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STUDIES ON SORTING AND CHARACTERIZATION OF PLUTONIUM PROCESS AND ROOM-GENERATED WASTES*

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ABSTRACT

Waste characterization of room and process-generated wastes contaminated with plutonium has been an important part of the waste management research and development programs at Los Alamos Scientific Laboratory. Preliminary information on the types of waste matrices and the degree of transuranic contamination from typical operations has been reported. Predicated on the accumulated data obtained thus far, laboratory experiments have been outlined to determine the extent and products of radiolytic degradation of the various waste matrices commonly found. Radiolysis experiments have shown that significant quantities of H_2, CO, and CO_2 can be generated when cellulosics are contaminated with 0.6 to 1.2 mg of heat-source plutonium per gram of substrate.

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Waste characterization studies are an important part of several of the waste management research and development programs administered by the Health Research Division of the Los Alamos Scientific Laboratory (LASL). Detailed information on wastes generated and sent to retrievable storage is required by both operational personnel and the research staff within the waste management program for a number of reasons. The identification of the waste matrix and the levels of contamination are of particular importance in order to optimize waste packaging, handling techniques, and storage facility designs. The design and operation of the new LASL Transuranic Contaminated Waste Treatment Development Facility requires information concerning the actual amounts of combustibles in the wastes, as well as the degree of transuranic contamination. A breakdown of combustible wastes into categories such as cellulosics, plastics, and rubbers is also needed for the design of the feed, combustion, and offgas filter train systems of the reduction facility.

A program to sort, identify, and assay actual wastes produced at plutonium-handling facilities at LASL is serving as a means of obtaining quantitative information on waste characterization. Such quantitative data, combined with the qualitative information obtained from questionnaires and personal interviews, have served as an input into the research and development programs involved in the effects of radiolysis on the waste matrix. Experiments to determine

the extent and products of radiolytic degradation of wastes and problems of internal corrosion from chemicals in the waste require identification of the commonly occurring components of the waste matrix. In addition, the identification of the waste types and volumes as a function of origin will be utilized by waste management personnel to analyze their practices for increased efficiency and decreased waste volumes, contamination levels and costs.

During the initial period of investigation, sorting and assay were confined to the process residues generated within the plutonium fabrication area of the Chemistry and Metallurgy Group (CMB-11) at DP West. The processes investigated included casting, machining, welding, assembly and dissassembly, as well as a variety of other experimental operations in fabrication and metal operations. Nine categories of materials were encountered during an initial three-month survey period in 1973. These include: non-plutonium metals, plastics, rubbers, cellulosics, glass, ceramics, insulation, graphite, and sweepings. The total weight, weight percent of the discard stream, and the total plutonium content of all materials discarded from the metal fabrication lines are given in Table I. Similar data for the portion of the material sent to 20-year retrievable storage are listed in Table II. The four waste matrices listed in Table II represented 81% of the bulk weight of material sorted but contained only 38% of the plutonium present as a contaminant. The average concentration of weapons-grade plutonium in the discarded material was 0.17 g of pluto-

nium per kilogram of waste or an equivalent activity of 12 μCi/g of waste. The assay of plutonium content was determined using a neutron counter to measure the spontaneous fission rate of the ^{240}Pu within the waste. Since the isotopic composition of the plutonium was known approximately, the total plutonium concentration could be calculated.

Beginning in October 1973, further studies were conducted to identify the composition of waste residue generated in the Plutonium Chemistry and Metallurgy Group areas which is assayed for 20-year retrievable waste storage. An overview of the composition of the retrievable wastes studied during October through December 1973 is given in Table III. During this study, the waste materials sorted averaged 51% noncombustibles and 49% combustibles on a weight basis. The plastic that was accumulated during the sorting operation consisted mainly of plastic bags from bag-out operations. Less than 10% of the plastic consisted of items such as tubing, laboratory plasticware, and metallographic mounts. The cellulosic material within the retrievable waste comprised only 11% of the total. Cellulosics which may have been in direct contact with plutonium are normally incinerated following assay in order to recover the plutonium. The remaining process solids following treatment were normally less than 1% of the retrievable waste stream on a weight basis. Since compaction of wastes is not practiced within the Chemistry and Metallurgy area, the packing density of

the metal and glass wastes was quite low.

Additional information concerning the general density of the waste categories was also desirable. Listed within Table IV is the typical weight-volume relationship for 20-year retrievable wastes generated at the Chemistry and Metallurgy Group areas. The volume percentages presented within the table were based on uncompacted waste packaged in 115-liter drums.

Studies are underway to relate the amounts and types of wastes generated to the specific unit operation such as ash leaching, ion exchange, and metal alloy processing. Data has been accumulated on the composition of waste originating from the ash leaching process at the DP West area. The information presented in Table V was obtained from the leach process in which off-site incinerator ash was leached three times with nitric acid and calcium fluoride. The metal shipping containers comprised most of the metal waste stream. The majority of the glass encountered resulted from a routine change-out of the dissolver pots.

Room-generated waste from scrap recovery operations was also sorted and assayed to determine its generation rate, composition, and level of contamination. In addition, the evaluation presented an opportunity to verify the administrative decision designating process-generated wastes as retrievable, i.e., >10 nCi/g transuranic content per AECM 0511, and room-generated wastes as nonretrievable. To facilitate the

sorting and assaying operations, a special glovebox train incorporating a modified FIDLER (Field Instrument for the Detection of Low-Energy Radiation) detector was constructed and installed within the DP West facility. The lack of sensitivity of the neutron counter utilized in assaying scrap (~1 μCi/g) prohibited its use as an assay unit in this study. On the other hand, the FIDLER-based system, in this case a flat-waste configuration monitor, has a sensitivity well below 10 nCi/g in short counting times although the accuracy is dependent on the density and configuration of the waste under investigation. Figure I shows the detector and an operator assaying sorted waste within one of the gloveboxes. The lower limit of detection for plutonium in typical loose trash is approximately 1 nCi/g.

During a period of three months approximately 100 boxes of room-generated waste were opened, sorted, and measured for contamination. No box of regular room trash was found to exceed a contamination level of 10 nCi/g. Room trash generated from special operations, for example, planned contaminated operations and scheduled maintenance of contaminated equipment such as valves and piping used to transfer plutonium solutions, was assayed to be greater than 10 nCi/g. A few of the waste boxes originating from the special operations contained materials which averaged approximately 700 nCi/g. Most of the contamination could be related to metallic equipment and materials (surgeons' gloves, booties and cheese-

cloth) in direct contact with the plutonium.

Specific attention was also given to the weight, source by area, and other descriptive information related to the room-generated trash. The density of the material was quite low since the boxes were not compacted. Presented within Tables VI and VII is the room-trash generation and density data for the three-month period and the typical composition of the waste sampled in November. The data within Table VII was the average composition of 28 of the 128 boxes of trash generated. As observed from the detailed breakdown of the composition, most of the waste was cellulosic in nature.

As mentioned earlier, one of the objectives of the study is to determine the types of waste which may present problems for interim storage. Prior knowledge and assessment of the problems to be encountered in interim storage can facilitate proper design of the storage container and facility with possible tradeoffs between these designs. Ongoing investigations by personnel of CMB-1, the Analytical and Instrumental Chemistry Group, have revealed that alpha radiolysis of waste can lead to significant production of certain gases. The effects of radiolysis and radiolytic products on various types of commonly found wastes are being studied by contaminating the matrices with plutonium nitrate and chloride at three different levels of alpha contamination and sealing the dried wastes in stainless steel cylinders equipped with gas pressure gauges.

The waste materials being investigated include cellulosics, Hypalon, isoprene, polyethylene, Tygon, neoprene, Lucite, vinyl bakelite, and polyvinyl chloride. The upper level of contamination corresponds to the criticality limit set for transportation of drums, 200 grams of weapons-grade plutonium per 210-liter drum. For these experiments, an equivalent amount of ^{238}Pu activity was substituted for ^{239}Pu and the concentration increased by a factor of 40 to simulate 20 years of radiolytic attack in six months. The upper limit of contamination was 1.2 mg of heat-source plutonium per gram of waste or approximately 6×10^8 dps/g. The other levels of contamination were one-half and one-tenth of the maximum value.

After 15.8 years of simulated time, 53% of the 30 samples have shown positive gas pressure. The earliest detectable pressure increase above atmospheric came at 0.11 simulated years. One sample of cellulosics in the strong contamination group has been sampled six times and the gas composition analyzed on a mass spectrometer. On the basis of 34 kg of waste in a 210-liter drum, the amount of gas produced in an equivalent of 15 years would be 1 470 liters. The gaseous radiolytic products which have been detected so far include H_2, CO, CO_2, and CH_4. Although a complete evaluation of the data will be performed upon the termination of the experiments, the preliminary data does indicate that the composi-

tion of the gases does vary with time. The gas pressure build-up, and the percent composition of the gases within a cylinder containing cellulosics as a function of time are illustrated in Figs. II and III. Both figures were constructed from data obtained by sampling the cylinder when the gauge pressure reached 15 psig, the approximate upper pressure limit of the gauge. The test cylinders will be opened and the wastes examined for chemical degradation products.

In summary, waste characterization studies of process and room-generated wastes contaminated with plutonium are currently being conducted to optimize waste management practices at LASL. Such studies are needed to assist in designing the waste reduction facilities and an interim storage complex for transuranic contaminated wastes, and to conceptualize waste/container/environment technology for permanent storage.

TABLE I

Pu METAL FABRICATION GLOVEBOX LINE WASTE AND SCRAP

FOR

JULY - SEPTEMBER 1973

Material Type	Net Weight, kg	Composition, Wt.% of Total	Pu, g	Average Activity, nCi/g
Metal (non-Pu)	128.2	35	56	3.1×10^4
Plastic and Rubber	70.0	19	34	3.5×10^4
Cellulosics	18.6	5	177	$68. \times 10^4$
Glass and Ceramics	54.5	15	6	0.8×10^4
Insulation	1.9	<1	<1	$<0.5 \times 10^4$
Graphite	92.9	25	95	7.1×10^4
Sweepings	2.5	<1	38	$108. \times 10^4$
Total	368.6		406	

TABLE II

MATERIAL SENT TO 20-YEAR RETRIEVABLE STORAGE

DURING

JULY - SEPTEMBER 1973

Material Type	Net Weight, kg	Composition, Wt.% of Total	Pu, g	Pu Content, Wt.% of Total	Average Activity, nCi/g
Metal	107.3	84	20	36	13×10^3
Plastic and Rubber	43.1	62	10	29	17×10^3
Glass and Ceramics	54.5	100	6	100	8×10^3
Insulation	1.9	100	<1	100	$<5 \times 10^3$
TOTAL	206.8		36		

TABLE III

COMPOSITION OF 20-YEAR RETRIEVABLE WASTE

FROM

ROUTINE CMB-11 OPERATIONS

OCTOBER THROUGH DECEMBER 1973

	Composition, Wt.%			
Material Type	Oct.	Nov.	Dec.	Average
Metal	25	22	32	28
Plastic	36	30	23	29
Rubber	5	8	10	8
Cellulosics	12	11	14	12
Glass	18	29	21	22
Process Solids	4	< 1	< 1	1

TABLE IV

TYPICAL WEIGHT-VOLUME RELATIONSHIP, 20-YEAR RETRIEVABLE WASTE, NON-COMPACTED, PACKAGED IN 115-LITER DRUMS

Material Type	Weight Percent	Volume Percent
Metal	32	22
Plastic	23	32
Rubber	10	10
Cellulosics	14	26
Glass	21	10

TABLE V

COMPOSITION OF PROCESS SOLID WASTE FROM ASH LEACH OPERATIONS

Material Type	Composition, Wt.%
Metal	23
Plastic	14
Rubber	7
Cellulosics	11
Glass	14
Solids	31

TABLE VI

ROOM TRASH GENERATION AND DENSITY DATA

	Oct.	Nov.	Dec.	Ave.
Boxes Generated	218	128	180	175
Volume (m^3)	14.8	8.9	12.2	12.0
Weight (kg)	1 262	749	1 038	1 016
Density (Mg/m^3)*	0.085	0.084	0.085	0.085**

*Megagrams/cubic meter

**Equivalent to 5.3 lbs/ft^3.

TABLE VII

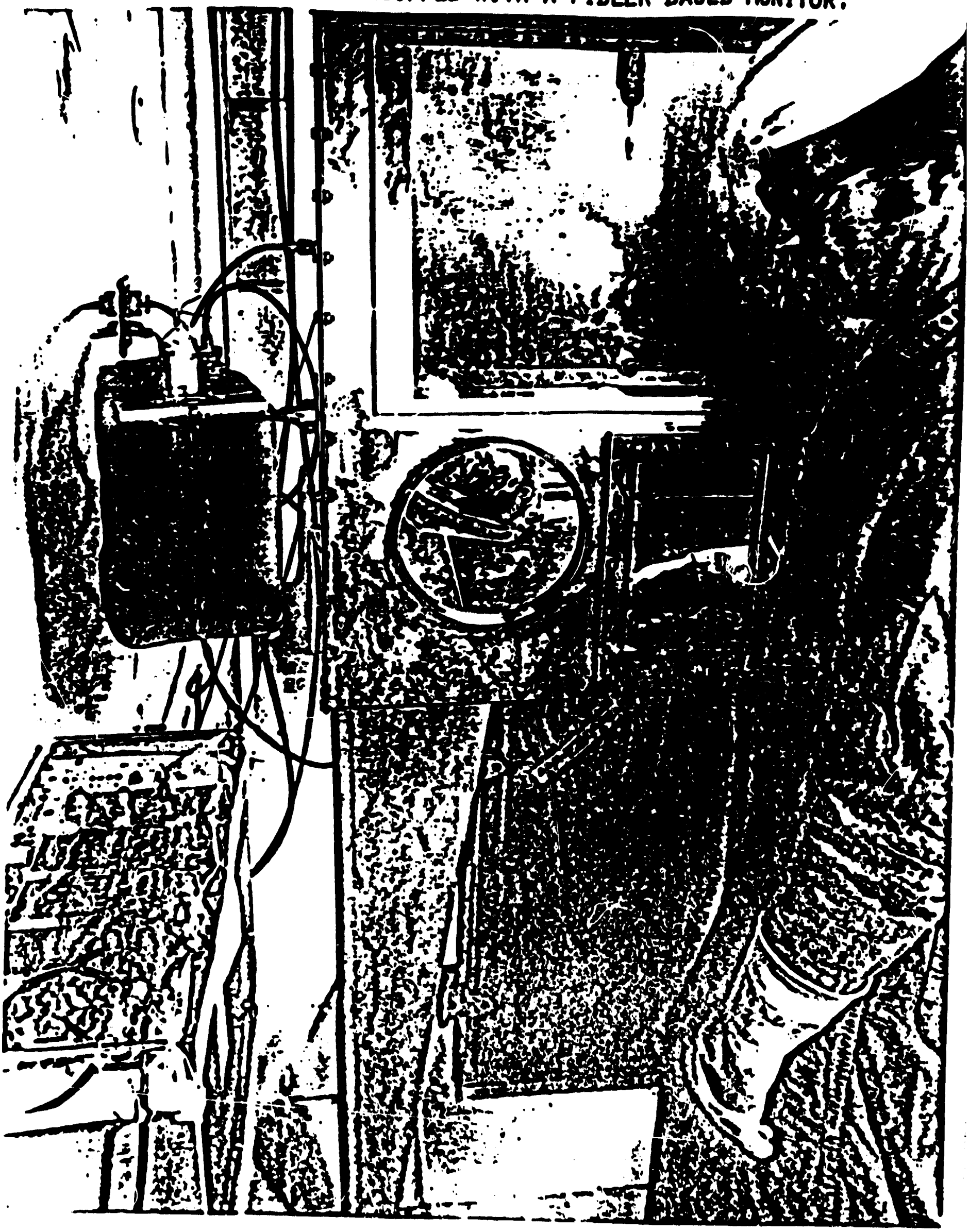
COMPOSITION OF CMB-11 ROOM TRASH

NOVEMRER 1973

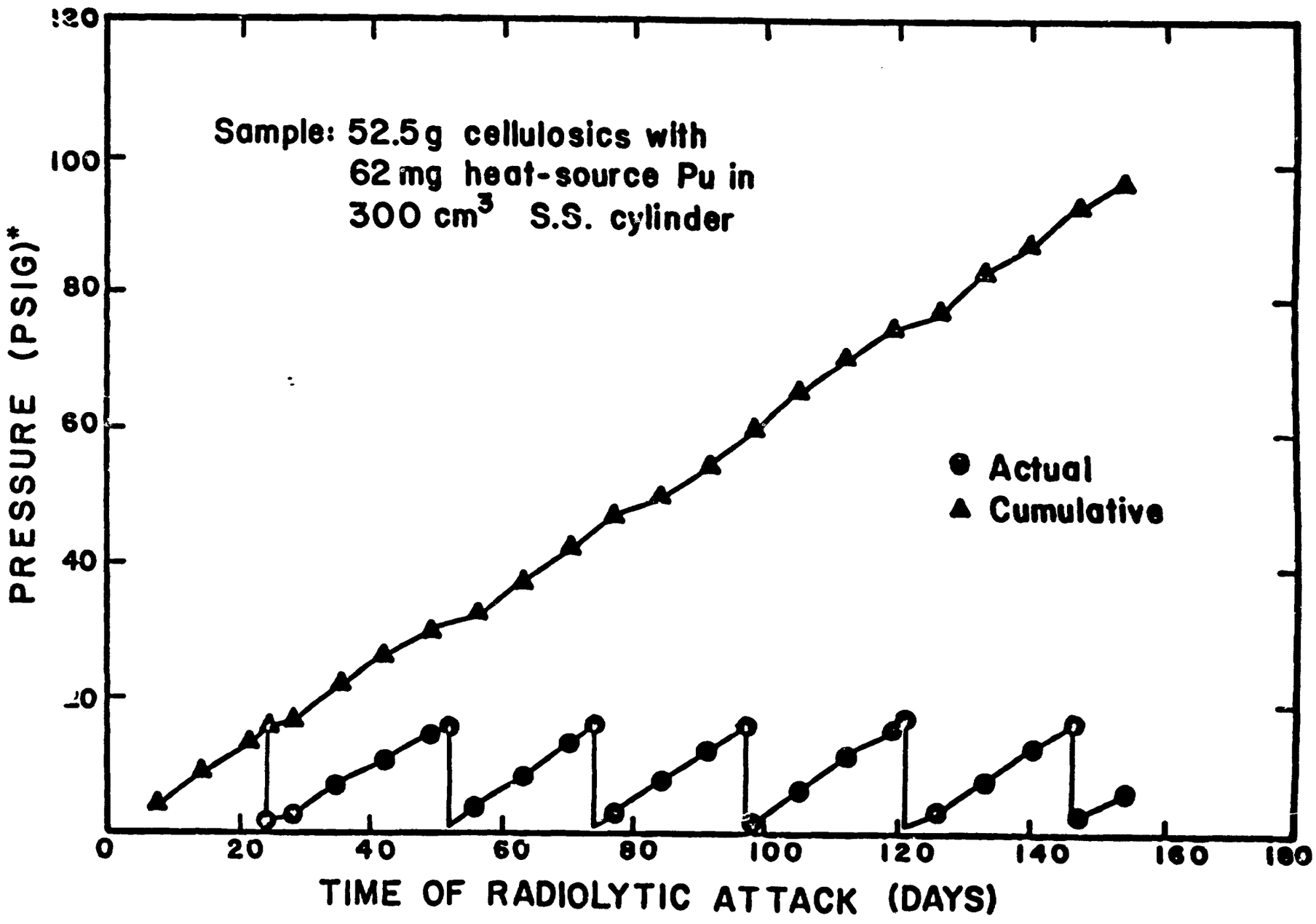
Material Type	Composition Wt.%	Total
A. Cellulosics		82
1. Chart paper, computer paper, other office type paper waste, surgeons' gloveboxes and tissue.	47	
2. Brown "Kraft" paper, masking tape	5	
3. Miscellaneous Paper	10	
4. Cheesecloth Mostly associated with decontamination work and wetted with cleaning solution.	6	
5. Clothing Booties, coveralls, caps, undershirts, shorts, and shower room cleanup including bars of soap and floor sweepings.	15	
B. Surgeons' Gloves Both Rubber and Plastic		4
C. Plastic Polyethylene bags, reagent bottles, bags from unpacking items such as face masks.		4
D. Styrofoam Packing materials and coffee cups		3
E. Glass Glass wool used for prefiltering room air exhaust, sample bottles.		1
F. Metal Flashlight batteries, wire, conduit, metal shavings, tin cans, knives, aerosol cans and aluminum foil.		5
	Total	100

LIST OF FIGURES

FIG. 1. OPERATOR ASSAYING TRANSURANIC CONTAMINATED WASTES IN A SPECIAL GLOVEBOX EQUIPPED WITH A FIDLER-BASED MONITOR.



...OF RADIOLYTIC GASES FROM CONTAMINATED CELLULOSICS AS A FUNCTION OF TIME.



*1 PSI IS EQUIVALENT TO 6.9 kPa

FIG. III. COMPOSITION OF GASES WITHIN A TEST CYLINDER AT VARIOUS SAMPLING TIMES.