NUREG/CR-0403
ORNL/NUREG/TM-246

High Temperature Testing of Smoke Detector Sources

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Prepared for the U.S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
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Under Interagency Agreement DOE 40-550-75

OAK RIDGE NATIONAL LABORATORY
OPERATED BY UNION CARBIDE CORPORATION · FOR THE DEPARTMENT OF ENERGY

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161

NUREG/CR-0403
ORNL/NUREG/TM-246

Contract No. W-7405-eng-26

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Manuscript Completed — September 12, 1978
Date Published — September 1978

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NRC FIN No. B0186

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N. H. Cutshall, I. L. Larsen, and F. N. Case

Radioisotope Department
Operations Division

Oak Ridge National Laboratory
Oak Ridge, Tennessee

ABSTRACT

The quantities of ^{241}Am released from smoke detector sources when heated to temperatures up to 1200°C have been measured under laboratory conditions designed to simulate actual fire conditions. Sources removed from smoke detector units purchased on the open market, as well as sources obtained directly from source manufacturers, were tested. In addition, complete smoke detector assemblies containing sources were tested to determine whether or not the decomposition products produced during combustion of smoke detector unit materials of construction caused significant changes in the quantity of ^{241}Am released from the source. The test methods and results are given in this report.

INTRODUCTION

Smoke detectors which provide early warning of incipient home fires have become extremely popular appliances with several million units now in use. The most widely used models operate on an air ionization principle that requires a source of ionizing radiation. Sealed sources containing americium-241 are most commonly used in home smoke detectors. Inevitably, some of these units will themselves be subjected to fires resulting in a potential for release of radioactive material. Procedures and criteria for evaluating the hazards associated with such releases are therefore being considered at national and international levels. The purpose of the work reported here was to determine the fate of radioactive material from smoke detector sources under simulated fire conditions and to evaluate procedures under consideration for testing the units.

Simulated fire tests are necessarily a compromise between the unlimited variety of situations occurring in real fires and the controlled, reproducible conditions required for scientific evaluation. Time and temperature sequences can easily be programmed and controlled. On the other hand, physical stress which may accompany or follow real fires is much more difficult to standardize.

Consequently, the tests have included observations of source conditions after simulated fires with attention given to possible effects of subsequent physical damage. In every case the primary focal point has been the possibility of release of radioactive material.

The radiation source is only a small component of a smoke detector (Fig. 1). It is both simpler and more precise to test the sources separately from the entire detector unit since the plastic and other combustible materials used in the smoke detector housing, circuit boards, etc. produce condensible fumes that greatly complicate the measurement of radionuclide release. However, testing of sources removed from the smoke detectors does not provide a realistic chemical environment during the test. Therefore tests have been conducted on sources alone, on sources and fragments of other detector components, and on whole, operating detectors.

PROCEDURES

Sources were procured by purchasing smoke detectors from commercial outlets and through direct contacts with source manufacturers. Although smoke detectors are marketed under a dozen or more brand names, americium-241 sources are manufactured by only two companies. No attempt has been made to determine which source manufacturer produced the sources in the smoke detectors purchased since several models may have used sources from either manufacturer. Individual sources obtained directly from each manufacturer have been tested. Detectors were assigned code numbers and were randomly allocated to the different procedural series. Three time/temperature sequences were used.

Standard 1-Hour Fire Tests. The standard 1-hour fire test follows a programmed time-temperature curve that is widely used for fire damage research.[1] The source was placed in a nickel boat and the boat inserted into a tube furnace lined with a Vycor tube. Temperature was increased from ambient to 927°C over a time period of 1 hour. The rate of heating was greatest initially and was continually decreased so that relatively high temperatures were maintained over much of the time period (Fig. 2). Air drawn through the tube during the entire test period was passed through a series of traps designed to capture any americium-241 that became airborne (Fig. 3). Two traps filled with nitric acid solution were followed by a membrane filter before the air was exhausted into a fume hood. When the final temperature was attained, the furnace was shut down and the entire apparatus allowed to cool. Air was drawn through the system during cooling. After the apparatus had cooled the traps were sampled, the filter was removed and the Vycor tube was rinsed with 1 molar nitric acid solution. In every test, the first trap solution, the acid used for rinsing the tube, and the filter were analyzed for contamination. The source remains and the nickel boat were smear tested for removable surface contamination by wiping them with a filter paper.

600°C NEA Test. The "600°C Test" is being considered as an internationally standardized procedure for evaluating smoke detector sources. In this test the sources are removed from their smoke detector housing and wiped with filter paper to determine if americium-241 is easily removable. Then each source is placed in a nickel boat and heated to 600°C in a Vycor tube passing

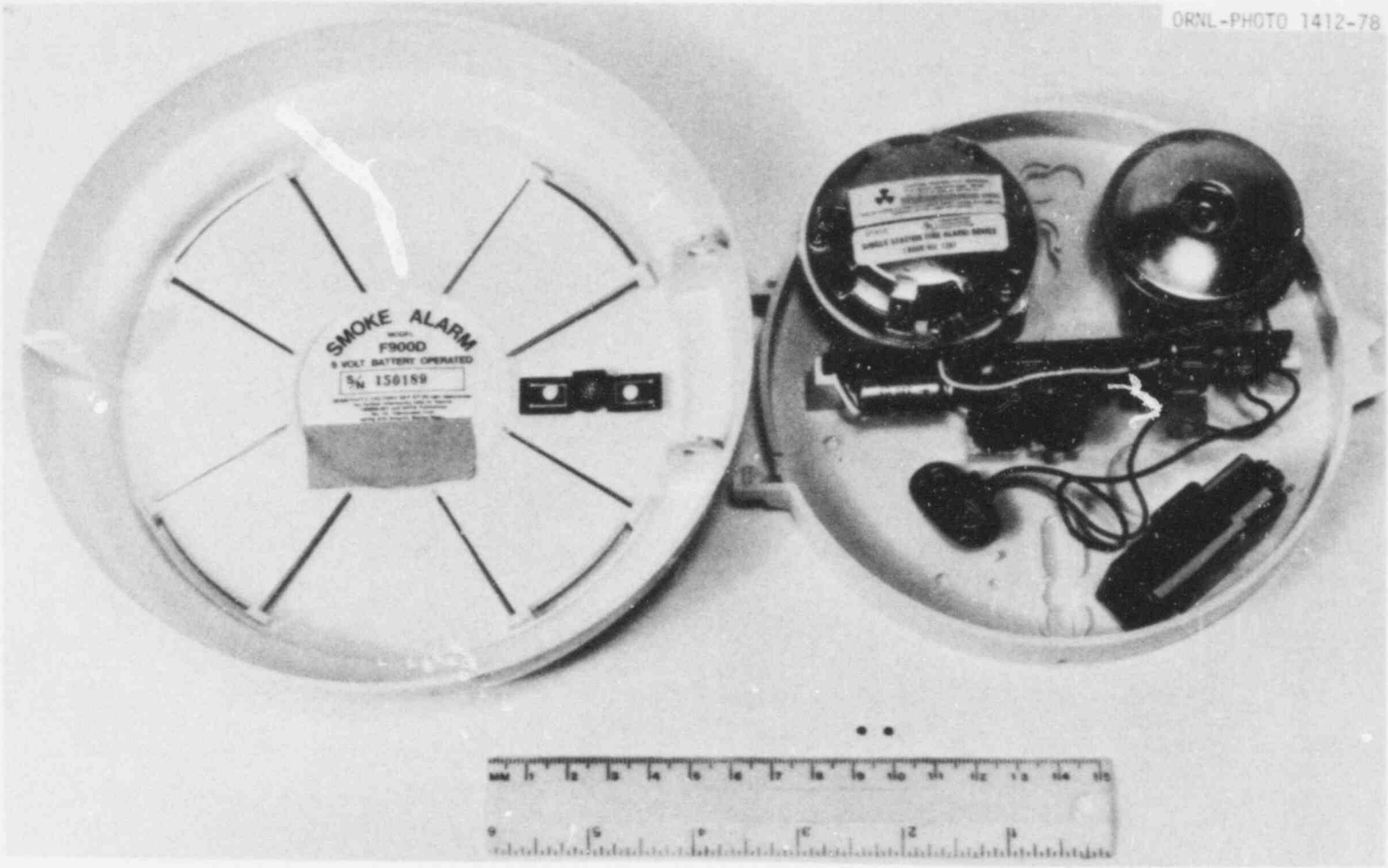


Fig. 1. Size Comparison Between Americium-241 Sources (just above the 9 and 10 markings on ruler) and Other Smoke Detector Components

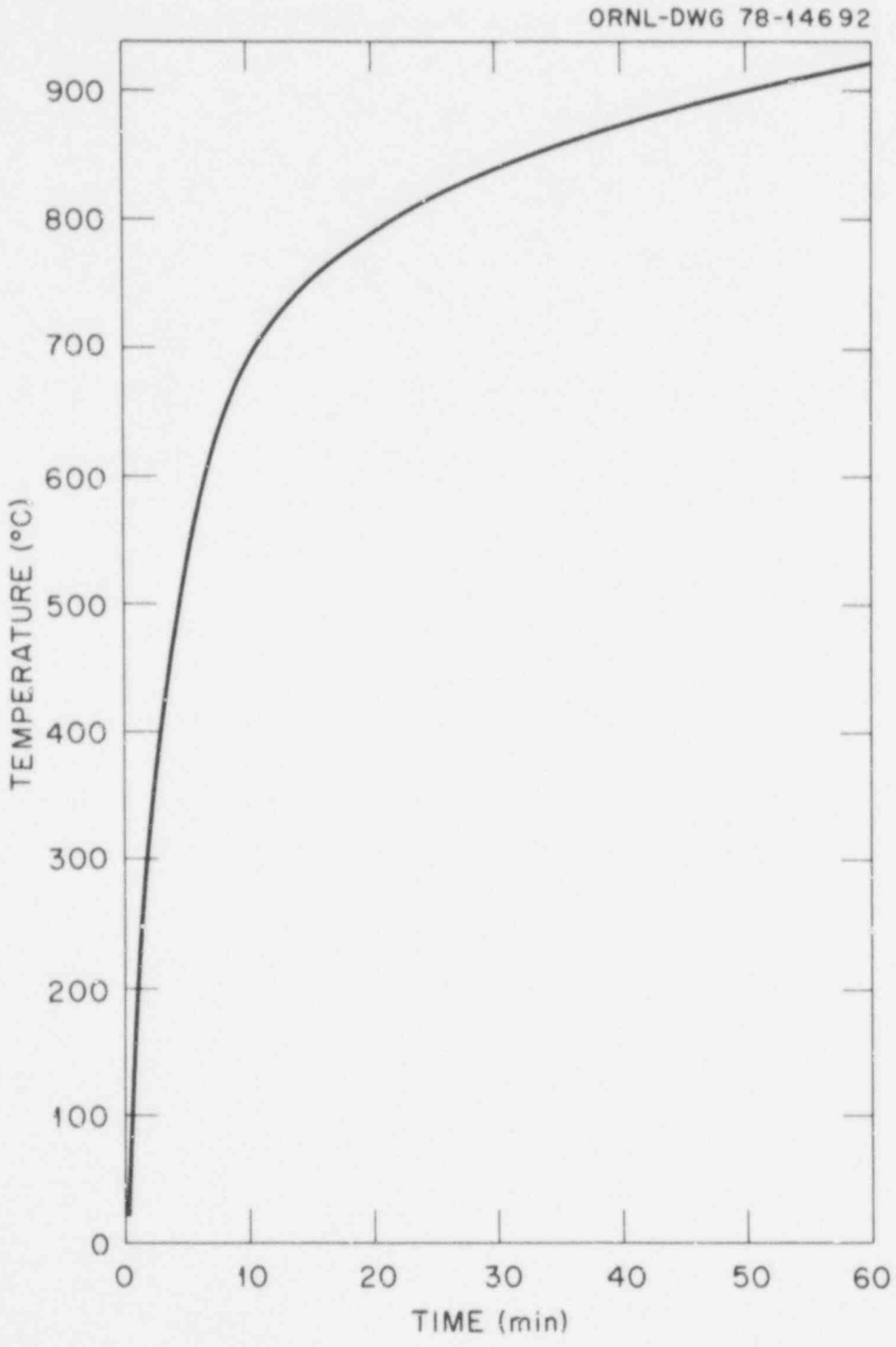


Fig. 2. One-Hour Time-Temperature Curve

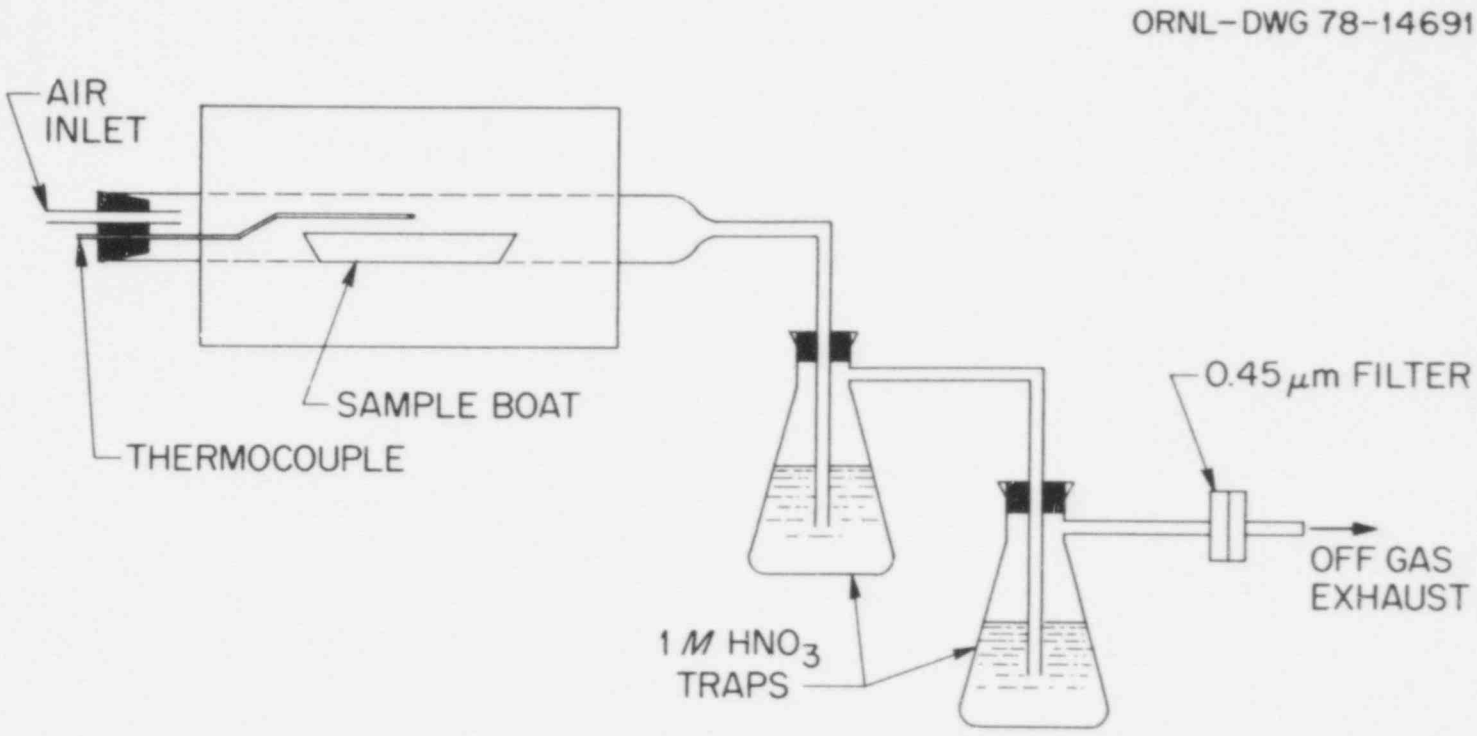


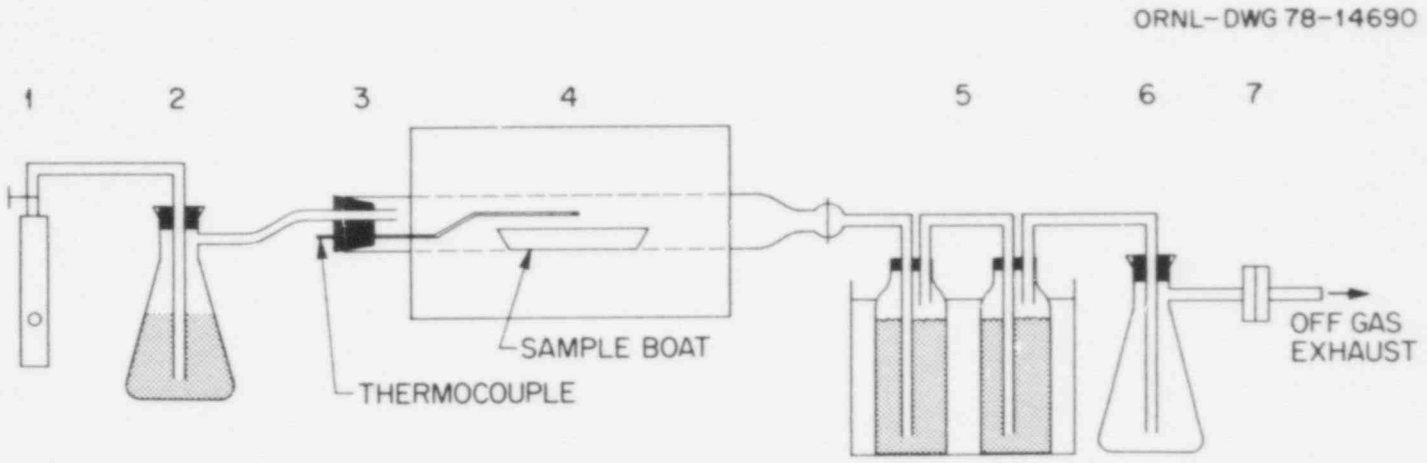
Fig. 3. 600°C Testing Apparatus

through a tube furnace. Air, drawn at 1 to 5 liters/min through the tube, passes through nitric acid traps and a membrane filter with 0.45 μ pore size before being released (Fig. 3). Temperature is maintained at 600°C for 1 hour and then gradually reduced. The cooled source is observed and wiped with a smear test paper. Trap solutions, air filters and smear papers are analyzed for alpha emitting americium-241. In addition the nitric acid used to clean the Vycor tube after each test is quantitatively sampled and analyzed. The tentative failure criterion for the 600°C test is ≥5 nCi of radioactivity on the wipe test after cooling.

1200°C NEA Test. In addition to the 600°C test, a more severe heating test has been devised.[2] Pretest treatment of the source was identical to the 600°C test, and the same air flow and trapping apparatus (Fig. 4) were used except that glacial acetic acid was substituted for the nitric acid in the traps and a dry trap inserted ahead of the filter. Some of the 1200°C tests involved not only sources but also plastic and composition parts of the smoke detector units. The smoke generated by these parts is better retained by glacial acetic acid than by 1 M nitric acid.

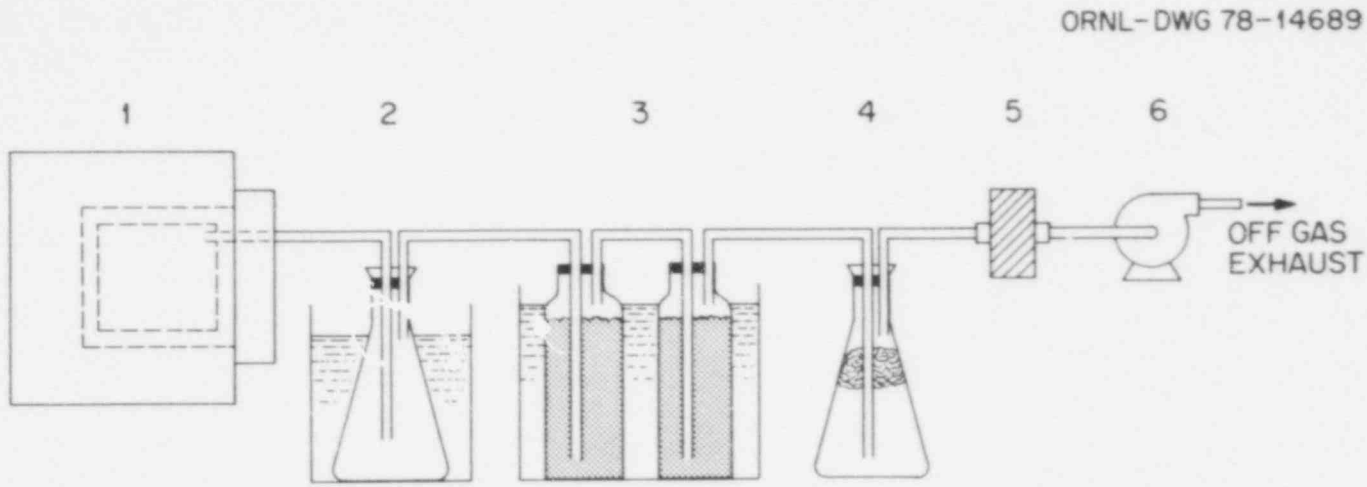
A quartz tube was used instead of Vycor and the nickel sample boat was placed into the furnace and heated to 1200°C for 1 hour. The heated source was then removed after cooling to 1000°C. Since the source materials melt at temperatures below 1200°C the residues were observed carefully after the test and the nickel boats were also analyzed for americium-241 that might have been transferred to them. Trap solutions, filters and wipe samples were analyzed as in the other tests. The suggested failure criterion for this test is ≥1% of the radioactive content lost from the source during the test.

Whole Smoke Detector Tests. Tests similar to the 1200°C test were performed on whole smoke detector units. Each smoke detector was placed inside a nickel container and heated in a muffle furnace. A 1/2-in. steel tube was inserted into the front of the nickel container through an orifice in the furnace door. The tube led to a condensing trap which was chilled with a mixture of methanol and dry ice (Fig. 5). Two glacial acetic acid traps were connected in series with the condensing trap. The acetic acid traps effectively retained the considerable amount of plastic volatiles emitted during combustion of the smoke detectors. Porcelain and glass beads inside the traps helped to disperse large bubbles. A dry trap containing glass wool as a prefilter was connected in series after the second acetic acid trap and was followed by a cartridge filter (type H ultra respirator filter for radionuclides, dust, fumes, and vapor) attached to a vacuum pump which was vented to an exhaust hood. Air was drawn through the system at approximately 10 liters/min. The temperature was raised to 1200°C over a 2- to 3-hour period and maintained for 1 hour at 1200°C with the exception of the first test. (During the first test the heating elements in the furnace burned out at a temperature between 1100°C and 1200°C.) After heating for 1 hour at 1200°C, the oven was turned off and allowed to cool and the nickel container was opened. Surface wipes were made around the exterior and interior of the americium-241 radioactive source-housing unit of the smoke detector remains. These wipes, along with the trap solutions, filters, and washings were analyzed for americium-241 content.



1. AIRFLOW METER
2. DESICCANT
3. 30 mm x 122 cm QUARTZ COMBUSTION TUBE
4. ELECTRIC FURNACE
5. GLACIAL ACETIC ACID TRAPS WITH GLASS AND CERAMIC SPHERES TRAPS IMMERSED IN A WATER BATH (18-20°C)
6. DRY TRAP
7. MILLIPORE FILTER (0.45 μm)

Fig. 4. 1200°C Test Apparatus for Smoke Detector Sources



1. MUFFLE FURNACE WITH NICKEL LINER
2. CONDENSING TRAP (DRY ICE AND METHANOL)
3. GLACIAL ACETIC ACID TRAPS WITH GLASS AND CERAMIC BEADS IN WATER BATH
4. DRY TRAP WITH GLASS WOOL FILTER
5. CARTRIDGE FILTER
6. VACUUM PUMP

Fig. 5. Test Apparatus for Whole Unit Testing of Smoke Detectors

Trap Efficiency Determinations. There is no *a priori* way to know the effectiveness of a trap series in recovering airborne contaminants. Therefore a test of the traps themselves was also conducted. In this test, 21 sources nominally containing 55 μCi total were heated in a nickel boat following the procedure for the 1200°C test except that a more elaborate trap system was used. Air drawn through the heating chamber over the sources was passed through four (rather than two) glacial acetic acid traps followed by a 1 molar nitric acid trap. From the wet trap series the air was passed through a dry flask, an activated charcoal filter, and, finally, a 0.45-μ membrane filter. All trap and filter contents were analyzed for americium-241.

In addition to the simulated fire tests, sources were subjected to a number of quality control and physical integrity tests.

Uniformity of Loading. Sources were analyzed in order to determine the precision of loading. Sources were positioned on either a Ge-Li detector or a NaI(Tl) scintillation detector and counted using a multichannel analyzer for a sufficient time period necessary to accumulate enough counts to produce a smooth Gaussian-like photo-peak displayed on the multichannel analyzer. The relative standard deviation for each batch, expressed as a percent of the mean ($\sigma/\bar{x} \cdot 100$), was then calculated (this value is designated $\sigma_T\%$). Some variability occurred due to variations in positioning of a source relative to the detector. To correct for this positioning variability, one source from each batch was counted a number of times, repositioned on the detector each time, and the relative standard deviation calculated ($\sigma_R\%$). The precision of loading ($\sigma_L\%$) was then calculated as follows: $\sigma_L\% = [(\sigma_T\%)^2-(\sigma_R\%)^2]^{1/2}$.

Surface Wipes. Each source within a batch was dry wiped with a filter paper using firm contact and the wipe analyzed for alpha emitters.

Cellophane Tape Tests. Each unmounted source within a batch was impressed between the adhesive sides of a folded piece of cellophane tape. The tape was then peeled back to see if any of the gold foil coverings could be removed. These tapes were then analyzed for alpha emitters.

Accuracy of Loading. Sources were dissolved in hot concentrated nitric acid and the gold foil residue then dissolved in hot aqua regia. The solutions were then analyzed either by counting the 60 keV gamma-ray and comparing results to that of a known standard or by measurement of alpha particle emission.

Measurements of Americium-241 Content in Samples. Americium-241 decays (half-life: 433 yrs) by alpha emission (5.48 MeV et al.) accompanied by a prominent gamma-ray (59.5 keV). Various methods can be used to determine americium-241. The Minimum Detectable Activity (MDA) defined[3] as "That amount of activity which, in the same counting time, gives a count which is different from the background count by three times the standard deviation of the background count" was determined for each detector used. Throughout the analysis, different time intervals have been utilized in counting the samples, ranging from a few seconds for determining the precision of loading of sources to 1000 minutes for low-level evaporated liquid solutions.

Alpha Scintillation Counters (light-tight sample holder, zinc sulfide phosphor, and photomultiplier tube coupled to a decade scaler) were used for determining the amount of radioactivity present on wipes, filters, and cellophane tape tests. MDA: 8. pCi ($8. \times 10^{-6}$ μCi). A similar type counter applicable for low-level activity has been used to analyze trap solutions and washings which were evaporated (1 ml) onto metallic discs before counting. MDA: 0.02 pCi ($2. \times 10^{-8}$ μCi).

Gas Flow Proportional Counters were used to analyze metallic discs containing 1 ml of evaporated liquid samples from trap solutions, Vycor tube washings, and some dissolved sources. MDA: 1.4 pCi (1.4×10^{-6} μCi).

NaI(Tl) Scintillation Detector and Multichannel Analyzer were used to measure the 59.5 keV gamma-ray from the decay of americium-241. This method was used for counting sources supplied by manufacturers, those removed from smoke detectors, dissolved sources prepared in plastic planchets or plastic petri dishes, and solutions evaporated under a heat lamp. It was also used for analyzing activity in the cartridge filters, glass wool filters, particulate debris, and some trap and washing solutions. MDA: 12 to 26 pCi (12 to 26×10^{-6} μCi), depending on counting time (60 to 10^3 min).

Samples were usually prepared in duplicate and the mean used to calculate the amount of radioactivity present in the sample. For conciseness, the error terms have been omitted. For those samples containing low-levels of radioactivity, considerable variability occasionally occurred among the duplicates, and in some instances by an order of magnitude. These difficulties may be due to the nature of the samples themselves; i.e., suspended particulate material in various trap solutions and the difficulty in homogeneous sampling.

RESULTS

The amounts and characteristics of americium-241 released from the sources are grouped according to the nature of the tests (Tables 1-10).

After the Standard 1-Hr Fire Test exposure, post-test wipes carried 10^{-3}-10^{-2} microcurie of americium-241, typically an order of magnitude higher than pre-test wipes (Table 1). Traps and filters in the air stream below the sources contained 10^{-4} microcurie or less of americium-241. Thus, while the tests usually resulted in an increased susceptibility to loss of americium by abrasion, the amount released as airborne contamination was very slight.

Table 1. Results of Standard One Hour Fire Tests (927°C)

(all values in microcuries)

Run No.	Stated Source Activity	Pre-test Smear	Post-test Smear	Airborne Contaminants
1	1.5	BDL	1.0×10^{-3}	4.1×10^{-5}
2	1.5	3.9×10^{-4}	8.2×10^{-3}	2.2×10^{-4}
3	1.5	BDL	2.2×10^{-3}	6.7×10^{-4}
4	1.5	4.3×10^{-4}	BDL	8.2×10^{-5}

BDL = Below Detection Limit. ($<8\times10^{-6}$ μCi).

1. Disc source in brass mount.
2. Strip source in metal circular stainless steel mount.
3. Disc source in brass mount plus representative parts (circuit board pieces, etc.).
4. Strip source on metal circular mount plus representative parts.

In the 600°C Tests the americium-241 that was released was primarily on the post-test wipes and not in the air stream (Table 2). With the exception of Run No. 1, all the radioactivity which became detached was below the failure criterion of five nanocuries (5×10^{-3} μCi). The single failure involved a disc source in a brass rivet-like mount. After the source was heated at 600°C for one hour, approximately 26 nanocuries (26×10^{-3} μCi) of radioactivity was removed using a wipe. In addition, after Runs 4B and 4D a greater amount of radioactivity was found on the wipes of the brass-mounted disc sources than for disc sources which had been removed from their brass mounts before being tested at 600°C, although the sources tested in Runs 4B and 4D did not fail the 5 nCi criterion. These sources were clearly more degraded when tested in the brass mounts (Fig. 6). Previous studies in Great Britain yielded similar results.[4] Another type of unmounted disc source is shown in Fig. a prior to being heated to 600°C and again in Fig. 7b after being heated to 600°C. A strip source before being tested at 600°C is shown in Fig. 8. Also included in the testing procedure was a run for 1 hour at 600°C and then reheating to 600°C for an additional 2 hours (Table 2). The amount of radioactivity released as an aerosol remained essentially the same, at least within the order of magnitude.

In the 1200°C Tests (Table 3) the total amounts of radioactivity released as airborne contaminants were greater than in the lower temperature tests, as high as 1.5×10^{-3} μCi. Releases were below the suggested pass-fail criterion--1% of the source activity. Several of the sources tested at 1200°C had been previously tested at lower temperatures. Increased amounts of radioactivity in the released airborne contaminants occurred with the higher temperature and in some instances by more than an order of magnitude (Table 4). A comparison of the various conditions of the 1150/1200°C tests is further illustrated in Table 5. No particular pattern is apparent under the various test conditions.

Table 2. Results of 600°C Fire Tests on ^{241}Am Smoke Detector Sources (all values expressed in μCi)

Run No.	Source Description	Source Activity	Pre-Test Smear	Post-Test Smear	Airborne Contaminants
1	Brass mounted disc source	1.5	BDL	*2.6×10^{-2}	4.7×10^{-5}
2	Strip source	0.4	BDL	BDL	5.3×10^{-5}
3	Unmounted disc sources (back-to-back)	5.0	1.1×10^{-4}	4.0×10^{-5}	3.3×10^{-5}
4a	Disc source removed from brass mount	1.5	BDL	8.5×10^{-5}	4.0×10^{-5}
4b	Brass mounted disc source	1.5	BDL	6.4×10^{-4}	
4c	4a reheated to 600°C for 2 hrs	1.5	---	4.7×10^{-5}	2.7×10^{-5}
4d	4b reheated to 600°C for 2 hrs	1.5	---	1.6×10^{-4}	
5	Disc source rivet mounted	4.3	2.7×10^{-5}	BDL	1.4×10^{-4}
6	Disc source in tin plated mount	0.35	BDL	1.5×10^{-5}	BDL
7	Disc source in tin plated mount	6.1	BDL	7.4×10^{-5}	BDL
8	Disc source in tin plated mount	1.0	BDL	6.3×10^{-4}	BDL
9	Disc source in stainless steel mount	1.0	BDL	BDL	3.8×10^{-5}
10	Disc source in nickel plated mount	1.0	BDL	BDL	3.4×10^{-5}

*Exceeds failure criterion of 5×10^{-3} μCi. BDL = Below Detection Limit.

Runs 1-5 were made on sources removed from commercially available smoke detectors.
Runs 6-10 were made on sources supplied by a source manufacturer.
Run 4: Two sources (4a and 4b) tested together at 600°C for 1 hr and again for 2 hr (4c and 4d).
Run 5: Industrial smoke detector (0.8 μ Millipore filter used).

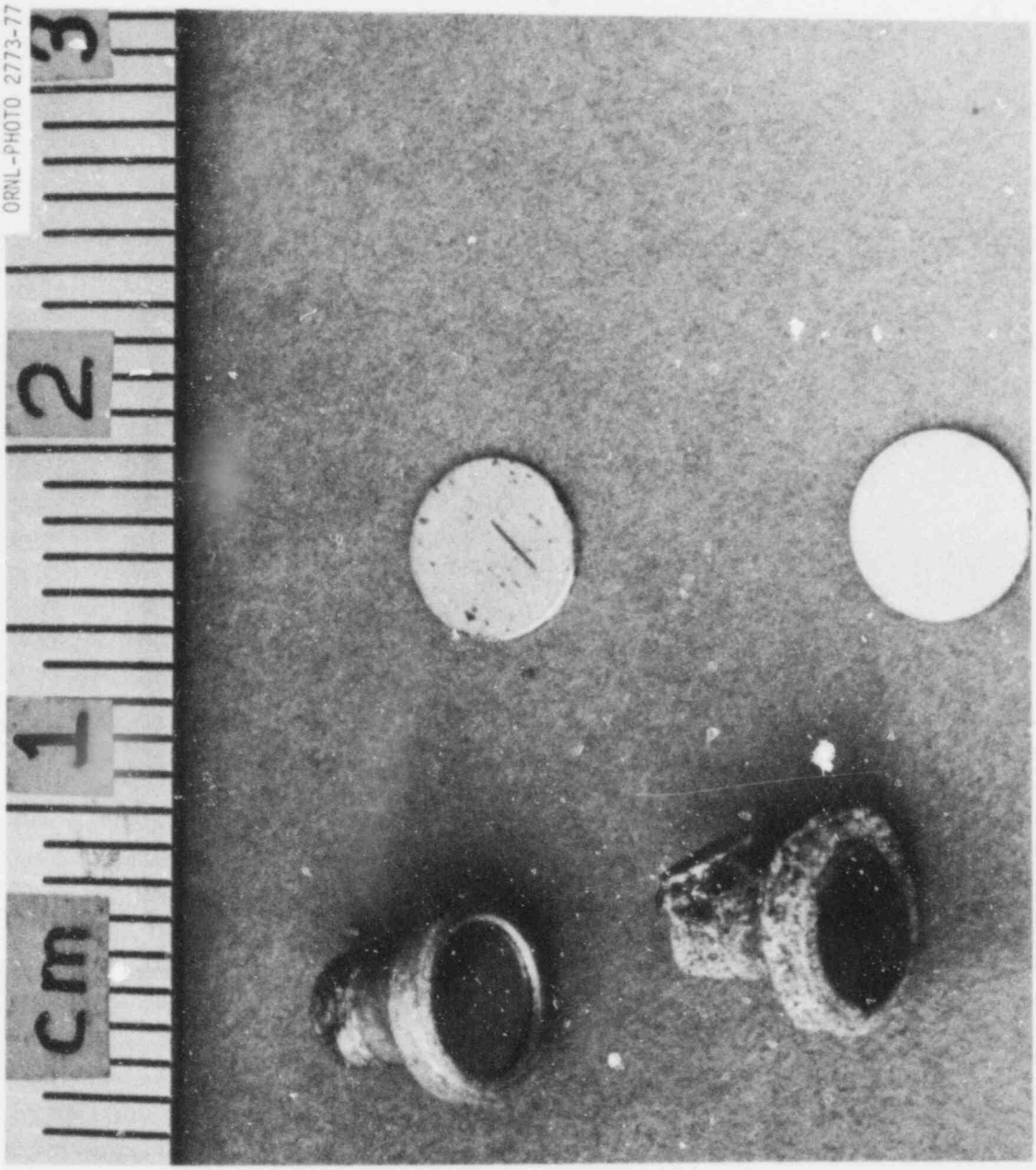


Fig. 6. Comparison Between Brass Mounted and Unmounted Sources Before and after Testing at 600°C
To_r: Untested Bottom: Tested

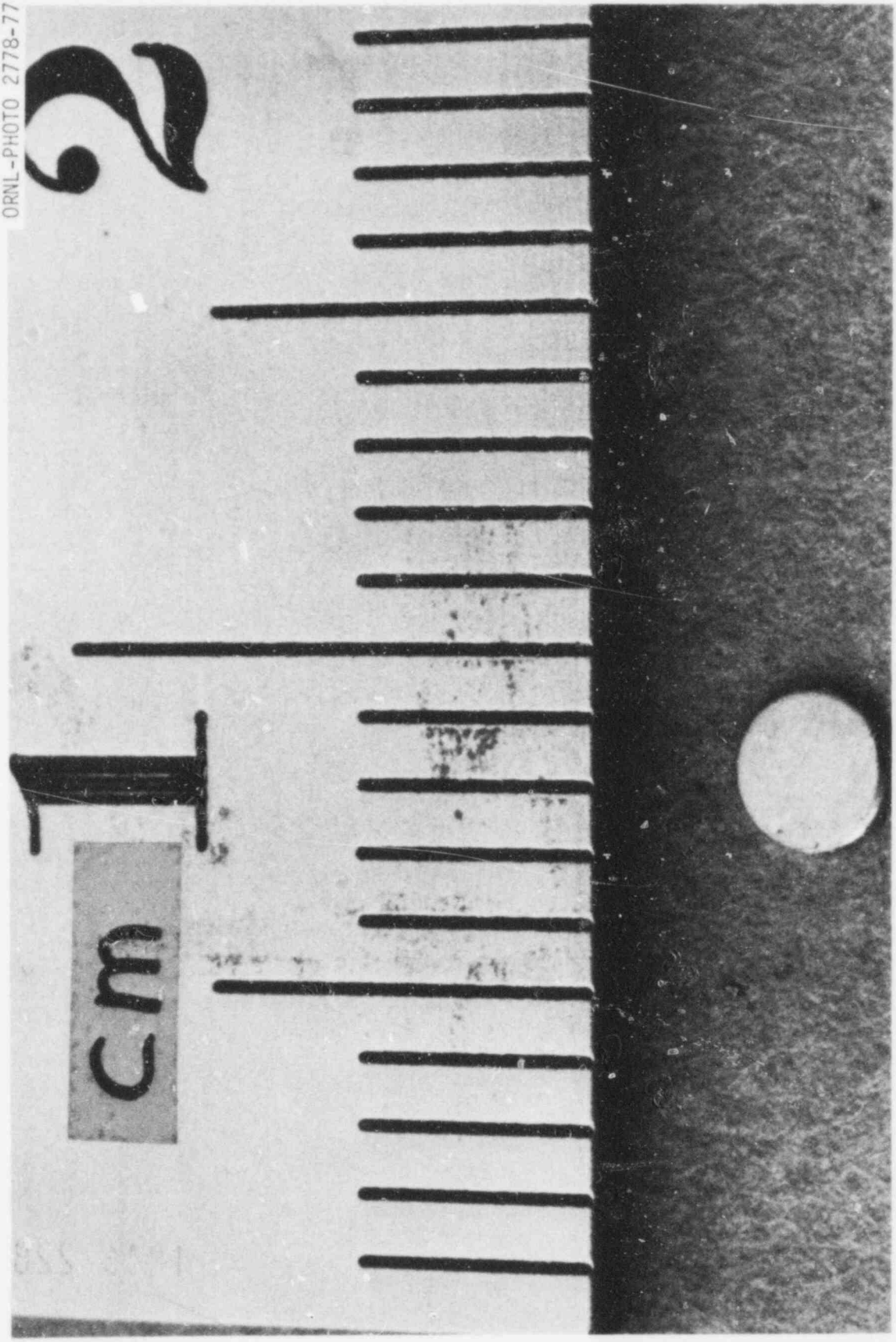


Fig. 7a. Unmounted Disc Source Prior to 600°C Test

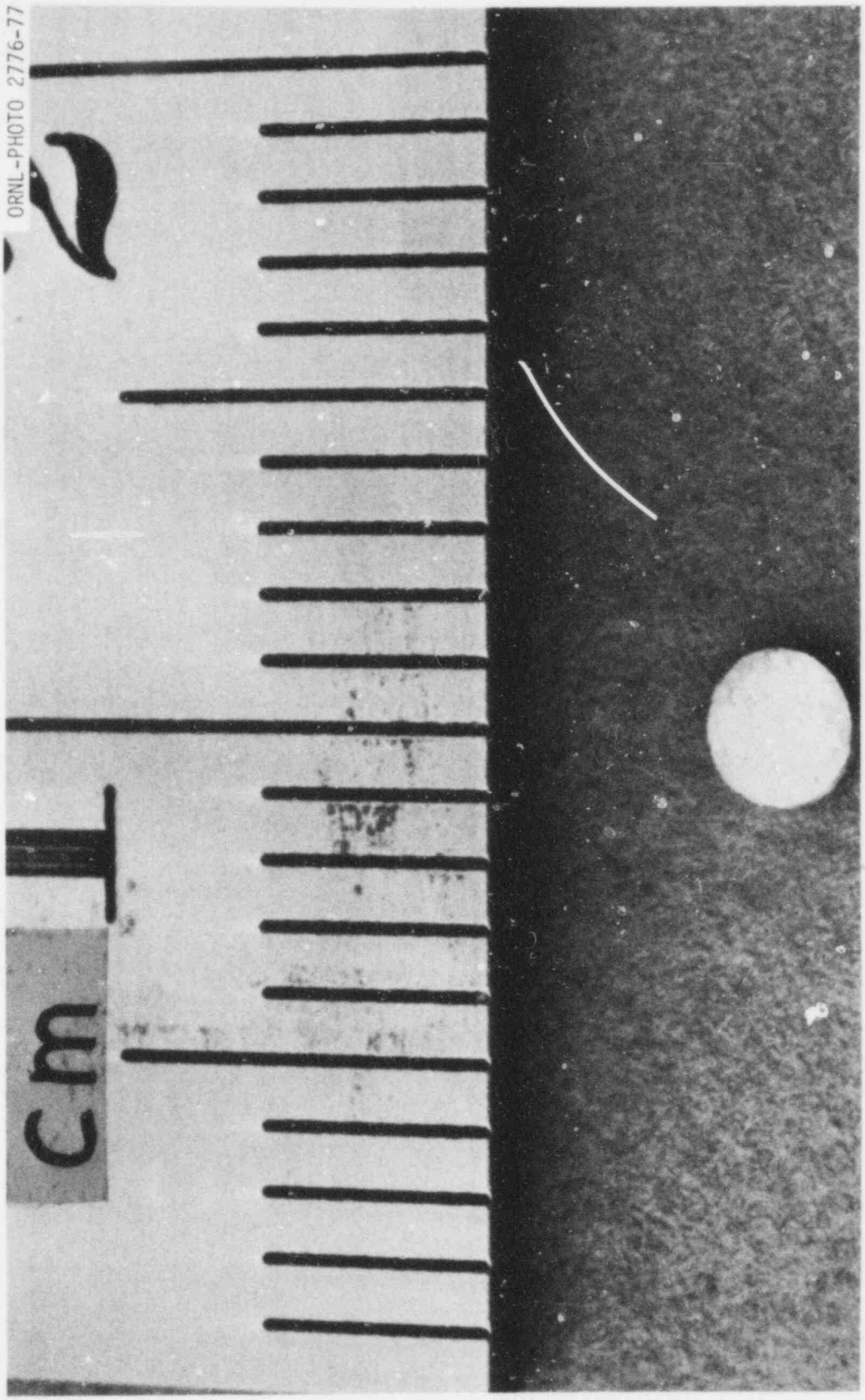


Fig. 7b. Unmounted Disc Source After Testing at 600°C

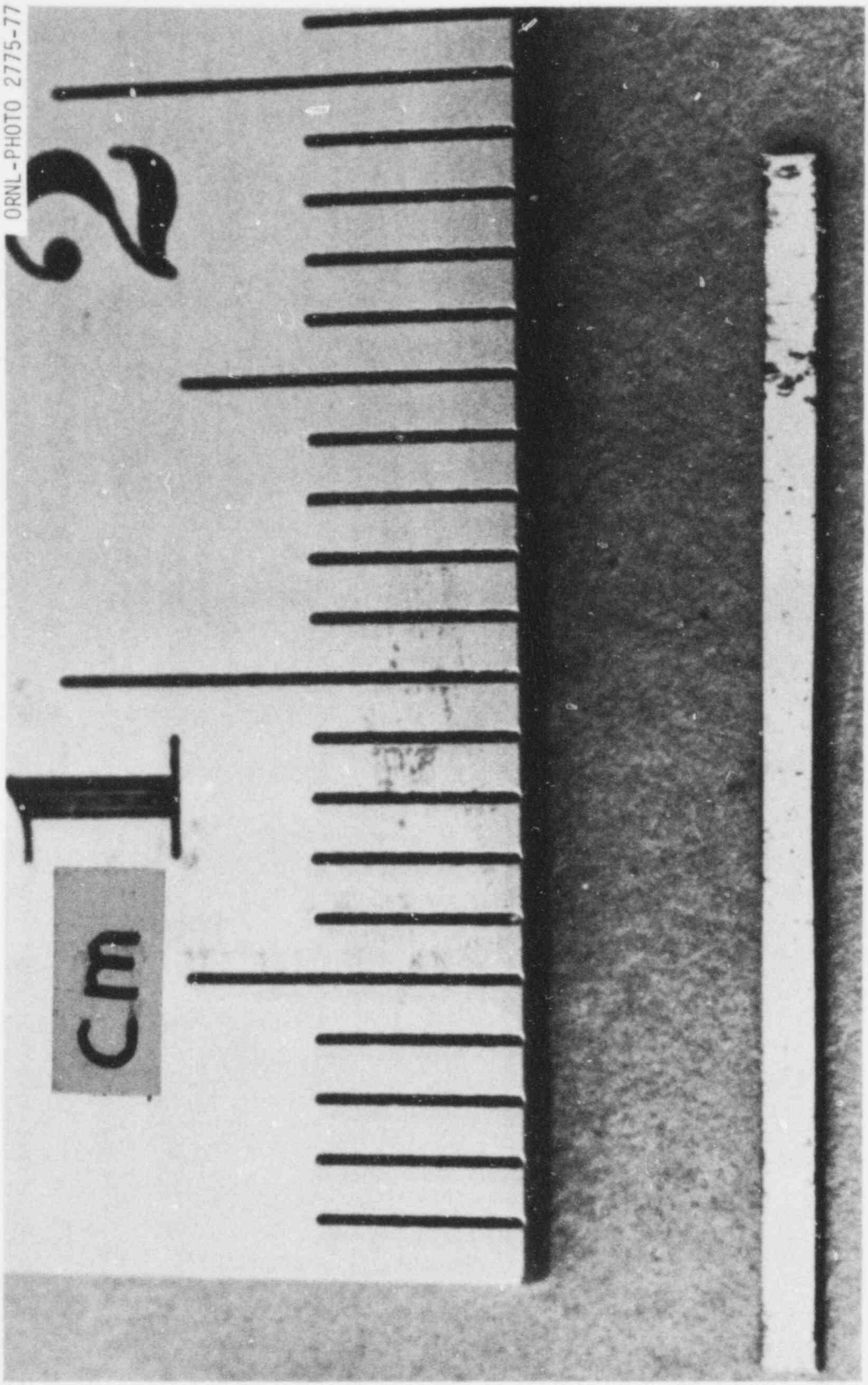


Fig. 8. Americium-241 Strip Source Before Testing

Table 3. Results of 1200°C Temperature Tests on ^{241}Am Smoke Detector Sources

(all values reported in microcuries)

Run No.	Source Activity	Pre-test Source Wipe	Post-test Source Wipe	Interior Sampling Boat Wipe	Airborne Contaminants
1	0.4(0.35)	BDL	2.4×10^{-2}	6.6×10^{-5}	7.2×10^{-5}
2	1.5	9.1×10^{-5}	2.9×10^{-3}	1.2×10^{-3}	1.1×10^{-4}
3	1.5	BDL	Disintegrated into particles when removed		
4	0.4(0.35)	BDL	5.4×10^{-4}	1.1×10^{-4}	4.9×10^{-4}
5	5.0	2.2×10^{-4}	7.5×10^{-3}	8.1×10^{-3}	6.8×10^{-4}
6	2.5	2.7×10^{-5}	1.9×10^{-4}	1.3×10^{-3}	7.7×10^{-5}
7	0.4(0.35)	1.4×10^{-5}	4.1×10^{-5}	9.9×10^{-5}	8.1×10^{-4}
8	5.0	BDL	6.5×10^{-4}	8.4×10^{-4}	5.7×10^{-4}
9	2.5	NA	1.6×10^{-2}	BDL	1.5×10^{-3}
10	1.5	5.7×10^{-2}	1.7×10^{-3}	BDL	2.6×10^{-4}
11	4.3	BDL	8.1×10^{-3} (disintegrated into particles when removed)	6.9×10^{-3}	6.5×10^{-4}

NA = Not Analyzed. BDL - Below Detection Limit. ($<8\times10^{-6}$ μCi).

1. Strip source (temperature attained only 1150°C; source previously heated to 600°C: Run 2, Table 2).
2. Disc source removed from brass mount (previously heated to 600°C: Run 4a and 4c, Table 2).
3. Disc source in brass mount.
4. Strip source.
5. Two disc sources mounted back-to-back.
6. Disc source.
7. Strip source with component parts.
8. Disc sources (back-to-back) with component parts (source previously heated to 600°C: Run 3, Table 2).
9. Disc source with component parts.
10. Strip source on circular metal mount with component parts (source previously heated to 927°C: Run 2, Table 1).
11. Rivet mounted disc source with component parts (previously heated to 600°C: Run 5, Table 2).

Table 4. Comparison of Airborne Release from Sources Tested at 600/927°C With Those Tested at 1150/1200°C

Source Description	Source Activity, microcuries	Microcuries of Americium-241 in Airborne Contaminants	
		600/927°C	1150/1200°C
Sources Not Previously Heated			
Brass mounted disc source	1.5	4.7×10^{-5} 4.1×10^{-5}	5.7×10^{-5}
Disc sources mounted back-to-back	5.0	3.3×10^{-5}	6.8×10^{-4}
Strip source	0.4	5.2×10^{-5}	4.9×10^{-4} (8.1×10^{-4})*
Source Previously Tested at 600/927°C and Retested at 1150/1200°C			
Disc source removed from brass mount	1.5	6.7×10^{-5}	1.1×10^{-4}
Strip source	0.4	5.3×10^{-5}	7.2×10^{-5}
Disc sources mounted back-to-back	5.0	3.3×10^{-5}	5.7×10^{-4}*
Strip source mounted on circular steel mount	1.5	2.2×10^{-4}	2.6×10^{-4}*
Rivet mounted disc source	4.3	1.4×10^{-4}	6.5×10^{-4}*

*Tested with component parts from smoke detector.

Table 5. Comparison Among Test Conditions at 1150/1200°C
(From Table 3)
(^{241}Am content expressed in μCi)

Run Number	Source Activity	Airborne Contaminants			
		Source Alone	Source and Components	Source Alone Previously Heated to 600/927°C	Source Previously Heated to 600/927°C Plus Component Parts
1	0.4			7.2×10^{-5}	
2	1.5			1.1×10^{-4}	
3	1.5	5.7×10^{-5}			
4	0.4	4.9×10^{-4}			
5	5.0	6.8×10^{-4}			
6	2.5	7.7×10^{-5}			
7	0.4		8.1×10^{-4}		
8	5.0				5.7×10^{-4}
9	2.5		1.5×10^{-3}		
10	1.5				2.6×10^{-4}
11	4.3				6.5×10^{-4}

In each of the three Whole Smoke Detector Tests (Table 6) the americium-241 sources disintegrated into non-recognizable forms or powder and became incorporated with the remaining debris. In detectors 1 and 2 most of the radioactivity was contained within the inner protective steel source mounting box of the smoke detector. In detector 3 the sources were mounted inside the smoke detector on an aluminum cap which melted and became unrecognizable. Wipes within the interior of the source mounting area were approximately an order of magnitude more contaminated than wipes made on the exterior of the mounting area or the trap solutions and filters.

In the Trap Efficiency Tests the total airborne americium-241 recovered was 600 pCi, of which 535 pCi was found in the first trap and 65 pCi in the fourth trap. No detectable americium-241 was found in any of the other traps or filters. The interior of the heating chamber contained 5.5 nCi of americium-241 outside the sample boat.

Table 6. Results of Whole Smoke Detector Tests at 1200°C

Americium-241 Activity, μCi

Run No.	Source Activity	Cold Trap	Liquid Traps	Dry Trap	Glass Wool Filters	Cartridge Filter	Post Test Wipes		Summation of Released Activity
							Exterior of Source Mounting Area	Interior of Source Mounting Area	
1*	5	6.5×10^{-5}	1.0×10^{-3}	NA	BDL	BDL	1.7×10^{-4}	8.3×10^{-3}	9.5×10^{-3}
2	3	7.1×10^{-4}	8.7×10^{-4}	3.1×10^{-4}	BDL	2.0×10^{-4}	4.6×10^{-4}	5.2×10^{-3}	7.8×10^{-3}
3	1.6	3.4×10^{-4}	4.1×10^{-4}	1.3×10^{-4}	BDL	BDL	7.0×10^{-5}	(destructed) see text	9.5×10^{-4}

*Temperature >1100°C reached before furnace malfunctioned and heating elements replaced.

BDL = Below Detection Limit. NA = Not Analyzed.
(see text)

1. Two disc sources; plastic housing.
2. Two rivet mounted disc sources; metal housing.
3. Two strip sources; plastic housing.

DESCRIPTION OF INDIVIDUAL DETECTORS

Smoke detector No. 1 contained two 5-mm-diam disc sources mounted back-to-back and had a total americium-241 activity of 5 μCi (Fig. 9). Figure 10 shows the remains of the smoke detector inside the nickel liner after being heated above 1100°C. The burned components remained approximately in the same relative position as shown before testing. (The source container is shown as the circular component in the lower part of the picture and the circular sound alarm is shown above the source container.) Wipes on the inside of the nickel liner indicate approximately 2×10^{-5} μCi and 1.7×10^{-4} μCi of americium-241 on the exterior of the charred metal source housing unit. Alpha wipes inside of the source container indicate 8.3×10^{-3} μCi americium-241. No recognizable forms of the source remained when interior of the mounting area was examined.

Smoke detector No. 2 (Fig. 11) contained two americium-241 sources separately mounted in polyethylene caps beneath the printed circuit board (Fig. 12). Each of these sources were contained in brass rivet-like mounts (see Fig. 6). Figure 13 shows the destructed smoke detector with the protective cover screen removed. Smears of the inside and outside of the nickel liner picked up 1.7×10^{-5} μCi of alpha activity. The exterior of the source containment area indicated approximately 1.1×10^{-4} μCi americium-241. Wipes of the alarm and batteries were 1.3×10^{-4} μCi. Wipes of the nickel liner bottom beneath the source area were 2.1×10^{-4} μCi americium-241. Wipes inside the protective metal box housing the polyethylene source holder caps indicated a greater amount of americium-241 activity: 5.2×10^{-3} μCi. The sources had disintegrated into particles that were unrecognizable in the debris.

Smoke detector No. 3 (Fig. 14) has two strip-type sources, one mounted in view on top of the aluminum cap and the other strip source mounted at right angles and beneath the aluminum cap surface (Fig. 15). Figure 16 shows the detector after being heated to 1200°C and Fig. 17 is the same detector with the protective wire screen removed. No trace of the aluminum source holder or sources could be found. Alpha wipes of the fused debris around the source area were low: 7.0×10^{-5} μCi americium-241. Analysis of the various destructed components of this smoke detector indicated that the dominant remaining radioactivity was associated with the inner debris underlying the fused detector housing. After removal of the destructed casing, the dominant radioactive portion of the remaining loose material was separated and then fractionated by sieving the material through standard Tyler sieving screens in order to determine the distribution of radioactivity with particle size. The weight of debris used, amount of radioactivity determined, and particle size distribution are summarized in Table 7. Americium-241 concentrations in the debris appear to be fairly uniform with the exception of the very coarse material and little (2.2×10^{-3} μCi) radioactivity was associated with the remaining very fine particulate material.

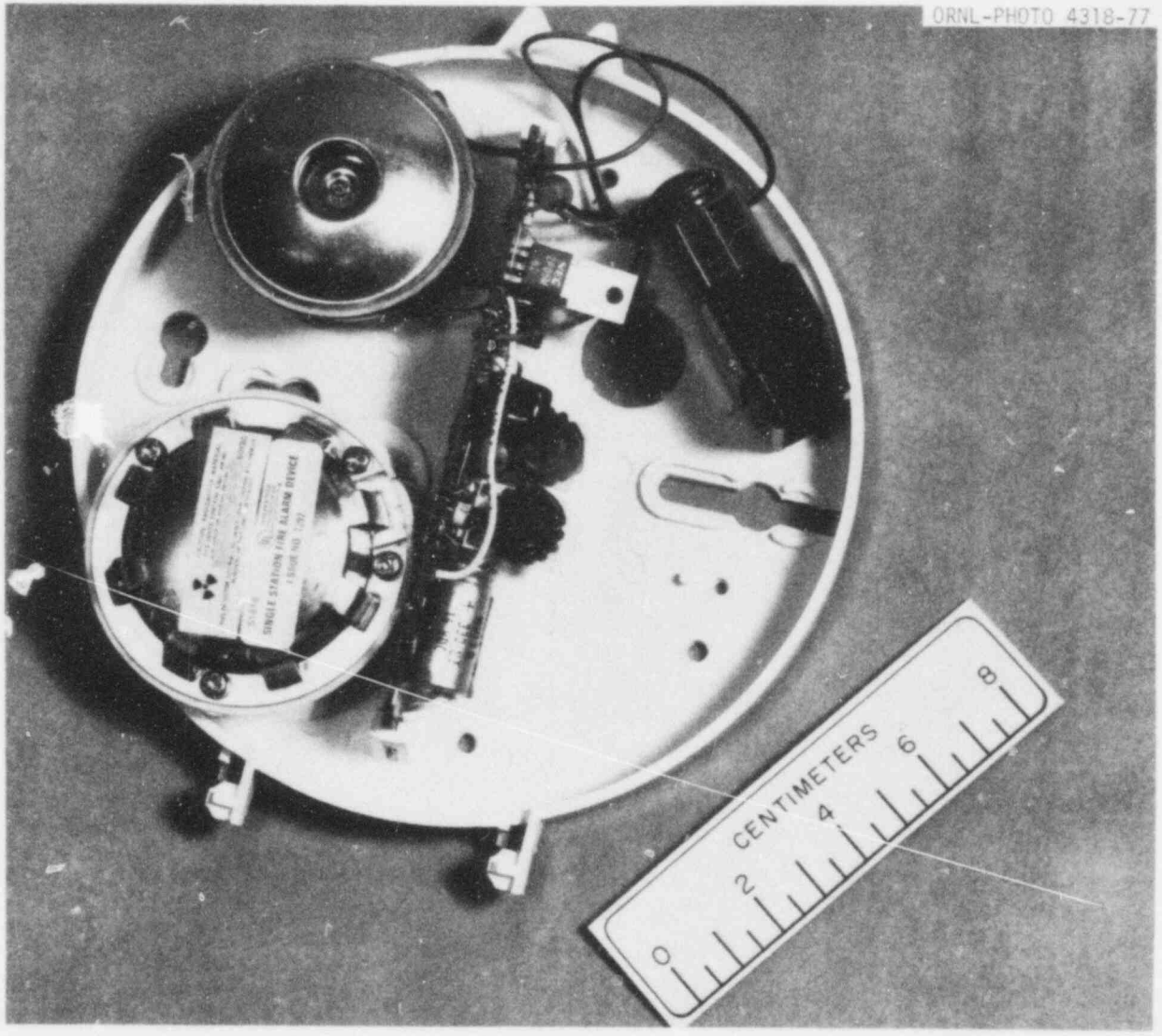


Fig. 9. Type 1 Smoke Detector Before 1200°C Test Showing Internal Components (cover case removed)

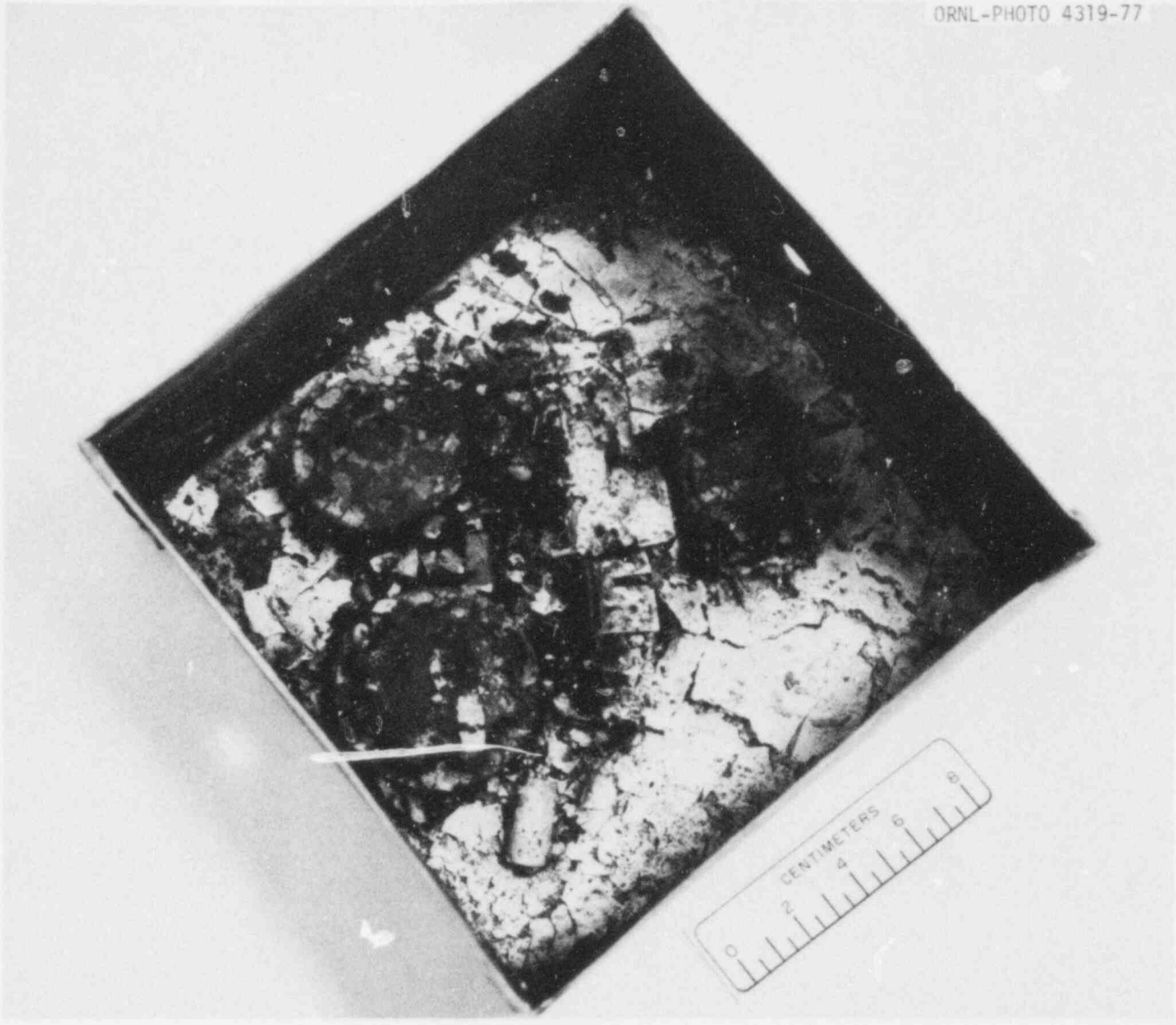


Fig. 10. Smoke Detector After Heating Greater Than 1100°C

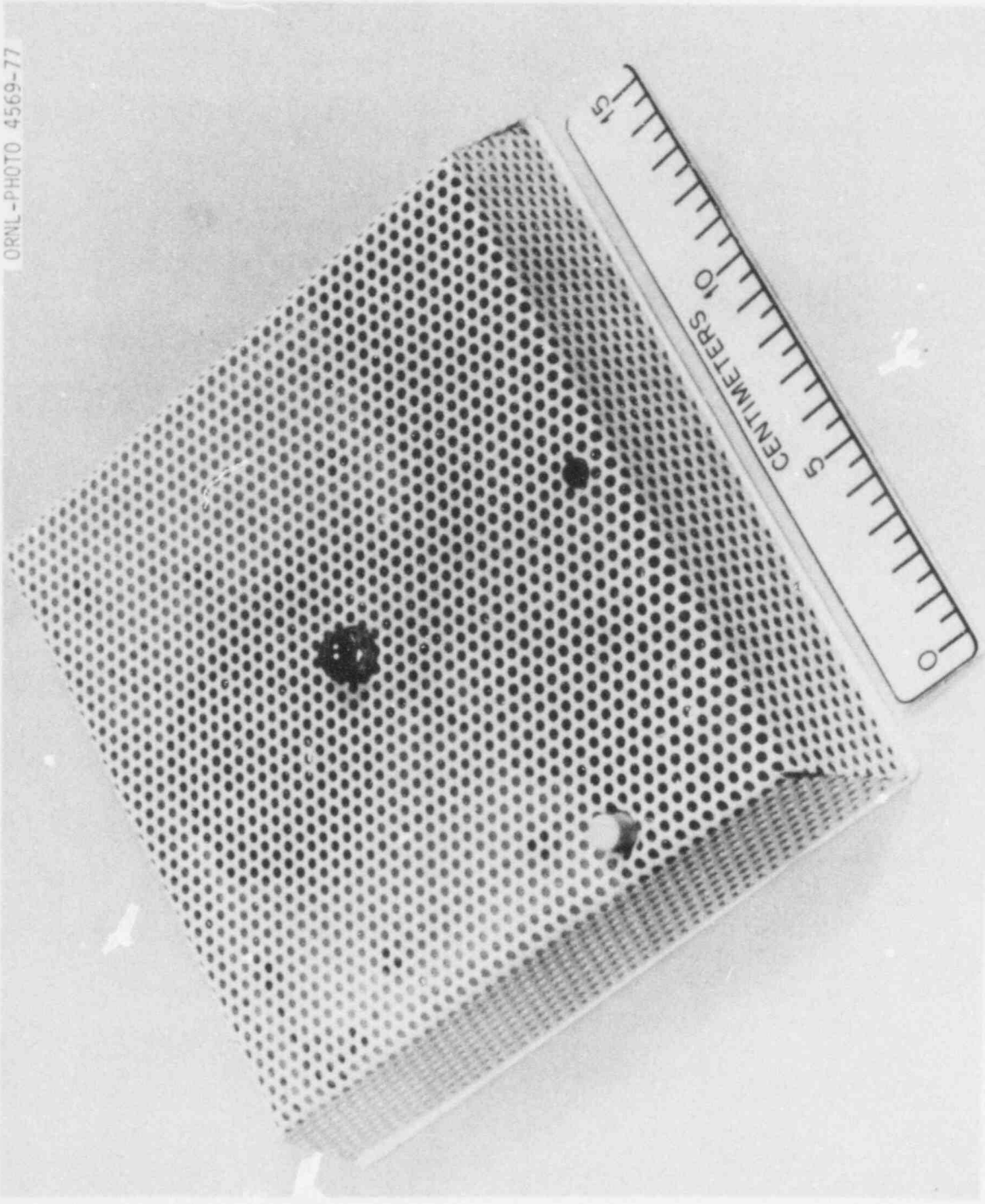


Fig. 11. Type 2 Whole Smoke Detector Tested at 1200°C

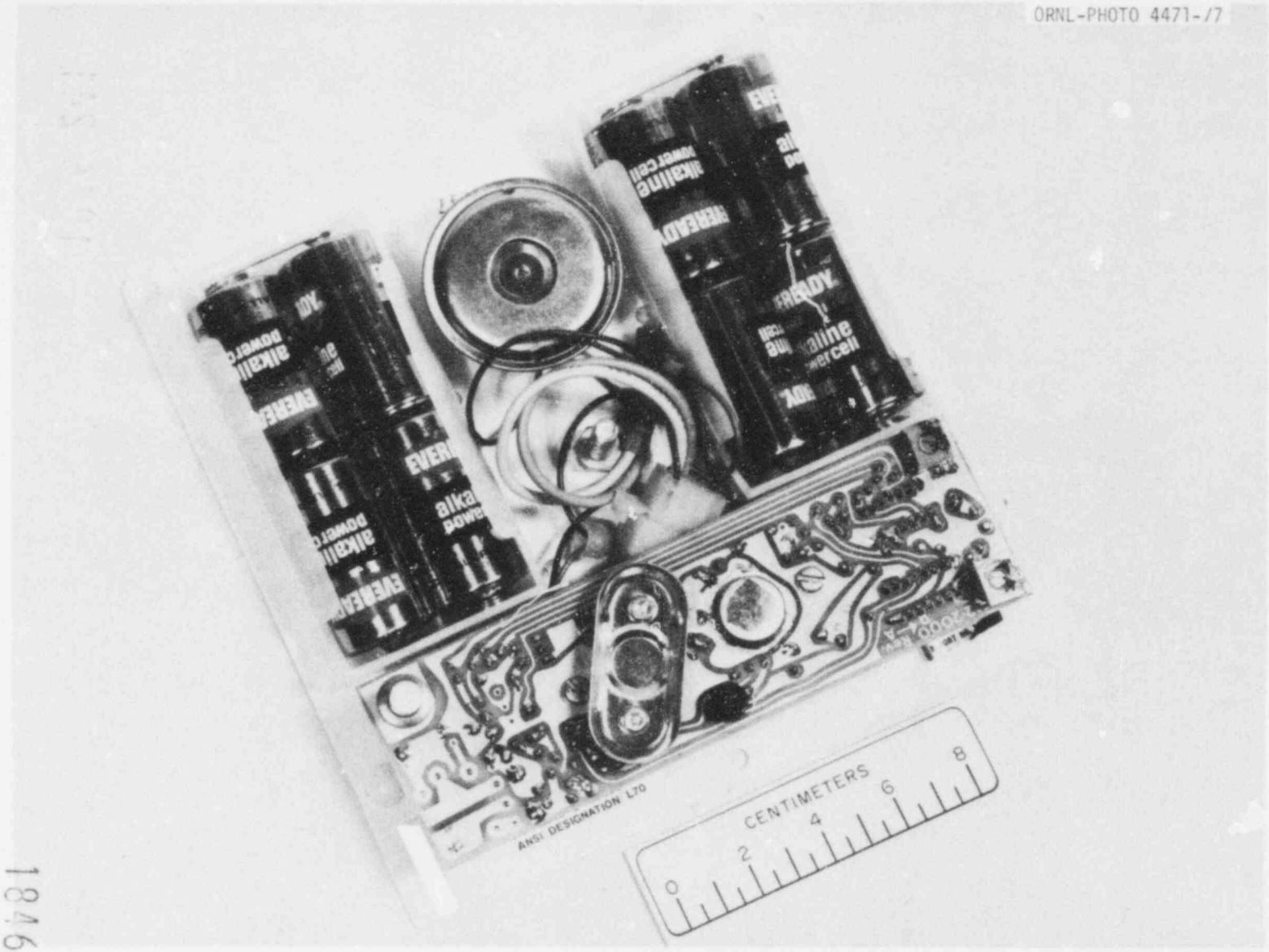


Fig. 12. Internal Components of Type 2 Whole Smoke Detector Tested at 1200°C

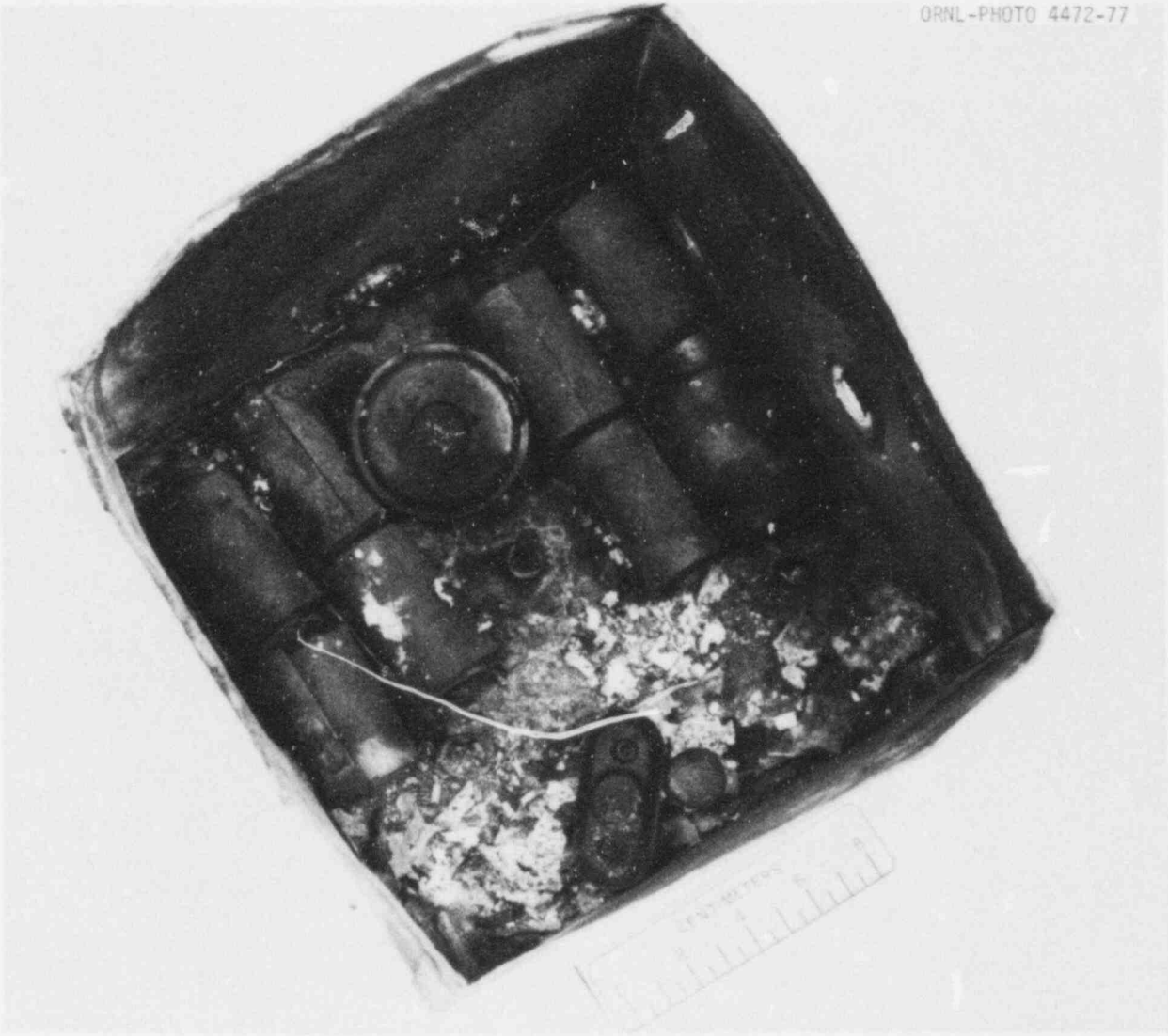


Fig. 13. Type 2 Whole Smoke Detector After Testing at 1200°C

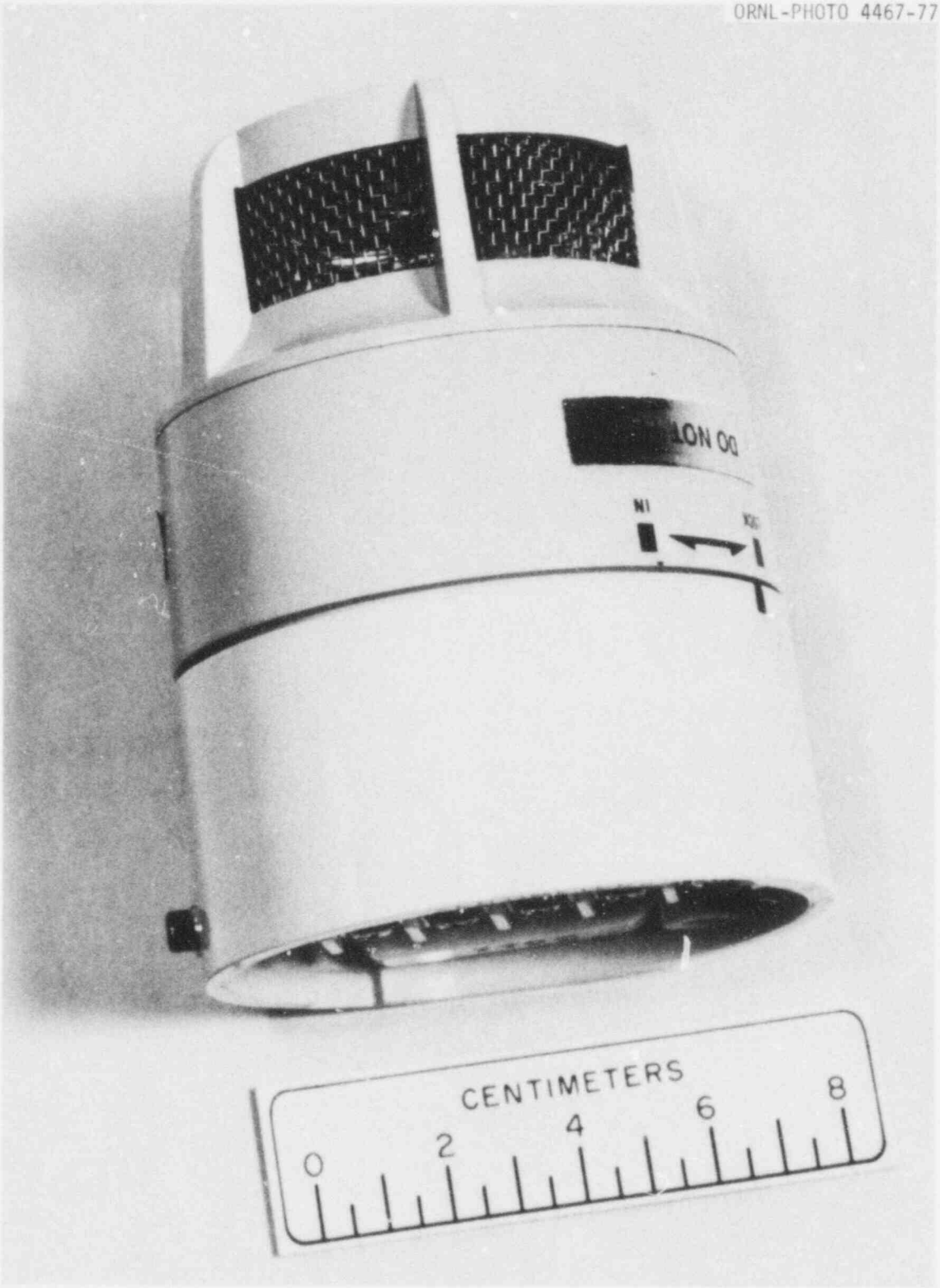


Fig. 14. Type 3 Whole Smoke Detector Tested at 1200°C

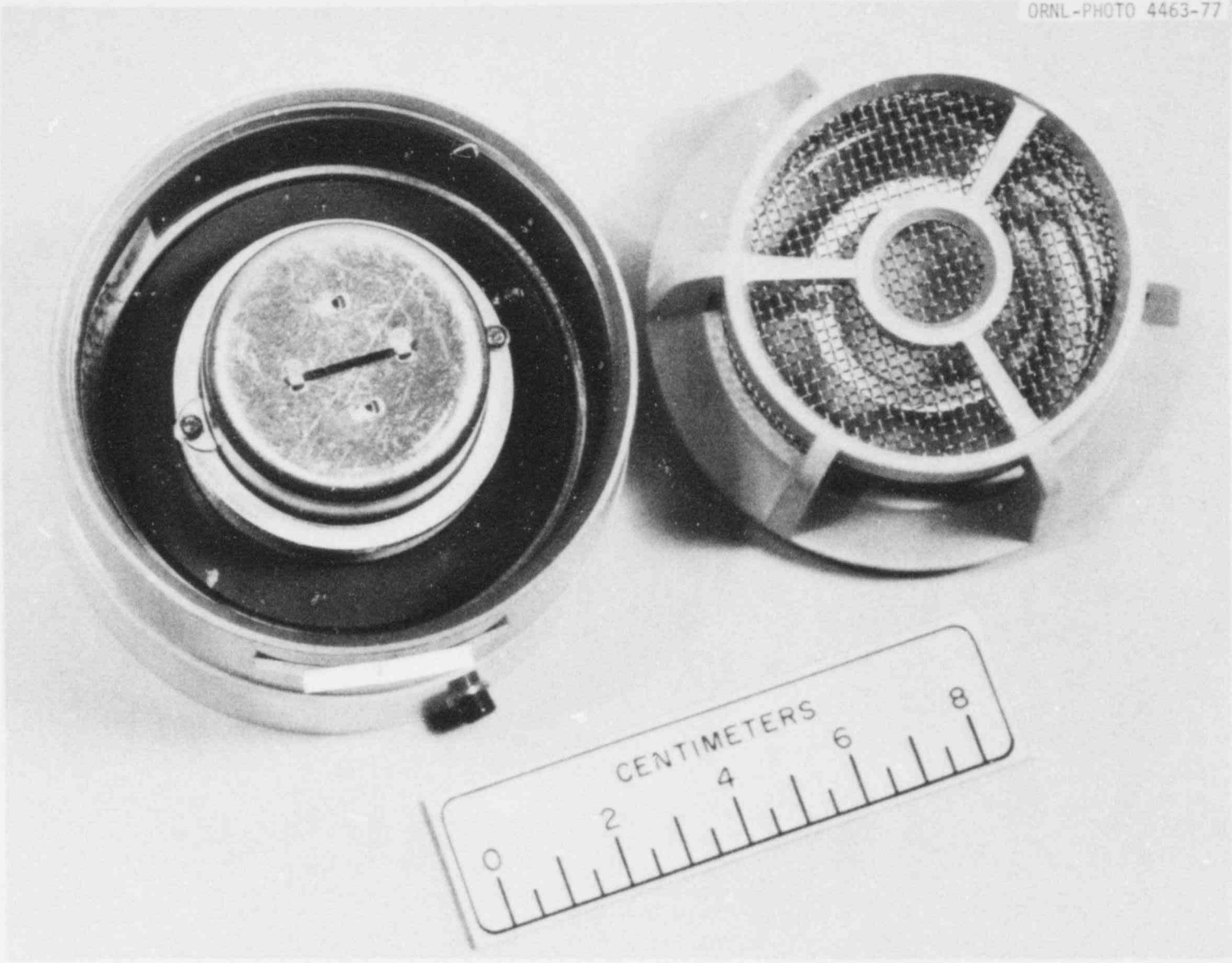


Fig. 15. Type 3 Whole Smoke Detector Before Testing at 1200°C
(cover removed showing arrangement of americium-241 strip sources)

Fig. 16. Type 3 Whole Smoke Detector After Heating to 1200°C

Fig. 17. Whole Smoke Detector After Heating to 1200°C (wire screen displaced in order to show inner damage)

Table 7. Distribution of Weight and Radioactivity in Loose Debris From Smoke Detector No. 3 Destruction Test at 1200°C

Sieve Size, microns	Weight of Debris Retained on Sieve, grams	% of Total Weight Analyzed	^{241}Am Activity, µCi*	% of Total ^{241}Am Activity	Concentration ^{241}Am, µCi/gram
2000	14.4	44.9	6.74×10^{-3}	1.91	4.68×10^{-4}
850	4.8	15.0	1.46×10^{-1}	41.4	3.04×10^{-1}
595	2.1	6.54	5.41×10^{-2}	15.3	2.58×10^{-2}
250	9.6	29.9	1.32×10^{-1}	37.4	1.38×10^{-2}
60	1.1	3.43	1.24×10^{-2}	3.51	1.13×10^{-2}
10	0.1	0.31	2.19×10^{-3}	0.62	2.19×10^{-2}
Sum:	32.1	100.08	3.53×10^{-1}	100.14	

*No correction made for self absorption of 60 keV gamma from ^{241}Am.

Following several of the 1200°C tests, attempts were made to obtain a mass balance from the results of the analysis of the various components (wipes, traps, washings, etc.). Sources, and in most instances the sampling boats, were dissolved in hot concentrated nitric acid followed by dissolution in hot aqua regia and analyzed for americium-241. A mass balance was then attempted (Table 8). In most cases the dominant radioactivity was associated with the residual sources and/or sampling boat. The results of the mass balance did not conform well to the listed source activity as stated by the manufacturer.

Integrity testing of the sources (Table 9) indicated a high reliability in performance. The precision of uniformity of loading within a batch of similar sources as supplied by two manufacturing firms ranged from approximately 3 to 9 percent with a median between 4 to 5 percent.

Alpha analyses of the surface wipes made on each of these sources were typically low, ranging from below detection limits (8×10^{-6} µCi) up to 560 pCi (5.6×10^{-4} µCi). All unmounted sources tested with cellophane tape indicated a range in activity from below detection limits (8×10^{-6} µCi) up to 550 pCi (5.5×10^{-4} µCi) with a weighted mean of approximately 2.3×10^{-5} µCi.

Accuracy of the americium-241 content of the sources (Table 10) varied from the manufacturer's stated activity level. Manufacturers' tolerances for these sources usually stipulated a given activity level or lower, but in some instances a deviation between 10 to 15% above the stated level for randomly selected sources was acceptable. Only six of the sources examined were found to be higher than indicated by the source manufacturer and of these only one exceeded 15%.

Table 8. Mass Balance on Sources Tested at 1200°C
(includes previous test results where applicable)

(values are given in μCi of ^{241}Am)

Run No.	Source Activity	Activity on Wipes	Airborne Contaminants	Dissolved Source and Boat	Summation of Activity	%Δ
2	1.5	4.3×10^{-3}	1.1×10^{-4}	7.3×10^{-1}	0.73	-51.3
3	1.5	*	5.7×10^{-5}	1.2	1.2*	-20.0
4	0.4(0.35)	6.5×10^{-3}	4.9×10^{-4}	4.9×10^{-1}	0.49	+22.5(+40.0)
5	5.0	1.6×10^{-2}	6.8×10^{-4}	3.8	3.8	-24.0
6	2.5	1.5×10^{-3}	7.7×10^{-5}	2.1	2.1	-16.0
7	0.4(0.35)	1.5×10^{-4}	8.1×10^{-4}	2.8×10^{-1}	0.28	-30.0(-20.0)
8	5.0	1.5×10^{-3}	5.7×10^{-4}	4.5	4.5	-10.0
9	2.5	1.6×10^{-2}	1.5×10^{-3}	2.1	2.1	-16.0
10	1.5(max)	6.7×10^{-2}	2.6×10^{-4}	6.2×10^{-1}	0.69	-54.0

*Source disintegrated into particles when removed from boat. Fragments dissolved and analyzed for radioactivity.

$$\%\Delta = \frac{(\text{summation activity} - \text{source activity})}{\text{source activity}} \cdot 100$$

\+ greater than stated source activity.
\- less than stated source activity.

Table 9. Results of Source Integrity Tests
(Americium-241 values expressed in mic.ocuries)

Run No.	Source Description	Source Activity	Number of Sources Tested	*Uniformity of Loading	Surface Wipes			Cellophane Tape Test		
					Low	High	Mean	Low	High	Mean
1	5 mm diam disc	0.35	28	5.4%	BDL	4.5×10^{-5}	6.4×10^{-6}	BDL	2.3×10^{-5}	4.5×10^{-6}
2	1/8" x 1/8" squares	0.9	30	2.8%	9.0×10^{-6}	5.6×10^{-4}	5.8×10^{-5}	BDL	5.5×10^{-4}	3.3×10^{-5}
3	5 mm diam disc	1.0	27	4.6%	BDL	4.6×10^{-5}	1.1×10^{-5}	BDL	9.5×10^{-5}	1.4×10^{-5}
4	3/32" diam disc	2.5	31	5.6%	2.2×10^{-5}	4.4×10^{-4}	8.0×10^{-5}	BDL	6.4×10^{-5}	8.6×10^{-6}
5	5 mm diam disc in tin plated source holder	6.1	30	3.0%	BDL	5.9×10^{-5}	6.0×10^{-6}	Not determined		
6	5 mm diam disc in tin plated source holder	0.35	30	7.1%	BDL	3.1×10^{-5}	2.8×10^{-6}	Not determined		
7	5 mm diam disc in tin plated source holder	1.0	30	4.5%	BDL	BDL	BDL	Not determined		
8	5 mm diam disc in stainless steel holder	1.0	30	4.8%	BDL	BDL	BDL	Not determined		
9	5 mm diam disc in nickel plated holder	1.0	30	4.9%	BDL	9.5×10^{-6}	2.7×10^{-6}	Not determined		
10	5 mm diam disc	0.2	24	3.5%	BDL	1.9×10^{-5}	2.1×10^{-6}	BDL	3.6×10^{-5}	6.0×10^{-6}
11	5 mm diam disc	0.45	24	4.3%	BDL	BDL	BDL	BDL	3.0×10^{-5}	3.2×10^{-6}
12	5 mm diam disc	0.7	23	5.6%	BDL	3.5×10^{-5}	1.2×10^{-5}	BDL	1.2×10^{-4}	2.7×10^{-5}
13	5 mm diam disc	2.2	24	3.7%	BDL	4.2×10^{-5}	8.2×10^{-6}	BDL	7.3×10^{-5}	2.2×10^{-5}
14	2.3 mm diam discs mounted back-to-back	5.0	19	6.8%	BDL	9.5×10^{-6}	3.8×10^{-6}	BDL	4.3×10^{-4}	7.9×10^{-5}
15	2.3 mm diam discs splits of back-to-back sources	2.5	10	8.9%	BDL	1.8×10^{-5}	7.8×10^{-6}	BDL	2.7×10^{-4}	8.5×10^{-5}

BDL = Below detection limit. ($\sim8\times10^{-6}$ μCi)
*Uniformity of loading expressed as an adjusted percent relative standard deviation (see text).

Runs 1-9: Sources supplied by Manufacturer No. 1.
Runs 10-15: Sources supplied by Manufacturer No. 2.

Table 10. Results of Americium-241 Analysis on Dissolved Sources

Run No.	Source Description	Americium-241 Content microcuri/source †Source Indication	Measured	%Δ
1	Strip source	0.4 (0.35)	0.28	-30.0(-20.0)
2	2.3 mm diam disc	2.5	2.3	-8.0
3	Strip source	0.4 (0.35)	0.35	-12.5(0.)
4	5 mm diam disc	1.5	1.65	+10.0
5	2.3 mm diam disc	2.5	2.45	-2.0
6	5 mm diam disc	1.5	1.55	+3.3
7	5 mm diam disc	0.35	0.34, 0.35	-2.9; 0.
8	1/8" x 1/8" squares	0.9	0.80	-11.1
9	5 mm diam disc	1.0	0.90	-10.0
10	3/32" diam disc	2.5	2.54	+1.6
11	5 mm diam disc in tin plated source holder	6.1	6.0, 6.0	-1.6,-1.6
12	5 mm diam disc in tin plated source holder	0.35	0.34, 0.31*	-2.9,-11.4
13	5 mm diam disc in tin plated source holder	1.0	0.99, 1.0*	-1.0,0.
14	5 mm diam disc in stainless steel holder	1.0	0.91, 0.88*	-9.0,-12.0
15	5 mm diam disc in nickel plated holder	1.0	0.98, 1.0*	-2.0,0.
16	5 mm diam disc	0.2	0.21	+5.0
17	5 mm diam disc	0.45	0.53	+17.8
18	5 mm diam disc	0.7	0.70	0.
19	5 mm diam disc	2.2	2.2, 2.4	0.,+9.1
20	2.3 mm discs mounted back-to-back	5.0	3.7, 4.9	-26.0,-2.0
21.	2.3 mm discs splits of back-to-back sources	2.5	1.9, 2.4	-24.0,-4.0

†Manufacturer's stated activity or as indicated on warning label.
*Sources removed from metal mount before analyzing.

$$\%\Delta = \frac{(\text{measured}-\text{indicated})}{\text{indicated}} \cdot 100$$

\+ greater than indicated value; - less than indicated value.
Runs 1-6: Sources removed from commercially available smoke detectors.
Runs 7-15: Sources supplied by source manufacturer No. 1.
Runs 16-21: Sources supplied by source manufacturer No. 2.

DISCUSSION AND CONCLUSIONS

Fire damage to smoke detector sources, tested either separately or with whole smoke detectors, consistently produces airborne contamination to the extent of 0.01% of the source or typically 0.1 nCi. As much as 0.1% may be airborne within a few centimeters of a 1200°C source, but condensation or settling within the test chamber itself normally removes this activity from the air. Sources exposed to high temperature, short of fusion, released 10^{-4} µCi to surface wipes and only those sources that are mounted in brass exceeded 10^{-3} µCi. Since the brass mounts apparently accelerated fusion or oxidation of the source material, it was suspected that solder might also attack the sources at high temperature. When sources were heated to 600°C with a bead of 50:50 (Pb:Sn) solder they were converted to a powdery residue. Thus, although the volatility of the sources is exceedingly low, even at temperatures as high as 1200°C, metallic fusion of the sources produces contaminated powders. Sources heated to 1200°C in whole smoke detectors apparently underwent similar oxidation and/or fusion with other components of the units. Consequently, the contaminated residue from whole smoke detector tests was powdered, whereas the residue from tests of sources only was typically a metallic bead. Even in the source only tests, however, as much as half the americium-241 is transferred from the metallic bead into the nickel boat containing the source.

In comparing results of these tests to proposed rejection criteria,[2] only the brass mounted sources would fail. The failure criterion for the 1200°C test is expressed as a percentage of source activity. Such a specification requires that the source activity be known and a criterion that specifies an absolute level of release would simplify testing. It would also mean that all sources would have to meet the same release criterion, regardless of the amount of activity contained.

Total recovery of americium-241 in test procedures declined with increasing complexity of tests and with increasing numbers of components to be analyzed. Thus, although overall recovery was above 95% for simply dissolving and analyzing sources, dissolving tested source beads and contaminated nickel boats plus leaching the test chamber yielded lower recoveries, averaging 75%. For this reason the trap efficiency tests are important in that they show that the majority of airborne activity was recovered in the first trap and the losses experienced in complex test series do not represent uncollected airborne activity. Rather, they are suspected to represent compounded slight losses at several steps in the preparation of samples for analysis.

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