

324
2-28-80

DR. 758

LA-7931-PR

Progress Report

MASTER

**Studies of Transuranic Waste Storage Under
Conditions Expected in the
Waste Isolation Pilot Plant (WIPP)**

Interim Summary Report

October 1, 1977—June 15, 1979

University of California



LOS ALAMOS SCIENTIFIC LABORATORY

Post Office Box 1663 Los Alamos, New Mexico 87545

REF ID: A66000

UNLIMITED

The four most recent reports in this series, unclassified, are LA-7478-PR, LA-7582-PR, LA-7649-PR, and LA-7775-PR.

This work was supported by the US Department of Energy, Division of Waste Management, Production, and Reprocessing.

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Department of Energy, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights.

**Studies of Transuranic Waste Storage Under
Conditions Expected in the
Waste Isolation Pilot Plant (WIPP)
Interim Summary Report
October 1, 1977—June 15, 1979**

Stanley T. Kosiewicz
Bruce L. Barraclough
Al Zerwekh

DISCLAIMER

This report was prepared as part of the work supported by the U.S. Department of Energy under contract number DE-AC02-77OR21400. The U.S. Government is authorized to reproduce and distribute reprints for government purposes not withstanding any copyright notation that may appear hereon. This report is not to be distributed outside the Laboratory.



1/84

STUDIES OF TRANSURANIC WASTE STORAGE UNDER CONDITIONS EXPECTED IN THE WASTE ISOLATION PILOT PLANT (WIPP)

Interim Summary Report
October 1, 1977—June 15, 1979

by

Stanley T. Kosiewicz, Bruce L. Barraclough,
and Al Zerwekh

ABSTRACT

This is the seventh quarterly report describing progress in an experimental program to determine the effects of radiolytic, thermal, and catalyzed thermal degradation of various forms of existing or developmental transuranic (TRU) wastes proposed for the Waste Isolation Pilot Plant (WIPP) emplacement. At the request of the WIPP technical program manager, this report is an interim summary of work done from October 1, 1977, through June 15, 1979. The major focus of the program has been on the gas generation potential of organic wastes produced by radiolytic and thermal degradation under simulated WIPP storage conditions. The effects of TRU contamination level, temperature, waste type, pressure, and exposure time on radiolysis are presented. In addition, results from preliminary experiments on processed-sludge dewatering are discussed.

A summary is presented here of the results of a detailed study of all retrievably stored TRU wastes present at the Los Alamos Scientific Laboratory (LASL) before January 1, 1978. The data indicate a gross volume for the LASL inventory of 1610 m³ with a total weight of nearly 1.21 x 10⁶ kg (1240 metric tonnes). The dominant radionuclide contents of the waste are plutonium (primarily ²³⁹Pu) and americium.

I. INTRODUCTION

This experimental program is designed to determine the effects of radiolytic, thermal, and catalyzed thermal degradation of various forms of existing or developmental transuranic (TRU) wastes proposed for Waste Isolation Pilot Plant (WIPP) emplacement. Waste matrices, experimental conditions, and specific experiments are selected after

periodic consultations and reviews with the Sandia Laboratories, Albuquerque (SLA) WIPP technical program manager to ensure meeting the continuing and developing needs of the program. An additional task to assess and characterize all forms of TRU wastes generated and retrievably stored at the Los Alamos Scientific Laboratory (LASL) from 1971 through the end of CY 1977 has been completed.

"Dose" and "dose rate" have occasionally been used interchangeably by readers of our reports. The words have quite different meanings, and are not interchangeable. We continue to use them to maintain uniformity with Bibler¹ of the Savannah River Laboratory (SRL) and other workers in the field of radiation chemistry.

Dose rate is the rate at which energy is deposited in a waste matrix by the radionuclide (plutonium) contaminant. In our reports, dose rate is expressed in nanocuries per gram (nCi/g) of waste. Also, we define a dose rate of 4×10^5 nCi/g to be equivalent to 1 X. *Dose rate* may also be equated to *plutonium loading*, *degree of plutonium contamination*, or the *plutonium concentration* (expressed in nCi/g) in the waste matrix. High dose rates imply highly contaminated material.

Dose is the total amount of energy deposited by the TRU contaminant in the waste matrix. Dose is the product of the dose rate and the elapsed time for an experiment. In our reports, dose is expressed in electron volts (eV) because this is the unit of energy used in the calculation of G(gas)

II. BACKGROUND

The WIPP is the first proposed national repository for defense-generated TRU wastes. Its planned location is 41.8 km (26 miles) east of Carlsbad, New Mexico (Fig. 1). TRU waste is waste other than legally defined high-level waste that contains more than 10 nCi of long-lived alpha-emitting radionuclides per gram of waste.² The sources of defense TRU waste are

- (1) currently produced waste,
- (2) retrievably stored waste,
- (3) buried waste, and
- (4) decontamination and decommissioning waste.

The first two sources of waste are relatively well-characterized, whereas the third is not. The Department of Energy (DOE) inventory of the first two sources is roughly 45 307 m³ (1 600 000 ft³) and the DOE inventory of buried waste is about 269 012 m³ (9 500 000 ft³). A large percentage of these wastes is combustible (20-25 vol%).³

Combustible wastes have the potential for generating various gases. During geologic storage, such gases could breach the repository and decrease

containment integrity. Mechanisms identified as potential sources of gas generation are

- (1) radiolytic degradation,
- (2) thermal degradation,
- (3) bacterial degradation, and
- (4) corrosion of metals.

Degradation products from the wastes may chelate the radionuclides, increasing the probability of mobilizing the products. Also, the combustible wastes may present a fire hazard, but only during the operating phase of the mine.

SLA has the major responsibility for technical aspects of the WIPP. SLA and its contract laboratories perform experiments in areas of major concern or where insufficient data exist. The WIPP TRU experimental program has been summarized by Molecke.⁴ Some of the other laboratories participating in the program are LASL, the University of New Mexico (UNM), and SRL. Experimental assignments were made based on previously established expertise.

In November 1977, an experimental program was contracted to LASL to study the radiolytic and thermal degradation of organic materials. These TRU wastes would be interred in the WIPP about 640 m (2100 ft) deep (Fig. 2). In addition to materials that typically compose TRU wastes, bitumen was included in the study because Europeans use it as a waste immobilization medium.

III. THERMAL DEGRADATION OF WASTE MATRICES

Thermal degradation of organic waste matrices under WIPP conditions and temperatures could generate gas. The waste matrices in the WIPP may be subjected to temperatures of 20, 40, or 70°C. The ambient WIPP temperature during its operating phase is 20°C. If heat-generating wastes are emplaced in the WIPP at 75 kW/acre loading, the temperature will rise to about 40°C approximately 200 yr after repository sealing. If heat-generating wastes are emplaced at 150 kW/acre loading, the temperature will peak at 70°C, then slowly decrease.

For our experiments, we included an overtest temperature of 100°C to obtain accelerated rate data. The TRU waste matrices in the WIPP are never expected to be subjected to 100°C.

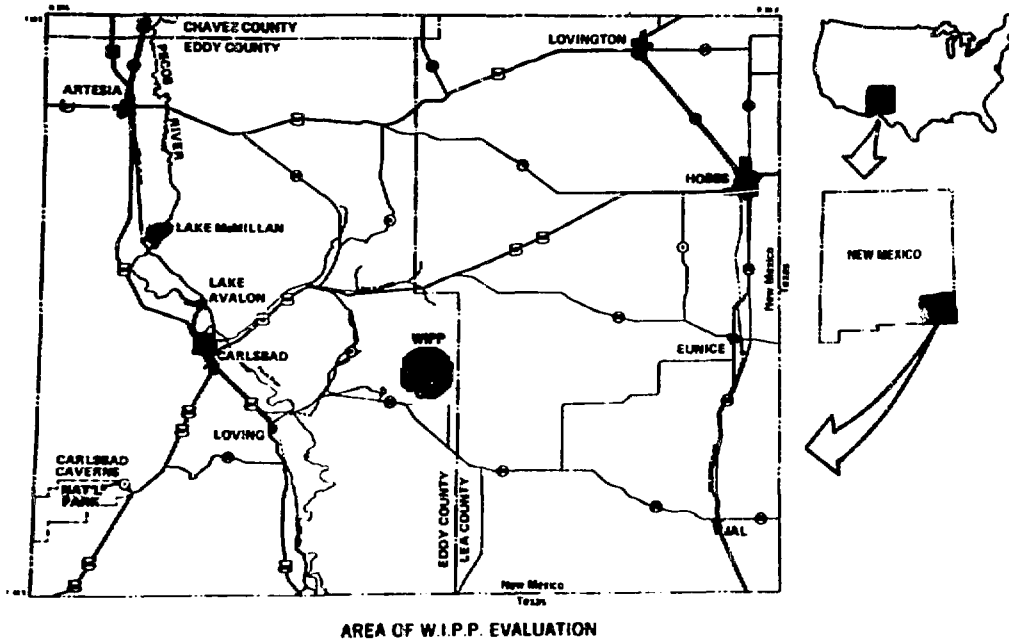


Fig. 1.
WIPP location.

Our thermal degradation studies consist of two phases: (1) determination of threshold decomposition temperatures for a broad range of common laboratory materials and (2) surveillance experiments on selected waste matrices at potential WIPP temperatures.

A. Threshold Decomposition Temperatures

Waste matrices to be tested are divided into three broad categories: cellulose, plastics, and rubbers. Materials from each of these categories were subjected to thermogravimetric analyses using aerobic (air) and anaerobic (nitrogen) atmospheres to determine threshold decomposition temperatures. The temperature at which a sample began to lose weight (exclusive of sorbed water) was taken as the threshold decomposition temperature. Previous experiments on TRU wastes⁹ showed that oxygen was

consumed rapidly to produce a relatively anaerobic atmosphere over the wastes. Consequently, the majority of the threshold decomposition experiments were performed in a nitrogen atmosphere.

To perform these studies, a Mettler TA-II thermobalance was used. Because this particular instrument was designed for high temperature work (>1000°C), it was modified to operate at lower temperatures. After modification, the instrument operated well at temperatures as low as 40°C. The experimental parameters used for the threshold decomposition studies were

- instrument: Mettler TA-II thermobalance.
- gas-flow rate: 10-20 cm³/min.
- temperature: 20-500°C.
- heating rate: 10°C/min. and
- sample size: 60-80 mg.

Threshold decomposition temperatures measured in air for various cellulose, plastics, and rubbers that are common in TRU wastes are presented in

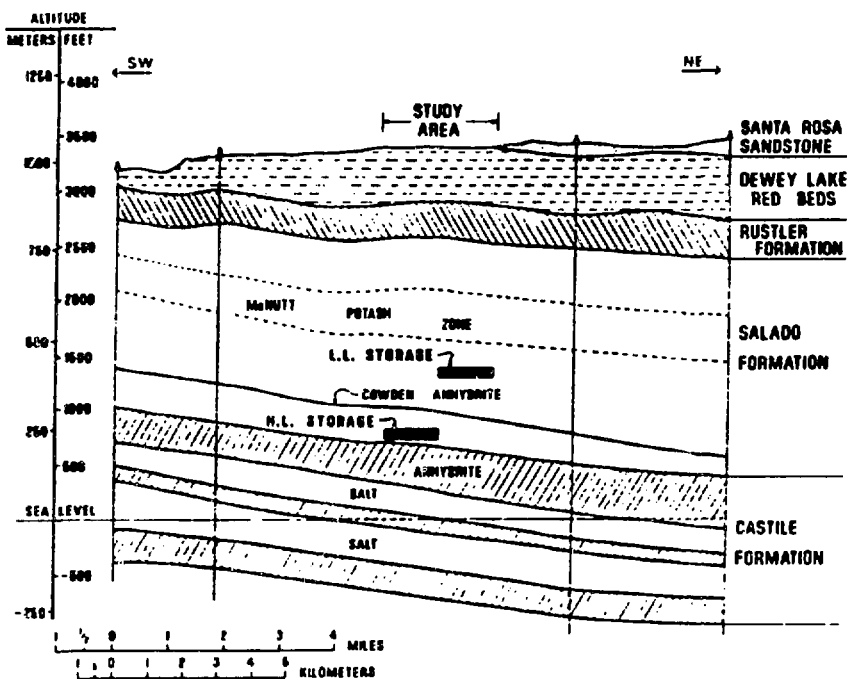


Fig. 2.
Geologic section through the Los Medaños area.

Table I. Although Pylox (polyvinyl chloride, PVC) gloves have the lowest temperature (150°C), it is substantially higher than the highest temperature expected in the WIPP (70°C). All of the cellulose evolved about 2-3 wt% water when heated through the 4-110°C range. This water was sorbed by the materials, not chemically bound. No other category of waste materials evolved water below 110°C. Hansen,⁶ however, proposed a reaction mechanism for the thermal oxidation of hydrocarbon polymers that produce water, and has observed oxidation of polyethylene at 80°C. Consequently, thermal degradation of TRU wastes at the maximum WIPP temperature (70°C) might produce water if oxygen were present.

Because several of the materials in each waste category have similar characteristics and threshold decomposition temperatures in air, only a few representative materials from each category were chosen to represent the entire category for additional studies. Because oxygen is rapidly consumed under waste storage conditions, decomposition likely

would occur under anaerobic conditions in the WIPP. Consequently, the only materials capable of water production are those that have chemically bound oxygen. The materials chosen for this portion of the work and their measured threshold decomposition temperatures in nitrogen are listed in Table II. Polyethylene is included because it constitutes a large percentage of typical wastes.

Anaerobic decomposition of paper starts at approximately the same temperature as it does in air. Unless the degradation mechanisms are considered, this fact may seem surprising. Schwenker and Beck⁷ conducted pyrolysis-gas chromatographic (GC) experiments to investigate degradation mechanisms of cellulose under different atmospheres. They concluded that the thermal degradation mechanism of cellulose was nonoxidative, and hence, independent of the atmosphere. Consequently, the threshold decomposition temperatures under aerobic and anaerobic conditions should be equal. For polymethyl methacrylate (Plexiglas or Lucite) and Nylon, the anaerobic decompositions appear to

TABLE I
THRESHOLD DECOMPOSITION
TEMPERATURES IN AIR

Material	Temperature (°C)
Cellulosics*	
Scott utility wipes	185
Kleenex tissues	185
Diaper paper (PE-backed)	190-195
Cloth (cotton twill)	185
T-shirt (cotton)	185-190
Cheesecloth	205
Wood	175
Fiberboard	185-190
Plastics	
Pylox gloves (PVC)	150
Tygon tubing (PVC)	175
Polyethylene	210
Polypropylene	195-200
Lucite [poly(methyl methacrylate)]	170-175
Teflon [poly(fluoroethylene)]	430-435
Rubbers	
Hypalon	165
Neoprene	175-180
Durasol/Neosol	180
Latex	195

*Water loss observed at 40-110°C.

TABLE II
THRESHOLD DECOMPOSITION TEMPERATURES
IN NITROGEN

Material	None	NaCl	Fe ₂ O ₃	FeCl ₃	Fe Powder
Utility wipes	185	180	185	170-195	195
Cotton	185	195	200	170-195	205
Lucite	150	155	170	185	160
Polyethylene	320	290	295	330	335
Hypalon	160	175	160	155	160

start at slightly lower temperatures than they do in air. Conley and Malloy⁶ state that the monomer is virtually the only pyrolysis product of polymethyl methacrylate in vacuum. They further suggest that pyrolysis occurs by "unzipping" the polymer by

radical formation. Consequently, pyrolysis of Lucite may be independent of the atmosphere. No information on Hypalon was found.

To determine if threshold decomposition temperatures would be lowered by materials that

might come into contact with TRU-contaminated wastes in the WIPP, selected simulated waste matrices were doped with four potential catalysts: sodium chloride, powdered iron, powdered iron oxide, and powdered ferrous chloride. The chemicals were applied to the waste matrices by shaking them with the materials. The loading ranged from 2 to 38 wt% because the chemicals adhered differently in accordance with the textures of the waste materials. The cellulose had higher loadings; the plastics and rubbers had lower. Catalyzed threshold decomposition temperatures are presented in Table II. No significant lowering of the temperatures was observed for any of the waste materials with any of the potential catalysts.

Even though the observed threshold decomposition temperatures are significantly higher than the maximum anticipated WIPP temperatures, the waste matrices could decompose very slowly during the long storage times in WIPP. Murphy⁹ states that cellulose decomposes even at room temperature at a very slow, but finite rate. The reason that no weight loss is observed on the thermobalance at low temperatures may be kinetic; that is, the weight loss may be so small and may occur so slowly that it cannot be observed instrumentally.

B. Surveillance Thermal Experiments

To increase the sensitivity of observation of thermal degradation, materials under test were sealed in stainless steel cylinders to allow evolved gases to accumulate for long periods. These experiments, as well as the radiolysis experiments discussed in Sec. IV, are essentially long-term surveillance studies. We measure small quantities of gas by their pressure rather than by their mass equivalent because this is easier. In addition, selected simulated waste matrices were doped with either sodium chloride or ferrous chloride. The number of catalysts originally considered for testing was reduced to these two because a literature search indicated that only they might increase gas evolution or lower decomposition temperatures in the WIPP. The catalysts were applied by shaking them with the waste materials. The loading level was about 2 wt%.

In two of our earlier reports,^{10,11} we stated that cellulose was decomposing at 70 and 100°C, and we attempted to quantify the apparent gas generation

rates. From the data presented in Table III, it is apparent that we cannot make quantitative statements on the rates of thermal decomposition of cellulose at or below 100°C. However, gas analysis data show qualitatively that degradation is, in fact, occurring. The experiment at 70°C with no plutonium loading generates a barely measurable quantity of gas, whereas at 40°C, an even smaller amount of gas evolution is observed. Presumably, the gas is generated from thermal degradation of the cellulose.

Even at low plutonium loadings (4×10^6 nCi/g and 4×10^4 nCi/g), temperature has a synergistic effect on radiolysis. In other experiments, we observe that for a 50°C increase in temperature, radiolysis increases by as much as 70%. Those experiments listed in Table III with higher dose rates (that is, more plutonium contamination) generate more gas than those with lower dose rates. Also, higher temperature experiments generate more gas than lower temperature experiments. Some variations or anomalies in the data at the lower temperatures and dose rates, as evidenced by a very small amount of gas for the 1 X -20°C experiment, reflect the problem of measuring slow rates for relatively short times. In addition, depletion of the oxygen initially present in the cylinders creates a partial vacuum that must be offset by evolved gases. Consequently, these data are in the region of our greatest experimental uncertainty.

The data for the experiments with a composite of 35% cellulose, 23% polyethylene, 12% polyvinyl chloride, 15% neoprene, and 15% Hypalon (Table IV) generally follow the same qualitative trends presented above. Plutonium loading does not affect gas generation rates as strongly for the composite as it does for the cellulose experiments. The experiment at 100°C with no plutonium generates more gas than any of the other experiments listed in Tables III-V. Molecke¹² states that bacterial degradation of hydrogenous waste can generate much more gas than thermal degradation can. Perhaps some of the data variations that we observe are due to gases evolved by bacterial degradation of waste matrices.

Table V lists similar data for polyethylene. These data show the smallest gas generation rates; not even qualitative trends are observed experimentally. The very low gas generation rates may be due to the partial vacuum created by oxygen depletion of the

TABLE III
DECOMPOSITION OF CELLULOSICS (PAPER)

Dose Rate ^a	Temperature (°C)							
	20		40		70		100	
	Elapsed Time (days)	Gas Rate ^b (mol/d-g)	Elapsed Time (days)	Gas Rate ^b (mol/d-g)	Elapsed Time (days)	Gas rate ^b (mol/d-g)	Elapsed Time (days)	Gas Rate ^b (mol/d-g)
1 X	484	0.06	427	0	255	0.5	43	2.0
					352	0.4	127	2.8
					450	0.3	233	1.6
							309	1.6
							402	1.5
0.1 X	392	0	387	0	200	0.3	61	1.3
					297	0.1	177	1.6
					395	0.1	293	1.3
							386	1.2
No Pu	---	---	416	>0	241	0.07	---	---
					338	0.05		
					436	0.08		

^a1 X = 4×10^5 nCi/g.

^bGas generation rates must be multiplied by 10^{-6} , for example, 0.06×10^{-6} mol/(d-g).

fill gas during low-temperature oxidation of the plastic.* More exposure time is required for these experiments.

Sodium chloride and ferrous chloride are potential catalysts that wastes may come in contact with in the WIPP. The sodium chloride comes from the salt in the mine, whereas the ferrous chloride comes from degradation of the steel drums containing the wastes. Experiments have been in progress for 8 to 11 months to determine whether these chemicals can act as catalysts in the thermal degradation of cellulose or polyethylene. Based on observable, but not yet quantifiable small increases in gas, the following tentative statements are made.

- (1) Sodium chloride may catalyze decomposition of polyethylene at 40°C.
- (2) Sodium chloride may catalyze slightly decomposition of cellulose (paper) at 40°C.
- (3) Ferrous chloride catalyzes decomposition of paper at 40°C.
- (4) Some very slight (thermal or bacterial) decomposition of paper occurs without plutonium or catalysts at 40°C.

The quantities of gas generated in these experiments are quite small, making these observations tentative.

IV. RADIOLYTIC DEGRADATION OF WASTE MATRICES

For our experiments on radiolytic degradation of waste matrices, we use conditions that represent "the worst case possible." That is, the conditions are more severe than any to be expected in real wastes, but are not necessarily the most extreme conditions that we could devise. Therefore, our data represent upper limits on the radiolytic gas generation potential of the TRU wastes.

The maximum amount of weapons-grade plutonium allowed per 210-l drum designated for interment in the WIPP is 200 g.^{10,14} Based on previous work, a 210-l drum can contain about 34 kg of combustible waste.¹⁰ These quantities of waste and plutonium produce a dose rate of 4×10^5 nCi/g waste, which is equivalent to 1 X by our definition. Warren

TABLE IV
DECOMPOSITION OF COMPOSITE^a

Dose Rate ^b	Temperature (°C)									
	20		40		70		100			
	Elapsed Time (days)	Gas Rate ^c (mol/d-g)	Elapsed Time (days)	Gas Rate ^c (mol/d-g)	Elapsed Time (days)	Gas Rate ^c (mol/d-g)	Elapsed Time (days)	Gas Rate ^c (mol/d-g)		
1 X	341	0.1	406	0	252	0.2	92	1.4		
	439	0.2			349	0.2	203	1.0		
					447	0.3	300	1.4		
							393	1.5		
0.1 X	392	0	278	0.07	296	0.1	105	1.1		
			376	0	394	0	189	1.3		
							281	1.1		
							314	1.3		
							379	1.1		
No Pu	---	---	359	0	371	0	62	2.9		
							91	2.5		
									166	1.5
									215	2.0
									259	2.8

^aComposite is 35% cellulose, 23% polyethylene, 12% polyvinyl chloride, 15% neoprene, and 15% Hypalon.

^b1 X = 4×10^6 nCi/g.

^cGas generation rates must be multiplied by 10^{-6} , for example, 0.1×10^{-6} mol/(d-g).

and Zerwekh¹⁶ also estimated that the average plutonium content in a 210-*l* drum of combustible waste at LASL was ≥ 10 g. Based on our LASL Waste Assessment inventory,¹⁷ the estimate on combustible content was fairly accurate, whereas the estimate of the average ²³⁹Pu content was high by about a factor of 10.

Using the preceding estimates, we designed many experiments using dose rates of 0.1 X or 1 X and temperatures of 20, 40, 70, and 100°C (Tables VI, VII). Materials under test were chosen from the general categories of cellulose, plastics, rubbers, and also a composite consisting of materials from the three waste matrix categories. The composite is a 35 wt% cellulose, 23% polyethylene, 12% polyvinyl chloride, 15% neoprene, and 15% Hypalon. This composition conforms to suggested guidelines.^{10,19} The isotopic compositions of the ²³⁸PuO₂ and ²³⁹PuO₂ used for contaminating the simulated waste matrices were presented in Refs. 10 and 11.

A. Low-Pressure Experiments

1. Waste Matrix Type. Previous Work by Zerwekh²⁰ has shown that cellulose and polyethylene evolve more gas than do rubbers during radiolytic degradation. Consequently, as shown in Tables VI and VII, more experiments were initiated with the two former materials to maintain our philosophy of "the worst case possible." We expect to extrapolate trends observed with cellulose and polyethylene to rubbers.

During the radiolysis experiments, simulated waste matrices are contaminated with plutonium dioxide and are then sealed in stainless steel cylinders with an internal volume of 300 cm³. Typically, the amount of waste matrix used is 50 g. Consequently, the quantity of waste matrix and the volume of the experimental cylinders are proportionate to an average drum of combustible waste. Usually, the cylinders are sealed with a pressure gauge and a valve allows sampling of evolved gases.

TABLE V
DECOMPOSITION OF POLYETHYLENE

Dose Rate ^a	Temperature (°C)							
	20		40		70		100	
	Elapsed Time (days)	Gas Rate ^b (mol/d-g)	Elapsed Time (days)	Gas Rate ^b (mol/d-g)	Elapsed Time (days)	Gas Rate ^b (mol/d-g)	Elapsed Time (days)	Gas Rate ^b (mol/d-g)
1 X	150	0.2	406	0	342	0.1	266	0.3
	248	0.3			440	0.2	363	0.4
							461	0.4
0.1 X	391	0	387	0	394	0	189	0
							286	<0.08
							384	0
No Pu	---	---	---	---	359	0.1	---	---

^a1 X = 4×10^6 nCi/g.

^bGas generation rates must be multiplied by 10^{-6} , for example, 0.2×10^{-6} mol/(d-g).

TABLE VI
NUMBER OF
RADIOLYSIS EXPERIMENTS*
AT 1 X

Material	Temperature (°C)			
	20	40	70	100
Cotton	x		x	
Cotton (moist)	x	x	x	
Paper	x	x	x	x
Paper (moist)	x	x	x	
PVC			x	
Lucite			x	
Neoprene			x	
Hypalon			x	
Hypalon (moist)	x			
Polyethylene	x	x	x	x
Composite	x	x	x	x
Composite (moist)		x		

*x = 1 experiment.

TABLE VII
NUMBER OF
RADIOLYSIS EXPERIMENTS*
AT 0.1 X

Material	Temperature (°C)			
	20	40	70	100
Cotton			x	
Paper	x	x	x	x
PVC			x	
Lucite			x	
Neoprene			x	
Hypalon			x	
Polyethylene	x	x	x	x
Composite	x	x	x	x

*x = 1 experiment.

The initial atmosphere in the cylinders is air at local atmospheric pressure [77.2 kPa, (11.2 psi)]

Polyvinyl chloride at a 1 X dose rate had an induction period of 8 to 10 wk and was producing 90+ mol% hydrogen, 4.5 mol% each of CO₂ and CO, and a small amount of CH₄. No chlorine species were detected by mass spectrometry. The CO₂ probably was produced by a reaction involving the oxygen initially present in the test cylinder. This hypothesis is supported by a second gas analysis that showed that no new CO₂ was produced. Bitumen, an additional waste matrix being studied, is discussed in Sec. VI.

2. Temperature Dependence of Radiolysis. In the last progress report,²⁷ we presented data on the effect of increasing temperature on the rate of evolution of gases by radiolysis. Recalculation of experimental data indicates that the initial increase in gas production for celluloses is about 70% for an increase in temperature from 20 to 70°C. At the high dose rates (160 X) we use in these experiments, thermal degradation contributes a negligible amount of gas relative to radiolysis. The magnitude of the increase in gas generation diminishes with time (Table VIII). Table VIII shows that the higher temperature experiment continues to produce more gas even at increased exposure times. Previously, we had

TABLE VIII

EFFECT OF TIME ON TEMPERATURE
DEPENDENCE OF RADIOLYSIS*

Elapsed Time (days)	Gas Evolution Increase for 50°C Temperature Increase (%)
9	70
18	60
27	53
36	48
45	42
54	38
71	29
89	28
116	31
143	31
160	32
188	32

*Celluloses contaminated with ²³⁹PuO₂ to 160 X

hypothesized that the effect might disappear with increasing exposure time if the waste matrix were more rapidly depleted. However, this disappearance has not occurred.

3. Dose Rate Dependency of Radiolysis. Most work on alpha radiolysis has been done at relatively high dose rates. An obvious advantage in using high dose rates is the rapid generation of data. In addition, the importance of other potential gas generation mechanisms (for example, thermal or bacterial degradation) relative to alpha radiolysis is diminished. The assumption usually is made that a dose rate dependency does not exist. Consequently, data from high dose-rate experiments are extrapolated linearly to low dose-rate conditions. However, most hydrogenous wastes have low contamination levels, probably $\leq 4 \times 10^6$ nCi/g waste. (See Sec. VI in Refs. 17 and 12.) In addition, high dose rates for short times have been used to try to simulate the effects of low dose rates for very long times. The validity of such extrapolations has yet to be proved.

G(gas) is the measure of radiolytic gas generation, and it is defined as the number of gas molecules produced per 100 eV of energy from the contaminant. For alpha radiolysis, *G(gas)* should not be considered as an intrinsic property of the material studied because it is a function of the intimacy of contact between the waste matrix and TRU contaminant. Our experiments should produce an upper limit on the radiolytic gas generation potential of the TRU wastes.

Bibler²¹ reports that no dose-rate dependency exists for concretes contaminated to dose rates from 1.4×10^{19} to 1.4×10^{20} eV/d/g waste. Zerwekh²⁰ performed experiments with dose rates ranging from 1×10^6 nCi/g to 15×10^6 nCi/g. His experiments were not directed at studying dose-rate dependency, nor did he comment on any dose-rate effects.

As we continue to collect more data from our experiments, a dose-rate dependency is possible. In these experiments, celluloses are contaminated with either ²³⁹Pu or ²³⁸Pu to dose rates from 4×10^6 nCi/g to 6400×10^6 nCi/g. This represents a plutonium contamination range of 1600 and probably the widest range ever studied for the alpha radiolysis of hydrogenous wastes.

G(gas) decreases with increasing dose (that is, the more energy deposited in the waste matrix, the smaller *G(gas)* becomes). This relationship probably

results from matrix depletion because the half-lives of ^{238}Pu and ^{239}Pu are sufficiently long that the activity levels of the radioisotopes remain essentially constant during an experiment. Zerwekh²⁰ also documents that $G(\text{gas})$ decreases with increasing time (dose).

In Ref. 17 a plot of $G(\text{gas})$ versus elapsed time for experiments with different dose rates was presented. $G(\text{gas})$ was shown to decrease more slowly for lower than for higher dose-rate experiments. This is a significant point because actual TRU waste contamination levels are low; hence, $G(\text{gas})$ will not decrease very rapidly. Consequently, TRU wastes will be sealed in the WIPP while they are still at their maximum gas generation potential, $G(\text{gas})_{\text{initial}}$.

$G(\text{gas})_{\text{initial}}$ can be obtained by extrapolation of plots of $G(\text{gas})$ versus time. A linear plot of $G(\text{gas})_{\text{initial}}$ versus dose rate indicates that $G(\text{gas})_{\text{initial}}$ may be dose-rate dependent (Fig. 3). We feel that the 10 X experiment with $G(\text{gas})_{\text{initial}} = 1$ is not valid because the cylinder valve leaked.

When $G(\text{gas})$ is plotted versus total dose or integrated energy deposited (Fig. 4), two separate groups of curves result. Data points from experiments with dose rates $\leq 40 \text{ X}$ ($160 \times 10^6 \text{ nCi/g}$) describe a curve that extrapolates to a $G(\text{gas})_{\text{initial}}$ of about 2.9. The experiments at 80 X ($320 \times 10^6 \text{ nCi/g}$) and 160 X produce curves that extrapolate to a

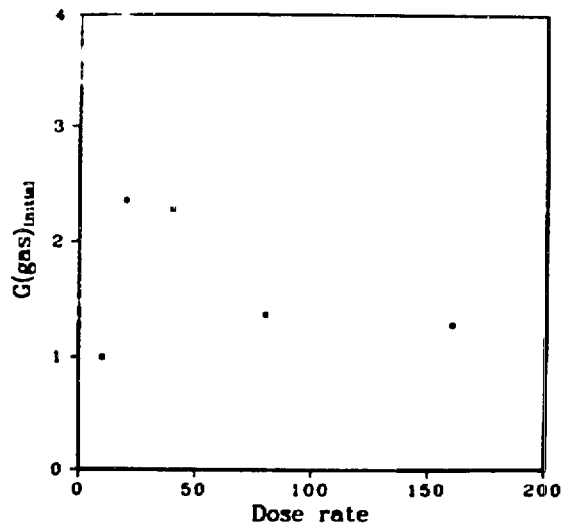


Fig. 3.
 $G(\text{gas})_{\text{initial}}$ vs dose rate

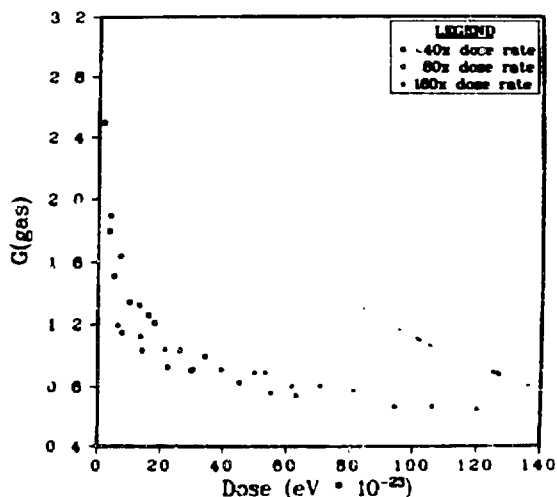


Fig. 4.
 $G(\text{gas})$ vs total dose.

$G(\text{gas})_{\text{initial}}$ of about 1.3. However, there could be a very sharp decrease in $G(\text{gas})$ during high dose-rate experiments for a region where no data points exist.

Experiments using pressure transducers and an initial atmosphere of argon will be initiated during the next quarter to determine more precisely the value of $G(\text{gas})_{\text{initial}}$. The pressure transducers will provide sensitive, continuous monitoring of the evolved gases, and an inert atmosphere will eliminate the effects of oxygen depletion that occurs early in our experiments.

If there is a discontinuity in $G(\text{gas})_{\text{initial}}$ between dose rates of 160 and $320 \times 10^6 \text{ nCi/g}$, we have no explanation for it. Plutonium-238 is used as the contaminant in all experiments with dose rates $\geq 10 \text{ X}$. Hence, the discontinuity is not caused by the use of ^{238}Pu in one experiment and ^{239}Pu in another. In addition, one operator and one batch of $^{239}\text{PuO}_2$ were used for all contaminations. Finally, the low dose-rate curve intersects the high dose-rate curves at a total dose of 40 to $50 \times 10^{20} \text{ eV}$.

4. Composition of Radiolysis Gases. Compositions of radiolytically generated gases are being determined to obtain information on the matrix depletion of cellulose. Evolved gases from all experiments with dose rates of 20 X to 160 X are plotted versus total dose in Fig. 5. An approximate ratio of the evolved gases is $\text{H}_2/\text{CO}_2/\text{CO} = 1.0/0.43/0.22$.

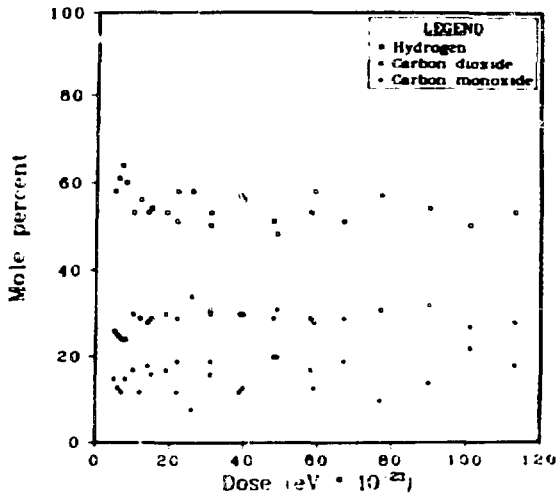


Fig. 5
Gas composition vs total dose

This corresponds to a molecular formula of $C_2H_2O_2$. However, we do not have a complete mass balance because we are not measuring evolved water that may be produced by radiolysis of celluloses. If any trend is observable in Fig. 5, it is a very slight increase in CO_2 and CO at the expense of H_2 . The composition of the evolved gases did not vary with dose rate. In addition, the gas composition is fairly constant for increasing dose, even though gas evolution decreases with increasing dose.

B. High-Pressure Experiments

After the WIPP is filled with wastes and then backfilled with crushed salt, the mine rock salt will creep plastically. When this happens, the overburden can crush the low-level TRU wastes isolated in the repository. This process also will seal the mine, trapping any gases that might evolve from the wastes. The overburden or lithostatic pressure on the TRU waste buried at 650 m (2100 ft) is expected to be about 1.5×10^4 kPa (150 atm). Because the WIPP salt is highly impermeable to gases,²² the gases probably would accumulate and pressurize in any residual void spaces.

We are obtaining information on component effects of the above-described crushing process from

three different types of high-pressure experiments. In the first experiment, radiolytically generated gases are allowed to accumulate and pressurize over the waste matrix to determine whether back reaction with the matrix or cross reaction with the evolved gases will occur. Such reactions, in effect, would decrease the net rate of gas evolution. The plutonium loadings (dose rates) used in these experiments should produce pressures of 1.4 to 2.1×10^4 kPa (200 to 300 psi) in 1 yr. In the second experiment, cylinders containing highly contaminated (160 X dose rate) waste matrices are pressurized to 1×10^4 kPa (1500 psi) with deuterium. These experiments are similar to the first type, but they should provide information more rapidly. A hydrogen isotope is used because hydrogen is one of the major gases evolved from the radiolysis of hydrogenous wastes. In the third experiment, a polyurethane liner is crushed against the waste matrix at WIPP lithostatic pressure. This experiment investigates whether crushing the matrix increases the contact efficiency between it and the plutonium contaminant with a subsequent increase in radiolytic gas production.

1. Self-Pressurization Experiments. Experiments are in progress in which radiolytically evolved gases are allowed to pressurize over waste matrices contaminated with $^{239}PuO_2$ to 160 X (640×10^6 nCi/g). These experiments are compared to others in which the pressures are relieved regularly. The evolved gases in self-pressurized cellulosic experiments thus far have generated about 1000 kPa (140 psi) of pressure. Table IX compares a self-pressurization experiment with one in which the pressure is relieved regularly. The data in this table show that the rate of gas evolution by radiolysis is decreasing as the pressure in the test cylinder is increasing. These results may corroborate those reported by Zerwekh.²³ From these experiments, we will continue to determine whether the trend develops further.

2. High-Pressure Gas Experiments. In two experiments, 1×10^4 kPa (1500 psi) of deuterium were placed over either polyethylene or celluloses contaminated to 160 X (640×10^6 nCi/g) with $^{239}PuO_2$. Although a deuterium pressure of 1×10^4 kPa does not equal WIPP lithostatic pressure, it should be high enough to provide information on the effects of

TABLE IX
COMPARISON OF SELF-PRESSURIZATION EXPERIMENT^a

Self-Pressurized Experiment		Comparison Experiment ^b		Comparison Self-Pressurized Experiment	
Elapsed Time (days)	Pressure (kPa)	Elapsed Time (days)	Pressure (kPa)	Δt (days)	ΔP (kPa)
78	585	78	524		-61
120	745	120	751	42	+6
190	965	190	1020	70	+55
211	1006	211	1110	21	+104

^a Cellulosics contaminated with ²³⁹PuO₂ to 100 N.

^b Pressure is regularly relieved at 95-100 kPa over ambient pressure.

high pressure on gas evolution rates. This deuterium pressure can be achieved conveniently without requiring specialized apparatus to assure safety. Deuterium is distinguished easily from any newly evolved hydrogen. For cellulosics, radiolytically evolved gases typically contain about 60% hydrogen. For polyethylene, hydrogen constitutes 95 + % of the evolved gases. Initially, in both experiments, the rates of hydrogen evolution decreased 50 to 60%. After four months, significant, additional pressure increase could not be detected. The observed decreases may result from

(1) suppression of additional hydrogen formation by the initial high pressure of deuterium.

(2) recombination of deuterium with the matrix accompanied by hydrogen formation to produce a net result of decreased gas generation, or

(3) very small leaks of gas from these relatively highly pressurized cylinders.

It would be remarkably coincidental, however, for both cylinders to leak gas at a rate that would produce roughly the same net decrease in hydrogen evolution.

Analysis of the gases suggest that the second mechanism is probably operative. However, the third mechanism cannot be discounted yet. Next quarter, a known amount of argon will be placed in the cylinders as a tracer. After gas analysis, calculations will be done to determine whether there is a mass balance of the argon in the cylinders.

Bibler¹ has previously allowed gases evolved from the radiolysis of concrete² to accumulate to pressures up to 1.4×10^6 kPa. He states that hydrogen is the only gas produced, that it evolves from the radiolysis of free water in concrete, and that this hydrogen pressure does not change the rate of gas evolution caused by the alpha radiolysis of the concrete. Zerwekh³⁰ reports that a pressure of about 0.7×10^6 kPa lowers the rate of gas generation for cellulosics.

3. WIPP Lithostatic Pressure on Solid Waste.

In this series of experiments, TRU-contaminated waste matrices are crushed at WIPP lithostatic pressure, 1.4×10^6 kPa (2100 psi), to determine whether more gas evolves because of increased contact between the TRU contaminant and the matrix. Initial experiments involved low dose rates (4×10^6 nCi/g), but they were terminated because of experimental difficulties. In these experiments, argon pressure was applied from the bottom of a test cylinder to compress a polyurethane liner against the TRU-contaminated waste at 1.4×10^6 kPa. Argon diffused through the liner, subsequently diluting any radiolytically evolved gases to very low levels. Two high-pressure cylinders were disassembled, their contaminated waste matrices removed, and the experiments reinitiated with much higher plutonium contamination levels.

The new experiments used $^{239}\text{PuO}_2$ to contaminate the matrices to a 160 X dose rate. After 39 days, analyses of the gases in the high-pressure cylinders showed that 25% of the test gases originated from alpha radiolysis. During that time, the internal volumes of the polyurethane liners decreased significantly (probably because of continued crushing of the polyurethane liner and the waste matrix). Such a volume decrease, even if accompanied by no gas evolution, would cause an increase in gas pressure and would appear to indicate that gases were evolving. Consequently, neither evolved gas pressure nor $G(\text{gas})$ can be plotted as a function of time or dose for these experiments to obtain $G(\text{gas})_{\text{initial}}$.

However, an experiment with a cellulose matrix has been sufficiently successful to determine within an order of magnitude the effect of 1.4×10^4 kPa of pressure on the waste matrix. The rate of gas generation was about 70×10^{-7} mol/day/g waste during the first 39 days of the experiment. This is about 30% of the gas generation rate of 220×10^{-7} mol/day/g for a comparison experiment at low pressure. Because of experimental uncertainties, close estimates cannot be made of the gas evolution rate for waste that is crushed at WIPP lithostatic pressure. This series of experiments was terminated because additional significant data probably would not be produced using the current high-pressure cylinder design.

V. SLUDGE DEWATERING

Rates of dewatering at 25, 40, 70, and 100°C were determined using samples of dewatered, LASL-processed sludge [$\text{Fe}(\text{OH})_3$ precipitant] that contains about 30% solids. These experiments were done using a Mettler TA-II thermobalance that was modified for low-temperature work and approximately 1-g sludge samples in a nitrogen flow of 10 cm^3/min . These conditions were chosen to represent the WIPP environment in which the salt in the mine acts as a desiccant. Figure 6 shows that at the highest anticipated WIPP temperature (70°C), the sludge is dewatered in 15 h. The rates of dewatering closely approximate the vapor pressures of water at the various temperatures (Table X). Consequently, rates of dewatering at other temperatures can be extrapolated readily from the data in Fig. 6. Although these experiments do not represent the true condi-

tions surrounding a drum of sludge, the following conclusion can be drawn from them. Once a sludge drum is breached, all the water in it will be absorbed quickly (on a geologic time scale) by the WIPP salt.

We have in progress an experiment that simulates placing sodium chloride in a sludge mine. A bottle with 15-g reagent grade sodium chloride was placed in a closed container of processed sludge. Periodically, the bottle was weighed to determine how much water had been absorbed by the salt. The data are shown in Fig. 7. From day 1 to day 7 the temperature was maintained at 20°C. From day 7 to day 14 the bottle was in an oven at 40°C, and a sharp increase in weight was observed. On day 14 the bottle was returned to 20°C. At 20°C, the rate of water absorption is fairly constant. There is water and undissolved salt standing in the bottle. This experiment is being continued to determine how much water will be absorbed by the salt.

VI. BITUMEN

Bitumen is used by the German Federal Republic and at the Eurochemic facility in Mol, Belgium, for immobilizing radioactive wastes before geologic isolation. In an evaluation of bitumen, ZoBell and Molecke²⁰ discussed microbial degradation and some properties of asphalts. However, little work has been reported on the alpha radiolysis of bitumen. Pretschner and Dagen²¹ worked with bitumen containing ^{210}Po . Using their data, we calculate a value of about 0.2 of $G(\text{gas})$ for their alpha radiolysis. Most of the gas produced was hydrogen.

We have initiated high dose-rate (160 X) experiments in which $^{239}\text{PuO}_2$ was put into molten bitumen. The alpha-radiolysis, gas-generation rate for bitumen is slower than for celluloses. We tentatively estimate $G(\text{gas})$ to be 0.5 to 1.0. Low dose-rate experiments will be initiated next quarter.

Thermogravimetric analyses show that bitumen begins to decompose in air at about 275°C and in nitrogen at about 290°C. The bitumen sample was provided by the WIPP technical program manager. It was manufactured by Industrial Fuel and Asphalt of Indiana, which supplies bitumen to the Werner and Pfleiderer Corp. (WPC) for use in their waste volume reduction (bitumenization) system as used at the Eurochemic facility.

TABLE X

VAPOR PRESSURES OF WATER AT DIFFERENT TEMPERATURES*

Temperature (°C)	Vapor Pressure (mm of Hg)	Time for Complete Dewatering (h)	Ratio of VP ^b at T ^c to VP at 100°C	Ratio of Times to 100°C
25	24	36.0	31.7	32.7
40	55	16.0	13.8	14.5
70	234	3.8	3.2	3.4
100	760	1.1	1.0	1.0

*See Ref. 23

^bVP = vapor pressure in 2nd column

^cT = temperature in 1st column

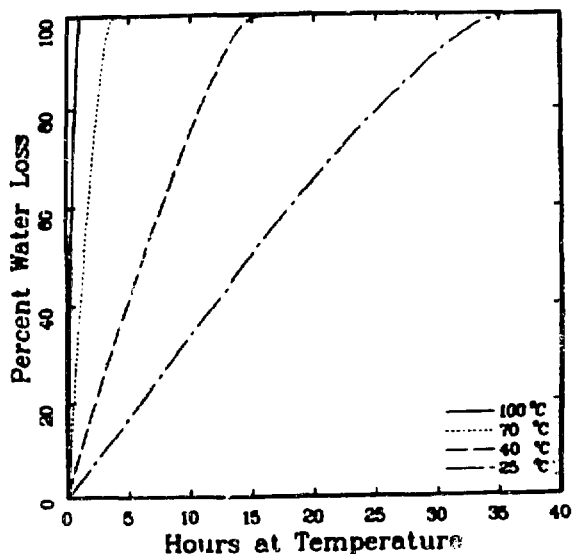


Fig. 6.
Dewatering of sludge.

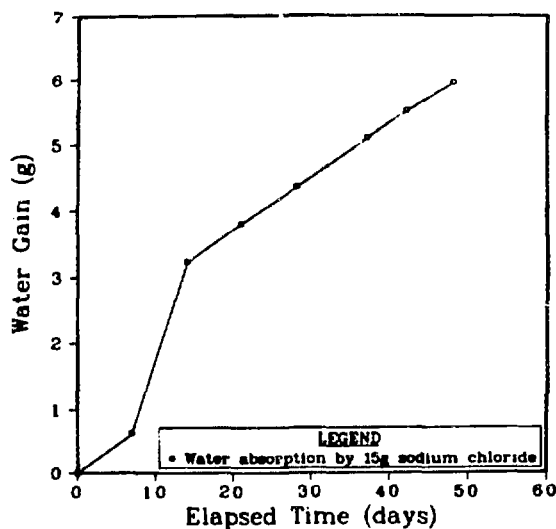


Fig. 7.
Water absorption by salt.

VII. ASSESSMENT AND CHARACTERIZATION OF LASL TRU WASTE INVENTORY

In 1970, the Atomic Energy Commission (AEC) directed that any solid wastes generated at AEC (now DOE) installations that were contaminated with ²³⁵U, its daughters, or any long-lived TRU isotopes should be segregated from other routine wastes and stored so that they could be readily

retrieved in an intact and surface-uncontaminated condition for at least 20 yr.²⁶ This directive was modified later to cover only those wastes contaminated to a level greater than 10 nCi/g of TRU waste.³ Wastes contaminated with TRU isotopes but below the 10 nCi/g level are to be handled as noncontaminated wastes.

LASL began compliance with the AEC directive in late 1970,²⁷ and since that time, stored wastes

have been accumulating on site awaiting final disposal at WIPP, which currently is in the developmental stage. To aid in the planning of WIPP, a study was undertaken to characterize in detail the total inventory of retrievably stored, solid, TRU-contaminated wastes that had accumulated at LASL until the beginning of CY 1978.

At LASL, the Health Research Division's Waste Management Group is responsible for the retrievable storage of all TRU wastes generated on site and for the acquisition and storage of data pertaining to these wastes. All records are entered and stored in a computer data base for ease of access and retrieval, and this data base is the source for practically all of the information on TRU wastes presented here. It should be remembered that the accuracy of all reported figures can be only as good as that of the data supplied to the Waste Management Group by the original waste generator, and that the extent to which all of these data are of uniform quality is unknown. Additionally, waste stream compositions and amounts of waste being generated are continually varying in response to changing programs, improved waste management techniques, etc., with the result that the information here is applicable only to those wastes stored before CY 1978 and it cannot be extrapolated indiscriminately for future waste additions to the inventory.

By January 1, 1978, there were 1610 m³ of TRU waste in retrievable storage at LASL, which is approximately 3.5 vol% of the national DOE inventory.²⁰ Figure 8 illustrates the yearly rate of TRU waste production, which from 1970 through 1977 averaged 230 m³/yr. This rate, however, is highly variable, and generally there has been a continuous trend toward larger waste volumes produced each year. It is expected that the final figure for 1978 will slightly exceed 375 m³, and 1979 and 1980 should show large increases in retrievably stored waste volumes, largely attributable to planned decontamination and decommissioning (D/D) activities. Much of this increased amount will be equipment and noncombustible building debris. Figure 8 would also seem to indicate that there were almost no combustibles (which currently constitute 21 vol% of the entire LASL inventory) among the stored wastes from 1971 through 1973. This absence is due to misunderstandings related to waste classification and segregation rather than to a near total lack of combustibles in the waste stream.

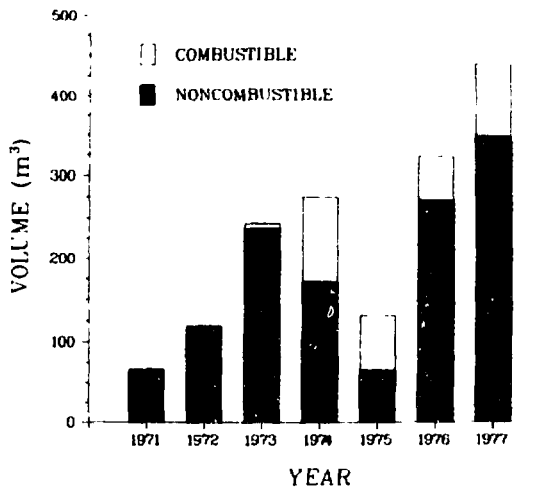
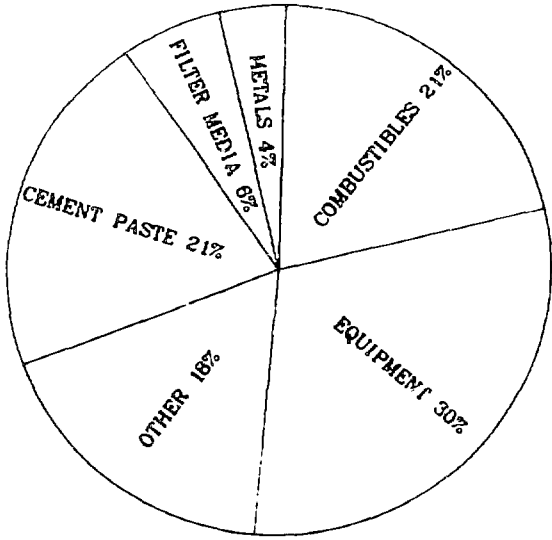


Fig. 8.
Generation rates of retrievably stored TRU waste at LASL.

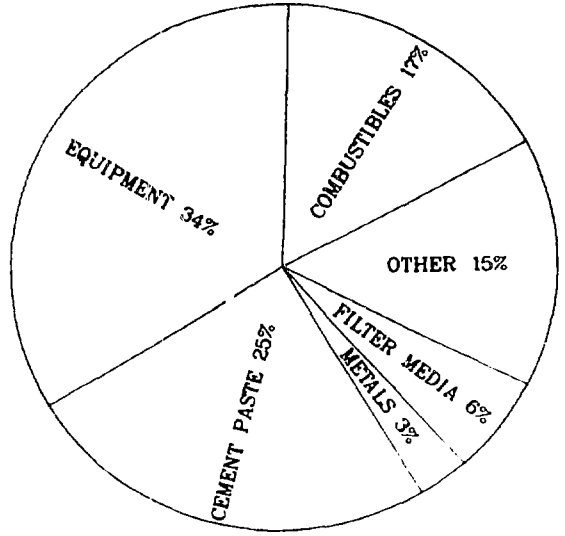
The 27 categories of TRU waste that were in retrievable storage at LASL by the end of CY 1977 are given in Table XI with the individual LASL waste code (WC) numbers, which will be used in the text and figures. The data have been separated into gross values for use in handling, storage, and transportation planning and into net values for use in experimental studies.

Waste volumes presented in Fig. 8 are quite accurate, whereas waste weights are not as well known because only 75% of the containers in storage are associated with gross weight data in the TRU waste data base. We have treated the volume data here as if the primary container contributes no excess to the total volume of the waste, (that is, we assume fill fractions equal 1.0). The difference between listed values for gross and net volumes is due solely to the cement casks, which are secondary-type containers (see below).

Ten of the 27 waste types account for over 90% of the inventory with cement paste, combustibles, and equipment constituting almost 75% of the total waste backlog (Fig. 9a and b). The same three waste types also constitute most of the total weight of the TRU inventory with cement paste in largest proportion by far (Fig. 10a and b). The percentage of cement paste in the TRU backlog will decline in the future because this waste type will not be generated after 1979.

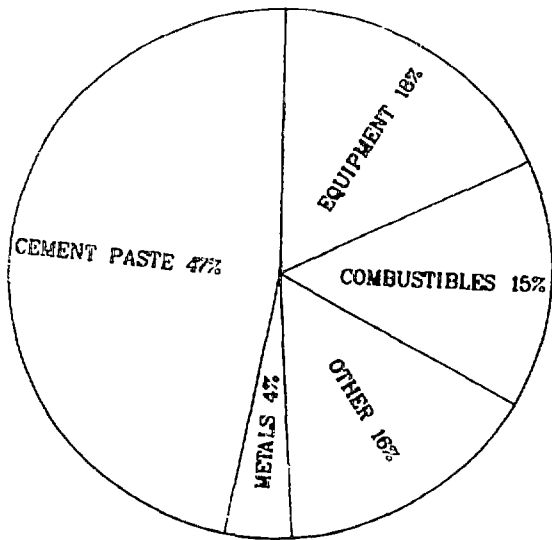


(a) Gross volume.

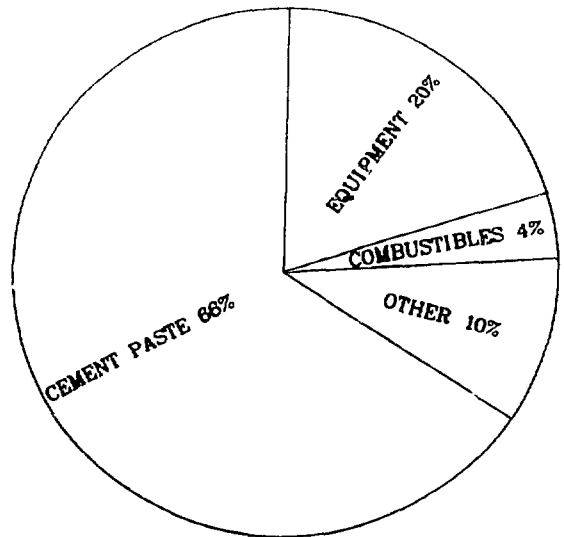


(b) Net volume.

Fig. 9.
Composition of LASL TRU waste inventory.



(a) Gross weight.



(b) Net weight.

Fig. 10.
Composition of LASL TRU waste inventory.

Table XI shows that cement casks have increased the weight of several types of waste (for example, WC 17, 18, 19, and 52) by as much as 1000 to 2000% and the excess volume contributed by the casks has increased gross volumes of some wastes from 200 to 500%. Containers of all types contribute one-third of the entire weight of the waste backlog.

Combustible wastes could present a short-term fire hazard during the operational phase of a waste repository. Additionally, wastes that evolve gases could pressurize and conceivably rupture a waste facility after the operational phase had ceased and all entrances had been sealed. This could lead to the return of radionuclides to the biosphere. Table XI gives an indication of the large number of waste types that theoretically could present a fire hazard or that could, under certain conditions, evolve gases. Possible mechanisms for this gas production include thermal degradation of combustible (and possibly other) wastes, corrosion of metals, and chemical and/or radiolytic attack of a wide range of waste matrices. Additionally, all containers used at LASL have either the potential for combustion (plywood crates) or for gas generation (metal containers and cement casks).

Densities for the various TRU waste types present in LASL storage are illustrated in Fig. 11 a and b. The average gross densities for most materials lie in the 250- to 400-kg/m³ range, whereas net densities occur most frequently between 125 and 275 kg/m³. Sludges, cement paste, and oils are the only wastes that radically depart from these ranges, with cement paste being the most dense material in the TRU inventory at 1600 kg/m³. The inventory averages for both gross and net densities are considerably higher than the averages listed above because of the large fraction of very dense cement present.

The overall density for all TRU wastes and containers is 770 kg/m³ (approximately one-third that of natural salt), which contrasts with a density of 546 kg/m³ (34 lbs/ft³) for all TRU wastes at Idaho National Engineering Laboratory (INEL).⁹ This difference probably does not result from more efficient packing of waste containers at LASL because reported densities for analogous wastes at INEL and LASL are quite similar. The difference probably is caused by the considerable fraction of cement present at LASL, both as a waste form and as a container type, which is largely absent at INEL.

All TRU wastes destined for retrievable storage at LASL have in the past been placed in one of five

types of containers (Table XII). Of these five types, the cement cask is the only container that is not a primary type, and it is the only one assumed here to contribute excess volume to the waste. Materials contaminated with ²³⁵U and ²³⁹Pu are first placed in 110-l drums, then the drums are sealed, in pairs, in the concrete casks. The 110-l drums have been used without secondary containment for waste storage, but now their use is restricted to cask storage. Because corrugated metal pipe (CMP) is used exclusively for the disposal of cement paste, its use will be discontinued when retrievable cement paste production is halted in 1979.

The volume and tare weights of the fiber-glass-reinforced plywood (FRP) crate listed in Table XII are those for a standard 1.22- by 1.22- by 2.13-m (4- by 4- by 7-ft) Rocky Flats FRP crate. Actually, only about 20% of all FRP crates in storage are of this type. There are more than twice as many that have dimensions of 1.22- by 1.22- by 2.44-m (4- by 4- by 8-ft). The volumes of the rest of the crates range from 0.5 to 49.8 m³, with the inventory average being 4.4 m³. Approximately 95% of all FRP crates in storage at the beginning of CY 1978 were filled with equipment and filter media, with equipment predominating.

Numbers of containers and their average weights for each waste type are presented in Table XIII. The average weights listed for FRP crates in Tables XII and XIII should be regarded somewhat skeptically, because only 14% of the crates in storage have an assigned estimated weight in the TRU waste data base.

More than 95% of all drums in storage weigh <100 kg, with the average for the 210-l drums being approximately 79 kg. This last figure is roughly 50% of the average value of 150 kg reported for drums stored at INEL.⁹ This difference probably is due to the large fraction of very heavy drums (180 to 275 kg) at INEL filled with Rocky Flats sludge, a waste type that is only a minor fraction of the inventory at LASL. However, amounts of sludge in storage should increase substantially at LASL in the future.²⁰ The maximum weights of 210-l drums, slightly over 300 kg, are similar at both installations.

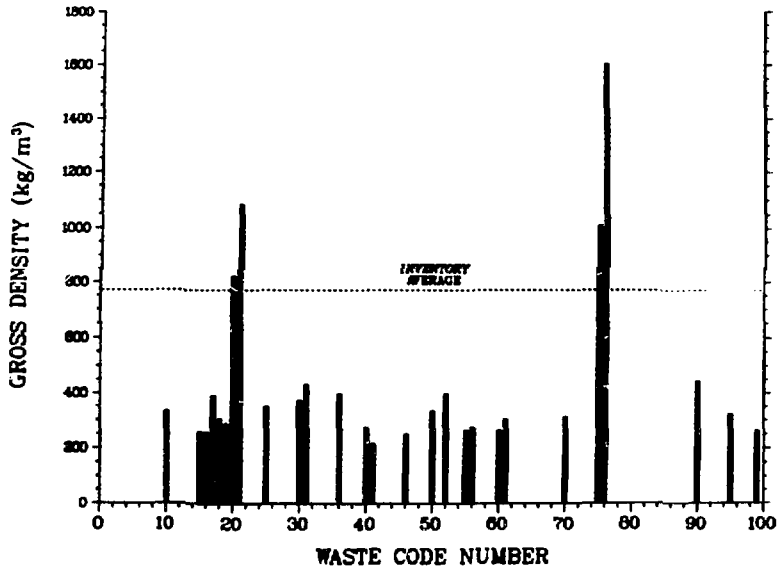
There is considerably more variation in the weights of FRP crates than of other container types because many of the crates have very different volumes and contain wastes of various densities. The heaviest items in the LASL TRU waste backlog are the 117 CMPs and two or three FRP crates²⁰

TABLE XI

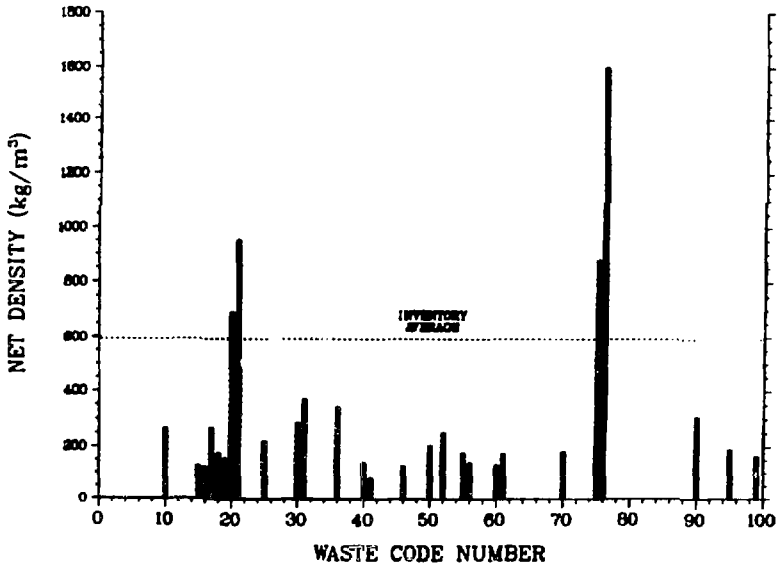
MATERIALS PRESENT IN LASL RETRIEVABLY STORED TRU WASTE

Waste Code	Description	Gross Volume (m ³)	Net Volume ^a (m ³)	Gross Weight (kg)	Net Weight ^b (kg)	Combustible	Gas Generation Potential ^c
10	Graphite	0.21	0.21	70	56	No	?
15	Mixed cellulose	70.96	62.26	27,410	7870	Yes	Yes
16	Plastic materials	7.18	5.44	3660	670	Yes	Yes
17	Rubber materials	16.50	3.45	18,370	930	Yes	Yes
18	Mixed paper, plastic, rubber, etc.	77.58	36.69	64,540	6430	Yes	Yes
19	Mixed combustible/noncombustible	57.90	25.71	49,370	4080	Yes	Yes
20	Hydrocarbon oils	1.47	1.47	1210	1010	Yes	Yes
21	Silicon-based oils	0.74	0.74	800	700	No	?
25	Leached process residues	52.82	52.82	18,540	11,650	No	Yes
30	Property-numbered equipment	110.50	100.49	51,300	28,860	No	Yes
31	Nonproperty-numbered equipment	380.42	377.82	166,620	139,990	No	Yes
36	Noncombustible building debris	31.70	31.70	12,680	10,880	No	?
40	Combustible hot-cell waste	2.94	2.94	810	420	Yes	Yes
41	Noncombustible hot-cell waste	4.06	3.19	1820	270	No	?
46	Skull and oxide	0.11	0.11	28	14	No	?
50	Metal crucibles, scrap, dies	36.18	33.57	14,690	6840	No	Yes
52	Other scrap metals	34.11	11.49	34,100	2900	No	Yes
55	Filter media	94.97	88.02	32,410	15,530	No	No
56	Filter media residue	0.63	0.63	170	90	No	?
60	Other combustibles	96.03	95.16	26,260	12,440	Yes	Yes
61	Other noncombustibles	167.84	96.50	122,410	16,710	No	?
70	Chemical waste	1.06	1.06	330	190	No	Yes
75	Chemical treatment sludge	9.66	9.66	9730	8440	No	Yes
76	Cement paste	344.19	344.19	577,850	548,490	No	Yes
90	Contaminated soil	1.47	1.47	650	450	No	No
		3.53	2.66	1990	500	No	No
95	Glass	4.90	4.90	1270	810	?	?
99	Unidentified material						
		1610	1394	1,239,090	827,240		

^aInternal volume of primary waste containers^bGross weight minus container weights^cThe potential of a waste to generate gas because of chemical, thermal, corrosive, or radiolytic degradation mechanisms



(a) Gross densities are for waste plus primary container (neglects cement casks).



(b) Net densities.

Fig. 11.
Densities of TRU wastes in retrievable storage at LASL.

TABLE XII
WASTE CONTAINER DATA

<u>Container Type</u>	<u>External Internal Volume (m³)</u>	<u>Tare Weight (kg)</u>	<u>Total Number</u>	<u>Vol% Inventory</u>	<u>Wt% Inventory</u>	<u>Average Gross Weight (kg)</u>	<u>Maximum Gross Weight (kg)</u>	<u>Maximum Surface Radiation (mrem/h)</u>
210-L drum (55 gallon)	<u>0.21</u> 0.21	28	1797	24	11	79	336	250
110-L drum ^a (30 gallon)	<u>0.11</u> 0.11	14	520	4	1	40	134	80
CMP ^b	<u>2.8</u> 2.8	232	117	20	45	4760	4768	Unk (<250)
FRP crate	<u>3.17</u> 3.17 (Variable)	273 (Variable)	129	36	18	1315	5000 ^c	70
Cement cask ^d	<u>1.10</u> 0.22	1163 ^e	248	17	24	1208	1320	Unk (<250)

^aNo longer used as a separate container.

^bUse will be discontinued in 1979.

^cEstimated. See text for explanation.

^dSecondary container only—contains two 110-L drums.

^eIncludes the weight of two 110-L drums.

TABLE XIII

NUMBERS AND AVERAGE WEIGHTS OF WASTE CONTAINERS IN STORAGE

Waste Code	Number of Containers						Average Gross Weight (kg)					Average Net Weight (kg)					
	210-l Drum	110-l Drum	Cask	CMP	FRP	Total	210-l Drum	110-l Drum	Cask	CMP	FRP ^a	210-l Drum	110-l Drum	Cask	CMP	FRP ^a	
10	1					1											
15	219	127	10			356	54	28	1194			26	14	28			
16	19	9	2			30	54	28	1194			26	14	27			
17			15			15			1225					59			
18	118	10	47			175	64	34	1205			37	19	35			
19	62	38	37			137	60	32	1201			33	17	35			
20	7					7	173					145					
21	3	1				4	227					199					
25	130	232				362	74	39				46	24				
30	94	11	12		19	136	79	41	1221		1515	60	32	63			1160
31	1	1	3		77	82			1234		2114			82			1816
36					6	6					2113						1812
40	14					14	58					30					
41	12	4	1			17	45	24				17	9				
46		1				1											
50	133	45	3			181	71	37	1212			43	22	45			
52	23	1	26		1	51	84		1227			53		55			
55	2	1	8		25	36	56		1196		908	37		39			603
56	3					3	58					30					
60	451	2	1			454	55	29				28	14				
61	354	30	82			466	64	33	1205			36	19	38			
70	4	"				6	66	34				38	20				
75	46					46	211					184					
76	79			117		196	234				4760	206				4528	
90	7					7	93					65					
95	10	3	1			14	67	35				40	21				
99	5	2			1	8	54	29				34	18				
	1797	520	248	117	129	2811											

^aEstimated. See text for explanation.

(which are much larger in volume), each weighing almost 5000 kg (5 metric tonnes).

Table XIV is a listing of all radionuclides recorded in the TRU waste inventory before the beginning of CY 1978. All isotopic data presented here is uncorrected for decay, and all curie numbers are for alpha particle radiation exclusively; for example, the beta contribution of ^{241}Pu has been neglected. Approximately 1400 (35%) of all radionuclide occurrences are in trace (nonaccountable) amounts, <0.1 g of plutonium and only suspected or trace quantities of others, such as ^{235}U and MFP.

The total curie content and heat production rate for the waste backlog are due mainly to ^{239}Pu , ^{241}Am , and ^{238}Pu , in that order. Technically, ^{238}Pu is not a TRU isotope, but most of the ^{238}Pu in storage at LASL is actually heat-source-grade plutonium that contains sufficient ^{239}Pu (~16%) to warrant treating ^{238}Pu as a TRU waste. At present, ^{238}Pu accounts for

approximately 90% of the total curie content and heat production in the waste backlog, but because of its relatively short half life ($t_{1/2} = 86.4$ yr), ^{241}Am will become the dominant TRU nuclide within 350 yr, and ^{239}Pu will become the most abundant TRU isotope within 1000 yr. The average heat production rate of 1.6 W/m³ is calculated by dividing the total heat content of 2602 W (Table XIV) by the total volume of waste, 1610 m³ (Table XI).

Table XV lists TRU isotope amounts and contamination levels by WC. The leached process residues (WC 25) are some of the most important wastes in the entire inventory in terms of plutonium content. These residues, which constitute only 3% of the total backlog volume, contain over 55% of the ^{239}Pu and approximately 45% of the weight of all plutonium in storage. Cement paste (WC 76) contains virtually all of the ^{241}Am , ^{235}U , and ^{238}U . The noncombustible material category (WC 61) contains

TABLE XIV
RADIONUCLIDE DATA FOR LASL TRU WASTE INVENTORY^a

TRU Isotope	Number Occurrences	Total Grams	Ci/g	Total Curies ^b	W/g	Total Watts
^{235}U	38	4,302	9.5×10^{-3}	41	2.7×10^{-4}	1
^{237}Np	2	3	7.1×10^{-4}	...	2.0×10^{-5}	...
$^{239}\text{Pu}^c$	2454	18,735	7.8×10^{-2}	1454	2.4×10^{-3}	45
^{241}Am	218	2,195	3.24	7112	1.1×10^{-1}	241
^{242}Cm	1	3.1×10^{-7}	3.3×10^3	...	1.2×10^2	...
^{244}Cm	1	1.2×10^{-5}	8.3×10^1	...	2.84	...
^{248}Bk	1	6.0×10^{-7}	1.7×10^3	...	1.24	...
^{250}Cf	1	2.5×10^{-4}	4.0	...	1.4×10^{-1}	...
Non-TRU isotope						
^{227}Ac	1	6×10^{-3}	7.3×10^1
^{227}Th	1	...	1.1×10^{-7}
^{230}U	307	9,761	2.1×10^{-6}	...	5.5×10^{-6}	...
^{238}U	27	4,354	3.3×10^{-7}	...	8.0×10^{-6}	...
$^{238}\text{Pu}^d$	921	4,832	1.47×10^1	71,030	4.8×10^{-1}	2315
MFP ^e	109
				<u>79,637</u>		<u>2602^f</u>

^aData uncorrected for decay.

^b α radiation only.

^c ^{239}Pu is weapons grade.

^d ^{238}Pu is heat-source grade.

^eMFP is mixed fission products.

^fThe total volume of waste is 1610 m³; the average heat generation is 1.6 W/m³.

TABLE XV

TRU ISOTOPE AMOUNTS AND CONTAMINATION LEVELS FOR LAST WASTE INVENTORY

Waste Cede	Total Grams Contaminant				Average Contamination Level (g nuclide/g waste)			
	²³⁸ Pu	²⁴¹ Am	²⁴⁰ Pu	Other	²³⁸ Pu	²⁴¹ Am	²⁴⁰ Pu	Other
10	8.7				1.6 x 10 ⁻¹			
15	804.32		234.7		1.6 x 10 ⁻¹		3.0 x 10 ⁻¹	
16	102.78		105.4		1.5 x 10 ⁻¹		1.6 x 10 ⁻¹	
17			120.1				1.3 x 10 ⁻¹	
18	302.75		593.03		4.7 x 10 ⁻²		9.3 x 10 ⁻²	
19	217.34		1314.3		5.3 x 10 ⁻²		3.2 x 10 ⁻¹	
20	1.31				1.3 x 10 ⁻²			
21	19.95		0.004		2.8 x 10 ⁻²		5.7 x 10 ⁻²	
25	10,514.5	0.15	0.3		9.0 x 10 ⁻¹	1.3 x 10 ⁻²	2.6 x 10 ⁻²	
30	107.6		76.8		3.7 x 10 ⁻²		2.7 x 10 ⁻²	
31	492		45.4		3.5 x 10 ⁻²		3.2 x 10 ⁻²	
36								
40								
41				0.006 ^a				2.3 x 10 ⁻²
46	9.4			9.0 ^b	6.7 x 10 ⁻¹			6.4 x 10 ⁻¹
50	1331.2		154.7		1.9 x 10 ⁻¹		2.3 x 10 ⁻²	
52	276.35		526.4		9.5 x 10 ⁻²		1.8 x 10 ⁻¹	
55	0.55		113.5		3.5 x 10 ⁻²		7.3 x 10 ⁻²	
56	5.62				6.3 x 10 ⁻²			
60	1265.01		13		1.0 x 10 ⁻¹		1.0 x 10 ⁻²	
61	2399.5	7.32	1520.5	4305 ^c	1.4 x 10 ⁻¹	4.4 x 10 ⁻²	9.1 x 10 ⁻²	
70	58.43				3.0 x 10 ⁻²			2.6 x 10 ⁻¹
75	3.808	0.006	0.063		4.5 x 10 ⁻²	7.1 x 10 ⁻²	7.5 x 10 ⁻²	
76	766.43	2187	1.76	14,106 ^d	1.4 x 10 ⁻¹	4.0 x 10 ⁻²	3.2 x 10 ⁻²	2.6 x 10 ⁻²
90	0.554				1.2 x 10 ⁻²			
95	46.51		6.9		9.2 x 10 ⁻²		1.4 x 10 ⁻²	
99	0.45			0.26 ^e	5.6 x 10 ⁻²			3.2 x 10 ⁻²
	18735	2195	4832					

^a ²²⁷Ac.

^b ²³⁵U.

^c ²³⁵U plus ²³⁷Np (3.0 g)

^d ²³⁵U plus ²³⁸U (4354 g)

^e ²³⁷Np.

all of the ^{238}U and about one-third of the total amount of ^{239}Pu . Combustible materials are contaminated with 50% of the ^{239}Pu and 22% of the total amount of plutonium in LASL TRU wastes.

Average, as well as maximum, contamination levels for the various waste materials vary by more than 5 orders of magnitude as shown in Fig. 12a and b. The average ^{239}Pu levels of several of the more highly contaminated waste types ranged between 1×10^6 and 5×10^6 nCi/g waste, whereas ^{239}Pu contamination levels were almost 2 orders of magnitude lower, ranging from 1×10^6 to 8×10^6 nCi/g waste.* Many of the waste types in these high-contamination ranges are materials that have a potential for radiolytic gas generation, such as WC 15, 16, 17, 18, and 19.

It was possible to determine radionuclide concentrations only for those containers that had a recorded gross weight (~75% of all containers). Consequently, some containers may have maximum levels of contamination higher than those shown in Fig. 12b. In addition, these maximum levels have been calculated by dividing the total amount of nuclide by the total net weight of each container. This would imply a uniform isotope distribution throughout the waste matrix. It probably would be more realistic to assume that the TRU elements are concentrated in only a small fraction of the total waste volume, possibly increasing the contamination levels by as much as an order of magnitude. Highest recorded plutonium concentrations are in the range of 1×10^7 to 3×10^7 nCi/g waste for ^{239}Pu and 10^6 to 10^8 nCi/g waste for ^{240}Pu .

Radiation dose rates at the surface of the primary container are given in Table XVI. As shown, 90% of all containers have a dose rate ≤ 1 mrem/h. Many of the 54 containers that measured at >10 mrem/h subsequently were placed in concrete casks, and consequently the dose rate for these wastes was considerably lowered. There were only five containers in the TRU waste inventory that exceeded the contact-handled limit of 200 mrem/h, and two of these were placed in concrete casks for shielding. CMPs are filled below grade with cement paste. Consequently, no surface dose rates have been measured for these containers but expected rates should be minimal because of dilution of ^{241}Am in a large volume of cement paste with consequent self-shielding. The highest known dose rate for an unshielded container

is 250 mrem/h for a 210-l drum of ^{241}Am contaminated waste from Lovelace Clinic, Albuquerque, sent to LASL for storage.

Certain TRU wastes currently in storage might have to be reprocessed before they can satisfy WIPP waste acceptance criteria when they are formalized. A plethora of strategies has been proposed for reprocessing these wastes. For instance, combustible materials could be greatly reduced in volume and rendered essentially inert by incineration in the controlled air incinerator (CAI) and then by immobilization of the ashes in one of a number of proposed waste matrices. Additionally, electro-polishing of equipment and scrap metals to remove surface contamination is also under consideration. This process might eliminate the necessity of having to dispose of large volumes of bulky equipment in an expensive waste isolation facility. However, there are certain waste forms in the LASL inventory that would be very difficult to reprocess and would probably have to be placed in the WIPP in an as-is condition. Cement paste stored in CMPs is the prime example of this type of waste form.

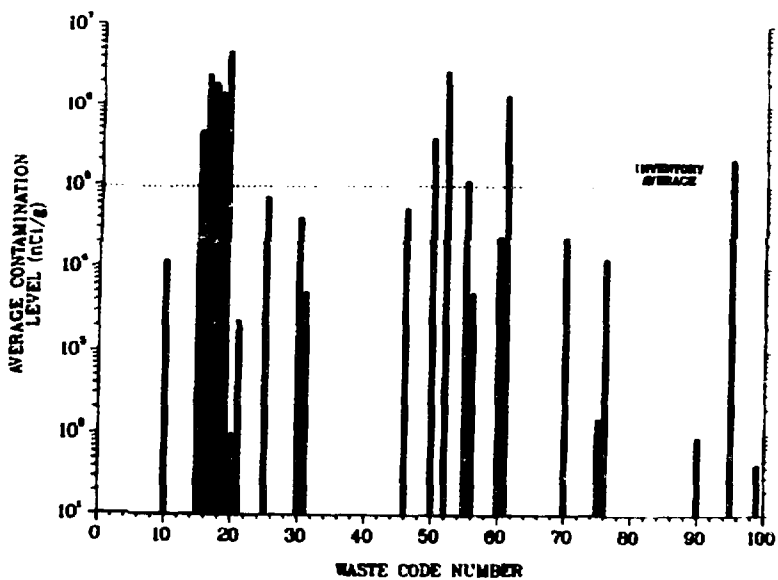
Published data for TRU wastes in storage at INEL² (which constitute about 75 vol% of the national DOE inventory) show that many of the physical and radionuclide characteristics of INEL and LASL wastes are roughly comparable. However, the plutonium loading of the LASL inventory is approximately 60 vol% greater than that at INEL, and the LASL combustible fraction is generally much more highly contaminated than comparable wastes at INEL. Additionally, the TRU backlog at LASL contains cement casks, oversized crates, and very heavy CMPs; all of which are absent at INEL. These differences might necessitate different planning, transportation, handling, and reprocessing strategies for LASL wastes than for those stored at INEL and other installations.

This is the final report in the LASL TRU waste assessment and characterization series.

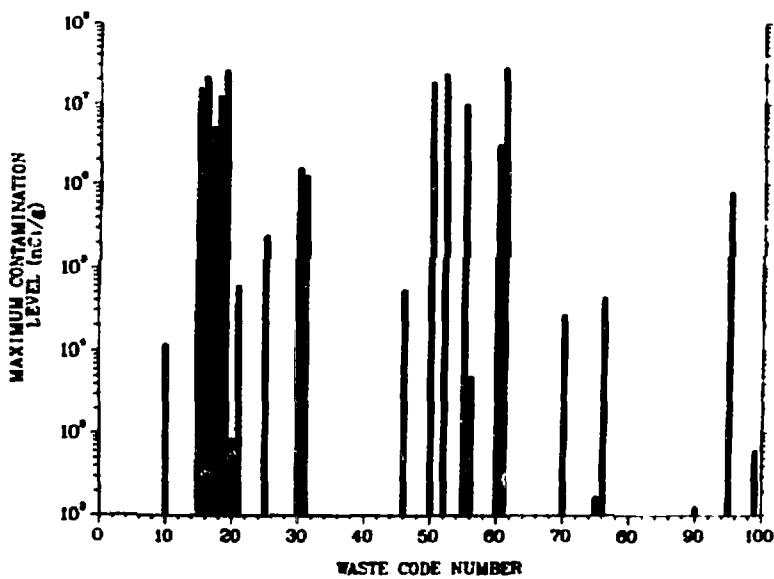
VIII. MILESTONES OF THE PROGRAM

Major milestones in the program continue to be achieved on schedule (Table XVII). By mutual agreement with the WIPP technical program manager, we continue to maintain program flexibility. This ensures timely response to the evolving needs of the WIPP experimental waste characterization program.

*For a detailed listing of contamination levels by nuclide, see Ref. 17.



(a) Average levels.



(b) Maximum levels.

Fig. 12.
Contamination levels of TRU wastes in retrievable storage at LASI.

TABLE XVI

CONTAINER SURFACE DOSE RATE

Surface Radiation* (mrem/h)	No. of Containers
<1	1218
1	1316
2	107
3	31
4	23
5	18
6	16
7	9
8	9
9	4
10	8
<hr/>	
	98%
11-50	12
51-100	17
101-250	5

*Uncorrected for decay. Radiation values were taken at the surface of the primary container. Many of the containers (7-10 meters) subsequently were placed in concrete casks, and the quantity of external radiation for these containers is much less.

IX. WORK PLANNED FOR NEXT QUARTER

The following work is planned for next quarter.

- Calibrate pressure transducers.
- Duplicate experiments using pressure transducers to determine more accurately whether a dose-rate dependency exists.
- Initiate low dose-rate experiments with bitumen.
- Initiate additional experiments with processed sludge and cement paste.

ACKNOWLEDGMENTS

The authors thank John Warren and Cynthia Murphy of LASL Group H-7 for their assistance in the TRU waste inventory assessment. We also thank Pamela Petersen, Nancy Shera, and Linda Willis for their assistance during preparation and assembly of the radiolysis experiments and during data collection and reduction.

TABLE XVII

MILESTONE CHART FOR S-124 (WIPP R&D PROGRAM)*

Quarter	FY 1978				FY 1979				FY 1980			
	1	2	3	4	1	2	3	4	1	2	3	4
Quarterly reports	XXXXXXXXXXXXXXXXXXXX								0	0	0	0
Literature search and update	XXXXXXXXXXXXXXXXXXXX								0	0	0	0
Computer literature search					XXXX							
Determine threshold decomposition temperatures	XXXXX											
Thermal degradation (catalyzed and noncatalyzed)												
1. Initiate experiments	XXXXXXXXXX											
2. Collection and interpretation of available data					XXXXXXXXXXXXXXXXXXXX				0	0	0	0
3. Termination and final conclusions												0
Radiolytic degradation (low-pressure)												
1. Initiate experiments	XXXXXXXXXXXXXXXXXXXX											
2. Collection and interpretation of available data					XXXXXXXXXXXXXXXXXXXX				0	0	0	0
3. Termination and final conclusions												0
Dose-rate dependency (low-pressure)												
1. Initiate experiments					XXXXXX							
2. Collection and interpretation of available data					XXXXXXXXXXXXXXXXXXXX				0	0	0	0
3. Termination and final conclusions												0
Radiolytic degradation (high-pressure)												
1. Design, fabricate cylinders	XXXXX											
2. Initiate experiments					XXXXXXXXXXXX							
3. Collection and interpretation of available data					XXXXXXXXXX				0	0	0	0
4. Termination and final conclusions												

Quarter	FY 1978				FY 1979				FY 1980			
	1	2	3	4	1	2	3	4	1	2	3	4
Rate of sludge dewatering												
1. Initiate experiments			xxx									
2. Collection and interpretation of available data			xxx									
3. Termination and final conclusions			xxx									
Rate of cement paste dewatering												
1. Initiate experiments							xxx					
2. Collection and interpretation of available data							xxx					
3. Termination and final conclusions							xxx					
Interface with Sandia rock mechanics laboratory permeability experiments												
							xxx					
LASL retrievable TRU waste inventory assessment												
1. Definition of goals			xxxxx									
2. Acquisition and compilation of data bases			xxxxxxxxxxxxx									
3. Interpretation and discussion				xxxx								
4. Termination and final conclusions							xxxxx					
Bitumen experiments												
1. Determine threshold decomposition temperatures							xxx					
2. Radiolytic degradation (low-pressure)							xxx	o				
a. Initiate experiments							xxx	o	o	o	o	
b. Collection and interpretation												
c. Termination and final conclusions												o
Pressure transducer equipment												
1. Order							xxx					
2. Introduce into radiolysis experiments							o					
Interim summary report to DOE HQ (Waste Acceptance Criteria Steering Committee)												
							xxx					
Final conclusions and recommendations												
												o

*X = Completed as scheduled.
o = Planned.

REFERENCES

1. N. E. Bibler, "Radiolytic Gas Production During Long-Term Storage of Nuclear Wastes," Savannah River Laboratory report DP-MS-76-51 (1976).
2. USAEC Manual Chapter 0511, Appendix B. USAEC, September 19, 1973.
3. Henry C. Shefelbine, "Preliminary Evaluation of the Characteristics of Defense Transuranic Wastes," Sandia Laboratories, Albuquerque, report SAND 78-1850 (1978).
4. M. A. Molecke, "Waste Isolation Pilot Plant Transuranic Wastes Experimental Characterization Program: Executive Summary," Sandia Laboratories report SAND 78-1356 (1978).
5. G. R. Waterbury and Al Zerwekh, "Transuranic Waste Research and Development Program (A412) Annual Report for the Period January 1 to December 31, 1976," Los Alamos Scientific Laboratory internal document, February 1977.
6. Ralph H. Hansen, "Thermal and Oxidative Degradation of Polyethylene, Polypropylene, and Related Olefin Polymers," in **Thermal Stability of Polymers I**, R. T. Conley, Ed. (Marcel Dekker, Inc., New York, 1970), Chap. 6, pp. 153-187.
7. R. F. Schwenker, Jr., and L. R. Beck, Jr., "Study of the Pyrolytic Decomposition of Cellulose by Gas Chromatography," *J. Polym. Sci. C2*, 331 (1963).
8. R. T. Conley and R. Malloy, "Vinyl and Vinylidene Polymers," in **Thermal Stability of Polymers I**, R. T. Conley, Ed. (Marcel Dekker, Inc., New York, 1970), Chap. 8, pp. 223-285.
9. E. J. Murphy, "Thermal Decomposition of Natural Cellulose in Vacuo," *J. Polym. Sci.* **58**, 649 (1962).
10. S. Kosiewicz, B. Barraclough, A. Zerwekh, and Benjamin J. Barnhart, "Studies of Transuranic Waste Storage Under Conditions Expected in the Waste Isolation Pilot Project (WIPP) July 1-September 30, 1978," Los Alamos Scientific Laboratory report LA-7582-PR (January 1979).
11. A. Zerwekh, S. Kosiewicz, and B. Barraclough, "Experimental Studies of the Degradation of RAD-Wastes for the Sandia Laboratories Waste Isolation Pilot Project (WIPP) April 1-June 30, 1978," Los Alamos Scientific Laboratory report LA-7478-PR (October 1978).
12. M. A. Molecke, "Gas Generation from Transuranic Waste Degradation: An Interim Assessment," Sandia Laboratories report SAND 79-0117 (rough draft, January 1979).
13. E. L. Christensen, "Waste Management at TA-55 CMB-11 Facility," Los Alamos Scientific Laboratory internal document, January 1978, App. D, p. 19.
14. H. Division Staff, "Guidelines for the Interim Storage of AEC-Generated Solid Transuranic Waste," Los Alamos Scientific Laboratory report LA-5645 (June 1974).
15. H-Division Staff, "Transuranic Solid Waste Management Research Programs, Quarterly Report, October-December 1973," Los Alamos Scientific Laboratory report LA-5614-PR (May 1974).
16. John Warren and Al Zerwekh, Los Alamos Scientific Laboratory, personal communication. June 1977.
17. S. T. Kosiewicz, B. L. Barraclough, and A. Zerwekh, "Studies of Transuranic Waste Storage Under Conditions Expected in the Waste Isolation Pilot Plant (WIPP) December 15, 1978-March 15, 1979" Los Alamos Scientific Laboratory report LA-7775-PR (May 1979).

18. Technical Alternatives Document, Department of Energy document ERDA-76-43, 2, Sec. 9, p. 3.
19. A. Zerwekh, Los Alamos Scientific Laboratory, unpublished data, February 1978.
20. A. Zerwekh, "Gas Generation from Radiolytic Attack of TRU-Contaminated Hydrogenous Waste," Los Alamos Scientific Laboratory report LA-7674-MS (June 1979).
21. N. E. Bibler, "Gas Production from Alpha Radiolysis of Concrete Containing TRU Incinerator Ash—Progress Report (April—July, 1978)," Savannah River Laboratory report (rough draft, 1978).
22. T. O. Hunter, "Technical Issues for WIPP," Proc. 19th Annu. ASME Symp. Geol. Disposal of Nucl. Waste, Albuquerque, New Mexico (March 1979), pp. 75-80.
23. **Handbook of Chemistry and Physics**, 45th Ed., R. C. West, S. M. Selby, and C. D. Hodgman, Eds. (The Chemical Rubber Co., Cleveland, Ohio, 1964) p. D 92.
24. C. E. ZoBell and M. A. Molecke, "Survey of Microbial Degradation of Asphalts with Notes on Relationship to Nuclear Waste Management," Sandia Laboratories report SAND 78-1371 (December 1978).
25. G. Pretschner and A. Dagen, "Untersuchung Zum Wasserstoff- und Methanbildung bei der Bestrahlung von Bitumen B-45 und Gemischen aus Bitumen B-45 und Feststoffen," Conf. -761260, SAAS-220 (December 1976), pp. 28-38.
26. Immediate Action Directive 0511-21, Appendix A, USAEC, March 20, 1970.
27. Margaret Anne Rogers, "History and Environmental Setting of LASL Near-Surface Land Disposal Facilities for Radioactive Wastes (Areas A, B, C, D, E, F, G, and T)," Los Alamos Scientific Laboratory report LA-6848-MS, Vol. 1 (June 1977).
28. J. E. Dieckhoner, Compiler, "Report for the WIPP Waste Acceptance Criteria Steering Committee: Detailed Characterization of DOE Stored TRU Waste," Office of Nuclear Waste Management, Division of Waste Production, Operations Branch (June 15, 1978).
29. J. Warren, Los Alamos Scientific Laboratory Group H-7, Personal Communications, August 1978.