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AEC RESEARCH AND DEVELOPMENT REPORT

IODINE RETENTION STUDIES

PROGRESS REPORT: JULY-DECEMBER 1969

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IODINE RETENTION STUDIES
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by

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ABSTRACT

The retention of iodine by activated carbon was measured by tests simulating predicted conditions during power surge and loss-of-coolant accidents. The iodine retention efficiency of carbon in the power surge test decreases significantly with on-line service in the confinement system, but remains higher than 99.85% after 3.8 years. Studies in the loss-of-coolant test are incomplete. Partial regeneration of the carbon in both tests increased the iodine retention efficiency.

A facility with a 5×10^7 rads/hr ^{60}Co source for determining the effect of intense gamma radiation on methyl iodide adsorption, particulate filter media, and other materials is nearing completion.

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INTRODUCTION

The activity confinement system for each Savannah River production reactor is designed to collect radioactive gases and particles that might be released in the unlikely event of a reactor accident. At the request of the AEC Division of Operational Safety, a continuing program is in progress at the Savannah River Laboratory to evaluate the performance of the carbon used in the confinement system for iodine removal under adverse operating conditions and to develop techniques to enhance its reliability and efficiency.

Previous reports summarize the progress from January 1965 to June 1969.[1-3] This report summarizes the iodine retention studies at the Savannah River Laboratory from July to December 1969.

SUMMARY

In a test that simulates a reactor power surge accident occurring just before partial regeneration of the carbon, the iodine retention efficiency of activated carbon decreased significantly with service but was within specification (more than 99.85%) for up to 3.8 years service. The iodine retention efficiency of carbon also decreased significantly with:

- Increased iodine loading
- Length of time since partial regeneration
- Increased entrainment of moisture
- Reduced packing density in the test bed

A test that simulates a reactor loss-of-coolant accident is being developed. Partial regeneration of the carbon in both tests increased the iodine retention efficiency.

The new ^{60}Co irradiation facility with a source intensity of about 5×10^7 rads/hr is nearing completion. This facility will be used for methyl iodide radiolysis and for irradiation of other materials such as particulate filter media.

DISCUSSION

RETENTION OF IODINE UNDER ACCIDENT CONDITIONS

Test Modifications

The iodine adsorption efficiency test,[3] used to determine whether iodine removal in the Savannah River confinement system was within specification (>99.85% removal), was modified to evaluate carbon performance under accident conditions. The original test measured the iodine adsorption efficiency of carbon and showed the effects of service, iodine loading, and partial regeneration on iodine adsorption efficiency. The modified test determines carbon performance under two simulated reactor accident conditions: a power surge and a loss of coolant. The term iodine retention efficiency is used in this report because both adsorption and desorption determine how much iodine is retained by carbon.

Power Surge Test

In a power surge accident the temperature of the reactor would increase rapidly and a burst of steam would be released to the ventilation air. After a brief interval, the fuel would begin to melt and fission products would be released in a steam atmosphere. Hence, in a power surge accident carbon in the confinement system would first be exposed to a steam—air mixture, then to a steam—air—fission product mixture, and then to ambient conditions.

Procedure

In the power surge test, the apparatus[2] with the test bed in place is heated to 80°C in about 10 minutes to equilibrate temperature. To minimize regenerative effects, no air is passed through the test bed during this heating. The apparatus is kept at 80°C to minimize moisture entrainment while a steam—air mixture at 80°C flows (at a face velocity of 65 ft/min) through the test bed for 10 minutes. Iodine is then loaded on the test bed by passing steam—air—iodine (tagged with ^{131}I) mixture through the test bed for 10 minutes. After the iodine is loaded on the test bed, ambient air at ∿80% relative humidity flows through the apparatus for 15 minutes. Any iodine that penetrates the test bed is collected on backup beds. The iodine retention efficiency is the ratio of the iodine remaining on the test bed to the iodine loaded on the test bed. Carbon can be evaluated by the power surge test either before or after annual partial regeneration.[2,3]

Results

Effect of Service

The iodine retention efficiency of the carbon in the Savannah River confinement system remains within specification before partial regeneration for about 3.8 years, as determined in the power surge test (Figure 1). This curve represents conservative values for iodine retention efficiency because annual partial regeneration treatment (for the "Freon"* leak test) improves the carbon quality.[3] In a single test on carbon regenerated after 56 months of service, the iodine retention efficiency improved from 99.32% to 99.94%. However, preliminary tests indicate that partial regeneration may be beneficial for only a few months. Iodine retention efficiency decreases with increased service because impurities such as SO_2 and organic matter are adsorbed and because oxides of nitrogen destroy active adsorption sites for iodine.[3]

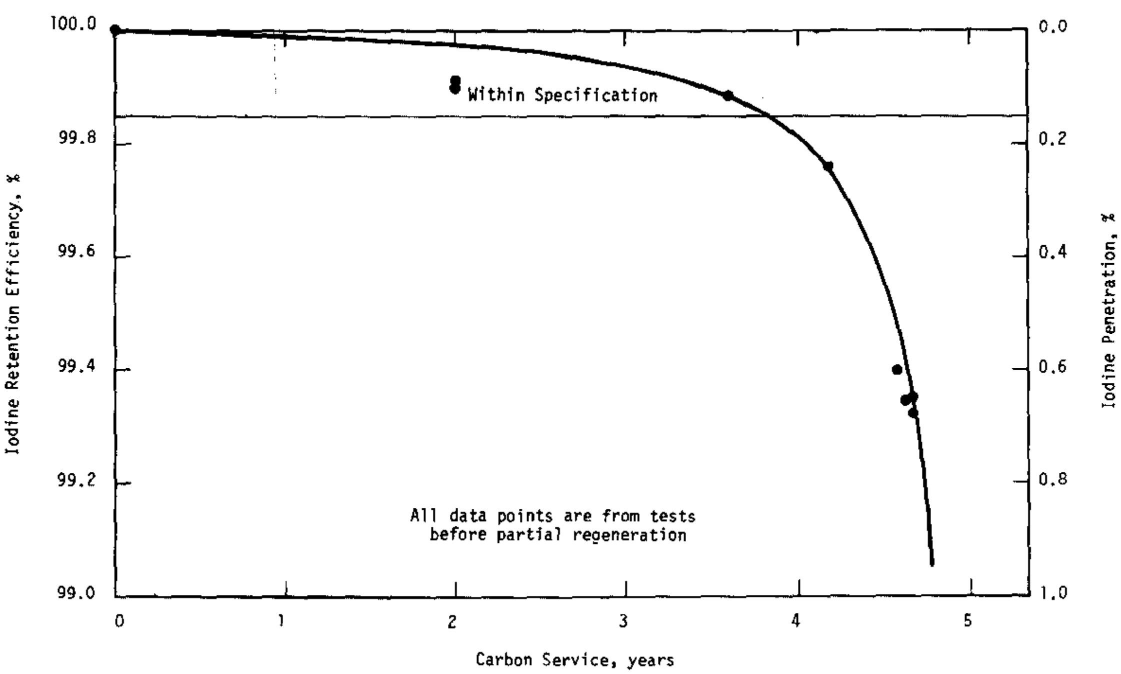

FIG. 1 EFFECT OF CARBON SERVICE ON IODINE RETENTION EFFICIENCY IN POWER SURGE TEST

* Trademark of DuPont.

In the power surge test, nearly all of the iodine penetrated the test bed during iodine loading (Figure 2). After the iodine is adsorbed on the carbon from steam—air at 80°C, ambient air flow at 80% relative humidity did not cause further iodine desorption.

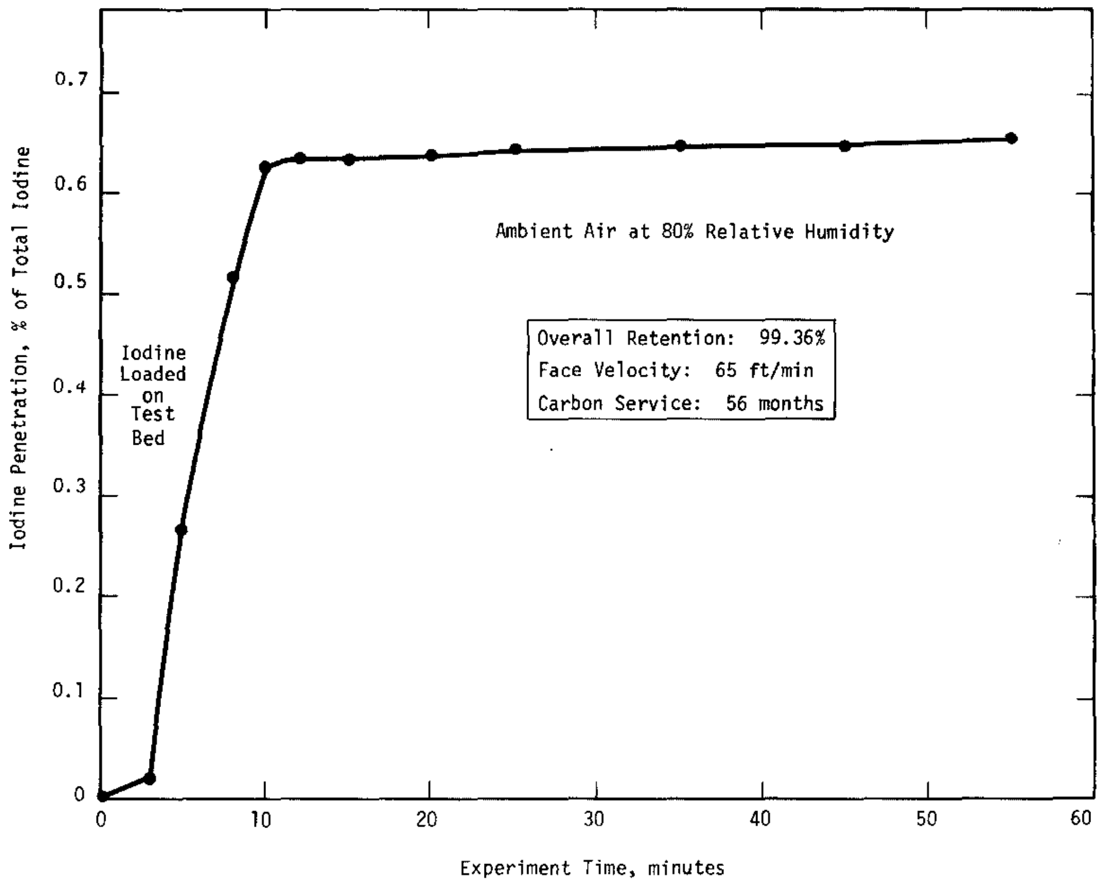

FIG. 2 IODINE PENETRATION DURING POWER SURGE TEST

As shown in Figure 3, the iodine retention efficiency of used carbon also decreases significantly as the iodine loading on the carbon increases. This effect is less pronounced with carbon having less service.[3]

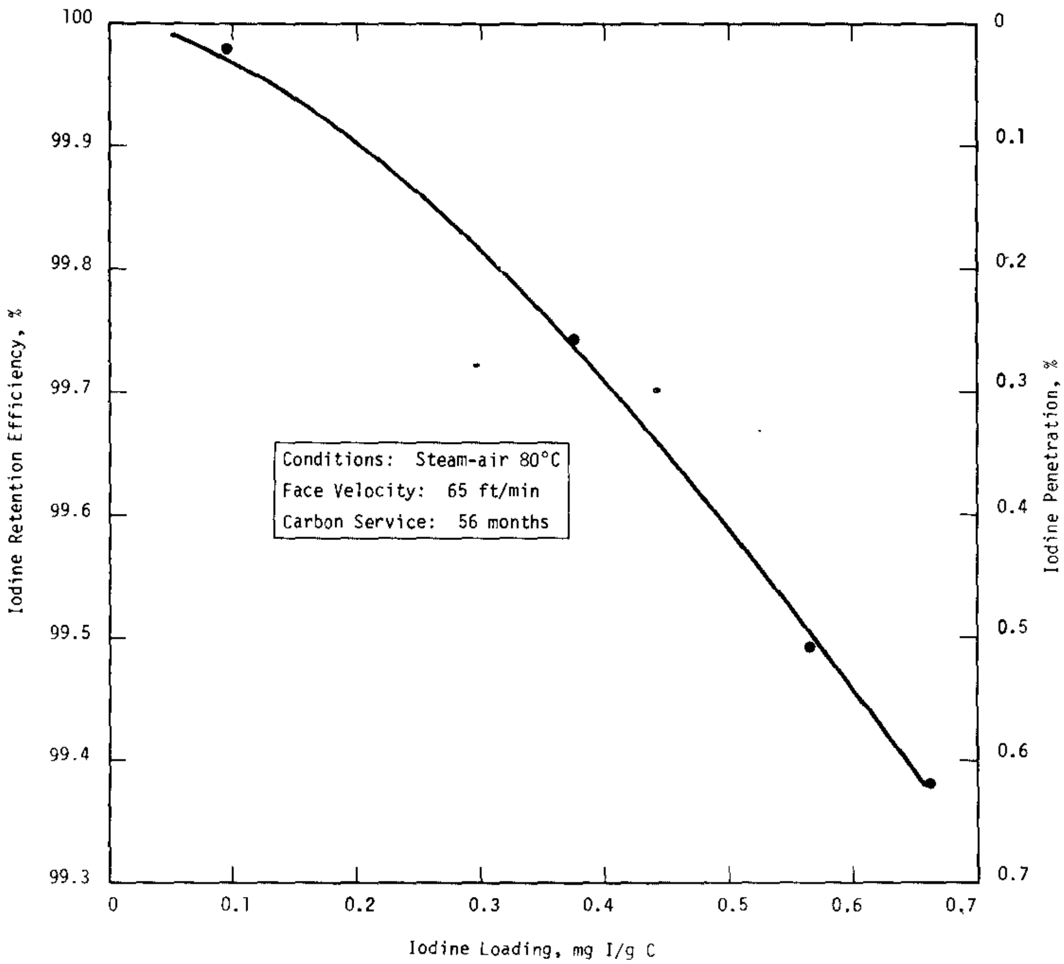

FIG. 3 EFFECT OF IODINE LOADING ON IODINE RETENTION EFFICIENCY IN POWER SURGE TEST

Effect of Entrainment

In the power surge test, water entrained in the test apparatus significantly decreases the iodine retention efficiency (Table I). When any of the components upstream of the test bed are below the temperature of the steam—air mixture (80°C), the steam condenses and water is entrained. The entrained moisture carries substantial amounts of iodine through the test bed and reduces the iodine retention efficiency significantly, as shown in Table I. In the experiments where visible entrainment occurred, the test bed was heated rapidly to 80°C, and the steam—air mixture at 80°C flowed through the test bed before the temperature inside the apparatus had reached 80°C; considerable condensation and entrainment resulted. The procedure was modified in subsequent tests so that the temperature inside the apparatus was 80°C before the steam—air mixture was added. With this modification entrainment was minimized, and the iodine retention efficiencies measured were more representative of those under actual accident conditions.

TABLE I

EFFECT OF ENTRAINED MOISTURE ON IODINE RETENTION EFFICIENCY

Conditions	Iodine Retention Efficiency, %	
	43 Months Service	56 Months Service
With entrainment	99.36	97.44
Minimum entrainment	99.89	99.06

In the test apparatus the volume-to-surface ratio of the upstream ductwork is 0.039 ft; in the confinement system this ratio is 2.95 ft. Condensation and entrainment increases with smaller volume-to-surface ratios. Therefore, the iodine retention efficiencies measured in the test apparatus are more conservative than those in the confinement system.

Effect of Reduced Packing Density

A decrease in density of the carbon packing significantly decreases the iodine retention efficiency of carbon. In a loosely packed bed, a bed with a lower than normal (0.62 inches H_2O) pressure drop across the test bed, iodine is not adsorbed by the carbon as readily as in a densely packed bed. As shown in Table II, iodine retention is reduced significantly if the packing density is reduced sufficiently to cause a 16% decrease in the pressure drop across the test bed.

TABLE II

EFFECT OF REDUCED PACKING DENSITY ON IODINE RETENTION EFFICIENCY

Packing Density as Indicated by Average Pressure Drop, inches H_2O	Iodine Retention Efficiency, %	
	35 Months Service	56 Months Service
0.62[a]	99.90	99.34
0.52	99.78	98.53

a Normal pressure drop across test bed.

Loss-of-Coolant Test

In a loss-of-coolant accident, coolant would drain suddenly from the reactor and the temperature of the fuel would increase rapidly. If the emergency core cooling system failed to operate promptly, the fuel would melt and release fission products to the ventilation air. Possible subsequent quenching of the hot fuel with water could release a burst of steam to the ventilation system. Hence, in a loss-of-coolant accident carbon in the confinement system would be exposed to fission products in ambient air possibly followed by a steam—air mixture.

Procedure

In the loss-of-coolant test, iodine (tagged with ^{131}I) is loaded on the test bed in 10 minutes in ambient air at a face velocity of 65 ft/min. The air flowing through the apparatus is heated to 80°C for 10 minutes, and then a steam—air mixture at 80°C flows through the test bed at a face velocity of 65 ft/min. In preliminary tests, steam—air flowed for 10 minutes. In later tests, this time was increased because iodine continued to penetrate the bed during steam—air flow. After the steam—air treatment air at 80% relative humidity flows through the test bed at a face velocity of 65 ft/min for 15 minutes. Iodine that penetrates the test bed is collected on backup beds. The iodine retention efficiency is the ratio of the iodine remaining on the test bed to the iodine loaded on the test bed. Carbon can be evaluated by a loss-of-coolant test either before or after the annual partial regeneration.[2,3]

Results

Although the development of the loss-of-coolant test has not been completed, preliminary tests show that the retention of iodine is satisfactory during iodine loading in ambient air (Figure 4). However, during the steam—air treatment at 80°C, iodine penetrates the test bed of carbon with 4.2 years service at a uniform rate for more than 5.5 hours. During the steam—air treatment, smaller water molecules may displace iodine in the pores of the carbon. After the steam is stopped and only air is passed through the apparatus (while it cools to room temperature) no more iodine penetrates the test bed. Additional tests are planned to further investigate iodine penetration.

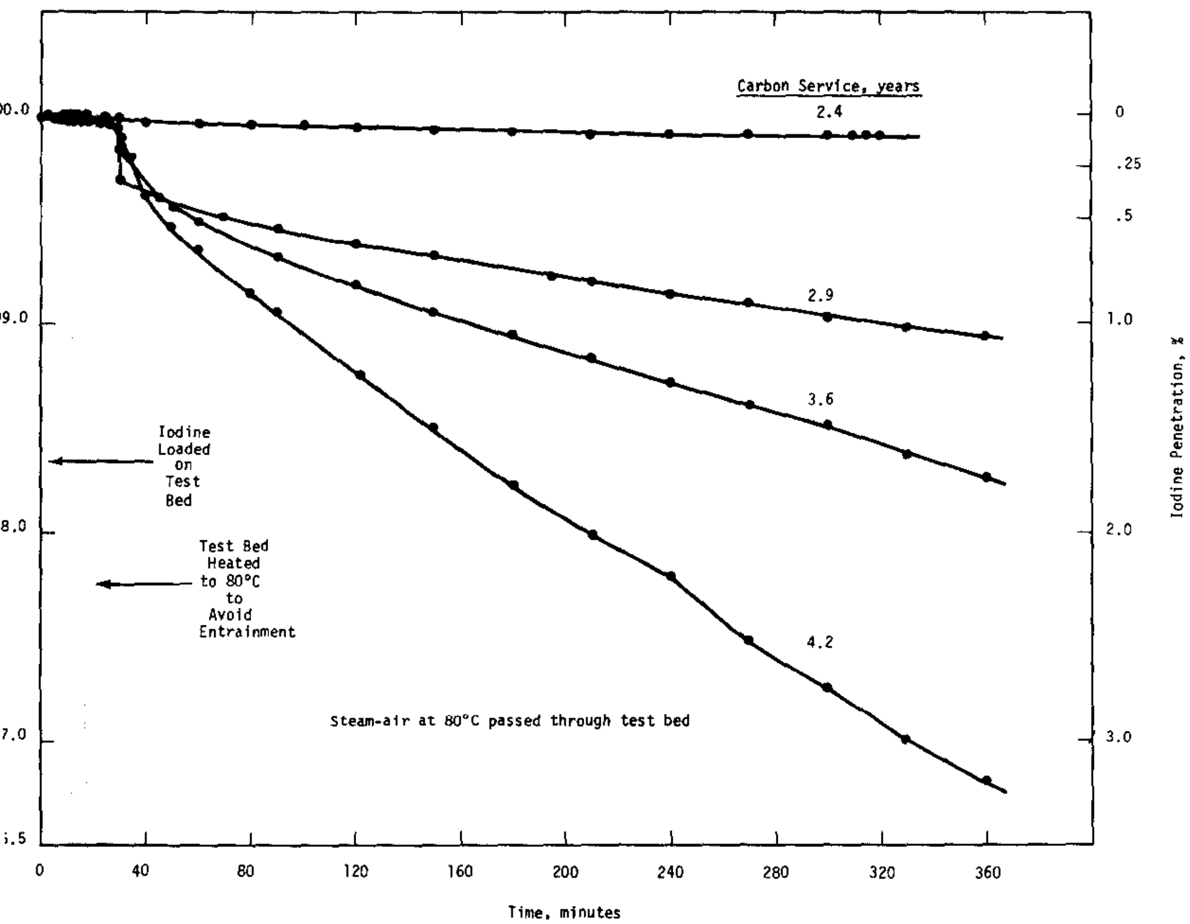

FIG. 4 IODINE RETENTION EFFICIENCY DURING LOSS-OF-COOLANT TEST

PARTIAL REGENERATION OF CARBON

As previously reported,[3] partial regeneration increases the iodine retention efficiency of carbon that has been in service. The improvement provided by partial regeneration was confirmed using the tests of iodine retention under accident conditions. As shown in Table III, the iodine retention efficiency is within specification (>99.85%) in both tests.

Further studies of partial regeneration will:

- Determine the effect of time after regeneration on iodine retention efficiency.
- Determine the effect of regeneration temperature on iodine retention efficiency.
- Determine the effect of regenerating agents (e.g., steam, air, and denitration agents, such as formaldehyde) on iodine retention efficiency.

TABLE III

EFFECT OF PARTIAL REGENERATION ON IODINE RETENTION EFFICIENCY

Test	Service, months	Treatment	Iodine Retention Efficiency, %
Power Surge	56	None	99.32
Power Surge	56	Partially regenerated[a]	99.94
Loss-of-Coolant	50	None	97.85
Loss-of-Coolant	50	Partially regenerated[a]	99.90

a Partially regenerated with air at 60°C flowing at 7.5 ft/min for 48 hours.

METHYL IODIDE RADIOLYSIS TESTS

Methyl iodide radiolysis tests are planned with the new ^{60}Co irradiation source. Figure 5 shows a diagram of the modified apparatus.

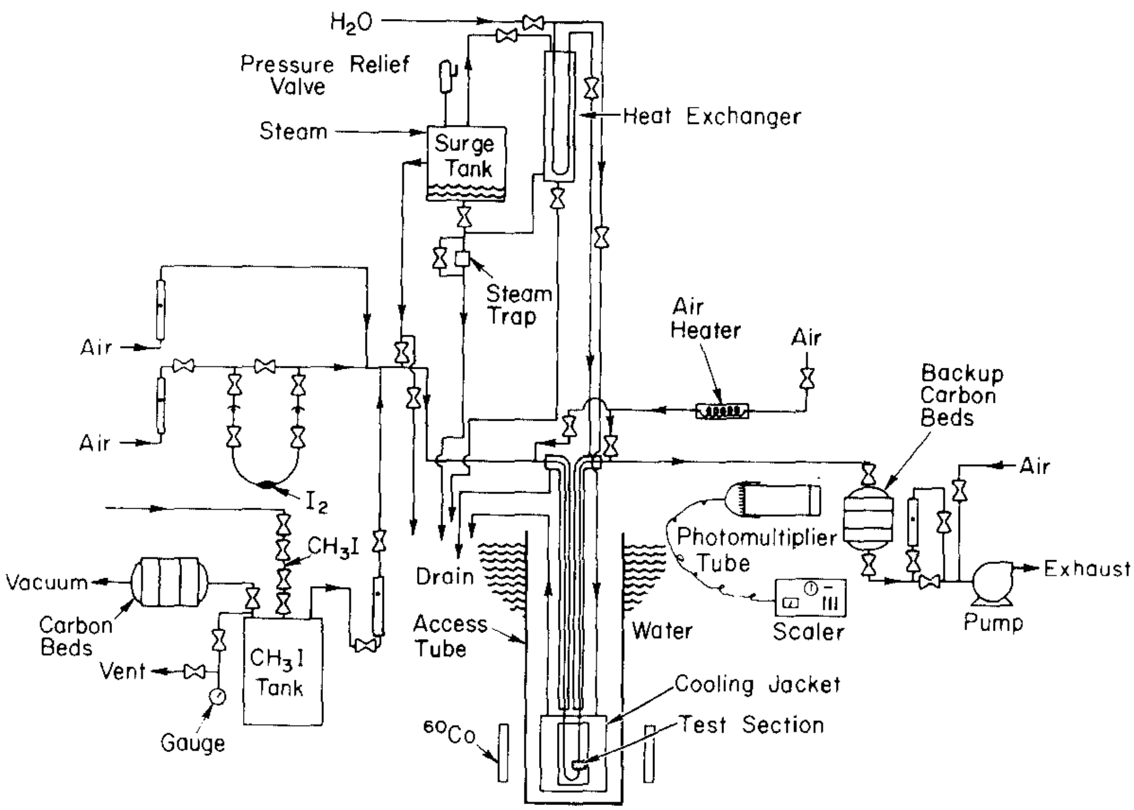

FIG. 5 METHYL IODIDE RADIOLYSIS APPARATUS

The first tests will provide data on gamma heating and dosimetry, necessary for predicting system behavior and gamma field strength in subsequent tests. Other tests will conform to one basic test sequence consisting of:

- Preheating the system for approximately 30 minutes with dry air at 65°C.
- Equilibrating the system for 30 minutes with steam-air flow at 65°C.
- Injecting elemental iodine into the system for 10 minutes.

- Injecting methyl iodide (tagged with ^{131}I) into the system, beginning 5 minutes after start of elemental iodine injection, and continuing for 1 hour in exponentially decreasing concentrations.

- Passing ambient air with no steam, elemental iodine, or methyl iodide through the system for 1 hour.

The effects of relative humidity, face velocity, absorbed gamma dose, iodine and methyl iodide loading on the test carbon, type of test carbon, and service life of the test carbon will be studied.

In addition, samples of materials used in confinement systems (e.g., filter media, gaskets, O-rings) will be irradiated in methyl iodide tests to determine the effect of gamma radiation on the physical properties of these materials. Measurement of physical properties after irradiation are planned by the Naval Research Laboratory.

REFERENCES

1. R. C. Milham. *High Temperature Adsorbents for Iodine - Progress Report: January 1965-September 1966.* USAEC Report DP-1075, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1966).

2. R. C. Milham and L. R. Jones. *Iodine and Noble Gas Retention Studies - Progress Report: October 1966-December 1968.* USAEC Report DP-1209, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1969).

3. R. C. Milham and L. R. Jones. *Iodine Retention Studies - Progress Report: January 1969-June 1969.* USAEC Report DP-1213, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1969).