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AEC RESEARCH AND DEVELOPMENT REPORT

CONFINEMENT OF AIRBORNE RADIOACTIVITY

PROGRESS REPORT
JULY - DECEMBER 1971

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L. R. JONES

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Savannah River Laboratory

Aiken, South Carolina

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CONFINEMENT OF AIRBORNE RADIOACTIVITY

Progress Report: July-December 1971

by

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ABSTRACT

Screening tests on 21 different iodine adsorbers were continued to aid in selecting a replacement carbon for the SRP confinement system. Carbon samples impregnated with I_2, KI_3, triethylenediamine (TEDA), KI, PbI_2, and Ag were subjected to high-intensity gamma irradiation, high-temperature desorption, and simulated service aging tests. Coconut-base carbons impregnated with either TEDA or TEDA plus KI were found to have the best overall iodine retention properties under the test conditions.

Two new test facilities for carbon weathering and high-intensity gamma irradiation were designed and partially constructed. Iodine adsorbers selected from the screening tests will be evaluated in the new test facilities.

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INTRODUCTION

The activity confinement system for each Savannah River production reactor is designed to collect halogens and particles that might be released in the unlikely event of a reactor accident. A continuing program is in progress at the Savannah River Laboratory to evaluate the performance of the confinement system for removing airborne radioactivity under adverse operating conditions and to develop techniques to enhance its reliability and efficiency.

Previous confinement system studies[1-9] at Savannah River have shown that elemental iodine retention on activated carbon is most strongly influenced by the operating temperature of the carbon beds, the moisture content of the air passing through the beds, the length of time the carbon has been in service, and the radiation exposure to which the beds are subjected after iodine loading.[2-9] Controlling parameters of radiation desorption phenomenon[9] are radiation intensity and duration, carbon type and service history, and the composition and relative humidity of the purge gas stream. Because the radiation-induced iodine release from carbon results primarily from the formation and subsequent desorption of organic iodides, iodine adsorbers impregnated with materials which will react with organic iodides are more effective for iodine retention than the unimpregnated carbon currently used in the confinement system. As a result of these findings, an extensive series of screening tests was undertaken to select a better iodine adsorber for use at Savannah River.

Some preliminary screening test results were summarized in a previous report.[9] This report summarizes the complete results from the test series and outlines the planned use of a new test facility where a wider range of temperature, humidity, and radiation exposure conditions can be obtained.

SUMMARY

Adsorption of iodine on activated carbon following a major reactor accident could result in radiation dose rates in the carbon beds in excess of 10^7 rads/hour. In a high-intensity radiation field, a portion of the adsorbed iodine is converted to organic iodides. This form of iodine is poorly retained by the unimpregnated carbon that is currently used in the SRP confinement system.

A series of screening tests was designed to evaluate available iodine adsorbers which could be used in the confinement system to replace the unimpregnated carbon. Performance of the adsorbers was tested under conditions of high-intensity irradiation, high temperature desorption, and accelerated weathering by NO_2. A total of 21 different adsorbers were tested including unimpregnated carbon; carbons impregnated with KI_3, PbI_2, I_2, triethylenediamine (TEDA), and TEDA plus KI; and inorganic adsorbers containing silver. Under the test conditions used, the coconut carbons impregnated with either TEDA or TEDA plus KI performed better than the other adsorbers as indicated in Table I.

New test facilities were designed and partially constructed that will permit further evaluation of these iodine adsorbers. Up to 40 carbon samples can be simultaneously exposed to reactor building exhaust air in a new carbon weathering facility. Iodine retention performance can be evaluated under a variety of temperature, humidity, and radiation intensity conditions in a new high-intensity gamma irradiation facility. Performance characteristics of other confinement system components (moisture separators, HEPA filters, gaskets, and adhesives) can also be evaluated in the new irradiation facility.

TABLE I

Screening Test Results

			Measured Iodine Penetration, %			
Mfgr.	Adsorber	Impregnant	Control[a]	Radiation[a]	NO_2[a]	High[a] Temp
A	coconut	None	<0.001	0.283[b]	0.061[b]	0.004
B	coconut	None	<0.001	0.299	-	-
A	coconut	KI_3	<0.001	0.017[b]	0.229[b]	0.052
A	coconut	KI_3	0.002	0.132[b]	-	-
B	coconut	KI_3	<0.001	0.014[b]	0.029[b]	0.028[b]
B	coconut	KI_3	<0.001	0.029	0.074	0.056
C	coconut	KI_3	0.001	0.030[b]	0.364	18.08
D	coconut	KI_3	-	0.045	0.552	2.412
E	petroleum	KI_3	0.011	0.058[b]	2.049[b]	6.484[b]
B	coconut	I_2	<0.001	0.021	0.043	0.070
A	coconut	PbI_2	0.009	0.046	0.117	-
A	coconut	PbI_2	-	-	0.029	-
B	coal	PbI_2	0.002	0.015	1.877[b]	-
B	coconut	PbI_2	<0.001	0.160[b]	0.433	0.084
B	coconut	KI + TEDA	<0.001	0.037[b]	0.046[b]	0.006[b]
B	coconut	TEDA	<0.001	0.003[b]	0.018[b]	0.003[b]
E	petroleum	TEDA	-	0.008	0.080	12.03
E	petroleum	TEDA	-	0.014	0.028	4.556
B	inorganic	Ag	-	0.168	-	-
B	inorganic	Ag	0.008	0.195	-	-
F	inorganic	Ag	0.002	0.011	-	-

a. Test conditions are described in text.

b. Average of replicate determinations.

DISCUSSION

SCREENING TESTS FOR IODINE ADSORBERS

Three screening tests and a control test were used to simulate a range of conditions to which carbon in the confinement system might be subjected. The three conditions are: 1) radiation exposure (as a result of adsorption and subsequent decay of radioiodine on the carbon), 2) high temperature (180°C) exposure after iodine loading (assuming partial failure of emergency core cooling), and 3) artificial weathering using NO_2 to simulate service weathering of the carbon.

Test Conditions

In all tests, 2-inch-diameter by 1-inch-thick test beds were used. Air filtered by a 1-ft thick carbon bed followed by a HEPA filter was used in all tests and passed through the test beds at a face velocity of 55 ft/min. Iodine loading was approximately 0.7 mg I_2/g adsorber for carbon samples and approximately 0.35 mg I_2/g test material for inorganic adsorbers.

Control and Radiation Tests

Radiation screening tests were run at an absorbed dose rate of 1.5×10^7 rads/hour in the carbon. The control test was run under the same temperature and humidity conditions as the radiation test, but outside the ^{60}Co radiation field. Elemental iodine was loaded onto the carbon test bed over a 60-minute period in an air stream at 80°C and 50% relative humidity. Iodine was then desorbed for 4 hours at 80°C and 50% relative humidity. Iodine was loaded by vaporizing freshly precipitated $^{127}I_2$ tagged with ^{131}I into a prefiltered air stream in the manner described in previous reports.[7,8]

High Temperature Test

Iodine ($^{127}I_2$ tagged with ^{131}I) was loaded onto the carbon test bed in 10 minutes in air at ambient conditions (23°C and ∿50% relative humidity) and desorbed for 4 hours with hot (180°C), dry air. The high temperature tests were run in the absence of the high-intensity gamma radiation field because the irradiation test apparatus was limited to temperature below 80°C.

NO_2 Test

Samples of test carbon were artificially weathered in an NO_2-air stream as discussed in Reference 8. Total NO_2 exposure was 91.1 mg NO_2/g carbon or the equivalent of 36 months of weathering in the SRP confinement system. Elemental iodine, tagged with ^{131}I, was loaded onto the test bed as described in the high temperature tests. Iodine was desorbed for 4 hours with 80°C air saturated with moisture. Because adsorbed NO_2 is rapidly desorbed at elevated temperatures, it was necessary to run the NO_2 weathering tests in a separate apparatus outside the radiation field.

Test Results

A total of 21 different iodine adsorbers were subjected to one or more of the screening tests. The primary objective of these tests was to select the best adsorbers for more detailed testing and eventual use in the confinement system. For this reason, some products that retained iodine poorly in one or more tests were not subjected to the complete series of screening tests.

All tests used new carbon as supplied by the vendors. Several general conclusions are indicated by the test data shown in Table I:

- The iodine retention efficiencies of coconut-base carbons impregnated with KI_3 varied widely from vendor to vendor, particularly in the high-temperature and NO_2 tests. In some cases the base carbon was the same, but different methods of impregnation were employed. Thus, the method of KI_3 impregnation appears to affect the performance of the product for nuclear applications.

- Petroleum-base carbons had consistently poorer iodine retention properties in the high-temperature test than most coconut-base carbons, regardless of the impregnant.

- Triethylenediamine-impregnated carbons were among the better performers both in the radiation test and the NO_2 test. One adsorber, a TEDA-impregnated coconut-base carbon, gave consistently better test results than all other adsorbers. A coconut carbon impregnated with TEDA and KI performed nearly as well as the carbon impregnated with only TEDA in most of the tests.

- The iodized carbons (KI_3, PbI_2, and I_2) are more sensitive to temperature and simulated weathering than either unimpregnated or TEDA-impregnated carbons (the exception being the TEDA-KI combination).

- Use of insoluble PbI_2 instead of highly soluble KI_3 appears to have no benefit for iodine retention under the conditions used in this test series.

- All the impregnated adsorbers performed better in the radiation test than the unimpregnated coconut-base carbons. This is consistent with earlier findings and the principal mechanism of iodine penetration in an intense radiation environment is formation and subsequent desorption of organic iodides.

EFFECT OF PURGE GAS COMPOSITION

Two additional tests were run on unimpregnated carbon in the radiation field to obtain more information on reaction mechanisms. Temperature, radiation field, and test duration were the same as those described earlier for the radiation screening tests except that nitrogen was substituted for air in the first experiment and dry air (<0.1% relative humidity) was used in the second experiment. Test results are summarized in Table II. Test results from the normal radiation screening test are included for comparison.

TABLE II

Effect of Purge Gas Composition on Iodine Desorption

Radiation Field, rads/hr	Purge Gas	Relative Humidity, %	Iodine Penetration,[a] %
1.5×10^7	Air	50	0.283
1.5×10^7	N_2	50	0.474
1.5×10^7	Air	<1	0.020

a. 1-hour loading, 4-hours desorption, unimpregnated carbon

If the principal reaction mechanism for organic iodide formation is a free radical mechanism, oxygen in the purge gas stream should reduce the organic iodide yield by reacting with iodide precursors. The test results show that more iodine is desorbed in N_2 gas than in air and indicate that the postulated free radical mechanism is correct.

Anhydrous air as a purge gas should inhibit organic iodide formation by restricting the supply of hydrogen to complete the reaction. Equipment limitations prevented running an anhydrous test; however, minimizing moisture with a dry air purge decreased iodine desorption more than one tenth. In the dry air test, approximately 70% iodine desorption occurred during the loading phase when air was being drawn through a glass frit containing moist iodine crystals. Desorption rates as a function of time are summarized in Figure 1.

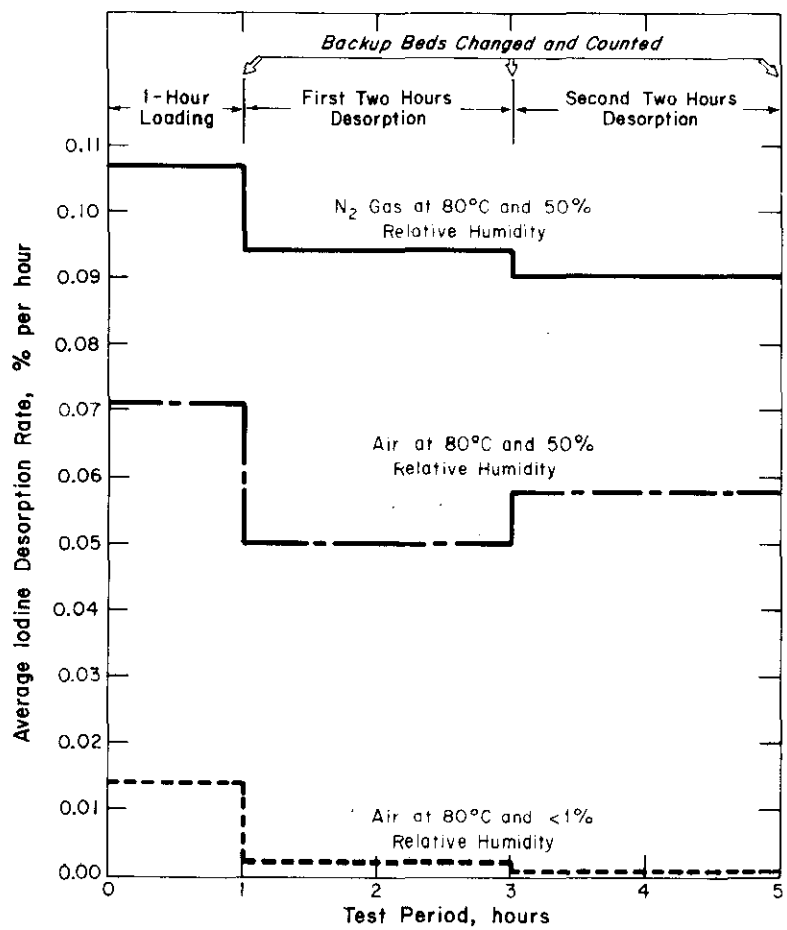

FIGURE 1. Effect of Purge Gas Composition on Iodine Desorption

NEW TEST FACILITIES

As a result of the preliminary screening tests, those adsorbers showing the most promise will be evaluated in a more detailed series of tests and the best adsorber will be selected for use in the reactor confinement systems. Limitations in test facility design and unavailability of service-aged impregnated carbons limited the scope of testing of potential replacement carbons as indicated in the previous report.[9] Two new test facilities were designed and are under construction that will provide the necessary diverse test conditions to properly evaluate the adsorbers selected from the screening tests. The new facilities are a carbon weathering (service aging) facility and a new ^{60}Co irradiation facility.

Carbon Weathering Facility

A test facility has been designed and partially constructed that will permit simultaneous exposure of up to 40 samples of carbon to reactor building exhaust air. The test facility provides for exposing groups of carbon samples at face velocities up to 3 times normal to provide accelerated aging data. Individual samples are housed in one-inch-thick beds that are divided into two compartments (Figure 2). The first compartment, containing approximately 100 g carbon, is a 3-inch-diameter baffled bed that can be placed directly in the new irradiation test apparatus without disturbing the carbon in the bed. The second compartment of the exposure bed contains approximately 160 g of carbon which can be repacked into smaller beds for other tests.

The test facility itself is adjacent to the confinement system compartments in one of the reactor areas and draws air from the exhaust plenum upstream of the confinement system. A moisture separator and particulate filter precondition the air in the same manner as the confinement system. Filtered air from the test facility is returned to the exhaust plenum for reprocessing in the confinement system thus avoiding any breach of confinement should any of the test carbons prove inferior in service.

An unimpregnated carbon (control) and a variety of carbons impregnated with I_2, KI_3, TEDA, and TEDA plus KI will be exposed in the new facility at normal flow and approximately double normal flow to aid in the selection of a new iodine adsorber for use in the confinement system. In addition, full sized beds containing carbon impregnated with both TEDA and TEDA plus KI have been installed in one confinement compartment in a different reactor building. The larger sized carbon samples available from the full sized beds will permit testing iodine retention efficiencies over a wider range of temperature, humidity, and radiation conditions.

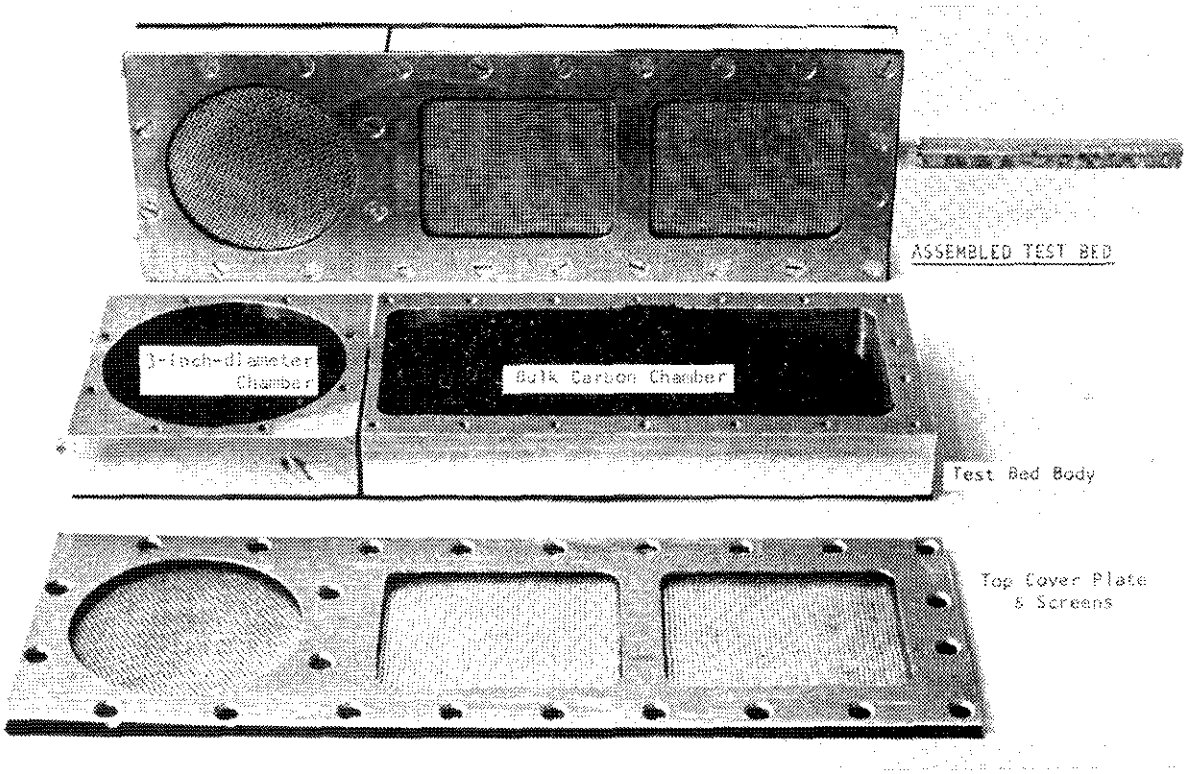

FIGURE 2. Carbon Weathering Facility Test Bed

New Irradiation Facility

Design capabilities of the new ^{60}Co irradiation facility were outlined in a previous report.[9] A schematic drawing of the new system is shown in Figure 3. Construction and checkout of the test assembly and the out-of-source components (hood, counters, etc.) were completed, the access tube was installed, and all 28 ^{60}Co slugs were loaded into the slug holder (1.64×10^6 Ci on 1/1/72). The measured dose rate at the center of the empty access tube was 4.7×10^7 rads/hr and 3.3×10^7 rads/hr in the center of the carbon test bed.

The slug holder can be positioned on a cylindrical storage rack away from the access tube to reduce shielding requirements when carbon irradiation tests are not in progress. An immersible can equipped with thermocouples and an air purge line was fabricated to fit inside the storage rack to facilitate static irradiation tests of various confinement system components when the slug holder is in the storage position. The gamma dose rate in this can is approximately 4.5×10^7 rads/hr. Coupons of gasketing material are shown hanging on the center spindle in the irradiation can in Figure 4.

The test apparatus and irradiation facility are shown in Figures 5 through 8. Figure 5 shows an exploded view of the test apparatus without its cooling water lines. Figure 6 is the assembled view with all lines in place. The slug holder is shown around the access tube in Figure 7 (the only light source in this photo is the Cerenkov radiation). An illuminated view of the irradiation facility is shown in Figure 8 with the slug holder positioned on the storage rack and the irradiation can in the center of the rack. The large tube on the left is the new 10-inch-diameter access tube. The two tubes on the right are the 6-inch-diameter access tubes of the older facility. Figures 7 and 8 were taken through 24 ft of water.

Test data obtained from the new irradiation facility will be reported in future progress reports.

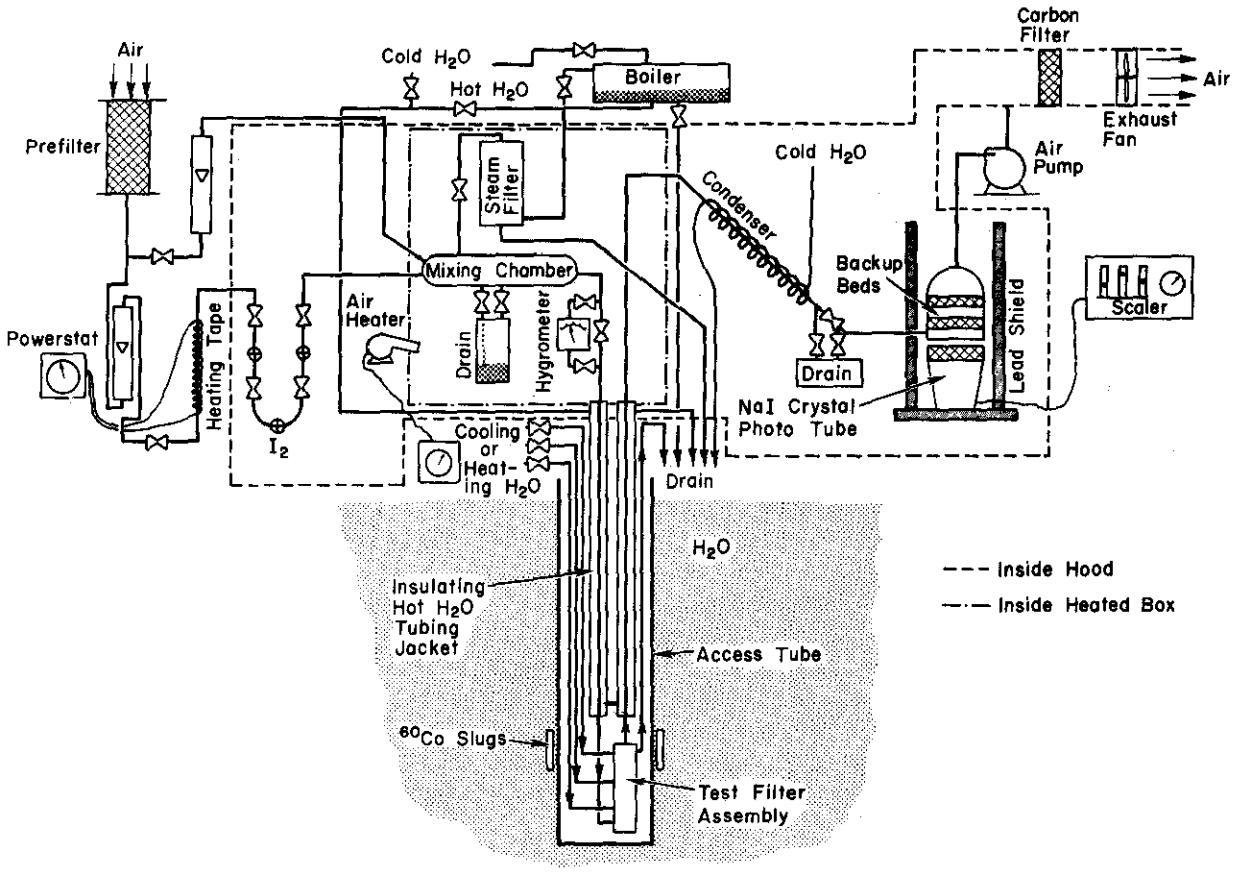

FIGURE 3. Schematic Diagram of New Irradiation Test Facility

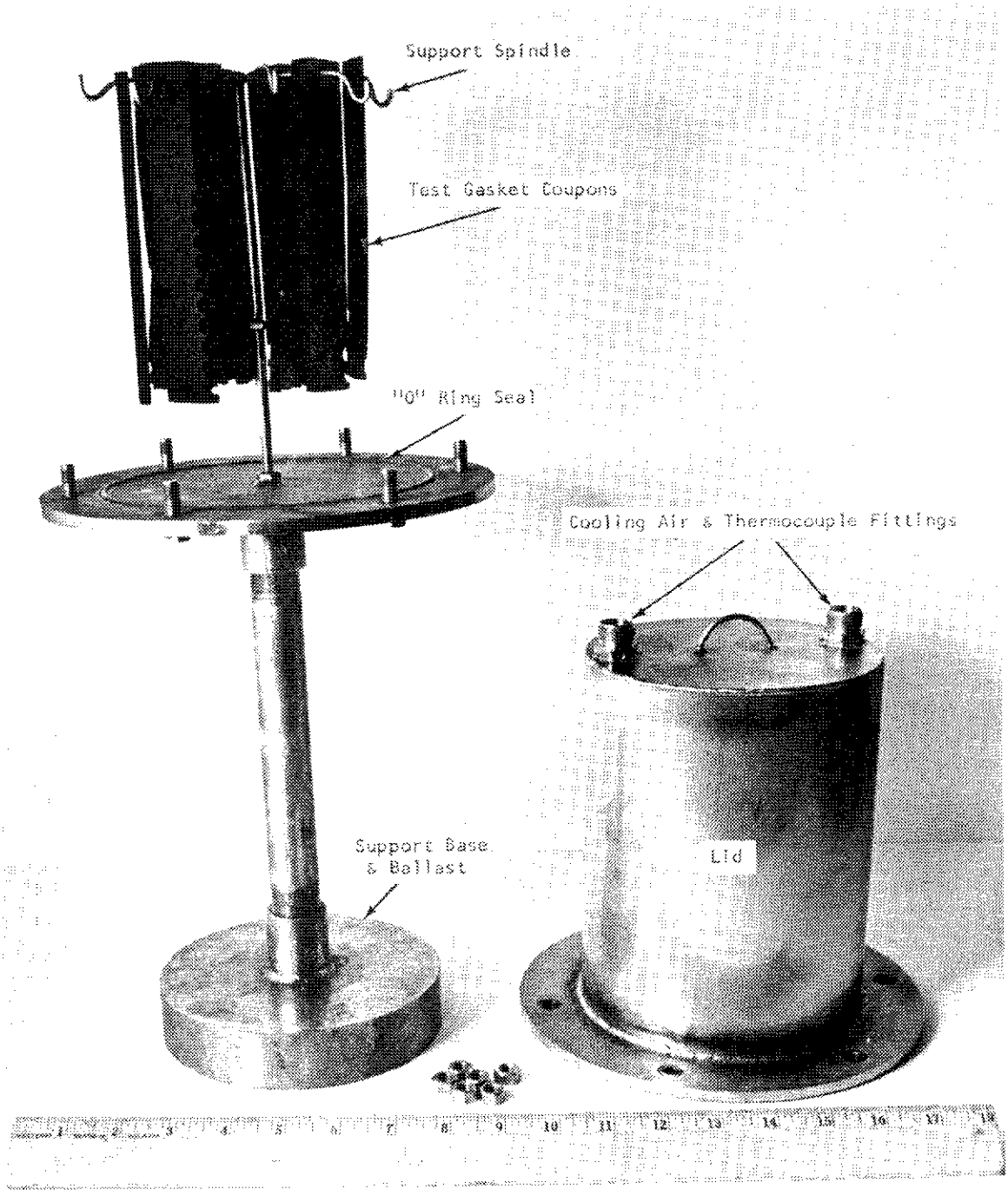

FIGURE 4. Irradiation Can with Gasket Coupons

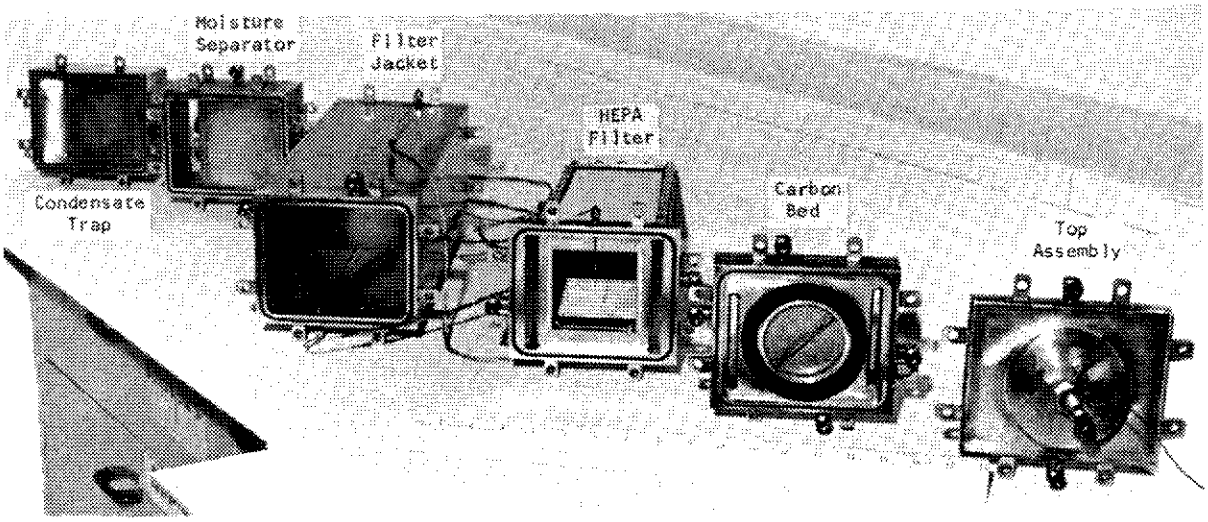

FIGURE 5. Exploded View of New Test Apparatus

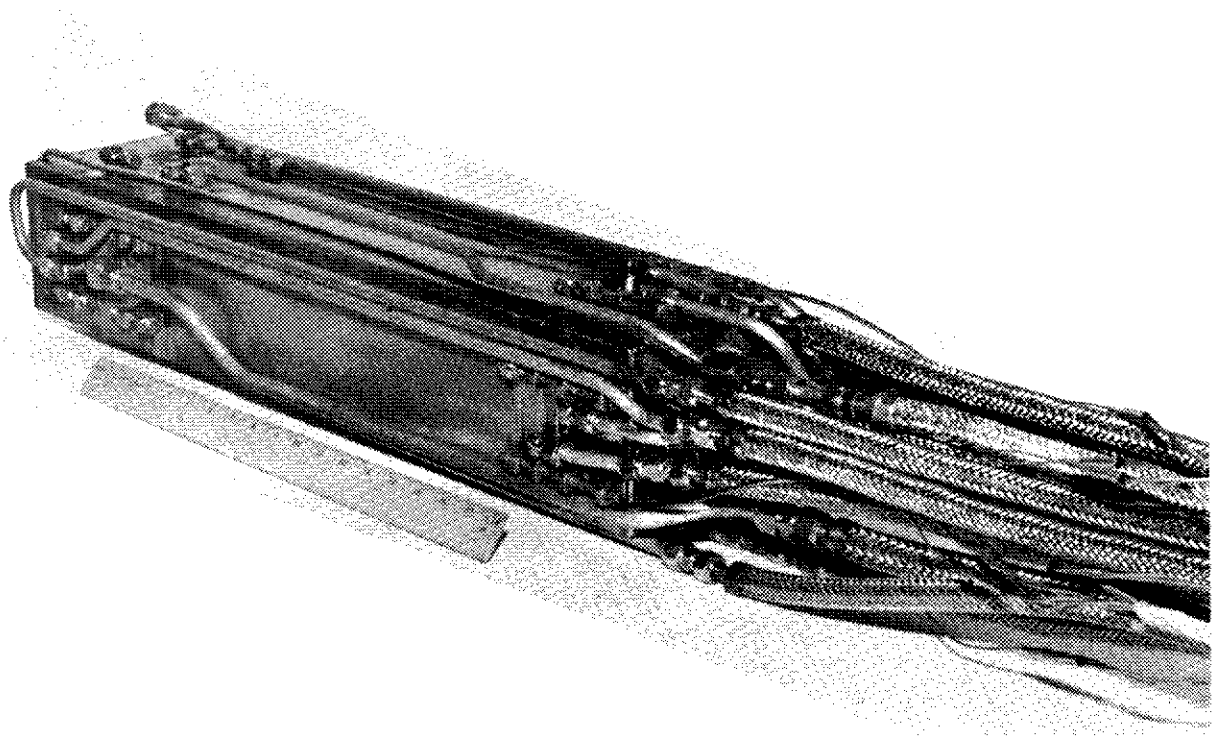

FIGURE 6. Assembled Test Apparatus

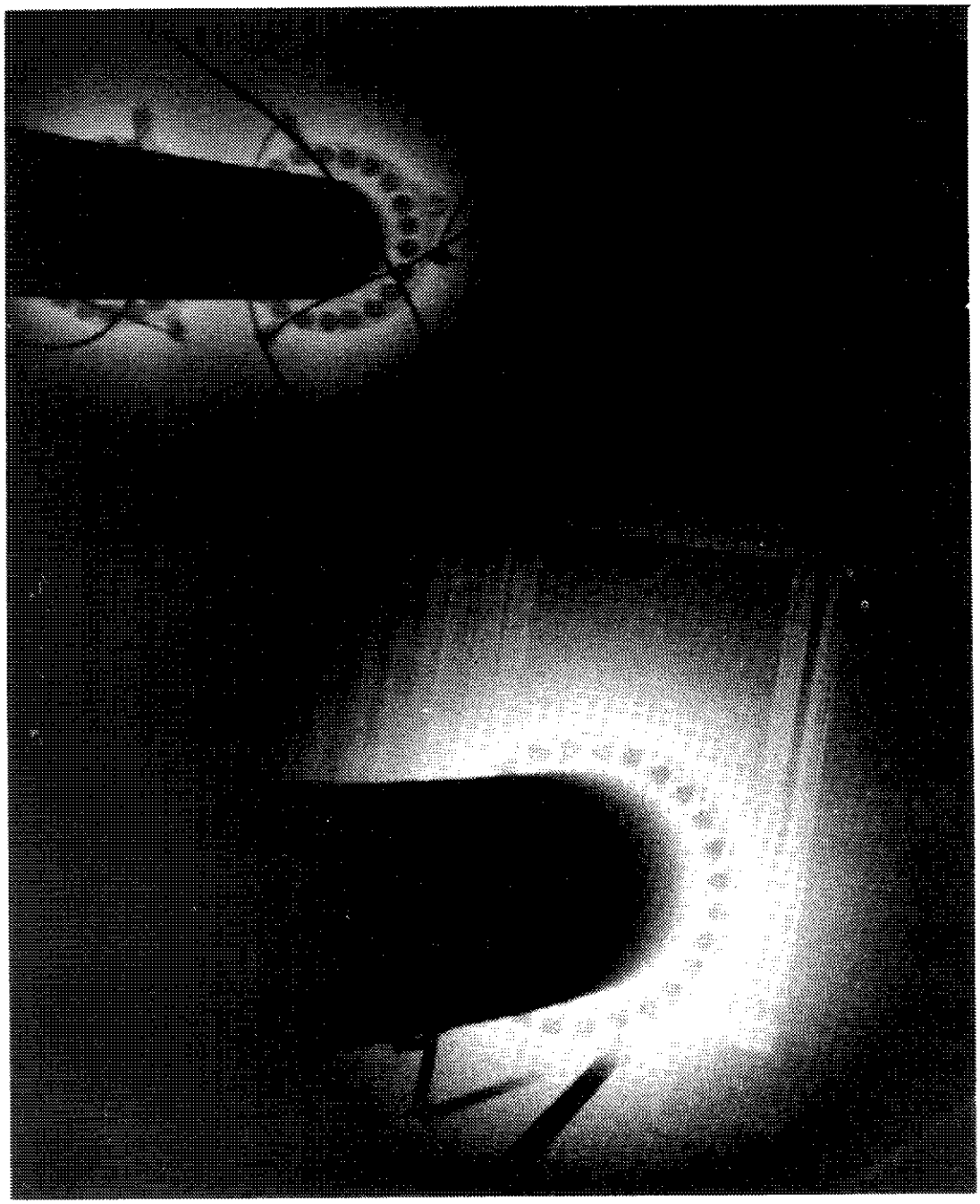

FIGURE 7. Access Tube with Slug Holder In Place

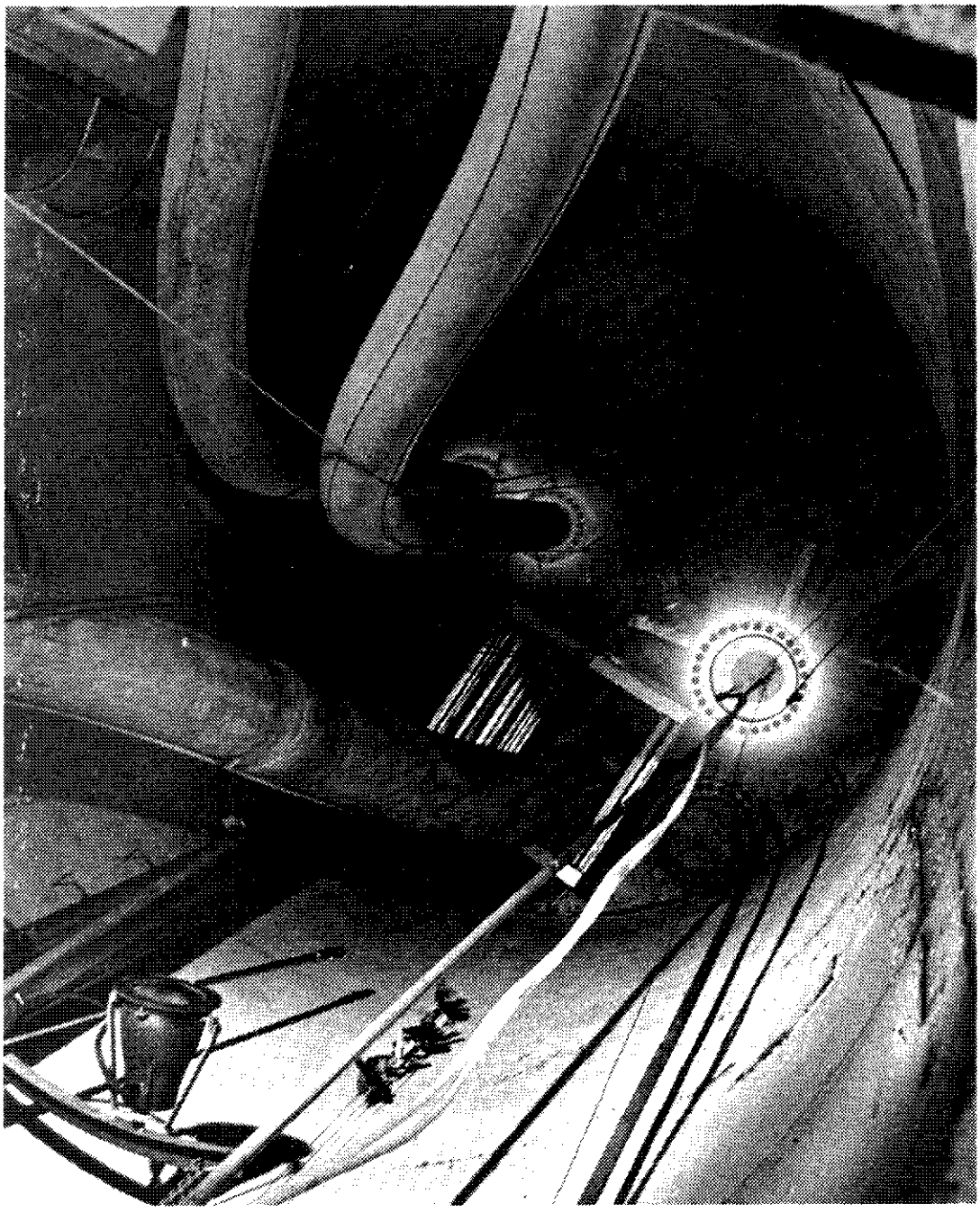

FIGURE 8. Slug Holder Positioned Around Irradiation Can

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