not differ significantly from those of Series 7. Hence, the presence of the organic compounds studied does not appear to increase the potential for formation of energetic compounds during drying.

Hence, the sludge samples synthesized in this study showed no evidence of formation of energetic compounds even in the presence of high quantities of several organic compounds. Despite the addition of conservatively large amounts of mercury or silver, no samples suggested an explosive or exothermic behavior.

#### **Simulated Sludge Combined with Energetic Standards: DSC**

One can not exclude the possibility that negative results in tests with simulated sludge simply reflect non-representative synthesis of the samples. Hence, the program also investigated

behavior of the simulated sludge after adding either mercury oxalate or mercury fulminate. This approach provided a means of characterizing the potential behavior of sludge and shock sensitive compound mixtures. These studies attempted to determine what concentrations of energetic compounds exhibited exothermic behavior, whether selected energetic compounds remained chemically stable in the sludge matrix, and if the presence of organic species in the mixtures propagated or increased the exothermic behavior of the mixtures.

Both mercury oxalate and mercury fulminate exist as shock sensitive compounds with sufficient stability to use as standards for such testing. Figure 9 and Figure 10 shows that the synthesized standards contained negligible impurities. Pure mercury oxalate and mercury fulminate, as prepared, decompose exothermically in air (145 J/g and 182 J/g, respectively) at temperatures of 225 and 167 °C, respectively (see Figure 11). Testing added each standard material to simulated sludge samples (see Appendix 1). Experiments added mercury oxalate and mercury fulminate into oven dried (50 °C) simulated sludge (Test Series 11 and 12, respectively), wet simulated sludge slurry (Test Series 13 and 14, respectively), and oven dried (50 °C) simulated sludge that contained a mixture of organic compounds (Test Series 15 and 16, respectively). Experiments used two or three concentrations of the specific energetic standard designated as either U (i.e., ultra low: 1 – 2.5 wt %), L (i.e., low: 4.9 – 9.3 wt %), or H (i.e., high: 13.6 – 21.5 wt %) to describe the concentration of the added standard. For example, Experiment 12H refers to a pre-dried simulated sludge containing a mercury fulminate concentration of 21.5 wt %.

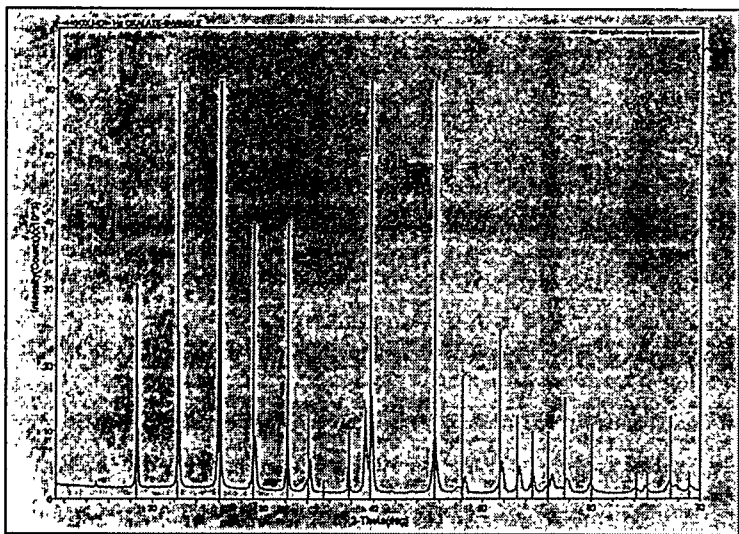


Figure 9. Diffraction patterns for pure mercury oxalate.

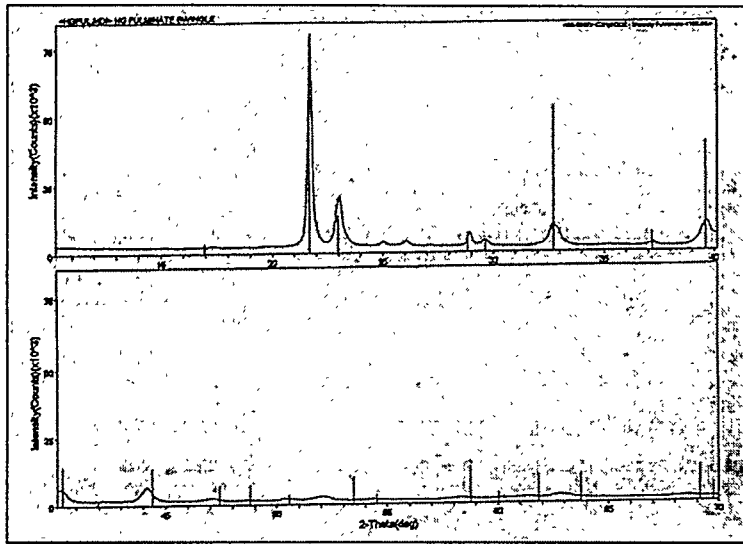


Figure 10. Diffraction patterns for pure mercury fulminate.

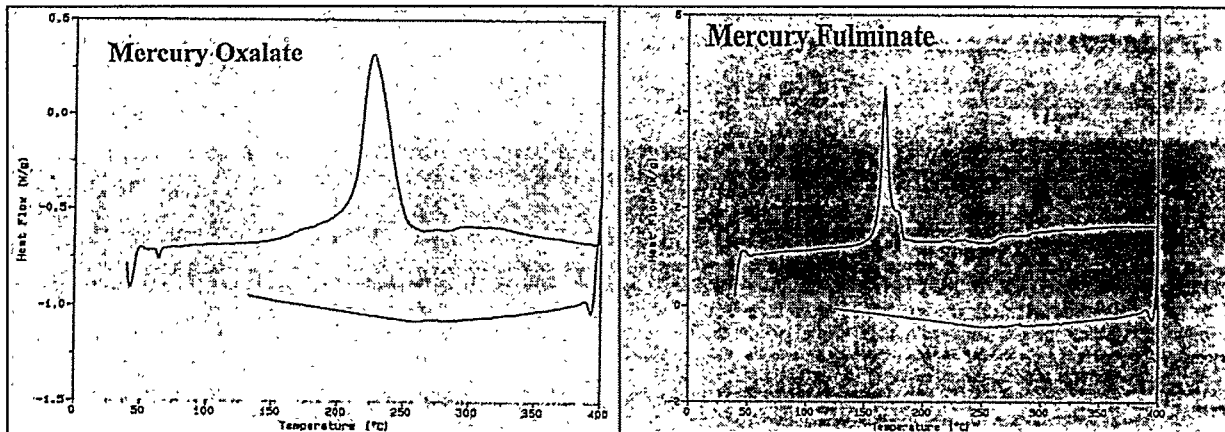


Figure 11. Calorimetry measurements for energetic standards.

Personnel subjected the prepared mixtures to both impact testing and DSC analysis. Test Series 11, 13, and 15 contained mercury oxalate. Figure 12 shows the DSC results for Experiments 11L and 11H. Recall pure mercury oxalate decomposes thermally at 225 °C. The DSC spectra for Experiments 11L and 11H, when compared with the DSC for the baseline simulated sludge as shown in Figure 7 (Experiment 1A), indicate that an exothermic reaction occurs at ~225 °C. The DSC spectra show that this exothermic reaction occurs in the midst of an endothermic reaction exhibited by the plain sludge. In other words, even though the energetic standard decomposes and gives off heat, the surrounding sludge absorbs the heat in a phase change reaction (most probably an aluminum oxide transition<sup>16</sup>). The net energy behavior remains endothermic at the concentrations tested.

Experiments 13L and 13H, while similar in composition, differ from Experiments 11L and 11H since they use wet alkaline sludge during preparation and not pre-dried sludge. Exposure of mercury oxalate to alkaline solutions reportedly destroys the energetic compound. Examination of Figure 13 shows that mercury oxalate still present in the sludge mixtures after 1 day of contact (i.e., the wet slurries containing the mercury oxalate dried overnight). Numerical analysis of the

DSC spectra provides further insight. The energies detected for decomposition of either mercury oxalate or mercury fulminate correlated linearly with the mass of added material. The calculated energies for these experiments (Table 6) indicate that the degree of exothermic behavior at 225 °C nearly matches that predicted for the amount of added mercury oxalate.

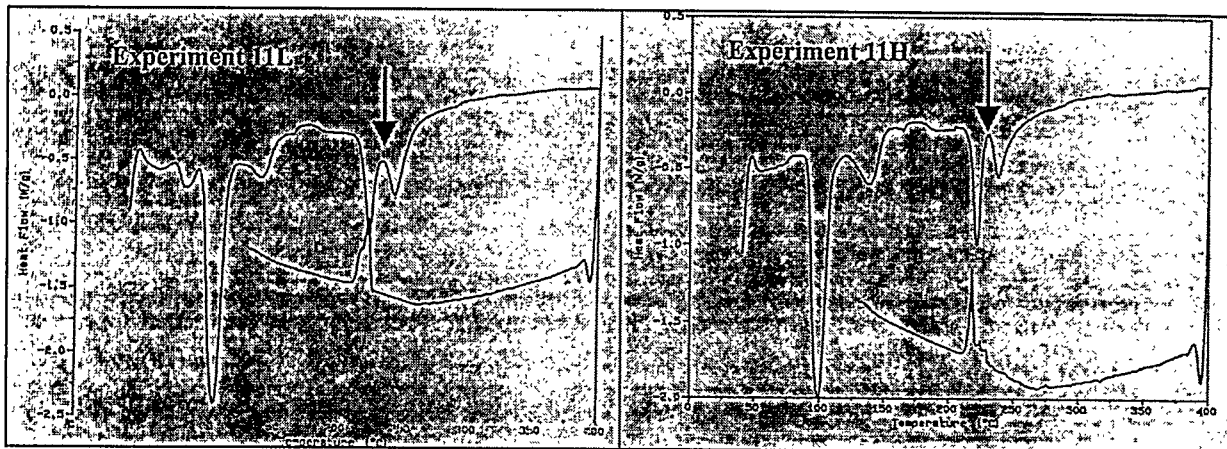


Figure 12. Calorimetry measurements of dry mercury oxalate added to dry sludge. (Arrows indicate location of exothermic peak for mercury oxalate.)

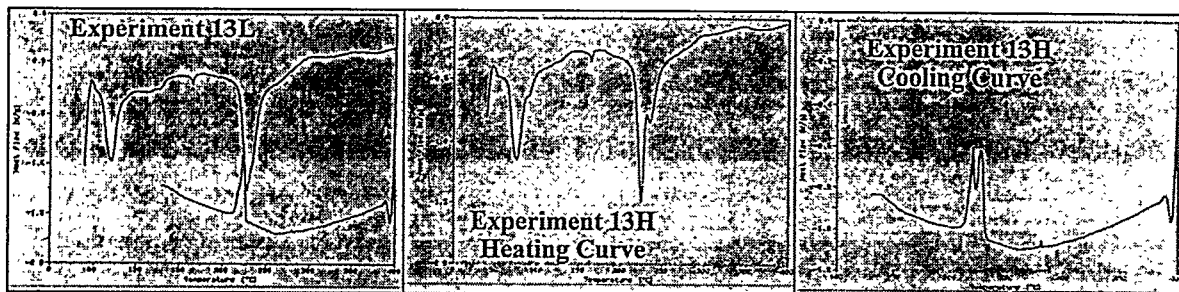


Figure 13. Calorimetry measurements of dry mercury oxalate added to wet sludge.

Sludge preparation for Experiments 15U, 15L, and 15H matched that of Experiments 11L and 11H except the simulated sludge matrix contained organic compounds (total of 3 wt %). Differential Scanning calorimetry results (shown in Figure 14) for 15L and 15H appear very similar to that observed in Experiments 11L and 11H (see Figure 12). Therefore, the presence of organic compounds in the sludge matrix does not increase the exothermic behavior of the material.



Table 6. Weight percent of explosives in simulated sludge mixtures.

Experiment #	Mixture	wt % (added)	wt % (DSC analysis)
11L	Dry sludge + oxalate	5.99	6.2
11H	Dry sludge + oxalate	14.9	20
12U	Dry sludge + fulminate	2.20	3.9
12L	Dry sludge + fulminate	9.30	9.53
12H	Dry sludge + fulminate	21.5	15.8
13L	Wet sludge + oxalate	4.90	2.68
13H	Wet sludge + oxalate	13.6	14.9
14U	Wet sludge + fulminate	1.68	0
14L	Wet sludge + fulminate	6.81	6.12
14H	Wet sludge + fulminate	17.5	17.0
15L	Dry + organic + oxalate	5.98	5.9
15H	Dry + organic + oxalate	22.6	19.8
16U	Dry + organic + fulminate	2.32	0
16L	Dry + organic + fulminate	8.75	6.4
16H	Dry + organic + fulminate	21.1	26.4

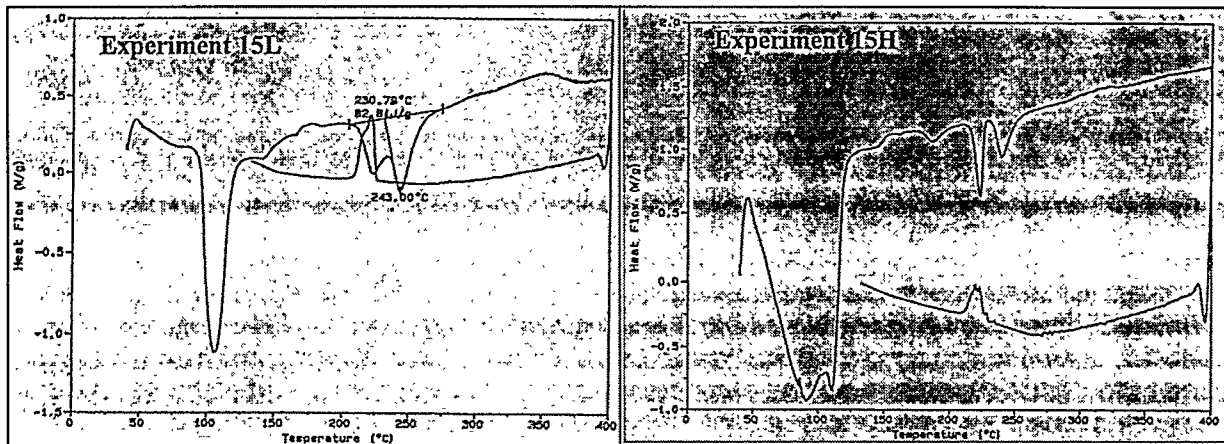


Figure 14. Calorimetry measurements of dry mercury oxalate added to dry sludge containing organic compounds.

Testing also examined the behavior of sludge with mercury fulminate added. Experiments 12U, 12L, and 12H examine the effects of mercury fulminate in pre-dried sludge (see Figure 15). Recall, mercury fulminate decomposes exothermically at 168 °C. An exothermic reaction occurred in all three experiments at ~168 °C. Furthermore, the energy released by the reaction increases with increasing concentration. However the level of energy released measured less than that needed to remove the water at low (i.e., <100 °C) temperature.



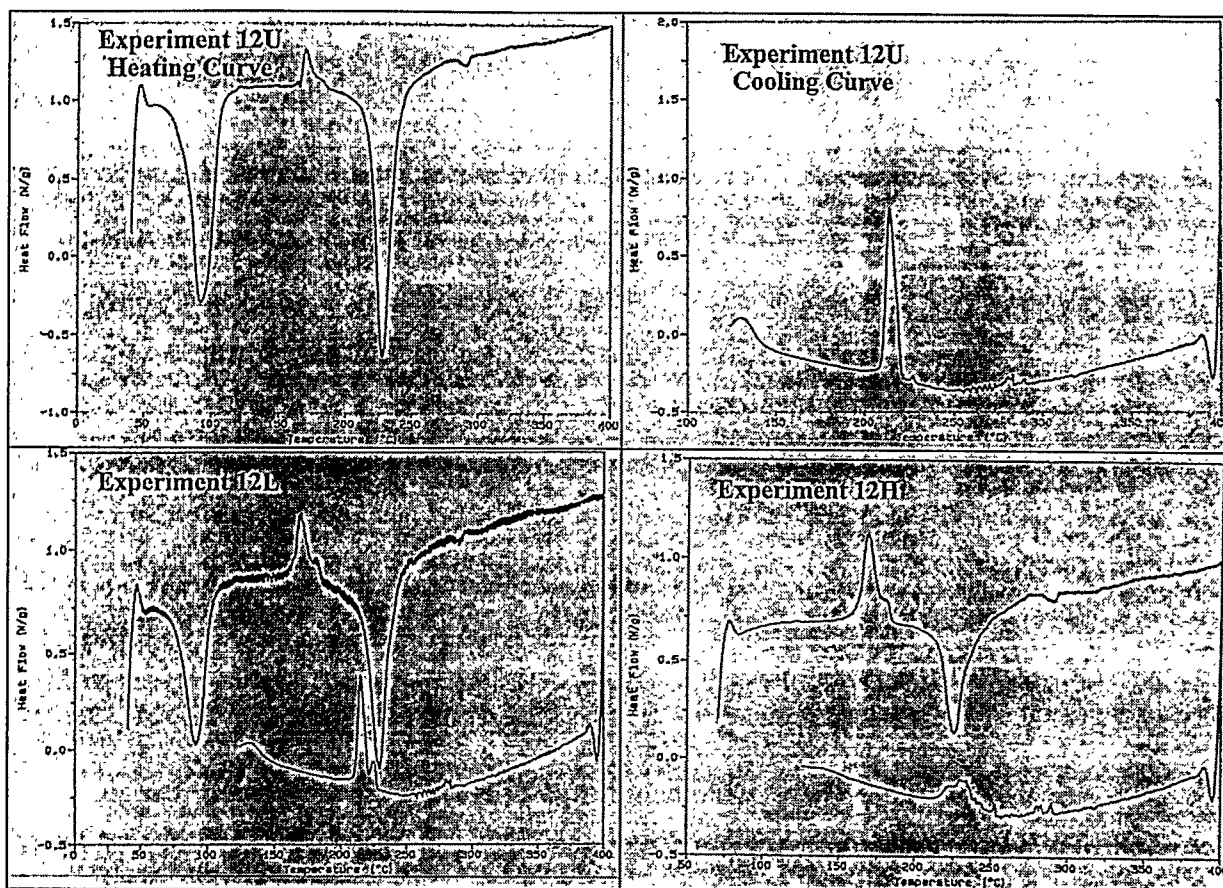


Figure 15. Calorimetry measurements of dry mercury fulminate added to dry sludge.

Experiments 14U, 14L, and 14H examine the influence of preparation using wet alkaline slurry. Again, literature suggests reaction of the fulminate with hydroxide will likely destroy the energetic material. Figure 16 exhibits the DSC results for the three experiments. Comparison of Figure 15 and Figure 16 indicates that the exothermic behavior remains essentially unchanged (given the slight difference in concentrations between the experimental sets). Numerical analysis of the data (Table 6) further confirms this visual observation. Therefore, short-term exposure (1 day) of mercury fulminate to slightly caustic solutions (0.22 M hydroxide) does not destroy the energetic material.

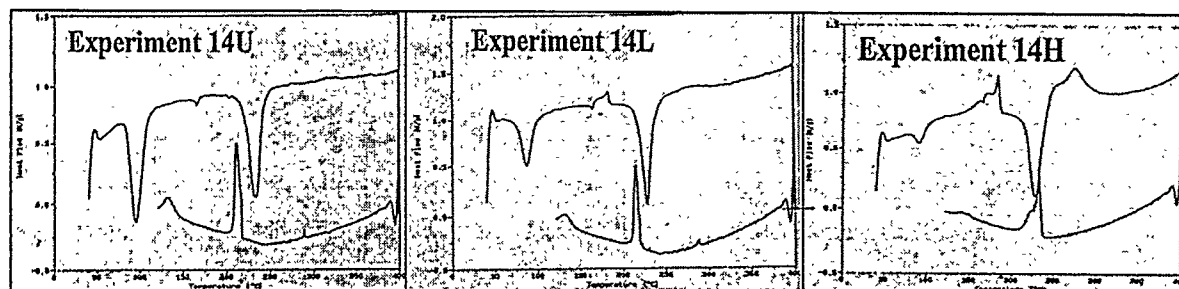


Figure 16. Calorimetry measurements of dry mercury fulminate added to wet sludge.

Lastly, Experiments 16U, 16L, and 16H investigate the influence of organic compounds on the energetic behavior of the mixtures (see Figure 17). Comparison of Figure 17 with Figure 15 indicates the presence of organic compounds (at 3 wt %) did not influence the exothermic behavior of the energetic material.

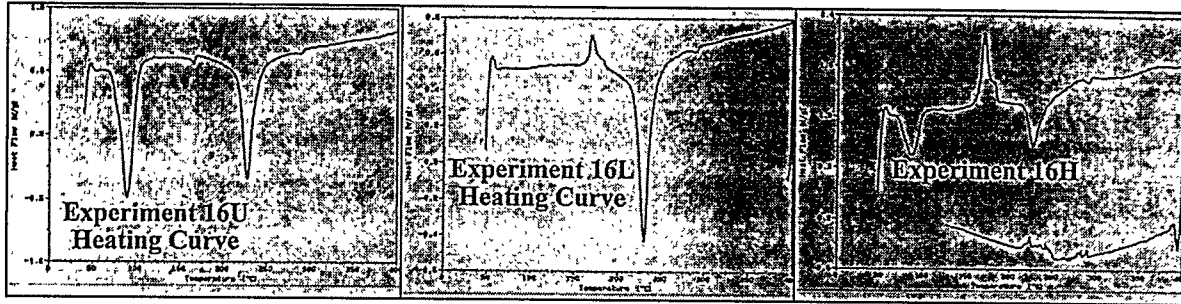


Figure 17. Calorimetry measurements of dry mercury fulminate added to dry sludge containing organic compounds.

#### Synergetic Interactions with Energetic Standards: DSC

The work described in the previous sections does not, however, fully investigate potential synergistic influences of energetic compounds with sludge and other components. For instance, one concern involves the potential for a limited amount of an energetic material to propagate a reaction into the larger amounts of organic compounds present. Alternatively, a small amount of a silver compound might cause the reaction of a mercury compound. Researchers conducted a few experiments to provide insight into these two scenarios.

Figure 18 examines the direct interaction of a mixture of mercury fulminate with a mixture of organic compounds (i.e., 30 vol % tri-n-butyl phosphate in paraffin, vendor supplied Ionac 604 resin, and permanganate digested Ionac 604 resin). The DSC shows evidence of direct combustion of the organic species in air as evidenced by the exothermic peaks centered near temperatures of 140, 250, 310, and 375 °C). The limited masses of organic compounds added in the simulated sludge – at concentrations well exceeding that expected in the tank farm waste – make observation of this behavior difficult in the presence of the sludge. Note the lack of a strong energetic peak near 167 °C (e.g., see Figure 11) typical for fulminate. The visual observation and numerical analysis of the spectra indicate the organic compounds suppress the reaction of the fulminate.

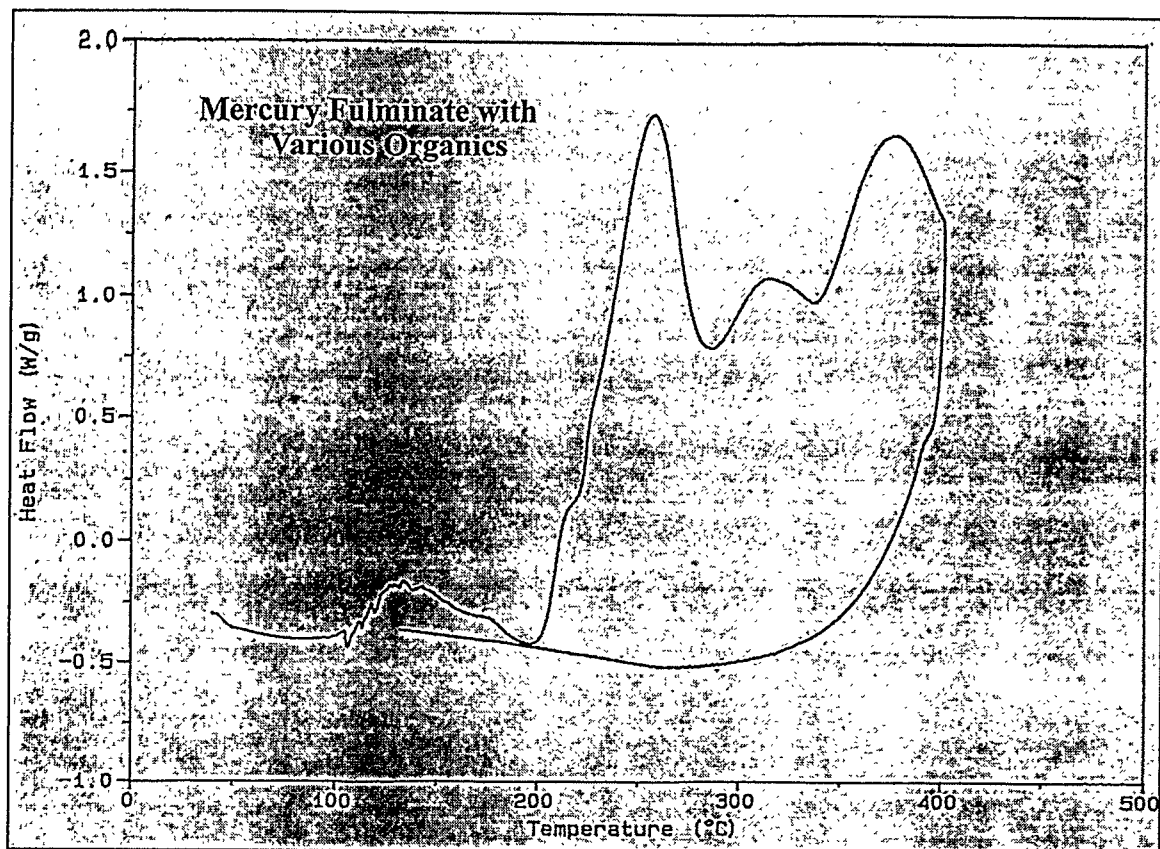


Figure 18. Calorimetry measurements for mixture of mercury fulminate with various organic compounds.

Figure 19 provides insight into the behavior that results for a nearly equal weight mixture of mercury oxalate and mercury fulminate. The exothermic reaction that occurs when the fulminate decomposes that also consumes the oxalate. Hence, mixtures of energetic compounds effectively lower the threshold amount needed to form a highly reactive mixture. The limited duration of this program did not allow investigation of whether coexistence of energetic compounds in sludge could result in significant exothermic behavior at relatively low concentrations. The study did not examine the behavior in combination with the simulated sludge. Presumably, sludge would dampen the potential for propagation of the reaction acting to disperse the energy from the initial exothermic reaction. The heat capacity, phase transition, and water content of the sludge would help dampen any interaction. However, testing did not verify this speculated behavior.

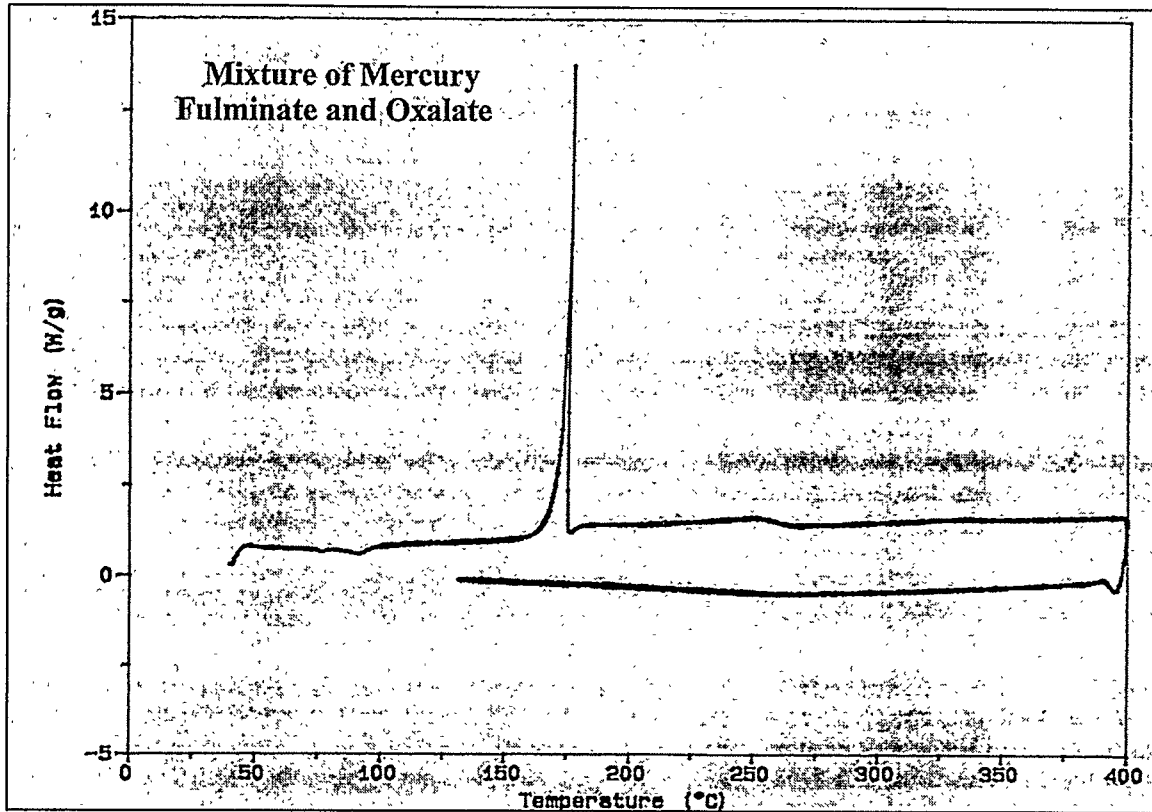


Figure 19. Calorimetry measurement for a mixture of mercury oxalate and mercury fulminate.

## CONCLUSIONS

While the experimental data does not preclude the existence of energetic compound in dry sludge, the information does suggest that such compounds will not likely form in the alkaline conditions. If formed, literature and expert knowledge suggest compounds like mercury oxalate and mercury fulminate will exhibit limited lifetimes due to reaction in the caustic environment. (These tests used a mild concentration of caustic – i.e., 0.22 M free hydroxide – and a short contact duration. At those limited conditions, the data show no evidence of decomposition.) Even with high concentrations of these compounds in sludge samples, the energy released during the sludge matrix dissipated the energy released during the reaction, so that the ASTM standard test did not provide positive indications of a reaction.

The composite data from these studies suggest that sampling of Tanks 12H and 15H with the impact device as currently designed<sup>17</sup> poses a low level of risk.

## Future Program Elements

The results of the current work suggest several areas that merit further study.

- Future work should examine the expected lifetime of selected energetic compounds in caustic media. Testing should likely include at least mercury oxalate, mercury fulminate, and silver nitride.
- Conduct additional calorimetry studies examining the behavior of simulated sludge with mixtures of energetic compounds. In particular, investigate the influence of the combined presence of silver nitride and mercury oxalate.
- Conduct vendor testing at larger scale to confirm these results and to provide information relative to propagation of any reaction into the organic compounds present.

## QUALITY ASSURANCE

Laboratory notebook WSRC-NB-96-667 (M. S. Hay, page 41) contains data from the characterization of samples from Tanks 8F, 12H, and 15H. Notebook WSRC-NB-98-00161 (K. L. Prettel, page 5) contains the recent DSC data as well as information on the synthesis of the simulated sludge. Notebook WSRC-NB-98-0238 (R. F. Swingle, pages 5-73) contains data from the impact testing. Notebook WRC-NB-97-0064 (W. R. Wilmarth, pages 26-28) contains the information on the synthesis of the mercury oxalate and mercury standards as well as the training records for personnel.

## ACKNOWLEDGEMENTS

Kevin Kalbaugh repaired the equipment and performed the calorimetry measurements on the Tank 15H sample. Bill Walker provided the digested resin added to the simulated sludge samples.

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- <sup>15</sup> L. Bretherick, HANDBOOK OF REACTIVE CHEMICAL HAZARDS, 3rd. Ed., Butterworths, 1985, London.
- <sup>16</sup> "14. Oxides and Hydroxides" (pages 1032-1034), "14.1 Aluminum Trihydroxide, Al(OH)<sub>3</sub>" (pages (1034-1035), and "14.2 Aluminum Oxide Hydroxide, AlO(OH)" (page 1035), in COMPREHENSIVE INORGANIC CHEMISTRY, Volume 1, First edition, J. C. Bailar et al., editors, Pergamon Press, New York, 1973.
- <sup>17</sup> T. A. Nance, Design Drawing: "Tank 12 Dry Sludge Sampling Device," R-R1-F-0051, Rev. 0, November 9, 1998.

**APPENDIX 1**

The following tables provide more detailed information regarding the experimental conditions and composition of materials.

**Table 7. Simulated sludge experimental design.**

Additive	Simulant ID							
	1	2	3	4	5	6	7	8
Mercury	-1	1	-1	-1	-1	-1	-1	1
Silver	-1	1	-1	-1	-1	-1	-1	1
Ionac 604 (undigested)	-1	-1	1	-1	-1	-1	0	0
Ionac 604 (digested)	-1	-1	-1	1	-1	-1	0	0
tri-n-butyl phosphate/ n-paraffin (30/70 vol %)	-1	-1	-1	-1	1	-1	0	0
Sodium oxalate	-1	-1	-1	-1	-1	1	0	0
<b>Temperature*</b>	A-D	A-D	A-D	A-D	A-D	A-D	A-D	A-D

\* A = 50 °C, B = 150 °C, C = 250 °C, D = 350 °C

**Table 8. Simulated sludge experimental design concentrations.**

Additive	Concentration (wt %)		
	-1	0	1
Mercury	0	NA	6
silver nitrate	0	NA <sup>a</sup>	0.0015
Ionac 604 (undigested)	0	0.75	3
Ionac 604 (digested)	0	0.75	3
tri-n-butyl phosphate/ n-paraffin (30/70 vol %)	0	0.75	3
Sodium oxalate	0	0.75	3

<sup>a</sup>NA indicates level not tested.

**Table 9. Experimental design of mercury oxalate and fulminate bearing simulated sludge compositions.**

Additive	Simulant ID							
	9	10	11L-H <sup>a</sup>	12U-H <sup>a</sup>	13L-H <sup>a</sup>	14U-H <sup>a</sup>	15U-H <sup>a</sup>	16U-H <sup>a</sup>
Mercury oxalate	1	-1	1	-1	1	-1	1	-1
Mercury fulminate	-1	1	-1	1	-1	1	-1	1
Inoac A641 (undigested)	-1	-1	-1	-1	-1	-1	0	0
Inoac A641 (digested)	-1	-1	-1	-1	-1	-1	0	0
Tri-n-butylphosphate	-1	-1	-1	-1	-1	-1	0	0
n-parrafin	-1	-1	-1	-1	-1	-1	0	0
Sodium oxalate	-1	-1	-1	-1	-1	-1	0	0
Temperature/Condition	Pure <sup>b</sup>	Pure <sup>b</sup>	50 °C <sup>c</sup>	50 °C <sup>c</sup>	50 °C <sup>d</sup>	50 °C <sup>d</sup>	50 °C <sup>e</sup>	50 °C <sup>e</sup>

<sup>a</sup>U, L, and H simulant ID labels refer to the approximate level of mercury oxalate or fulminate added to the simulant where U = ultralow, L = low, and H = high.

<sup>b</sup>Mercury oxalate and fulminate tested as pure components (i.e., not blended with sludge).

<sup>c</sup>Mercury oxalate and fulminate mixed with simulated sludge previously dried at 50 °C (dried sludge simulant 1A).

<sup>d</sup>Mercury oxalate and fulminate mixed with simulated sludge slurry and then dried at 50 °C (wet sludge simulant 1A).

<sup>e</sup>Mercury oxalate and fulminate mixed with organic-bearing simulated sludge previously dried at 50 °C (dried sludge simulant 7A).

**Table 10. Experimental concentrations of mercury oxalate and fulminate in simulated sludge compositions.**

Simulant ID	Concentration (wt %)	
	Mercury Oxalate	Mercury Fulminate
9	pure	NA <sup>a</sup>
10	NA	Pure
11L	5.99	NA
11H	14.9	NA
12U	NA	2.20
12L	NA	9.30
12H	NA	21.5
13L	4.90	NA
13H	13.6	NA
14U	NA	1.68
14L	NA	6.81
14H	NA	17.5
15U	1.58	NA
15L	5.98	NA
15H	14.9 (2 <sup>nd</sup> batch 22.6)	NA
16U	NA	2.32
16L	NA	8.75
16H	NA	21.1



Table 11. Listing of impact tests.

<u>Sludge Sample Tested</u>	<u>Number of Impact Tests</u>	<u>Results</u>
9 (Pure Mercury Oxalate)	23	Minimum Level of Detection by Smoke – 3 mg total mercury oxalate
1-A	6	No reaction detected
7-D	5	No reaction detected
8-A	6	No reaction detected
11-L	5	No reaction detected
11-H	5 (4*)	No reaction detected
13-L	5	No reaction detected
13-H	5	No reaction detected
15-L	5	No reaction detected
15-H	5 (3*)	No reaction detected
10 (Pure Mercury Fulminate)	16	Minimum Level of Detection by Smoke – 2 mg total mercury fulminate; sparks observed in 3 tests
11-U	2	No reaction detected
2-A	2	No reaction detected
12-U	6	No reaction detected
12-L	6 (4*)	No reaction detected
12-H	6 (6*)	No reaction detected
16-U	6	No reaction detected
16-L	6 (3*)	No reaction detected
16-H	6 (6*)	No reaction detected
14-U	7	No reaction detected
14-L	6	No reaction detected
14-H	6 (6*)	No reaction detected

\* Numbers in parentheses indicate number of impact tests where mercury oxalate or fulminate amounts met or exceeded minimum observable detection limits assuming sludge contained at least 15 wt % of either fulminate or oxalate.

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