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LEACHING PROPERTIES OF SOLIDIFIED TRU WASTE FORMS

MASTER

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DEPARTMENT OF NUCLEAR ENERGY

BROOKHAVEN NATIONAL LABORATORY
ASSOCIATED UNIVERSITIES, INC.

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1. PROGRAM DESCRIPTION

Approximately 12×10^6 ft^3 of defense TRU wastes are in storage or burial at various DOE installations, principally at the Idaho National Engineering Laboratory (INEL), Hanford, and the Savannah River Plant. (Continued waste generation rates of 2×10^5 ft^3/yr are estimated.) These wastes will ultimately be moved and placed into a geologic repository for disposal. While there is some question as to whether existing TRU wastes will be disposed in their current form or first undergo some additional volume reduction process, it is evident that volume reduction will be required for future TRU wastes. Since the bulk of these wastes consists of contaminated combustible solids including such items as protective clothing, rubber boots and gloves, paper, rags, filter aids, wood, and ion exchange resins, volume reduction assumes considerable importance relative to the reduction of waste shipment volumes, conservation of repository capacity, lowering of disposal costs, and meeting repository criteria directed at restricting the amount of combustible material in disposal. There are a number of volume reduction options available for combustible TRU wastes, the most viable option currently being some form of incineration. While incineration does result in a large volume reduction for combustible solid wastes, the ash that results is highly dispersible. As such, the immobilization of incinerator ash by solidification producing a solid monolithic waste form is desirable.

Safety analysis of solidified TRU contaminated incinerator ash waste forms requires an estimate of the ability of these waste forms to retain activity in the disposal environment. The primary mechanism for the potential release of this activity is assumed to be by leaching. The experimental program conducted at Brookhaven National Laboratory will determine the leaching properties of TRU contaminated incinerator ash waste forms using hydraulic cement, urea-formaldehyde, bitumen and vinyl ester-styrene as solidification agents. The data obtained will indicate the relative activity retention abilities of these waste forms and provide the basis for long-term release projections from full-scale waste forms. The leach rates of the following radionuclides: ^{239}Pu, ^{235}U, ^{237}Np, ^{241}Am, ^{137}Cs, and ^{90}Sr are of interest. Waste form leach rates for these radionuclides will be determined where practical although experimental expediency may dictate the use of other isotopes of these elements and/or availability considerations preclude the use of one or more of these elements.

Three types of leaching tests will be conducted to provide sufficient data for long-term predictions under a variety of conditions. Initially, leaching data will be obtained by use of an IAEA/ISO leach procedure.(1,2) In this method, the entire external surface area of the waste form is exposed to a leachant solution at 25°C which is sampled and completely replaced at twenty-four hour intervals for the first five days, after which the leachant is replaced once per week for the following eight weeks and then once per month. Static leach tests will also be performed at both 25°C and 70°C. In these tests, the leachant solution is not changed and is only sampled once. For a given waste form and leachant type, six leach specimens will be prepared (for each temperature) to allow leachant sampling at 2 days, 4 days, 8 days, 16 days, 106 days, and 341 days. The final leaching test procedure will be a flow rate study where activity release will be determined for three leachant flow rates of from 10^{-3} g/min to 1 g/min, which covers the range of observed aquifer flow rates. All leach test procedures will include some replication.

For each of the leaching procedures, five generic groundwaters will be employed as follows:

(1) Salt brine whose formulation is based on an analysis where a salt rock core taken at the 2725 foot level of AEC #3 at the WIPP facility was dissolved in deionized water. This salt brine composition is shown in Table 1.

(2) Simplified sodium dominated groundwater, 30 meq/liter NaCl.

(3) Simplified calcium groundwater, 30 meq/liter $CaCl_2$.

(4) Simplified bicarbonate groundwater, 30 meq/liter $NaHCO_3$.

(5) Deionized water.

2. WASTE FORM FORMULATIONS

Solidification agents should incorporate relatively as much incinerator ash as possible in the final waste form to maximize packaging efficiency and reduce associated disposal costs. However, the waste form ash content should not be so high as to either impose substantial difficulties in the solidification process or to compromise waste form properties. The amount of ash that can be reasonably incorporated in the waste form is dependent on the composition of the ash and its interaction with the solidification agent. Simulated incinerator ash compositions have been formulated for various waste producing sites. These compositions vary considerably and have been shown to result in different compositional phase diagrams for acceptable solidification with portland type I cement.(3) In addition, simulated waste compositions while similar chemically to actual wastes, may not adequately simulate the physical form or chemical distribution of actual incinerator ash. As such, this program utilizes an actual incinerator ash resulting from the incineration of a mixed combustible waste representative of plutonium handling and processing facilities.

Waste forms are formulated using incinerator ash waste produced by the rotary kiln incinerator at Rocky Flats. This incinerator has been used to burn a non-radioactive mixed combustible waste formulation that is representative of the low level wastes produced at Rocky Flats. The waste feed composition and incinerator operating parameters employed are shown in Table 2. A 40 kg batch of this ash was received at BNL at the end of February 1979. A sample of this ash was taken for analysis by DC arc emission spectroscopy. The results of this analysis and one performed at Rocky Flats are shown in Table 3. For both sets of analyses, confidence limits are approximately ± 10% for concentrations above one weight percent and ± 50% for lower concentrations (except where indicated). Both analyses are in reasonable agreement. Differences may be due to inhomogeneities in the ash samples analyzed. The ash composition determined from the Rocky Flats analysis has also been used to calculate a cumulative composition weight percentage assuming that cations have been completely converted to stoichiometric oxides. This calculation provides a cumulative composition of 90.7% for all species identified.

However, more than one oxidized state may exist for some cationic species. In addition, visual observation of the ash suggests that the ash contains some metallic species in partially unoxidized form.

The rotary kiln ash as received has a low bulk density of approximately 0.23 g/cm^3. It is flocculent in nature (similar to cigarette ash), gray-white in color but exhibits color gradations and has carbon present as distinct particulate. Small pieces of wire and other partially unburned residues are also present. In the presence of water, the ash has a pH of 5.5-6. The characteristics of the as-received ash are significantly different from the simulated ash composition used previously.

While it was desired to formulate specimens using the rotary kiln ash, a source of activity was needed for the leaching experiments. It was decided to obtain a quantity of dual chamber burner incinerator ash from Rocky Flats for this purpose. This incinerator is used to burn highly contaminated combustibles prior to an acid leaching plutonium recovery operation. Two samples of this ash, with a total mass of 318 grams, was received in March. This ash contains 28.9 grams of plutonium. As indicated in the ash assay shown in Table 4, both ash samples are similar with 94% of the plutonium present (on a weight basis) as Pu-239. A small quantity of Am-241 is present in both ash samples. The total plutonium activity content is 12.38 Ci (0.0389 Ci/g ash), of which 13.4% is Pu-239 and 83.0% is Pu-241. Small quantities of this "hot" ash are added to the "cold" rotary kiln ash during leach specimen fabrication. This is preferable to the simple addition of plutonium since the plutonium incorporated in the waste form will be in the same chemical and physical state resulting from incineration. The relatively small quantity of "hot" ash used will not appreciably alter the average chemical composition of the incinerator ash incorporated into leach specimens.

Prior to the receipt of the actual incinerator ashes, waste form formulations were investigated using the simulated Rocky Flats plutonium recycle incinerator ash composition shown in Table 5 for solidification with portland type I cement, urea-formaldehyde, bitumen and vinyl ester-styrene. Waste forms were produced containing a wide range of ash contents with each solidification agent. These formulations were immersion tested in distilled water for three months to determine the approximate maximum ash content which resulted in waste forms that would be expected to maintain their integrity during leach testing. An approximate upper limit of 50 wt.% simulated ash was found for portland type I cement, urea-formaldehyde, and bitumen. Vinyl ester-styrene could incorporate 75 wt.% ash although some difficulties with its promoter-catalyst system was noted.

Formulational work using the "cold" rotary kiln ash with cement solidification indicated that it behaves considerably differently than the simulated ash composition used earlier. This difference is largely attributable to the physical structure of the ash. It has a large particle size with significant contained porosity as indicated by its apparent water absorption. However, the water required to saturate the ash is weakly held since it separates on mixing. The ash as-received is difficult to mix directly with cement (independent of the order of addition) unless sufficient water is added to satisfy the ash's absorptive requirements. When sufficient water is added, a minimum of mixing often resulted in water separation and an excessively wet mixture. To minimize these tendencies, the ash was ball milled. This not only decreased the ash particle size and porosity, but also resulted in a more homogeneous ash with a bulk density of approximately 0.37 g/cm^3. The resulting ash had lower (although substantial) water requirements for cement formulations and was also easier to work with. The cement waste form formulation which was developed for the experimental program consists of 30 wt.% ash, 30 wt.% cement and 40 wt.% water. The ash content of this formulation is lower than that developed for the simulated ash because of the significantly higher water content requirements of the actual ash.

These differences in ash behavior also affect solidification with urea-formaldehyde, bitumen and vinyl ester-styrene. As such, the "cold" ash will be ball milled prior to incorporation in all binders. The formulation with urea-formaldehyde (UF) is 34% ash, 44% UF, 17% water and 5% sodium bisulfate catalyst solution by weight. Initial UF leaching specimens did not solidify. This is apparently due to the alkalinity of the "hot" ash, which can be compensated for by the addition of more acid catalyst.

3. TRU HANDLING AND LEACHING FACILITY

3.1 Facility Cell

The transuranic handling and leaching facility has been constructed inside the former Irradiation Cell in Building 830. The inside dimensions of this hot cell are 14'x20'x14' high. The walls are 4½ feet thick, constructed of medium density concrete (240 lbs/ft^3), with the exception of the wall containing the access door. This wall is 6½ feet thick and is composed of ordinary construction grade reinforced concrete with a density of 150 lbs/ft^3. The access door which is lead encased in steel, weighs approximately 25 tons and closes the main cell opening which is five feet wide and seven feet high. Additional access is available through ceiling and wall plugs. The ceiling is made of ordinary construction grade reinforced concrete and is 6½ feet wide thick. Two cell walls each have a 52 inch thick high density lead shielding glass window. In addition, this cell was equipped with two through the wall manipulators and a completely enclosed General Mills arm manipulator. The Irradiation Cell is connected to an adjacent Preparation Cell by a 16 feet deep water canal.

While the extensive shielding capability provided by the irradiation hot cell is not necessary for the transuranic leaching experiments proposed, the space was available and it does serve as an effective means of isolating the work from general laboratory facilities. Some modification of the cell interior was required to make this space suitable for a leaching facility. The cell was surveyed to determine the level of existing contamination, if any. No significant contamination was detected (none was expected since the cell was only used for irradiation experiments using sealed sources). The General Mills arm was removed as were two canal elevator lift and transfer assemblies. The elevator openings to the canal and a center floor opening to an enclosed water pool were covered with stainless steel plates. The interior of the cell was then completely repainted. A fire retardant wooden interior doorway was constructed to permit restricting access to the facility during the working day when the lead shielding door is open. While the cell has existing gamma radiation monitoring equipment and alarms, an alpha air monitoring unit was installed (Eberline Model Alpha-3). In addition, both photoelectric and ionization chamber smoke detectors have been installed.

The cell has its own temperature controlled air recirculation system which contains two levels of HEPA filtration. The facility will probably be operated at 100% recirculation although the percentage of air which is recirculated can be varied. Any portion of air which is not recirculated is connected to the main laboratory stack inlet where an air flow rate of 700 linear feet per minute moves the exhaust air to the stack which is approximately ¼ mile from the inlet. The initial level of HEPA filtration is located at the cell exhaust duct while a second HEPA filter is located in the basement machine room at the main stack exhaust inlet.

3.2 Specimen Preparation Glove Box

A two compartment stainless steel glove box with an air lock entry assembly for each compartment has been modified and placed in the leaching facility to provide capability for remote fabrication of transuranic contaminated leaching specimens. This glove box had been previously used at laboratory for plutonium handling operations. The unit was thoroughly decontaminated prior to receipt in Building 830. Each of the two compartments is 33 inches long x 24 inches wide x 33 inches high. They are separated by a stainless steel barrier containing an 11½ inch diameter door. Each compartment has lighting, electrical supply outlets and its own internal HEPA filter in addition to a pair of glove ports. There is an airlock assembly for each glove box compartment. The airlocks are 19 inches long x 19 inches wide x 20 inches high. The airlocks are each separated from the main glove box compartments by an 11½ inch diameter door. Both airlocks also have an 11½ inch diameter door to the outside, a pair of glove ports and a plexiglass endplate to permit viewing. The glove box is supported on a table assembly which permits adjustment of the glove box height.

Air from the glovebox is exhausted through two levels of HEPA filtration. The first level is the internal HEPA filter in each compartment as previously mentioned. These filters are both connected to a double series HEPA filter outside the glove box through a two inch diameter manifold. This is in turn connected to an exhaust pipe leading into four inch diameter polyethylene flex tubing which is fed through existing ductwork to an exhaust blower in the basement machine room. The blower is not normally used since the main stack supplies sufficient negative pressure for most glove box uses. The glove box is operated under a negative static pressure of approximately one inch of water. A bagging facility is installed on the "hot" side airlock. An air bleed and HEPA filter is used to bring the box pressure to atmospheric when bagging is performed.

Specimen preparation procedures have been developed under which one side of the glove box is treated as a "hot" compartment in which the specimens are fabricated while the other compartment is "cold". The "cold" compartment is used to remove solidified leach specimens from their preparation containers and place them into leach containers. These leach containers must be uncontaminated to allow their removal from the glove box to the leaching apparatus.

Problems were encountered with the operation of the glove box during the preparation of the first set of leaching specimens (ash, cement and UF). During transfer to the "cold" side of the box (which took place 24 hours after specimen preparation), significant contamination and airborne activity were noted. This was determined by surface smears and air sampling from the "cold" compartment. Health physics personnel would not permit the removal of the leach containers until the "cold" side of the box was decontaminated. Also, additional modifications to the glove box were required for further operation. The contaminated ash is apparently highly dispersible with minute particles remaining suspended in the air in the "hot" side of the box for considerable periods. Since the glove box was operated under static negative pressure conditions, this airborne contamination was able to move to the "cold" compartment during the transfer operation. The glove box has been modified to allow an air flow from the "cold" side into the "hot" side during transfers and, if desired, during procedures in the "hot" compartment. The exhaust blower on the recirculation air line is used to provide an increased air flow through HEPA filters and into the glove box when this is desired. A small suction hood is also now used over the "hot" container during specimen preparation. Improved in-box decontamination facilities are also being provided. An improved air sampler on the "cold" compartment airlock has been installed.

3.3 Leaching Facilities

A semi-automated apparatus has been designed for performing IAEA/ISO leach tests. This technique requires that the leaching solution be changed and completely replaced at frequent intervals (daily for the first week, once per week for the following eight weeks and subsequently once per month). Since the leachant solutions are expected to contain quantities of plutonium removed from the leach specimens, it was decided to build an apparatus which would require a minimum of direct manual handling of the specimens and solutions. Leaching equipment was designed and constructed to allow the leaching experiment to be performed in the following manner. The leach specimen is placed into a one liter capacity polymethylpentene screw cap container. This material was chosen because it is autoclavable and has a low potential for reacting with and "plating out" species of interest on the container walls. The specimen is supported on a plexiglass stand which makes essentially the entire surface area of the leach specimen accessible to the leachant. The container is connected to a ¼ inch o.d. valved inlet manifold, through which the desired amount of leachant can be added to any individual leach container. The manifold and ¼ inch i.d. ball valves are type 304 stainless steel. A separate inlet manifold is used for each leachant type. The inlet manifold also has a recirculation return line to prevent clogging of the inlet manifold as well as provide a mixing action in the leachant storage container.

Leachant is stored in steel drums with 27 gallon polyethylene liners. A small Flotec gear pump is used to provide the motive force for leachant transfer. After the appropriate period of leaching, the leachant in the leach container is stirred and sampled using a Eppendorf pipette. After sampling, the valve to the exhaust manifold is opened. Since the exhaust manifold is maintained at a negative pressure, opening the ball valve for the desired leach container removes the leachant under a suction action. A teflon suction tube is positioned in the leachant container which allows removal of essentially all of the leachant. The exhaust manifold is 3/8 inch O.D. type 304 stainless steel which drains to a forty gallon capacity glass lined mild steel tank with a fluid level indicator. This tank when full can be drained directly into a feed line to a 2,000 gallon capacity pair of buried waste holding tanks (outside of Bldg. 830) which are periodically emptied by tank truck. The leachant exhaust manifold leads to a plexiglass measuring cell apparatus before emptying into the waste holding tank. The measuring cell allows determination of leachant pH, conductivity and (if desired) Eh and oxygen content.

This apparatus may not however be compatible with a leach procedure recently proposed for DOE contractors in TRU programs. This procedure requires periodic removal of "plated" activity from the leach container. However, the procedure can be followed using a two container method with somewhat increased handling.

The static 25^{o}C leach tests will be conducted at ambient temperature in the leach facility. The IAEA/ISO procedures specify 25^{o}C±5^{o}C while ambient temperature in the leach facility is approximately 21^{o}C±1^{o}C. The facility temperature will be monitored during testing. A specially modified incubator with excellent temperature stability has been ordered for the 70^{o}C static leach tests.

4. LEACHANT ANALYSIS

Quantitative alpha spectrometry typically employs a solid state silicon surface barrier detector to count specimens electroplated onto stainless steel planchets. The resolution of such a detector is of the order of 50 KeV which permits simultaneous analysis of alpha emitters with similar energies. However, the sample preparation required in this technique is very labor intensive. The leaching experiments to be conducted at BNL will produce more samples for analysis than can be conveniently prepared in this manner. As such, other analytical techniques were investigated and liquid scintillation counting (LSC) was chosen as the primary analytical technique.

Liquid scintillation counting has been utilized for alpha counting since 1954.(4,5) This method is attractive due to the ease of specimen preparation, the commercial availability of remote sample changers, and the 100% efficiency usually obtained in alpha counting. (Alpha particles are counted at 100% efficiency in LSC provided the activity is dissolved in the scintillation solution. The efficiency is lowered by a geometry factor if the activity plates out on the container walls or is suspended on a solid matrix.) While energy resolution is not as good as can be obtained with other methods, different alpha emitters can be counted simultaneously provided their respective alpha particle energies are sufficiently different. Peak resolution in LSC is a function of a number of factors but alpha particles of approximately 5 MeV generally can be resolved if peak energy differences are of the order of 0.5-0.7 MeV. As such, some alpha emitting radionuclides may possibly be analyzed simultaneously since appreciable primary particle energy differences exist: ^{239}Pu (5.147 MeV), ^{241}Am (5.482 MeV), ^{237}Np (4.787 MeV), ^{235}U (4.354 MeV).

A Searle Model 6892 automatic liquid scintillation counter was purchased for this program. It employs a bidirectional automatic conveyor sample changer which accepts up to three hundred samples. The detector consists of two 2-inch diameter bi-alkali photomultiplier tubes which are situated 180° apart in a metal chamber with specular aluminum reflectors in a 2-inch thick lead shielding housing. There are two variable simultaneous sample analysis channels with two additional fixed level channels for external standard source (ESR) analysis.

Analysis channel baseline and width, counting time, preset count terminators and sample changing mode can be independently determined and incorporated into internally stored program instructions. Data output is through a Teletype Model 43 printer.

5. EXPERIMENTAL RESULTS

Experiments were initiated to determine the extent of "plate out" occurring for ^{239}Pu in the semi-automated leaching cells. These cells are constructed primarily out of polymethylpentene which was selected for its low "plate out" potential and thermal stability (autoclavable). "Plate out" is expected to be strongly dependent upon the chemistry of the system. As such, experiments were performed using each of the five leachant solutions and each solidification agent, except urea-formaldehyde. One inch diameter x two inch long cylindrical specimens of portland type I cement, bitumen, and vinyl ester-styrene, each containing 50 wt.% simulated ash were leached for three weeks in 250 milliliters of leachant without leachant renewal. Also leached was a specimen of portland type I cement (water/cement = 0.3 by weight) containing no ash. In addition to "plate out" experiments, the resultant leachant solutions were also employed in studies to determine counting efficiency and chemical quenching (if any) in LSC analysis.

After leaching, ten milliliters of each of the solutions (except brine) were placed into 125 ml polymethylpentene containers (of the same manufacturer as the one liter leaching cell containers). The pH of these leachant solutions was determined. The pH range was 7.5-10 for bitumen and vinyl ester-styrene leachants and 12-13 for cement and cement (no ash) specimens. These leachant solutions were then each spiked with twenty lambda of a 0.5 $\underline{M}$ HNO_3 solution of ^{239}Pu. After a contact time of 18 hours, the leachants were each sampled and counted. A loss of 40-60% of the initially contained ^{239}Pu was observed. Investigation of "plate out" as a function of time was not conducted.

The leachants, prepared as described previously, were used to determine counter efficiency and the stability of counting samples. These leachants should represent worse case situations for the IAEA/ISO test and should contain large quantities of dissolved solids removed from the leach specimens.

Leachant analysis to date has utilized Packard Insta-Gel as the liquid scintillation cocktail. This cocktail is able to form a stable counting media with relatively large quantities of aqueous samples. A seven milliliter aliquot of each leachant solution was mixed with ten milliliters of Insta-Gel and allowed to stand for two weeks. Stable counting samples were obtained with all leachants except salt brine. While other leachants formed a stable gel phase with Insta-Gel, the salt brine mixture separated into two phases. A stable counting sample was not obtained with as little as one milliliter of brine leachant in ten milliliters of Insta-Gel. A stable sample was produced using a 1:25 dilution of the brine prior to combining seven milliliters of diluted brine with ten milliliters of Insta-Gel. While a sample which separates into two phases can be counted, the counting efficiency can decrease substantially. A low-level x-ray detector will be used for salt brine leachant analysis.

Alpha particles are counted at 100% efficiency in LSC provided the activity is uniformly distributed in the scintillation solution and no chemical quenching effects occur. Counting efficiencies were determined using each of the leachants spiked with americium-241. Seven milliliters of leachant was combined with ten milliliters of Insta-Gel for each counting specimen. Counting efficiencies were determined relative to the known quantity of americium-241 added to each LSC vial. Salt brine leachant samples (which were unstable, having separated into two phases) exhibited a range of counting efficiencies of 63.7% to 79.6% with an average of 70.0%. The other leachants had an average counting efficiency approaching 100.0%. Some lower counting efficiencies were determined as indicated in Table 6. It is unclear, however, whether the lower calculated counting efficiencies are due to slight chemical quenching effects or pipeting errors. It appears, in most cases, that pipeting error is most likely since lower efficiencies do not seem to be associated with any particular leachant or waste form type.

It was also noted that small quantities of a precipitate were present in leachants derived from portland cement waste forms. The precipitate quantity varied from 0.08 to 0.47 grams, with the larger precipitate quantities present in the salt brine leachant. This precipitate was collected for analysis to determine whether it is likely to occur in IAEA/ISO leaching experiments and if it may be expected to remove activity from leachant solutions. This precipitate was found to be calcium carbonate. It is not believed that it will remove substantial quantities of plutonium from the leachant.

A set of scoping specimens was prepared for IAEA/ISO leaching at the end of March. These were the first IAEA/ISO leach specimens whose primary purpose was to determine if sufficient activity had been added to leach specimens to allow the desired level of detectibility in the leachant. A portland type I cement specimen and urea-formaldehyde specimen were prepared, each containing 13.5 mCi plutonium, and placed into 500 milliliters of distilled water leachant. In addition two 0.33 gram ash specimens (12.8 mCi) were prepared for leaching. The UF specimen did not solidify due to insufficient catalyst. Contamination of the glove box "cold" side was noted, as previously mentioned, when these specimens in leach containers were to be transferred to the leaching apparatus. Since leachant samples could not be obtained until health physics requirements were satisfied (33 days), the resultant test is actually a static test. Subsequent LSC analysis of the leachant showed a fractional activity release of 1.6×10^{-8} for the cement specimen with 9.4×10^{-7} fractional release for the ash. These correspond to fractional activity release rates of 4.9×10^{-10} day^{-1}, and 2.9×10^{-8} day^{-1} for the cement and ash respectively. The leachant containers have not been examined for "plate out".

While problems have impeded the initiation of leach testing, it appears that these problems have been solved. It is expected that current scheduling commitments can be met, although it may be useful to put more initial emphasis on the static procedure than the IAEA/ISO method.

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TABLE 1

Composition of Salt Brine Leachant (derived for AEC #8, 2725 foot level core)

	Composition (mg/liter)
$NaCl$	287,000.
Na_2SO_4	6,200.
$Na_2B_4O_7 \cdot 10H_2O$	16.0
$NaHCO_3$	14.0
$NaBr$	520.0
KCl	29.0
KI	13.0
$MgCl_2$	40.0
$CaCl_2 \cdot 2H_2O$	3,300.
$FeCl_3$	6.0
$SrCl_2 \cdot 2H_2O$	33.0
Rb_2SO_4	1.6
$CsCl$	1.3
Total Dissolved Solids	297.2 g/liter
pH (adjusted)	6.5

TABLE 2

Rocky Flats Rotary Kiln Incinerator Ash Operating Parameters

A. Feed Composition

	Weight %
Polyvinyl chloride	7.9
Polyethylene	22.8
Neoprene	18.8
Tributyl Phosphate	1.0
Kerosene	9.5
Paper	40.0
	100.0

B. Feed Rate

Solids	6.8 kg/hr
Liquids	0.45 liter/hr

C. Operating Temperature

Kiln	$1000^{\circ}C \pm 100^{\circ}C$
After-Burner	$1400^{\circ}C \pm 100^{\circ}C$

D. Operating Time

∿ 150 hrs

TABLE 3

Rocky Flats Rotary Kiln Incinerator Ash Composition

Constituent	Weight Percentage RF Analysis	 BNL Analysis
C	10.4 ± 30%	13.1
Al	18	18
Si	15	>10
Zn	2.6	2.6
Ti	1.8	4.8
Fe	1.4	0.3
Ca	1.2	5
Ba	0.94	0.1
Mg	0.65	1
Li	0.5	<0.001
Cr	0.12	0.05
Na	0.1	0.5
P	0.1	-
SO_4^{-2}	0.1 ± 0.05	-
Ni	0.08	0.1
Mn	0.07	0.03
Cl^-	0.06	-
Cu	0.05	0.08
K	0.05	0.1
Pb	0.05	0.03
W	<0.05	-
Sr	0.02	0.05
Co	0.01	-
Mo	0.01	-

TABLE 3 (Con't'd)

Rocky Flats Rotary Kiln Incinerator Ash Composition

Constituent	Weight Percentage RF Analysis	 BNL Analysis
Zr	0.01	-
Cs	<0.01	-
Te	<0.01	-
B	0.005	-
Sb	0.005	-
V	0.005	-
As	<0.005	-
Cd	<0.005	-
Nb	<0.005	-
Ge	0.001	-
Hg	<0.001	-
Sn	<0.001	-
Ta	<0.001	-
Th	<0.001	-
Be	0.0005	-
Bi	<0.0005	-
In	<0.0005	-
Ga	<0.0001	-
Rb	-	<0.01
Ag	-	0.0005

TABLE 4

Dual Chamber Burner Incinerator Ash TRU Assay

ASH SAMPLE 1

Isotope	Quantity, grams	% Total Plutonium	Activity, Ci
Pu-238	0.002	0.01	$3.49\text{x}10^{-2}$
Pu-239	17.290	93.81	$1.06\text{x}10^{0}$
Pu-240	1.078	5.85	$2.44\text{x}10^{-1}$
Pu-241	0.057	0.31	$6.51\text{x}10^{0}$
Pu-242	0.004	0.02	$1.56\text{x}10^{-5}$
TOTAL PLUTONIUM	18.431	100.00	$7.85\text{x}10^{0}$
Am-241	0.023	-	$4.09\text{x}10^{-3}$

ASH SAMPLE 2

Isotope	Quantity, grams	% Total Plutonium	Activity, Ci
Pu-238	0.001	0.01	$1.74\text{x}10^{-2}$
Pu-239	9.851	93.83	$6.04\text{x}10^{-1}$
Pu-240	0.611	5.82	$1.38\text{x}10^{-1}$
Pu-241	0.033	0.31	$3.77\text{x}10^{0}$
Pu-242	0.002	0.02	$7.79\text{x}10^{-6}$
TOTAL PLUTONIUM	10.499	99.99	$4.53\text{x}10^{0}$
Am-241	0.031	-	$3.86\text{x}10^{-3}$

TABLE 5

Simulated Rocky Flats Plutonium Recycle TRU Ash Composition

Material	Weight Percentage
Al_2O_3	3.69
BaO	0.59
B_2O_3	1.67
CaO	3.64
Cr_2O_3	2.36
CuO	0.64
Fe_2O_3	5.85
PbO	0.54
MgO	2.90
MnO_2	0.10
NiO	0.50
P_2O_5	0.59
KOH	0.50
PbO_2	2.61
SiO_2	45.00
NaOH	0.89
V_2O_5	0.64
SnO	0.15
TiO_2	2.17
C	25.00

TABLE 6

LSC Counting Efficiency (Am-241)

Leachant	Leach Specimen[(a)]	Counting Efficiency, %
Salt Brine	portland cement	79.6
	bitumen	63.4
	vinyl ester-styrene	63.7
	portland cement (no ash)	73.4
Sodium Dominated Groundwater	portland cement	91.8
	bitumen	94.1
	vinyl ester-styrene	99.3
	portland cement (no ash)	100.1
Calcium Dominated Groundwater	portland cement	99.4
	bitumen	95.5
	vinyl ester-styrene	102.6
	portland cement (no ash)	95.3
Bicarbonate Dominated Groundwater	portland cement	100.5
	bitumen	102.7
	vinyl ester-styrene	100.3
	portland cement (no ash)	102.2
Deionized Water	portland cement	101.9
	bitumen	100.7
	vinyl ester-styrene	95.2
	portland cement (no ash)	102.9

(a) except where indicated, 50 wt.% simulated ash waste