

*Hydrogen Venting Characteristics of
Commercial Carbon-Composite Filters and
Applications to TRU Waste*

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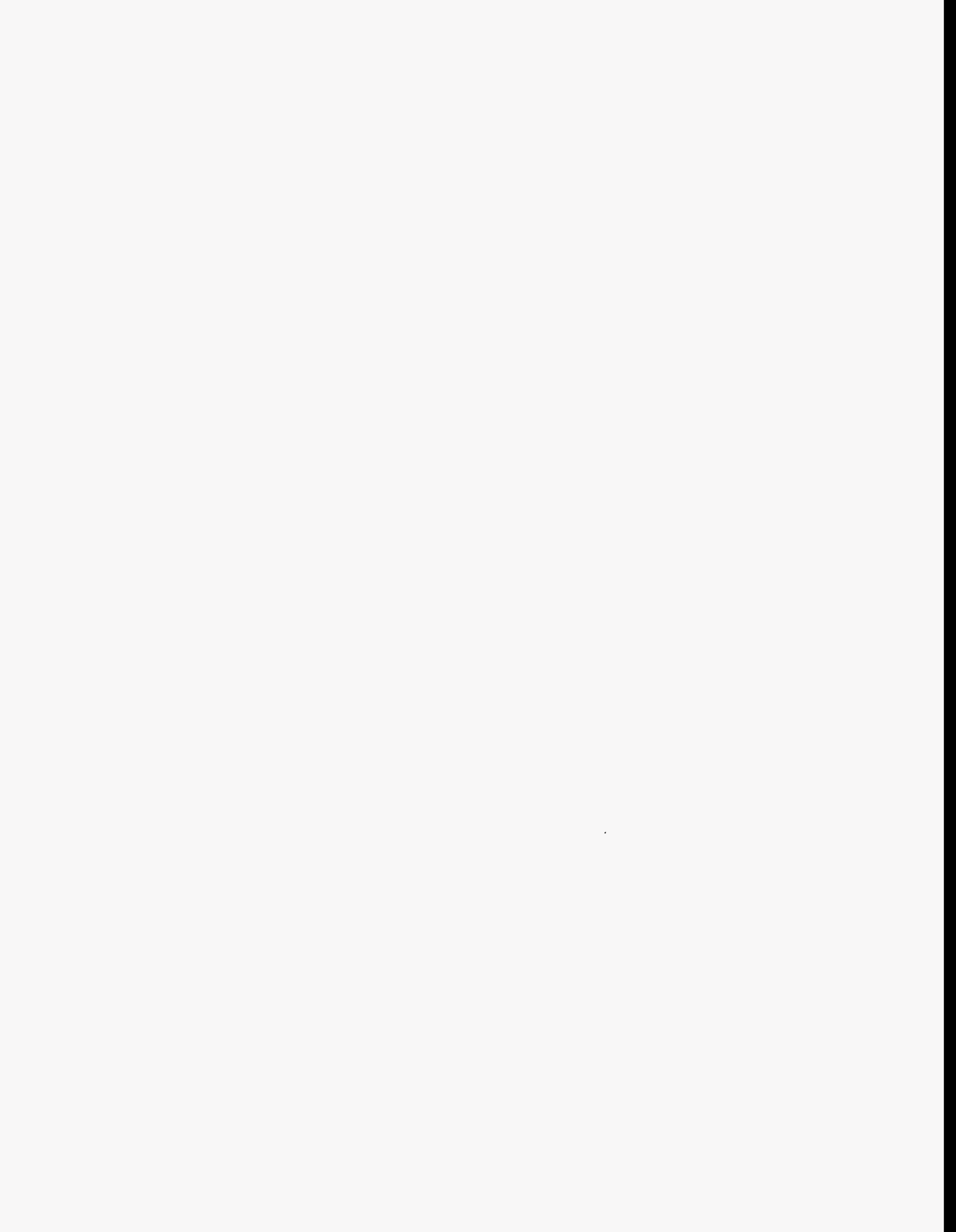
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HYDROGEN VENTING CHARACTERISTICS OF COMMERCIAL CARBON-COMPOSITE FILTERS AND APPLICATIONS TO TRU WASTE

E.L. Callis, John H. Cappis, Miles C. Smith, Robert S. Marshall

ABSTRACT

The generation of hydrogen (by radiolysis) and of other potentially flammable gases in radioactive wastes which are in contact with hydrogenous materials is a source of concern, both from transportation and on-site storage considerations. Because very little experimental data on the generation and accumulation of hydrogen was available in actual waste materials, work was initiated to experimentally determine factors affecting the concentration of hydrogen in the waste containers, such as the hydrogen generation rate, (G-values) and the rate of loss of hydrogen through packaging and commercial filter-vents, including a new design suitable for plastic bags. This report deals only with the venting aspect of the problem. Hydrogen venting characteristics of two types of commercial carbon-composite filter-vents, and two types of PVC bag closures (heat-sealed and twist-and-tape) were measured. Techniques and equipment were developed to permit measurement of the hydrogen concentration in various layers of actual transuranic (TRU) waste packages, both with and without filter-vents. A test barrel was assembled containing known configuration and amounts of TRU wastes. Measurements of the hydrogen in the headspace verified a hydrogen release model developed by Benchmark Environmental Corporation. These data were used to calculate revised wattage limits for TRU waste packages incorporating the new bag filter-vent.

1.0 INTRODUCTION

A significant concern of the Laboratory's transuranic (TRU) waste program involves the limitations prescribed in the TRUPACT-II SARP (Reference 1), restricting the quantity of radioactive material that a waste package may contain. These material loading limitations, known as wattage limits, were implemented to prevent flammable gases (primarily hydrogen), from building up to combustible concentrations. Hydrogen gas is generated in TRU waste through the interaction of radiation with hydrogenous materials, such as paper and plastics, that make up the waste matrix. Wattage limits are calculated by modeling the generation of hydrogen within a waste package and its subsequent diffusion into the annular space of the TRUPACT-II shipping cask.

Approximately thirty percent of the existing TRU waste at the Laboratory exceeds the wattage limits in the TRUPACT-II SARP. Additionally, the wattage limits are restrictive enough to virtually eliminate high specific activity wastes, such as ^{238}Pu , from being shipped to the Waste Isolation Pilot Plant (WIPP) for disposal. These considerations and concerns are, of course, applicable to the on-site storage of wastes also.

With this in mind, we set about determining how to increase the radioactive material loading in TRU waste drums, while maintaining hydrogen below the lower flammable limit of five percent in the innermost confinement layer. Two approaches presented themselves. The first approach

was to re-evaluate the hydrogen generation rate, or G-value, to determine if the values used in the TRUPACT-II SARP were accurate or overly conservative, as was suspected. Preliminary results of this effort have been discussed elsewhere (Reference 2,3). The second approach, and the subject of this report, was to determine ways to enhance the diffusion of hydrogen out of waste drums thereby lowering concentration within the drum. In this report we discuss the results of our experiments to determine the hydrogen diffusion rates for materials and commercial filter-vents used at the Laboratory in TRU waste packaging, including a new filter suitable for venting bags and cans. Additionally, hydrogen concentration data from existing waste containers are presented. This information indicates that hydrogen production within TRU waste drums is indeed a potential safety concern in unvented containers. Finally, as a solution to the problem of restrictive wattage limits we present a revised wattage limit table for wastes packages incorporating the new bag filter vent.

2.0 HYDROGEN DIFFUSION COEFFICIENT MEASUREMENTS

Experimental: Hydrogen diffusion rates through various barriers were determined by mass spectrometric measurements of the loss rate of hydrogen through the barrier at ambient pressure. A 2-liter stainless steel high vacuum chamber (Fig. 2-1) was constructed using a commercially available ultra high vacuum (UHV) nipple with 6-inch flanges. Inlet and outlet ports for flushing and sampling were installed on one end, and a means for mounting the item to be tested was installed on the opposite end. Drum and bag carbon composite filters, (Figs. 2-2 and 2-3) manufactured by Nuclear Filter Technology of Colorado, were mounted on the chamber using a 5-inch diameter copper disk in which the central section had been milled to 0.020 inch thickness with a 1.25-inch diameter hole to accommodate both the bag-type and drum filters. The copper disk was mounted to the chamber using a bolted flange and Viton O-rings for a gas-tight seal.

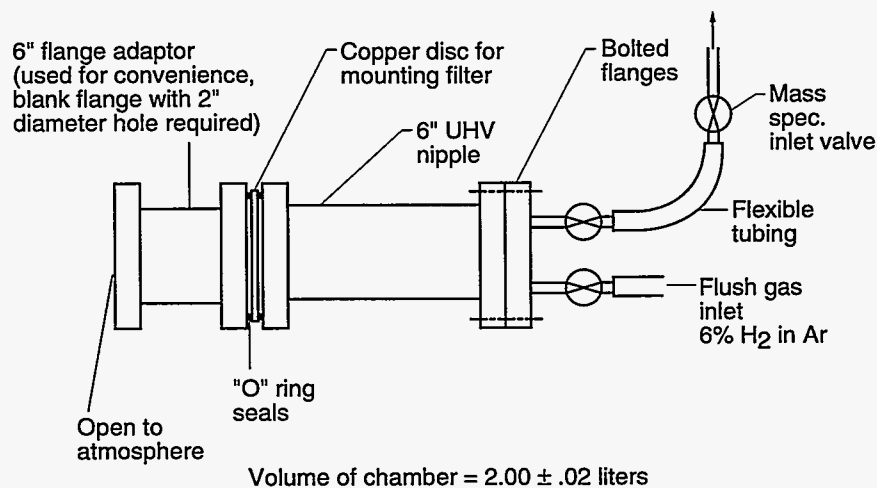


Fig. 2-1. Carbon-composite filter testing chamber.

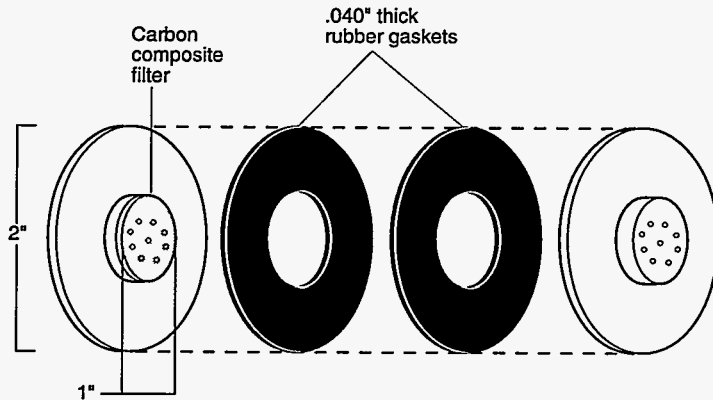


Fig. 2-2. NFT 030 filter.

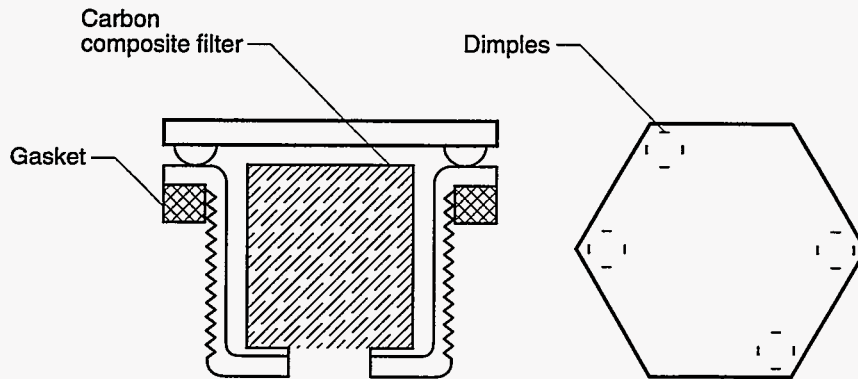


Fig. 2-3. NFT 013 filter.

Hydrogen diffusion tests were also performed on 14-mil PVC bag-out bags with open ends that were either heat-sealed by the manufacturer, or used "horse-tail" (twist and tape) seals prepared by three technicians who normally perform this task. The "horse-tail" seals were made in the usual manner by tightly twisting approximately a 25-cm length of bag, wrapping the twisted segment tightly with PVC tape, cutting through the center of the twisted and taped length with a knife, taping over the cut end, and finally retaping over the stub end. For these bag-end tests, a larger diameter (15.13 inch) flange was fabricated from aluminum so that the bags would fit snugly over the flange. A 1-inch wide rubber strip was placed around the bag at the flange to prevent damage to the bag by a worm-drive clamp used to secure a seal between the bag and flange. The modified chamber is shown in Fig. 2-4. Fig. 2-5 shows a photo of the apparatus with a bag installed. The lower part of the photo shows the copper discs for mounting the bag or drum filters.

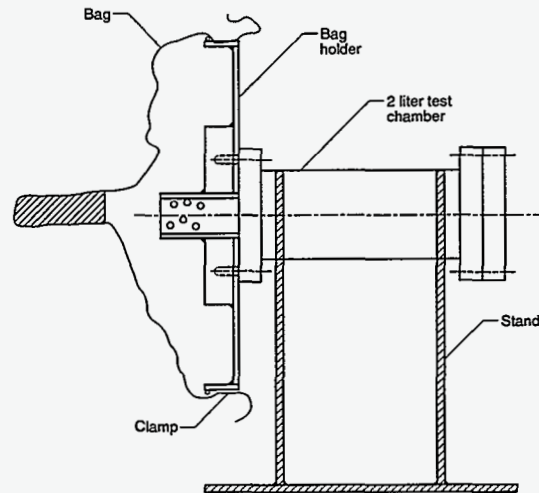


Fig. 2-4. Chamber modified for bag testing.

A commonly available 6% H_2 -94% Ar mixture of gas was used. After mounting the test item, the chamber was flushed to obtain a hydrogen concentration of 4-6%. The absolute value of the H_2 concentration was not critical because the rate-of-change of H_2 concentration was all that was needed. After shutting off the flush gas, the outlet valve was opened for 1-2 minutes to assure equilibrium between the pressure inside and the pressure outside the chamber. Next, the chamber was left undisturbed for 15 minutes to 2 hours (depending on the item being tested) prior to sampling, in order to further assure ambient pressure equilibration.

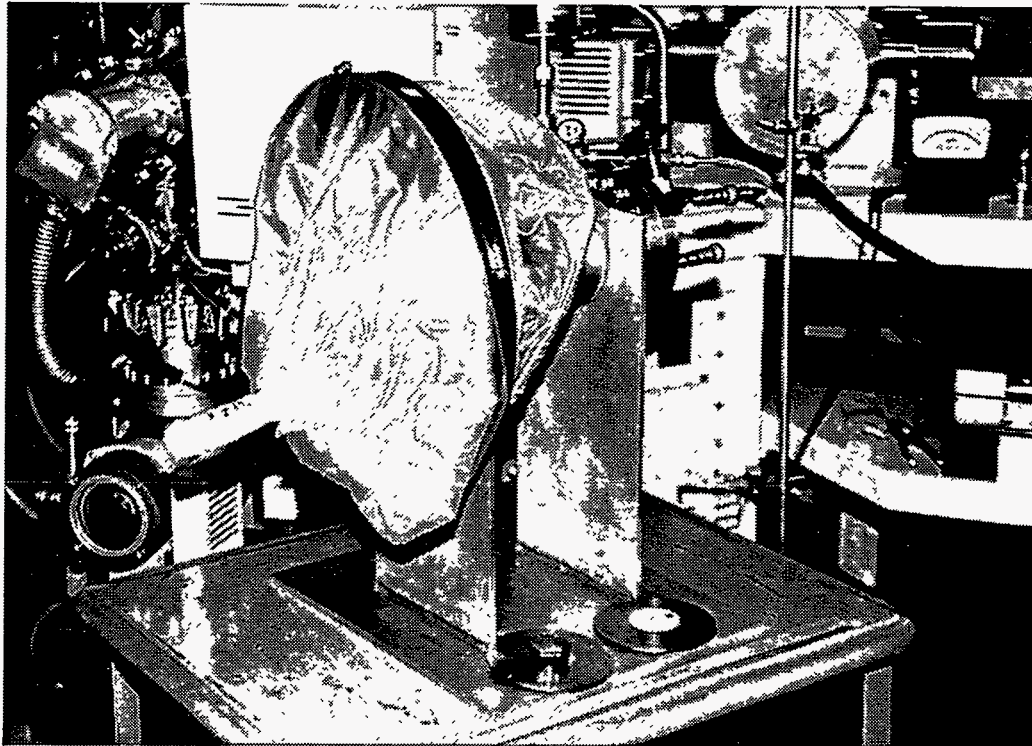


Fig. 2-5. Hydrogen diffusion testing apparatus with bag installed.

Sampling was performed by attaching one of the valved outlets of the chamber directly to the inlet of the mass spectrometer with a short length of flexible tubing. The sampling volume was kept low (about 7 cc or $< 0.35\%$ of the chamber volume) to avoid altering the system ambient pressure significantly. By evacuating the sampling line up to the chamber valve with the mass spectrometer inlet pumps, closing the inlet valve to the mass spectrometer, briefly opening then closing the chamber valve, and finally opening the mass spectrometer inlet valve, samples were taken. This method (as opposed to syringe sampling and sample injection, for example) assures the integrity of the sample admitted into the instrument.

A minimum of 4 samplings were made on each item tested, at intervals of 15 minutes to several hours, depending on the rate of loss of H_2 . The leakage rates for the filters were quite high, so 4 samplings over a 60-minute period were adequate to determine the diffusion coefficient. The leakage rate for the horse-tail sealed bags and bag-ends was two orders of magnitude lower, so 2-3 day periods were needed to record significant changes in the H_2 concentrations.

The mass spectrometer chosen was a Finnegan MAT-271 magnetic sector instrument designed for high precision measurements of gas composition. This instrument is equipped with both Faraday and electron multiplier detectors, a heated inlet system, and a computer controlled data acquisition and control system. The mass resolution is adjustable from 250 to 3000. These data were all taken at low resolution (250).

For H₂ concentrations in the 4-6% range, the measurement precision was about 0.2% relative. This high precision permits the measurement of relatively small changes in H₂ concentration, as is necessary at low diffusion rates.

Calculations

For the conditions of these experiments, where a fixed volume of gas is equilibrating with the atmosphere in which the H₂ concentration is essentially zero, we have:

$$dF/dt = -\lambda F(t)$$

where $F(t)$ = molar concentration of H₂ in the chamber
 λ = constant for a given barrier and chamber volume, at constant temperature and pressure.

This equation integrates to the familiar relation:

$$F(t) = F_0 e^{-\lambda t}$$

where F_0 is the initial H₂ concentration.

This relationship can also be expressed as:

$$\ln F(t) = \ln F_0 - \lambda t$$

Hence, by linear regression analysis of $\ln F(t)$ vs. time, the constant λ (slope) can be determined.

The diffusion coefficient for a particular item can be defined as:

$$D = \frac{\text{H}_2 \text{ flow across barrier}}{\text{Concentration differential across barrier}}$$

If the number of moles of gas in the test chamber is M , then:

$$D = \frac{(dF/dt) * M}{F(t)} = \frac{-\lambda F(t) * M}{F(t)} = -\lambda M$$

The value of M is calculated as:

$$M = (V_c/K) * (P_a/P_{stp}) * (T_{stp}/T_a)$$

- where,
- V_c = Volume of chamber, liters
 - P_a = Ambient pressure, typically 582 Torr at Los Alamos
 - P_{stp} = Standard pressure (760 Torr)
 - T_a = Ambient temperature, typically 296 °K.
 - T_{stp} = Standard temperature, 273.15 °K.
 - K = molar volume constant = 22.414 liters/mole at STP.

The coefficient D, has dimensions of moles/sec./mole fraction.

Measurement Results. The diffusion coefficients for drum and bag types of carbon filters, NFT 013 and NFT 030 were measured. Table 2-1 lists the diffusion coefficients determined for the drum filters and some values previously measured by others. Note that the mean NFT- 013 (drum) filter diffusion coefficient (8.5E-6) is approximately three times greater than that reported in the TRUPACT-II SARP (NuPac 1992) for the same filter type.

Table 2-1. Diffusion Coefficients of NFT 013 Drum Filters

Date	Filter ID	H2 Diff. Coeff. (moles/sec/mole fraction)
2/09/93	NFT LA563	8.31E-06
2/10/93	NFT LA914	8.37E-06
2/12/93	NFT LA908	8.44E-06
2/12/93	NFT LA902	8.94E-06
	Mean	8.52E-06
	Std. Dev.	2.88E-07
	% RSD	3.38%
	Range	7.40%

Values Previously Reported: Liekhus (1994, Ref. 4): 1.17E-5 to 7.4E-6
 Trupact II SARP (Ref. 1): 3.6E-6

Table 2-2 lists the results for the NFT 030 bag filters. There are no studies previously performed on the NFT 030 filter in the literature for comparison. * A histogram of the measured values is shown in Fig. 2-6, and indicates a somewhat normal distribution. Note that these filters were probably from three different batches prepared by the manufacturer and the reproducibility appears to be good (range of 12.6% for 12 filters).

Table 2-2. Summary of Hydrogen Diffusion Coefficient Measurements of NFT 030 Bag Filters

Date	Filter ID	H2 Diff. Coeff. Moles /sec/mole fraction
2/09/93	NFT INL-381, LANL 1	1.105E-5
2/10/93	NFT INL-381, LANL 2	1.116E-5
2/10/93	NFT INL-381, LANL 4	1.170E-5
2/5-8/93	NFT INL-381, LANL 6	1.130E-5 ± .032(n=4)
6/29/93	NFT 3/93, LANL 13	1.128E-5
6/28/93	NFT 3/93, LANL 14	1.145E-5
5/5/93	NFT 3/93, LANL 15	1.217E-5 ± .009(n=3)
6/28/93	NFT 3/93, LANL 16	1.075E-5
6/25/93	NFT 4/93, LANL 25	1.092E-5
6/29/93	NFT 4/93, LANL 26	1.125E-5
6/29/93	NFT 4/93, LANL 27	1.121E-5
6/29/93	NFT 4/93, LANL 28	1.105E-5
	Mean	1.127E-5
	Std Dev	0.037E-5
	%RSD	3.3%
	Range	12.6 %

*We have since learned of similar measurements made in 1990 by S.H. Peterson, et al. in "Determination of Flow and Hydrogen Diffusion Characteristics of Carbon Composite Filters Used at the Waste Isolation Pilot Plant," (P.O. 75WRS369171Z), Westinghouse STC, Pittsburgh, Pa., 1 March 1990 (not published in open literature). We subsequently obtained three of the actual filters that were measured by Peterson and performed tests on them at LANL. See Appendix 1.

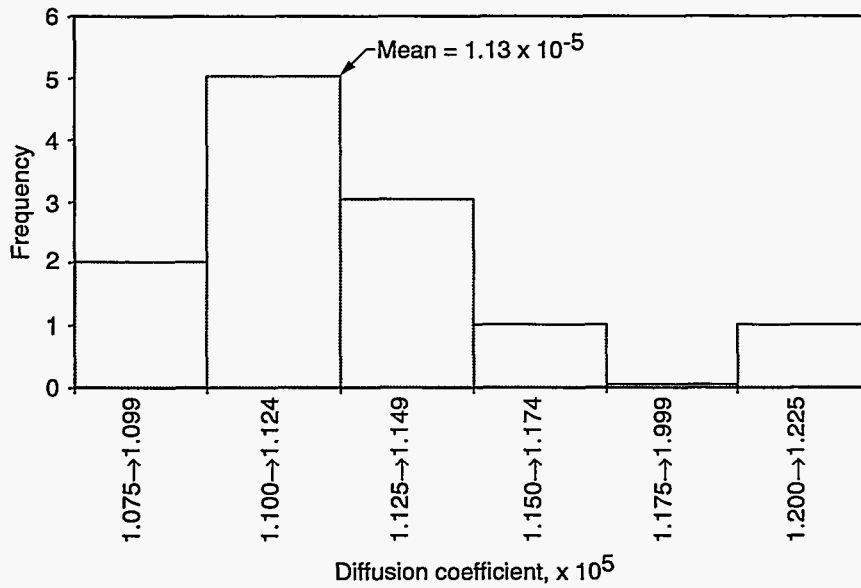


Fig. 2-6. Histogram of diffusion coefficients measured for NFT 030 filters.

The results of the diffusion tests made on the "horsetail" and heat-sealed bag-out bags are shown in Table 2-3. These data include permeation through $\sim 2800 \text{ cm}^2$ of bag, in addition to the actual seals. The relatively wide range (128%) and large standard deviation (36%) of diffusion coefficients for the "horse-tail" sealed bag-ends is to be expected, and reflects variation in the technique used by the three technicians. Note that the mean diffusion coefficient value of $2.85\text{E-}7$ for the heat sealed bag ends is about one-half of the value reported in the TRUPACT-II SARP (NuPac 1992). However, the observed mean diffusion coefficient for the "horse-tail" sealed bag ends ($7.6\text{E-}7$) agrees well with the value reported in the same document.

Table 2-3. Summary of 14-mil PVC Bag/Horsetail Closure Measurements

Horsetails (Twist and Tape Closure)

Start Date	Bag ID	Diff. Coeff. (moles/sec.mole fraction)	Liekhus*
10/05/93	T.L.-6	7.00E-07	2.5 ± .7E-7 (N=2)
10/18/93	T.L.-5	8.35E-07	
10/21/93	T.L.-6A	8.35E-07	
10/27/93	T.L.-5A	1.20E-06	
10/25/93	J.J.-1	2.22E-07	
11/01/93	Tim-4	9.17E-07	
11/29/93	J.J.-2	7.10E-07	
12/01/93	Tim-3A	6.63E-07	
	Mean	7.60E-07	
	Std. Dev.	2.8	
	%RSD	36%	
	Range	128%	

Heat-Sealed Bags

Start Date	Bag ID	Diff. Coeff.	Surface Area, sq. cm	Liekhus*
11/09/93	Bag #1	2.90E-07	2800	2.04 ± .07E-7 (N=2)
11/16/93	Bag #2	2.80E-07	2825	

Precision and Accuracy. Measurement precision was estimated by performing replicate measurements on two of the NFT 030 filters. These data are shown in Table 2-4. The one relative standard deviation repeatability on these measurements was better than 3% in both cases, which is quite adequate for this work, and is of the same order as differences between filters.

*See Reference 4.

Table 2-4. Replicate Hydrogen Diffusion Coefficient Measurements for the LANL 6 and 15 NFT 030 Bag Filters

Date	Filter ID	Replicate Number	H2 Diff. Coeff. (moles/sec/mole fraction)
2/5/93	NFT INL-381, LANL 6	1	1.105E-5
2/8/93		2	1.143E-5
2/8/93		3	1.170E-5
2/9/93		4	1.104E-5
		Mean	1.130E-5
		Std Dev.	0.032E-5
		% RSD	2.8%
5/05/93	NFT 3/93, LANL 15	1	1.226E-5
5/21/93		2	1.209E-5
6/28/93		3	1.217E-5
		Mean	1.217E-5
		Std. Dev.	0.009E-5
		% RSD	0.70%

Four sources of possible bias were evaluated. The volume determination of the bag-out bag ends and horsetail sealed bag ends was the most prominent. This volume was determined by injecting a known amount of He into the chamber/bag and, after equilibration, the He concentration in the bag was measured. An estimated margin of error for these measurements is $\pm 10\%$. Another possible source of bias could be caused by a leak in a chamber or in sampling lines to the mass spectrometer. The use of high-vacuum quality components minimized this possibility and no leakage was detected during various tests. Lack of equilibration of the chamber pressure with ambient could also cause bias. Equilibration was verified by the linearity of the $\ln H_2$ concentration vs. time plots. Figure 2-7 shows the results from following the loss of H_2 during a bag filter test for a much longer than normal period of time. Non equilibrium would be evidenced by curvature in the $\ln H_2$ concentration vs. time plots. In general, these plots showed no significant deviations from linearity, with the internal precision of the slope determinations being typically less than 1%.

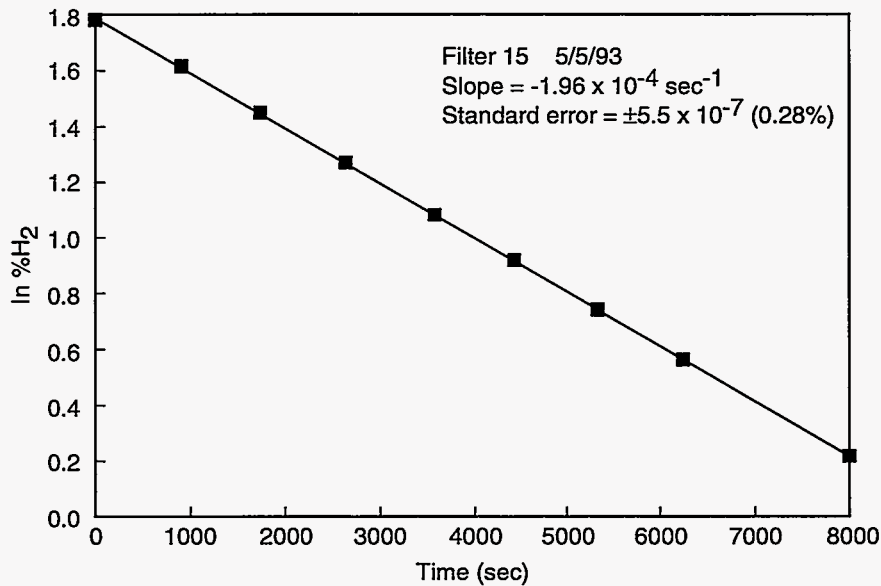


Fig. 2-7. Plot of ln (% H₂) vs. time for an extended interval.

Finally, there is some speculation (Reference 4) that the volume of gas in a test volume may influence the diffusion coefficient determination. Obviously, if this was the case, the measurements would not be valid. With our test chambers, we evaluated this effect by comparing the diffusion coefficients calculated from measurements taken with a filter installed on a 55 gallon drum with those taken of the filter installed on the 2-liter chamber described previously. For this test, two sources of leakage must be considered; the barrel itself, as well as the filter. For a sealed barrel (no filter), a coefficient of 1.1E-6 was obtained. For the barrel with filter installed, a value of 1.03E-5 was measured. The coefficient for the filter alone is the difference in these two values (9.2E-6), and does not differ significantly from the value measured using the 2-liter chamber (8.5E-6). Therefore, we are quite confident that these measurements relate to real-world situations.

3.0 APPLICATIONS TO TRU WASTE

The relatively high diffusion rates observed for the bag filters makes them potentially useful for venting hydrogen and other flammable gases generated by TRU waste. The current wattage limits in the TRUPACT-II SARP (NuPac 1992, Reference 1) are somewhat limiting due to the restricted diffusion rates of the bags used in TRU waste packaging. Therefore, we felt that the use of bag filters would significantly increase the wattage limits for TRU waste packages. The next two sections discuss an experimental and a calculational effort we used to quantify the impact that use of the bag filter would make on wattage limits for TRU waste.

3.1 TESTS ON EXISTING WASTE AT LANL

Sampling and Analysis of Existing Waste Cans

Summary

A specific, three-layer packaging configuration was used at the LANL plutonium facility for the containment and removal of ^{238}Pu -contaminated waste generated in glove boxes. To establish a criteria for safely storing these cans on-site (relative to the gas generation occurring in the packages), a sampling procedure was developed to measure the gas content in each of the layers. Two basic matrices, cellulose and polyethylene, were predominate in the waste. Of primary concern was the production and retention of H_2 to potentially combustible levels.

For this study, samples were taken from twelve waste containers, ten with a primarily cellulose matrix and two with a primarily polyethylene matrix. The gas content was determined using mass spectrometric techniques. Hydrogen content in the various layers ranged from 0.02 to 20.41%. These results contraindicated storage of these packages as they were currently configured. As a result, the package exhibiting the largest H_2 retention capability was modified by the use of carbon composite filters on the cans and bags. By coupling this data with other filter flow studies and modeling presented in this paper, modification of the packages for eight of the cellulose waste cans was made and they were placed into standard 55-gallon barrels. Head-gas monitoring of the test barrel was conducted for 122 days, and it was observed that the H_2 concentration agreed well with the model predictions.

This section will review the packaging configuration, describe the sampling apparatus and its use, present the gas content distributions from measurements, and document the results of adding a filter to a specific waste package can and bag. The next section will include the configuration of the test drum, measurements from the test drum, and results from the working standards samples analyzed in conjunction with these samples.

Packaging

The typical packaging configuration, Fig. 3-1, consisted of three layers: 1) an inner slip lid metal can, 2) a 12 mil polyethylene bag sealed with a horse tail, and 3) an outer slip lid metal can. Waste materials were added to the inner can until it was full, then the slip lid was put into place. The slip lid was either taped with plastic tape circumferentially at the slip lid interface with the can, or it was held in place with two pieces of tape placed radially across the surface and down the sides of the can. The inner can was then placed in a bag which was sealed with a horse tail closure, or with a horse tail closure on each end of the bag. The bag was put into the outer can (volume approximately 2 gallons), and the slip lid was put on and held in place with tape applied either circumferentially or radially, as described for the inner can.

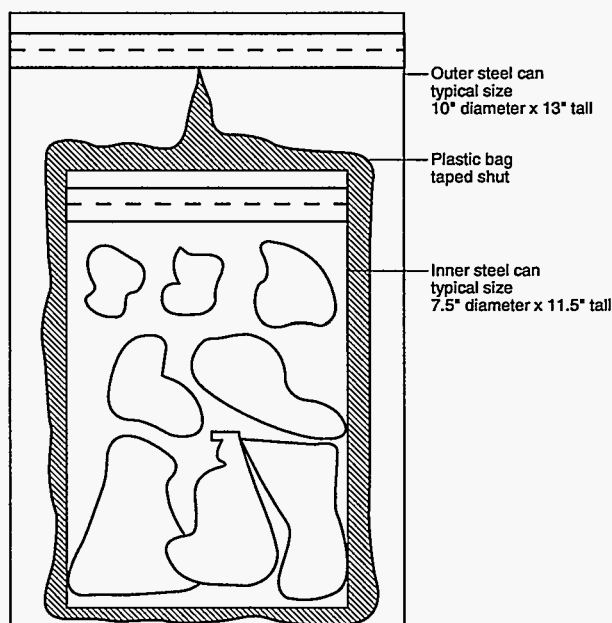


Fig. 3-1. Typical waste packaging of ^{238}Pu contaminated waste from LANL plutonium facility glove boxes.

Equipment and Supplies

- a. Can penetrator and sampling device (Fig. 3-2, the head-gas sampler for “bagout cans” and Fig. 3-3 - a more detailed view of the can penetrator head).
- b. Sample valving and manifold (Fig. 3-4).
- c. Stainless steel (20 gauge) beveled sampling needle connected to a stainless steel valve.
- e. Stainless steel sample cylinders, 25 or 50 ml fitted with stainless steel bellows-sealed valves.
- f. House vacuum source. Measured to 24 inches Hg.
- g. Vacuum grease.
- h. Plastic bags, 1 and 2 quart sizes.
- i. Argon gas.
- j. Flexible vacuum tubing.
- k. Finnigan-Mat 271 Gas Mass Spectrometer with a hydrogen ion source.
- l. Bag filters (NFT 030).
- m. Drum filters (NFT 013).
- n. Standard 55 gallon waste drum.
- o. Apparatus for sampling drum headspace, (Fig. 3-5).
- p. Matheson standard gas with 509 ppm H_2 and 514 ppm He in N_2 .
- q. Tank gas with a nominal 6.6% H_2 in Ar.
- r. Septa, 1/2" diameter, 3/16" thick
- s. Double stick tape.
- t. 2" vinyl tape.
- u. Ratchet wrench with 3/4" socket.
- v. Caulking cord, "Mortite," 1/4" diameter.

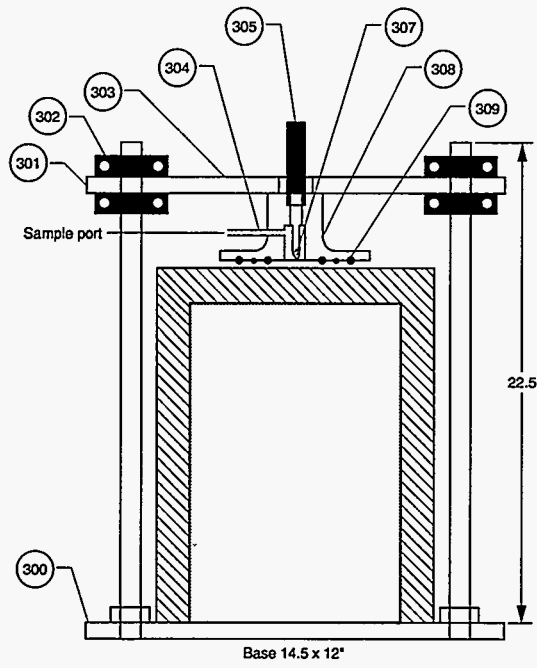


Fig. 3-2. Head-gas sampler for "bagout" cans.

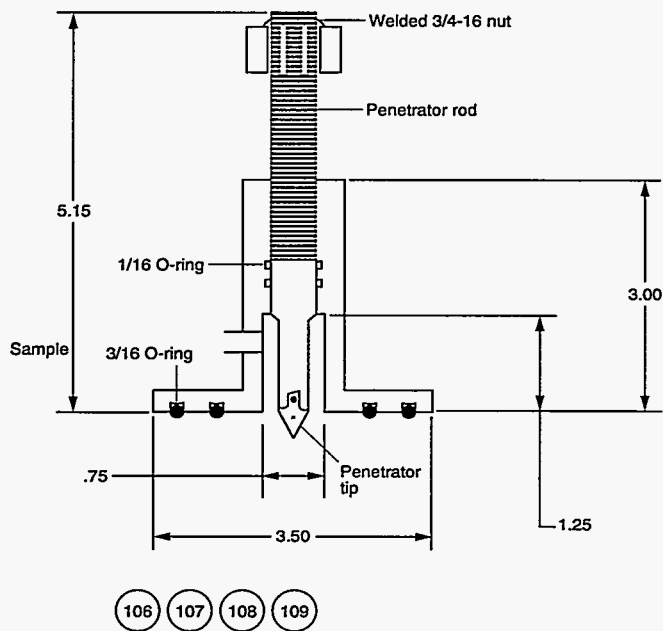


Fig. 3-3. Penetrator head.

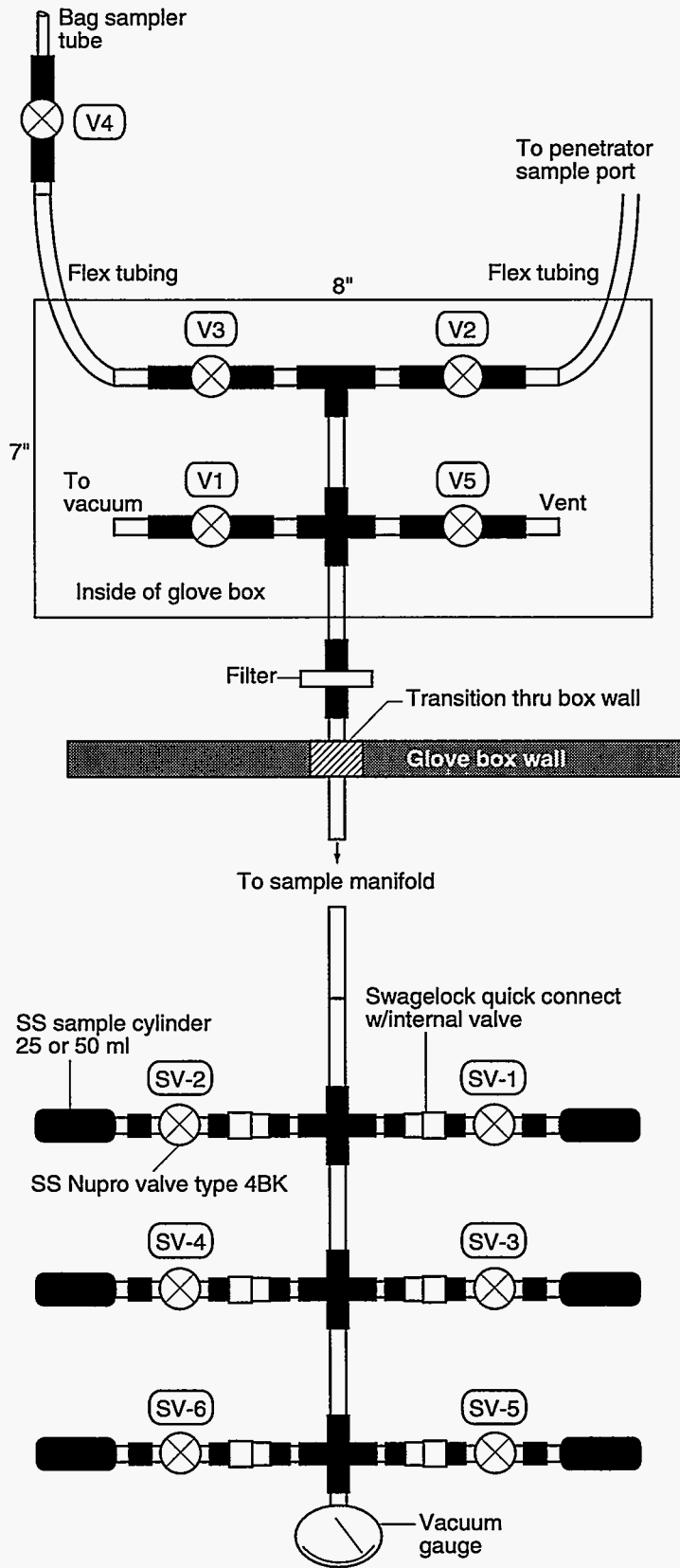


Fig. 3-4. Head-gas test in waste cans - sample valving.

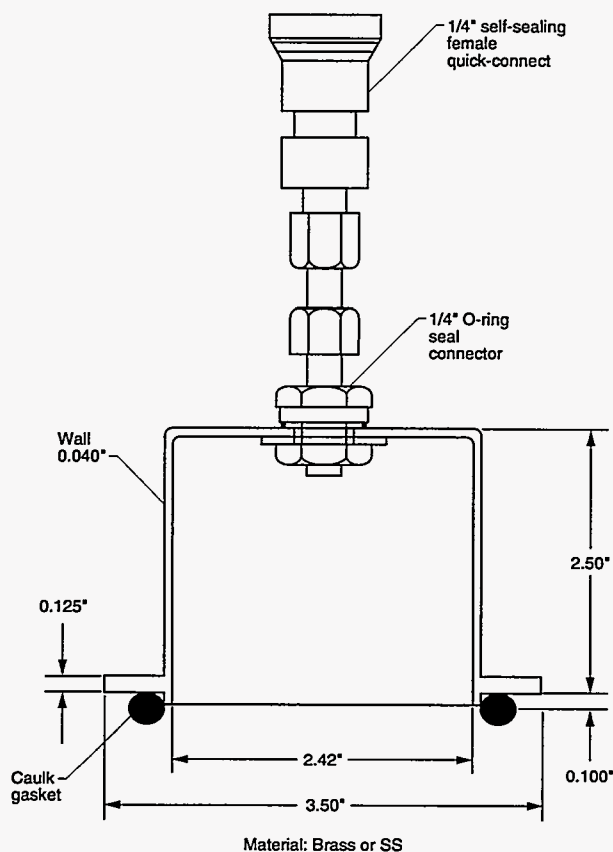


Fig. 3-5. Thru-filter drum headspace sampling device.

Experimental - Waste Can Gases

All sampling for existing waste packages was conducted in a glove box inside the LANL Plutonium Handling Facility (TA-55). The can penetrator and sampling device, Figs. 3-2 and 3-3, and sample valving portion of the sample valving and manifold, Fig. 3-4, were assembled inside the glove box. A standard transition coupling, which was designed into the glove box assembly, was used to connect the inside of the box sample valving to the manifold located outside the box. A 0.5 um filter was installed in the gas sampling line to trap any particles which might enter the line with the sample.

The stainless steel sampling chambers, Fig. 3-4, were evacuated to high vacuum on the sample handling manifold of the mass spectrometer and the valve on each was closed so each container remained at high vacuum. The containers were connected to the manifold system, Fig. 3-4, outside the glove box using quick-connect fittings. Up to six sampling tubes were connected at a time.

For the valving system inside the glove box, Fig. 3-4, flexible thick-walled 3/8" I.D. vacuum tubing was attached to the stems from valves V1, V2, and V3. The opposite end of the tubing from valve V1 was attached to the house vacuum source, the end from valve V2 was attached to the sample tube on the penetrator head assembly, and the end of the tubing from V3 was attached

to the valve stem connected to the bag sampling assembly. A light coating of high-vacuum grease was put on the outside surface of each of the stems before sliding the flexible tubing over them, and a hose clamp was used to secure the connections.

The following steps were taken in the assembly of the penetrator head, Fig. 3-3, for sampling the headspace gases of the cans. Each of the two 1/16 "O" rings was greased with high-vacuum grease and installed into its groove. The penetrator rod was screwed into the head until the tip was well beyond the sealing surface, making sure the 1/16 "O" rings were seated. A penetrator tip was attached to the penetrator rod using a set screw to secure it into place. The penetrator rod was screwed out of the head until the penetrator tip was inside the head well. The penetrator head 3/16" "O" rings, located on the plate making contact with the can lid, were lightly greased and inserted into the grooves. Fig. 3-6 shows a photo of the penetrator components.

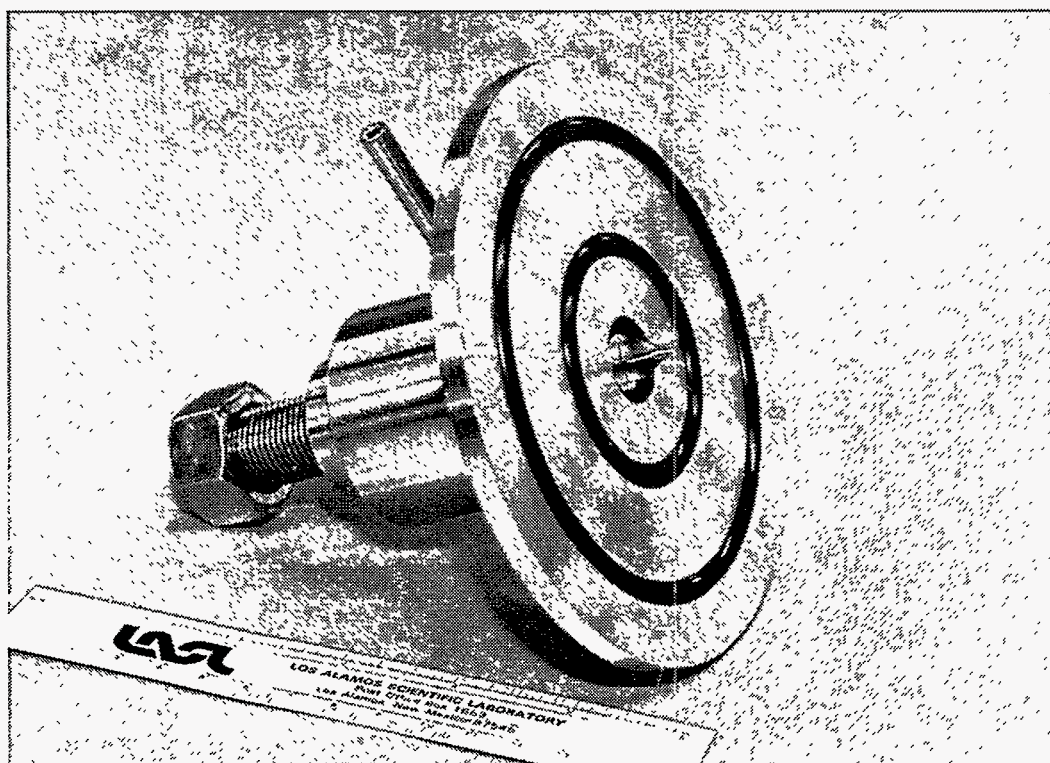


Fig. 3-6. Waste can penetrator components.

A set screw was used to attach the penetrator head (308) firmly to the mounting plate (301) of the head-gas sampler for "bagout cans", (Fig. 3-2). The penetrator head mounting plate was attached to the sampling base using the knurled nuts (302) which turned freely on the threaded support rods. The other end of the flexible tubing was attached in a similar manner to the valving assembly at valve V2. A light coating of vacuum grease was put on the 3/16" "O" rings (309) located on the surface of the penetrator head, which mated with the can surface. The knurled nuts (302) were adjusted to position the penetrator head in a manner that gave enough space between the base plate (300) and the penetrator head (308) to insert the can being sampled. The

penetrator rod (305) was screwed out to make sure the penetrator tip (307) was not protruding beyond the surface of the penetrator head mating surface.

To eliminate any potential source of heat or spark, two steps were taken in the design of the penetrator. First was the use of a very fine thread on the penetrator rod so that insertion of the tip into the metal can proceeded very slowly. The metal deforms and then slowly tears under the pressure. Second was the designing of the penetrator screw so a ratchet wrench could be used to screw the penetrator into place. Use of the ratchet forced the operator to move the screw in steps, which prevented rapid puncturing and allowed any heat build-up from the friction of each step to dissipate through the large heat sink of the lid itself.

The sample can was placed on the base plate and the position adjusted so a smooth, undented and unscratched surface was under the 3/16" "O" rings. In several cases this meant turning the can upside-down on the plate and puncturing the bottom to obtain the sample. Once the can was in position, the knurled nuts (302) below the plate supporting the penetrator head were screwed down the support rods until the penetrator head was resting on the can surface. The upper knurled nuts (302) were then snugged down to level the penetrator head and turned a final half-turn. A plastic bag with a hole for the penetrator rod to pass through was draped over the entire apparatus and a flexible tube from the argon source was taped to inside of the bag. Fig. 3-7 shows the assembled apparatus ready for sampling.

To check for air in-leakage during sampling, the plastic bag was filled with argon to displace the air around the sealing surfaces of the penetrator and slip lid interfaces of the cans. Because the Ar/N₂ atmospheric ratio is not altered by reactions in the waste, in-leakage of Ar could be detected and corrected for. If the Ar/N₂ ratio varied from natural after sampling, a leak was indicated, either around the "O" ring seals or at the slip lid junction. The excess argon was subtracted from the measurements so the subsequent gas distribution was indicative of the head-gas before any external gases entered the waste package.

Significant leakage (up to 20% Ar) was detected in a few cases. The main source of leakage is believed to be at the can lid o-ring seal. In retrospect, a caulking-cord seal would probably have provided a better seal because of surface irregularities in the can lids.

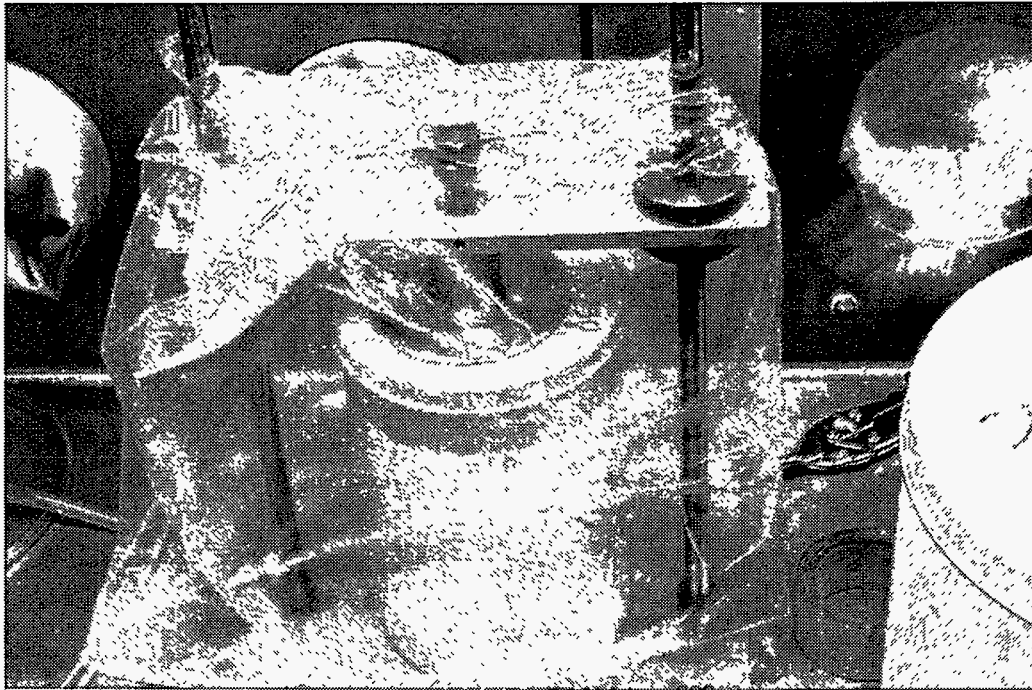


Fig. 3-7. Assembled apparatus for piercing and sampling waste cans.

After set-up, the steps for obtaining the samples were as follows:

Outer Can Sampling

1. Open valve V1 (to vacuum) and observe vacuum gauge on manifold. Once maximum vacuum is achieved, open valve V2 (to penetrator head) and again observe vacuum on gauge. Upon reaching maximum vacuum, close valve V1 and observe vacuum gauge for several minutes to be sure system is not leaking.
2. While making vacuum test, flood bag with argon and continue a slow flow of argon into bag during sample taking.
3. Use the ratchet wrench on penetrator screw nut, Fig. 3-2 (305), and turn penetrator screw clockwise while observing vacuum gauge on manifold. When penetration is made, quickly close valve V2, maintaining a partial vacuum in manifold tubing. This prevents further evacuation of the waste container when the sample cylinder is opened and so reduces the amount of cover gas drawn into the system.
4. Slowly open valve, SV, Fig. 3-3, on appropriate sample cylinder and allow sample to expand into cylinder. Close SV valve.
5. Wait 1 minute and take a second sample of the same gas into another cylinder, as in step 4.
6. With valve V2 closed, remove flexible vacuum tubing from sample penetrator, loosen upper knurled nuts (302) so penetrator support plate (303) can be raised high enough to remove can. Use lower knurled nuts (302) to hold support plate in place.
7. Remove the can from the sample assembly and remove the slip lid from the can so the plastic bag is accessible.

Bag Sampling

8. Use double-stick tape to attach a septum to the plastic bag.
9. Open valve V1 (to vacuum) and observe pressure gauge on manifold until maximum vacuum is obtained, then open valve (to bag penetrator) and again pump to maximum vacuum.
10. Arrange bag with argon around can containing bag and continue to flood the area to be sampled with argon.
11. Insert beveled sampling needle tip halfway through the septum and open valve V4. When maximum vacuum is obtained, close valve V1, then insert the tip the remainder of the way through the septum and into the bag. Using the small needle size, the sample will very slowly expand into the manifold system. Close valve V4 at about 10 inches of vacuum and take two samples using the same procedure as in steps 4 and 5.
12. Close valve V3 and remove beveled sampling needle from bag.

Inner Can Sampling

13. Open bag and remove inner can.
14. Place inner can on the can sampling device using the same set-up procedures used for the outer can.
15. Sample inner can using steps 1-6.
16. Turn off argon cover gas supply.
17. Remove sample cylinders. (Health physics personnel to be present).

Sample measurements were made using the same Finnigan-MAT model 271 mass spectrometer used for the diffusion measurements discussed earlier in this paper. Two gas mixtures, a Matheson standard gas with 509 ppm H₂ and 514 ppm He in N₂ and a tank with a nominal 6.6% H₂ in Ar were analyzed in parallel with each batch of samples as a quality control for the measurements.

Experimental - Test Barrel

A 55-gallon test drum was prepared to evaluate the mathematical model* for predicting H₂ concentrations using diffusion coefficients and G-values** measured at LANL. The drum was loaded with a total of 7.1 grams of ²³⁸Pu amassed in either of the cellulose waste items measured in the penetration experiments. The loading configuration with the amount of ²³⁸Pu per item is illustrated in Fig. 3-8. The quantity of ²³⁸Pu in each can was verified by calorimetry measurements. The Y(x) values for each space represent the independent volumes used in the modeling equation for predicting the gas distribution, particularly hydrogen accumulation. Each of the waste cans was fitted with a NFT 030 carbon filter and inserted into a polyethylene bag with a "horse-tail" on each end of the bag. The bagged cans were placed inside the 55-gallon barrel which was lined with two 14-mil PVC bags. The liner bags were folded over, but were not taped. A NFT 013 carbon filter was installed into the lid of the drum.

*Information received from Miles Smith, Benchmark Environmental Corp., 4501 Indian School Rd. N.E., Ste. 105, Albuquerque, N.M. 87110

**Information received from E.L. Callis, CST-8, MS G740, Los Alamos National Laboratory, Los Alamos, N.M. 87545

To sample the drum headspace, the custom-built head-gas sampler, Fig. 3-5, was used. This device, which is simply a small chamber with an attached evacuated sample cylinder, is sealed over the drum filter vent and allows non-intrusive sampling of the drum headspace. Because of the small volume of the sampling chamber ($\approx 200\text{cc}$) and the fairly rapid H_2 flow characteristics of the filter vent, the H_2 level in the chamber equilibrates with the headspace in a manner of minutes. Heavier gases, of course, require longer equilibration times. An o-ring formed from caulking cord was placed around the raised flange on the bottom and an evacuated sample tube was connected to the quick disconnect on top. The assembly was placed over the carbon filter on the drum lid and pressed down to compress the caulking cord and achieve a good seal. After a minimum time of four hours, to allow equilibration of the gas between the headspace gas in the barrel and sample device volume, the valve on the sample cylinder was opened, the gas expanded into the cylinder, and the valve closed. Analyses were done on the MAT 271 mass spectrometer.

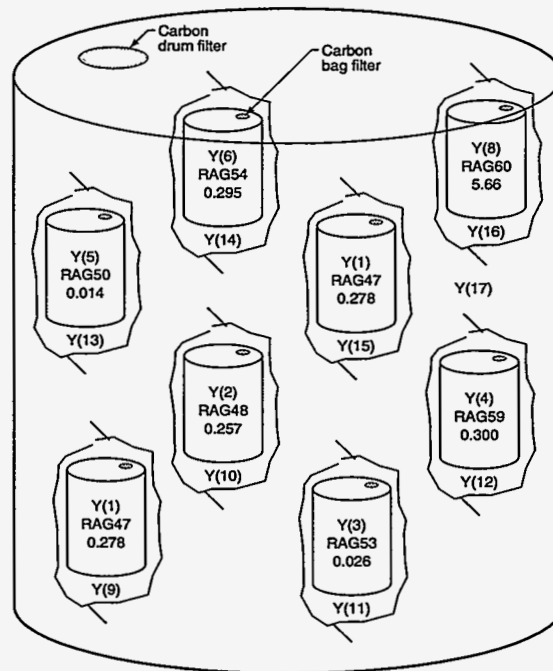


Fig. 3-8. Configuration of test drum used to verify H_2 content prediction model.

Results and Discussion

Table 3-1 gives a summary of the results obtained from each of the three layers for ten cellulose matrix and two polyethylene matrix bagout waste packages. All packages had been assembled at least six months before the penetration experiments were performed, with some of them being over one year old. Results are expressed as mol % of H_2 , CH_4 , N_2 , CO , O_2 , Ar , and CO_2 measured in each layer. Rags 49 and Rags 60 exhibited the highest degree of H_2 retention with 20.4 % in the outer can and 17.0% in the bag respectively. With the exception of Rags 50, which contains only 14 mg of ^{238}Pu , O_2 content is depleted relative to atmospheric levels. Conversely, CO_2 is elevated relative to atmospheric levels in all instances except for Rags 50. Whenever the

H₂ is greater than 1.25%, measurable levels of CO are obtained for a cellulose matrix. For CO in a polyethylene matrix, a definitive pattern was not observed. All of these gas species are consistent with what is being observed in other current work at this Laboratory* and in previous work by others (Reference 2, 3). The reduction of O₂ present in the various layers would seem to reduce the possibility of obtaining a combustible mixture from the H₂ produced.

*Information received from E.L. Callis, CST-8, MS G740, Los Alamos National Laboratory, Los Alamos, N.M. 87545

**Table 3-1. Gas Compositions Measured in ²³⁸Pu Waste Packages.
All packages were at least 6 months old.**

Sample ID	mg ²³⁸ Pu	Layer	%H ₂	%CH ₄	%N ₂	%CO	%O ₂	%Ar	%CO ₂
Rags 49	700	Outer Can	20.41	0.27	46.78	6.90	4.48	0.55	20.61
		Bag	11.23	0.07	59.20	4.76	11.07	0.71	12.96
		Inner Can	10.50	0.06	59.63	4.87	11.23	0.71	13.00
Rags 56	600	Outer Can	2.93	0.03	74.50	2.48	11.45	0.89	7.72
		Bag	3.52	0.04	74.72	3.58	8.48	0.89	8.77
		Inner Can	4.68	0.03	72.68	4.91	3.55	0.86	13.29
Rags 57	226	Outer Can	1.57	0.04	76.90	1.81	11.72	0.92	7.04
		Bag	2.01	0.04	77.75	2.71	8.77	0.93	7.79
		Inner Can	2.31	0.04	77.40	3.21	5.82	0.92	10.30
Rags 47	278	Outer Can	1.21	0.09	79.82	0.00	13.96	0.95	3.97
		Bag	1.57	0.10	81.84	0.45	11.12	0.97	3.95
		Inner Can	1.32	0.16	81.69	0.43	11.23	0.97	4.20
Rags 48	257	Outer Can	1.43	0.04	77.00	1.61	12.82	0.92	6.18
		Bag	1.17	0.09	79.81	0.53	13.20	0.94	4.26
		Inner Can	1.31	0.08	79.31	0.75	11.47	0.96	6.12
Rags 50	14	Outer Can	0.02	0.08	78.43	0.00	20.38	0.93	0.16
		Bag	0.03	0.08	78.60	0.00	20.16	0.94	0.19
		Inner Can	0.02	0.08	78.62	0.00	20.13	0.94	0.21
Rags 53	26	Outer Can	0.29	0.09	78.99	0.00	18.08	0.94	1.61
		Bag	0.32	0.09	79.29	0.00	17.61	0.94	1.75
		Inner Can	0.30	0.09	79.18	0.00	17.76	0.94	1.73
Rags 54	295	Outer Can	2.88	0.12	75.22	1.83	10.15	0.98	8.82
		Bag	3.26	0.13	75.60	2.96	7.75	0.90	9.40
		Inner Can	2.76	0.13	76.06	2.30	9.75	0.91	8.09
Rags 60	5660	Outer Can	6.24	0.20	58.46	4.32	13.30	0.70	16.78
		Bag	17.04	0.81	33.23	19.16	4.10	0.39	25.27
		Inner Can	12.92	0.47	44.88	13.59	8.09	0.54	19.51
Rags 59	300	Outer Can	5.11	0.03	75.71	3.45	5.01	0.90	9.79
		Bag	6.46	0.04	75.69	4.46	1.49	0.90	10.96
		Inner Can	5.94	0.04	75.72	4.12	3.17	0.90	10.11
PLS 131	200	Outer Can	2.50	0.00	80.55	0.00	6.37	0.96	9.62
		Bag	2.69	0.00	82.29	0.00	4.04	0.98	10.00
		Inner Can	2.33	0.00	82.09	0.00	5.01	0.98	9.59
PLS 132	800	Outer Can	1.65	0.00	82.61	0.65	6.71	1.21	7.17
		Bag	2.12	0.00	84.68	0.99	2.72	1.01	8.48
		Inner Can	1.86	0.00	83.95	0.87	4.48	1.00	7.84

Because Rags 49 yielded the largest volume of H₂, it was used to test the effectiveness of adding the NFT 030 carbon filter to the lid. The inner can, as originally found in the package, was taped with two layers of 2" plastic tape. In all subsequent experiments with this sample, the same taping procedure was used. The results of these experiments are summarized in Fig. 3-9 and Table 3-2. Initially, H₂ concentrations of greater than 10% were observed in all layers. After the first sampling, a new lid was put into place on just the inner can and a sample taken after ten days, which showed a buildup of H₂ to 1.2%. The experiment was repeated, but left for 42 days before sampling with the same H₂ increase to 1.2%. These values confirmed the observations made of the other cans in these penetration experiments, showing the leak rate of the unvented slip lid cans to H₂ diffusion is highly variable and cannot be assumed to leak H₂ freely.

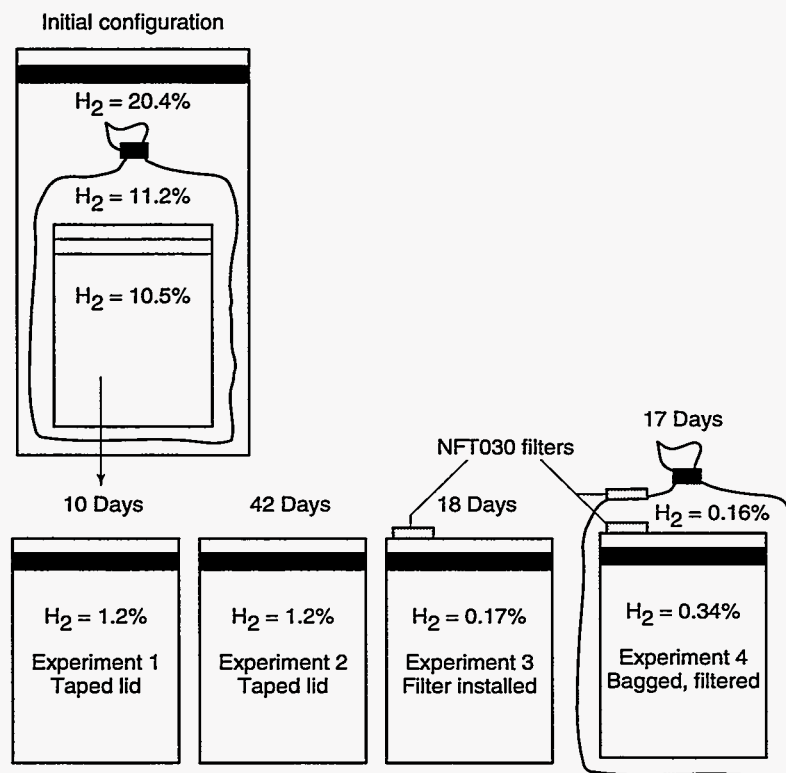


Fig. 3-9. Hydrogen levels measured in waste package (Rag 49) with various layers of confinement (data in Table 3-2).

Table 3-2. Waste Package (RAG 49) Gas Composition Measurements Before and After Installation of Carbon-Composite Filter Vents
(contents \approx 700mg ^{238}Pu on cheesecloth)

Exp. No.*	Samp ID **	Equil. Time/ Days	%H ₂	%CH ₄	%H ₂ O	%N ₂	%CO	%O ₂	%Ar	%CO ₂
1-a	IC	10	1.21	0.016	0.12	79.56	1.70	10.25	0.95	6.32
1-b	IC	10	1.19	0.018	0.067	79.53	1.73	10.37	0.95	6.21
2-a	IC	42	1.17	0.011	0.056	77.32	1.90	9.70	0.92	8.99
2-b	IC	42	1.08	0.013	0.13	77.32	1.77	10.49	0.92	8.40
3-a	IC	18	0.17	0.029	0.07	78.02	0.41	18.57	0.93	1.98
3-b	IC	18	0.16	0.038	0.11	77.94	0.32	18.95	0.93	1.66
4-a	VB	17	0.16	0.040	0.06	78.39	0.28	18.84	0.93	1.36
4-b	VB	17	0.16	0.041	0.08	78.30	0.34	18.85	0.93	1.36
4-c	IC	17	0.36	0.032	0.12	78.07	0.68	16.18	0.93	3.75
4-d	IC	17	0.32	0.038	0.46	77.97	0.61	16.86	0.93	3.26

* Experiment #1 - Inner can was resealed on 12/08/92, resampled 12/18/92. Two layers yellow tape.

Experiment #2 - Inner can was resealed on 12/18/92, resampled 01/29/93. Two layers yellow tape.

Experiment #3 - Inner can was resealed on 01/29/93, resampled 02/16/93. Lid had carbon filter. Taped.

Experiment #4 - Inner can resealed and bagged, both with carbon filters at 1030 on 02/16/93, resampled on 03/05/93. Taped.

** IC=INNER CAN VB = VENTED BAG

To test the effectiveness of the NFT 030 filters, one was fitted on an inner can lid and used to reseat the Rags 49 sample. After 18 days, the H₂ in the can was 0.17%, about a factor of 10 lower than in the unvented case. As a final step, the can was resealed with a filtered lid and put into a polyethylene bag which had been fitted with a NFT 030 filter and closed with a single horse-tail. The H₂ concentrations were 0.34% in the can and 0.16% in the bag after 17 days. Note in Table 3-2, the O₂ was depleted and the CO₂ enriched similar to what had been seen in closed systems (References 2, 3, 5, 6). The limit of these reactions with a filter in place was not studied in these experiments.

The gaseous distribution in the headspace of the test barrel for a test period of 122 days is given in Table 3-3. In the first few days the H₂ concentration rose rapidly to about 0.8%, and then changed fairly slowly. The data is plotted in Fig. 3-10 for comparison with a model[†] for predicting the H₂ levels developed in conjunction with this experiment. The agreement of the

[†]Information received from Miles C. Smith, Benchmark Environmental Corp., 4501 Indian School Rd., N.E., Ste. 105, Albuquerque, NM 87110

measured vs. predicted is reasonably good, considering the nature of the experiment. The amount of scatter is larger than one might expect, and may be due to temperature and atmospheric pressure variations over the 3 month period, which have not been investigated. In retrospect, much more frequent samplings should be made in an experiment of this type to facilitate tracking short-term variations. In general, however, these measurements validate the use of the bag filters as a means of reducing the buildup of H₂ in TRU waste packages.

Table 3-3. Test Barrel Gas Composition Measurements

Elapsed Time Days	%H₂	%CH₄	%N₂	%CO	%O₂	%Ar	%CO₂	~H₂O
0.17	0.030	<0.001	77.96	0.04	20.82	0.97	0.10	0.23
4.17	0.653	<0.09	76.88	0.38	19.04	0.94	2.02	0.31
11.11	0.750	<0.10	76.81	0.76	17.47	0.94	3.17	0.50
18.11	0.750	<0.10	76.62	0.95	16.64	0.94	3.97	0.30
23.98	0.934	<0.09	76.34	1.20	15.66	0.94	4.84	0.15
35.13	0.811	<0.01	75.98	1.16	15.73	0.98	5.34	0.20
48.12	0.808	<0.01	76.30	0.95	15.11	0.95	5.89	0.21
56.11	1.499	<0.01	75.54	1.64	13.06	0.92	7.32	0.25
67.11	0.972	<0.01	75.72	1.37	14.30	0.92	6.71	0.29
122.07	0.735	<0.01	76.46	1.13	14.38	0.95	6.33	0.086

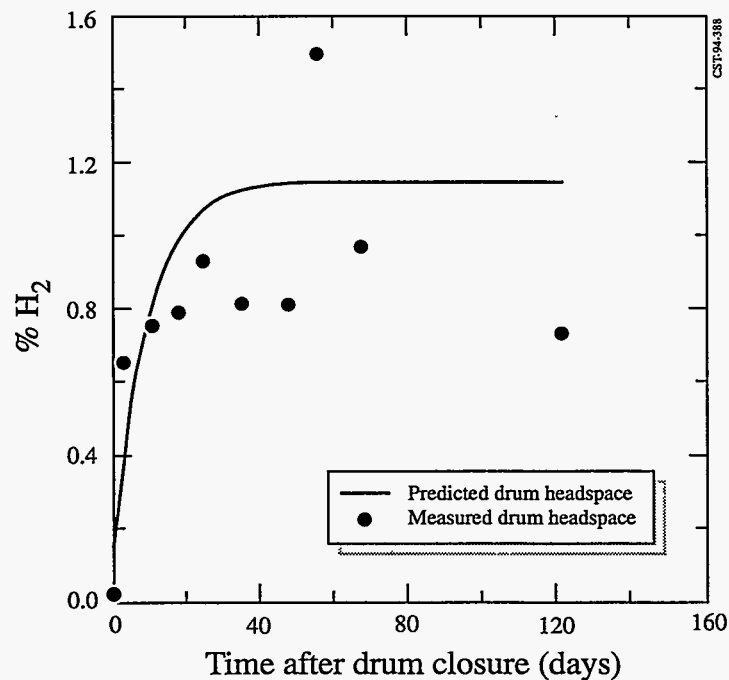


Fig. 3-10. Predicted and measured H₂ concentration in test drum headspace.

Quality Assurance Tests

To verify the operation of the MAT-271 mass spectrometer, two gas mixtures, a Matheson standard containing 509 ppm H₂ in N₂ and a tank mixture with a nominal value of 6.64% H₂ in Ar were analyzed in conjunction with the samples. The results are given in Table 3-4. For an H₂ concentration of 509 ppm the long term precision of the measurement at the 95% confidence interval was ± 54 ppm with a mean bias of +13 ppm. For the H₂/Ar mixture concentration of 6.64%, the long term precision (%RSD) of the measurements was $\pm 1.5\%$, with no significant bias.

Table 3-4. Penetration and Barrel Samples Analyzed by QA Standards

Certified Value	509 ppm H ₂	Nominal Value	6.64% H ₂
Date	H ₂ ppm	Date	H ₂ %
12/07/92	521	12/09/92	6.74
02/16/93	530	03/08/93	6.58
03/08/93	487	03/24/93	6.80
03/24/93	525	08/31/93	6.70
08/30/93	534	09/28/93	6.56
08/31/93	520	10/01/93	6.58
09/15/93	530		
09/28/93	509		
10/01/93	590		
Mean	527	Mean	6.66
Std. Dev.	± 27	Std. Dev.	0.10

3.2 CALCULATION OF REVISED TRUPACT-II WATTAGE LIMITS

This section shows the impact on the decay heat limits of TRUPACT-II shipping categories resulting from the use of filtered bags. The analyses presented here assume that each bag in a TRU waste package is filtered with the bag filter. As discussed previously, the diffusion characteristic of the bag filter at room temperature is 1.12×10^{-5} mol/s/mol fraction. This corresponds to a resistance of 89,286 s/mol. As explained in Appendix 3.6.12 of the TRUPACT-II SARP (NuPac 1992), two different values of a confinement layer resistance are needed (one value for Waste Type I and a second value for Waste Types II and III). Our experimentally determined value represents the resistance of the bag filter for Waste Types II and III. The filtered bag resistance value for Waste Type I we calculated to be 123,726 s/mol, using the equations listed in Appendix 3.6.12 of the TRUPACT-II SARP (NuPac 1992). Table 3-5 summarizes the confinement layer resistances used in the analyses.

Table 3-5. Confinement Layer Resistances

Confinement Layer	Waste Type I Resistance (s/mol)	Waste Type II and III Resistance (s/mol)
Drum Filter	729,000	526,000
SWB Filter	375,000	270,000
TDOP Filter	375,000	270,000
Inner Bag	2,399,000	1,792,000
Liner Bag	214,000	214,000
SWB Liner	126,000	126,000
Punctured Drum Liner	19,600	19,600
Filtered Bag	124,000	89,000

Maximum allowable gas generation rates and decay heat limits for each shipping category were calculated using equations (4) and (5) of Section 3.4.4.4.3 of the TRUPACT-II (NuPac 1992) respectively. In calculating the effective resistance to the release of hydrogen, the values of the resistance of the filtered bag listed in Table 3-5 above were used in place of the resistances of the liner bag and inner bag confinement layers. Table 3-6 summarizes the results of the analyses that we performed. In Table 3-2 the decay heat limits listed in Table 3.4.4.4-1 of the TRUPACT-II SARP are presented for each shipping category along with the decay limits that were calculated assuming each bag is filtered. The last column of Table 3-6 lists the corresponding percentage increases in the decay heat limits for each shipping category as result of venting each bag with a single filter.

Table 3-6. Decay Heat Limits and Increases in Decay Heat Limits with Filtered Bag Use

Payload Shipping Category	Original SARP Decay Heat Limits Watts per Generator	Decay Heat Limits With Filtered Bags Watts per Generator	Percentage Increase in Decay Heat Limit
I.1A0	0.2059	0.2059	0%
I.1A1	0.1796	0.1899	6%
I.1A2	0.1593	0.1761	11%
I.1A3	0.0465	0.1643	253%
I.2A0	0.2534	0.2534	0%
I.2A1	0.2211	0.2337	6%
I.2A2	0.1961	0.2168	11%
I.2A3	0.0573	0.2022	253%
I.2A4	0.0418	0.1894	353%
I.3A0	0.8237	0.8237	0%
I.3A1	0.7185	0.7595	6%
I.3A2	0.6372	0.7045	11%
I.3A3	0.1862	0.6570	253%
I.3A4	0.1358	0.6155	353%
II.2A0	0.2250	0.2250	0%
II.2A1	0.1923	0.2101	9%
II.2A2a	0.1680	0.1971	17%
II.2A2	0.0868	0.1971	127%
II.2A3	0.0561	0.1856	231%
II.2A4	0.0414	0.1753	324%
II.1A5	0.0328	0.1662	406%
II.1A6	0.0272	0.1579	481%
II.2AM	40.0	40.0	0%
III.1A0	0.1125	0.1125	0%
III.1A1	0.0962	0.1051	9%
III.1A2a	0.0840	0.0985	17%
III.1A2	0.0434	0.0985	127%
III.1A3	0.0280	0.0928	231%
III.1A4	0.0207	0.0877	324%
III.1A5	0.0164	0.0831	406%
III.1A6	0.0136	0.0790	481%

**Table 3-6. Decay Heat Limits and Increases in Decay Heat Limits with Filtered Bag Use
(continued)**

Payload Shipping Category	Original SARP Decay Heat Limits Watts per Generator	Decay Heat Limits With Filtered Bags Watts per Generator	Percentage Increase in Decay Heat Limit
I.1B0	0.1456	0.1456	0%
I.1B1	0.1320	0.1374	4%
I.1B2	0.1206	0.1301	8%
I.1B3	0.0426	0.1235	190%
I.2B0	0.1792	0.1792	0%
I.2B1	0.1624	0.1691	4%
I.2B2	0.1485	0.1601	8%
I.2B3	0.0524	0.1520	190%
I.2B4	0.0391	0.1446	270%
I.3B0	0.5824	0.5824	0%
I.3B1	0.5278	0.5496	4%
I.3B2	0.4826	0.5202	8%
I.3B3	0.1702	0.4939	19%
I.3B4	0.1272	0.4701	270%
II.1B0	0.1711	0.1711	0%
II.1B1	0.1515	0.1623	7%
II.1B2a	0.1359	0.1544	14%
II.1B2	0.0774	0.1544	100%
II.1B3	0.0520	0.1473	183%
II.1B4	0.0391	0.1407	260%
II.1B5	0.0314	0.1348	330%
II.1B6	0.0262	0.1293	394%
II.2BM	40.0	40.0	0%
III.1B0	0.0855	0.0855	0%
III.1B1	0.0757	0.0812	7%
III.1B2a	0.0680	0.0772	14%
III.1B2	0.0387	0.0772	100%
III.1B3	0.0260	0.0736	183%

**Table 3-6. Decay Heat Limits and Increases in Decay Heat Limits with Filtered Bag Use
(continued)**

Payload Shipping Category	Original SARP Decay Heat Limits Watts per Generator	Decay Heat Limits With Filtered Bags Watts per Generator	Percentage Increase in Decay Heat Limit
III.1B4	0.0196	0.0704	260%
III.1B5	0.0157	0.0674	330%
III.1B6	0.0131	0.0646	394%
I.1C0	0.9127	0.9127	0%
I.2C0	1.1234	1.1234	0%
1.3C0	3.6510	3.6510	0%
II.1C0	1.0201	1.0201	0%
II.1C1	0.7026	0.7026	0%
II.1C2	0.5358	0.5358	0%
II.1C3	0.1222	0.4585	275%
II.1C4	0.0690	0.4007	481%
III.1C0	0.5100	0.5100	0%
III.1C1	0.3513	0.3513	0%
III.1C2	0.2679	0.2679	0%
III.1C3	0.0611	0.2292	275%
III.1C4	0.0345	0.2003	481%

We performed a sensitivity analysis to evaluate the impacts of multiple filters on decay heat limits assuming from one to five bag filters on each bag. The analyses were performed for the two shipping categories in each Waste Type that have the lowest allowable decay heat limits. For Waste Type I, the shipping category of interest was I.2A4 while for Waste Types II and III, the shipping category of interest is III.1A6. Effective resistances used in the decay heat limit calculations were based on dividing the resistance values for the filtered bag listed in Table 3-5 by the appropriate number of filters. The results of the sensitivity analyses are summarized in Table 3-7 and depicted graphically in Fig. 3-11 (for Waste Type I) and in Fig. 3-12 (for Waste Type II and III).

Table 3-7. Decay Heat Limits vs. Number of Filters on Each Bag

Number of Filters	Decay Heat Limit (W) Shipping Category I.2A4	Decay Heat Limit (W) Shipping Category III.1A6
0	0.0418	0.0136
1	0.1894	0.0790
2	0.2168	0.0928
3	0.2278	0.0985
4	0.2337	0.1017
5	0.2374	0.1037

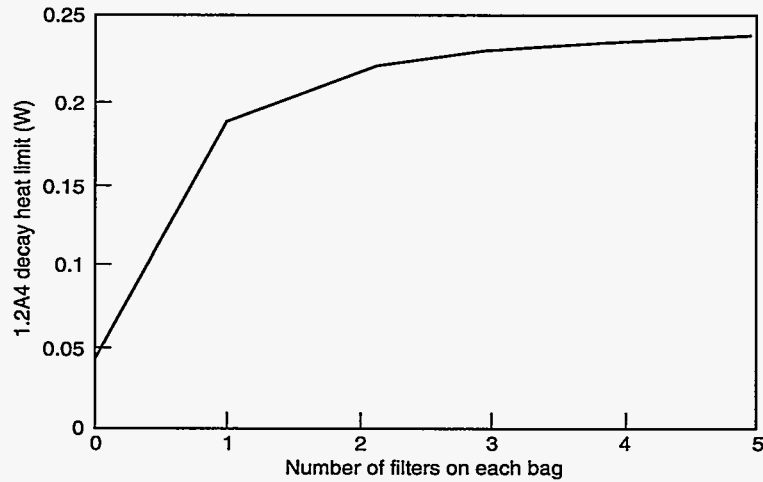


Fig. 3-11. Shipping category I.2A4 decay heat limit vs. number of filters on each bag.

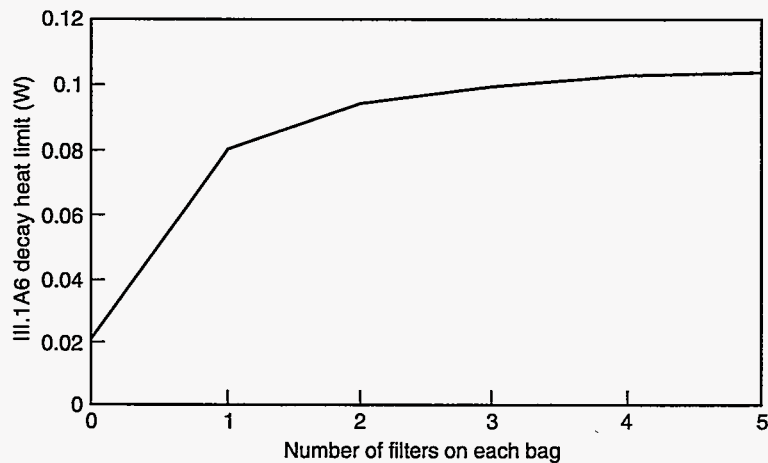


Fig. 3-12. Shipping category III.1A6 decay heat limit vs. number of filters on each bag.

4.0 CONCLUSIONS

Our measurements on actual waste packages demonstrate that commonly used waste packaging materials (bags and cans) offer significant resistance to hydrogen venting and that the use of carbon-composite filters are effective in reducing the buildup of H₂ to potentially combustible levels.

We evaluated the impact of the use of filtered bags on decay heat limits for various waste categories. The analysis assumed that each bag is filtered with a single filter whose diffusion characteristic was experimentally determined to be 1.12×10^{-5} mol/s/mol fraction. The percentage increase in heat limits ranges from 4 percent in the case of shipping categories with one liner bag to 481 percent for the case of shipping categories with six layers of bags. Sensitivity analyses

have been performed to evaluate the impacts on decay heat limits that with multiple filters on each bag. The sensitivity analyses indicate that minimal improvements in the allowable decay heat limits would be obtained by using more than two filters per bag. It should be noted that decay heat limits for the "d" shipping categories (i.e., The experimental bin shipping categories) were not calculated because the bin program has been canceled. Based on the analyses presented in this report, allowable decay heat limits can be dramatically increased for those shipping categories with multiple layers of inner bags if these bags are filtered. The use of filtered bags would require that additional shipping categories be developed and an amendment to the TRUPACT-II certificate of compliance.

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Appendix 1. WH-05, 07, and 08 Filter Testing

In November, 1995, after a draft of the present report was circulated, the authors learned from Dr. Murthy Devarakonda* of previous measurements taken of similar filters, performed by Peterson, et al.,** in 1990. The hydrogen diffusivity values reported by Peterson were significantly lower (by more than a factor of two) than the values reported in the present work on newer filters. To try to resolve the apparent discrepancy, Dr. Devarakonda was able to supply three of the original filters, a WH-05, 07 and 08, tested by Westinghouse in 1990, for comparison testing at LANL.

Table 1 shows a summary of these measurements. As a control, we also re-measured one of the filters that we had previously measured in 1993 (LANL 28), obtaining essentially the same result. Figure 1 shows a typical leak rate plot from which the coefficient is determined and indicates the good precision with which these measurements can be made ($< 0.5\%$). Our results on filters WH-05, 07, and 08 are about 40 % higher than those reported by Peterson. However, this is much better agreement than the factor of 2 between our measurements on later filters (apparently manufactured 6/93), and these filters, apparently manufactured 11/89. Obviously the filters are different.

The first (and only) difference that was apparent was that the older filter housing (WH-05) has only 5 holes, while the newer one has 9 holes (see Figure 2). To determine what effect the number of holes might have on the diffusion characteristics, an experiment was performed in which some of the holes in one of the newer filters (LANL 28) were sealed, and the diffusion coefficient measured. These data are listed in Table 1 and plotted in Figure 3.

These results, rather surprising, show that the diffusion rate is directly proportional to the number of holes (or total cross-sectional area). We had assumed that the filter itself would be the controlling factor.

Hence, it appears that differences in the filter housings are enough to account for most of the discrepancy between the data. The remaining 40 % or so could be due to actual differences in the filter media. In addition, this work indicates that the diffusion rate could probably be increased significantly by increasing the total cross sectional area of openings in the filter housing by increasing the number of holes in the filter housing, or by other means.

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** S.H. Peterson et al., "Determination of Flow and Hydrogen Diffusion Characteristics of Carbon Composite Filters Used at the Waste Isolation Pilot Plant," (P.O. 75WRS369171Z), Westinghouse STC, Pittsburgh, Pa., 1 March 1990 (not published in open literature).

Diff. Coeff., m/sec/mol fract.

Filter ID	This Work	Peterson, et al.
LANL 28 (6/93 LANL value = $1.105E - 5$)	$1.08E - 05$	
WH-05 (11/89)	$6.16E - 06$	$4.32E - 06$
WH-07 (11/89)	$6.30E - 06$	$4.17E - 06$
WH-08 (11/89)	$5.58E - 06$	$4.32E - 06$

Hole Sealing Tests, LANL 28

Holes Left Open	Diff. Coeff., mol/sec/mol fract.	
9	$1.08E - 05$	(All open)
5	$6.68E - 06$	Same number of holes as old filters (11/89). Note similarity in value.
3	$4.28E - 06$	
0	$3.78E - 07$	

Table 1. January '96 Filter Measurements Summary

FILTER WH-08 11/89

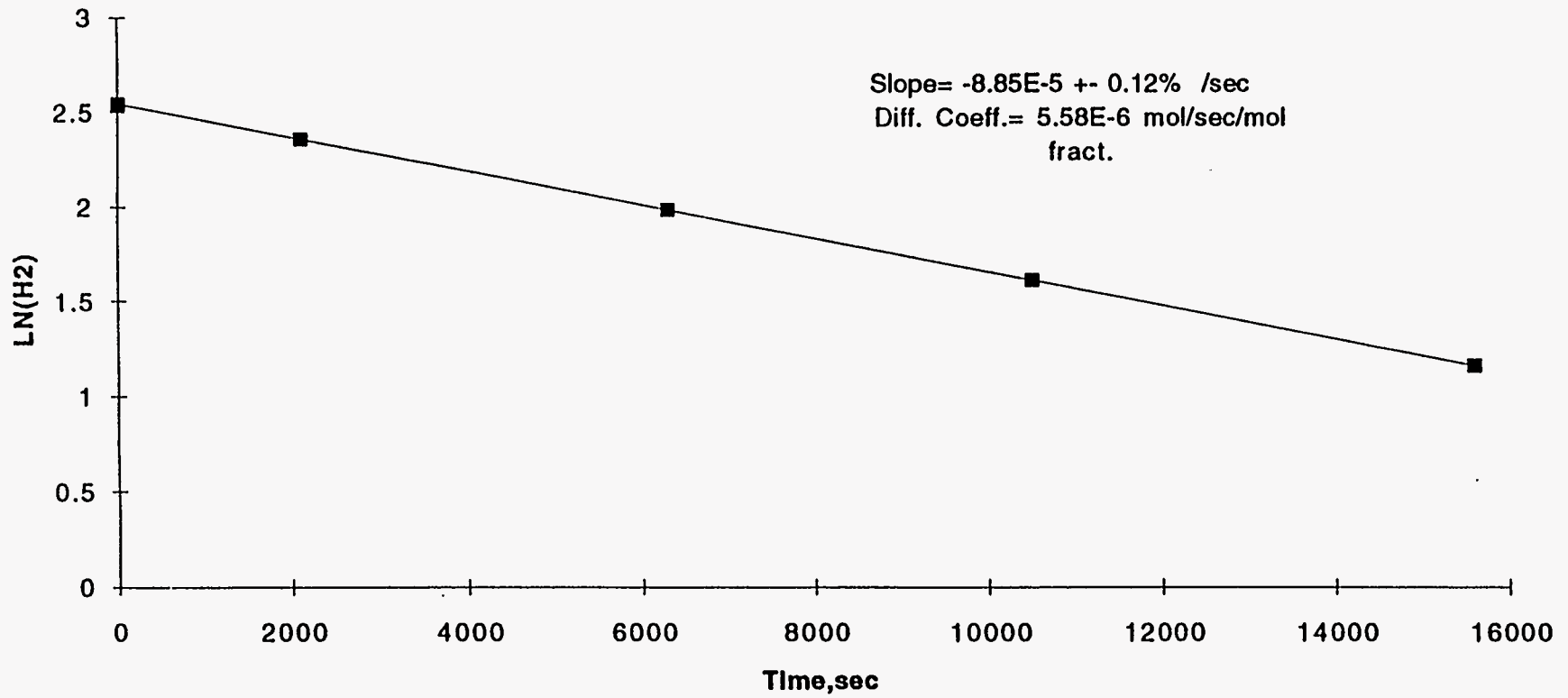


Figure 1. WH-08 Filter (11/89)

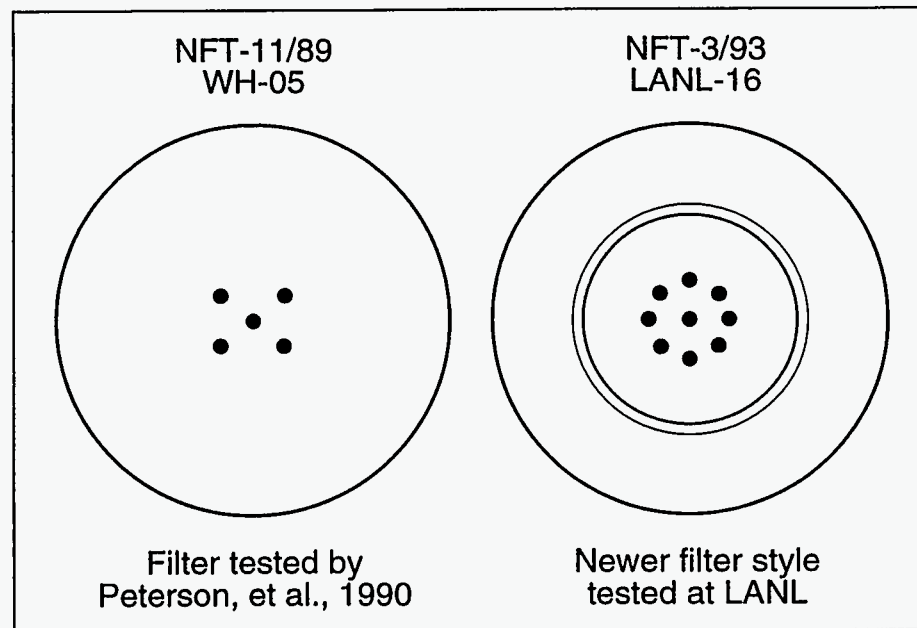


Figure 2. WH-05 filter and LANL-16

Relative diffusion rate VS # of open holes in filter
Filter # LANL 28 Jan 1996 Data

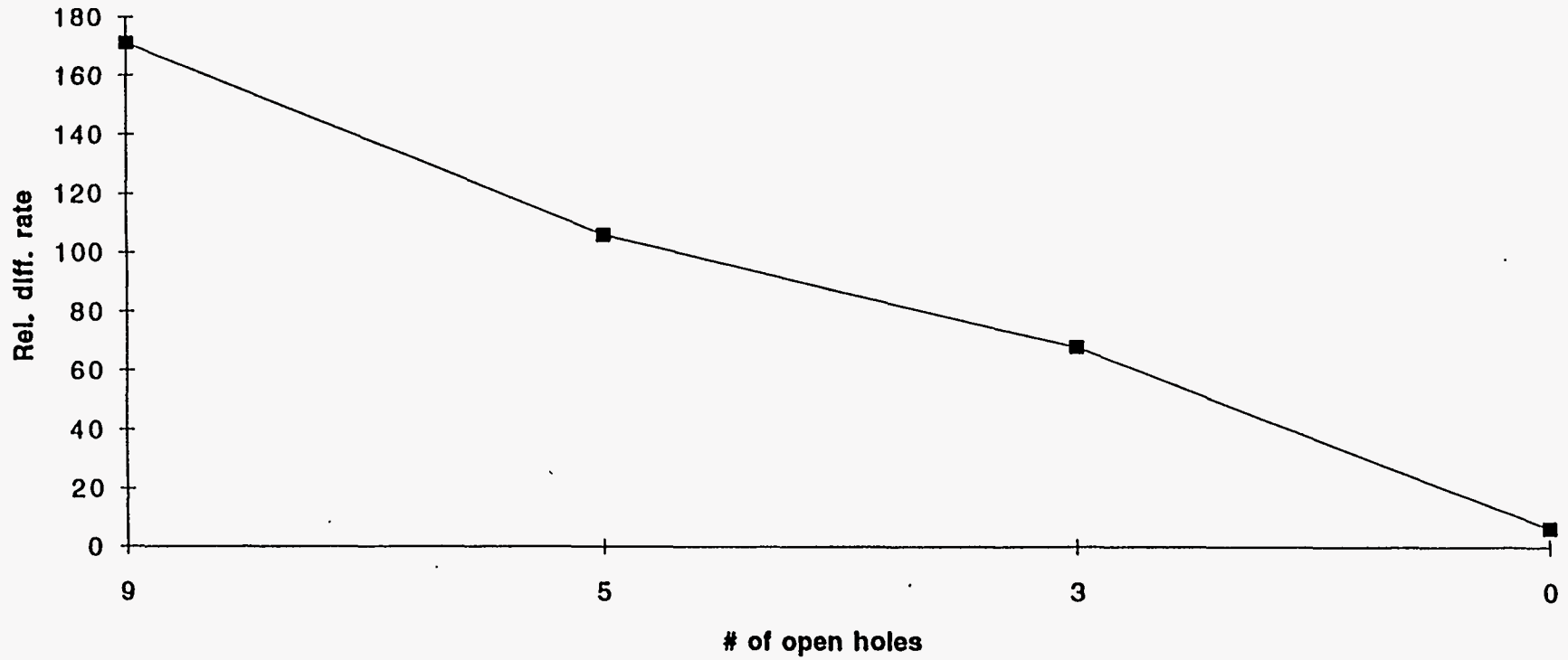


Figure 3. Relative diffusion rate vs. number of open holes in Filter LANL 28 (Data from 1-96)

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