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Progress Report: January - December 1974

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PREPARED FOR THE U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION UNDER CONTRACT AT(07-2)-1

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by

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ABSTRACT

Several commercial activated carbons, marketed for iodine removal in reactor off-gas cleanup systems, were evaluated for iodine penetration at elevated temperatures (4 hr at 180°C), and the penetration data varied widely. Subsequent chemical analysis of the samples indicated a strong correlation between the atom ratio of iodine to potassium (I/K) in the carbon and the high-temperature performance data. Carbons containing excess iodine (I/K >1) performed consistently poorer than carbons with excess potassium (I/K <1). Excess alkalinity (as indicated by a high pH of a water extract of carbon) was also shown to be required for efficient iodine retention. Improved iodine retention was obtained for three carbons (coconut, coal, and petroleum base) with proper adjustment of the I and K content and the pH of the carbon in laboratory impregnations.

Iodine penetration tests were also performed on several carbons in an intense gamma radiation field ($>10^7$ rads/hr). Test data show that carbons intentionally exposed to high concentrations of DOP aerosol performed as well as unexposed carbons.

Studies of the rate of evaporation of elemental iodine from aqueous solutions indicated that significant quantities of I_2 might be expected to become airborne within a short period of time (5 hr) after release to open ponds. Addition of sodium thiosulfate to the solution substantially reduced the evaporative loss of iodine; however, the effects of high-intensity radiation fields on iodine-thiosulfate solutions remain to be evaluated.

Small HEPA filters containing filter media of the type used in the Savannah River confinement system were exposed to reactor building air and a high-intensity radiation field. Following this exposure, they were tested for flow performance under simulated accident conditions. Radiation exposure slightly impaired the performance of new filters and improved the performance of service-aged filters. Service aging effects on filter performance were far more significant than radiation effects.

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INTRODUCTION

The airborne activity confinement systems for the Savannah River production reactors are designed to collect halogens and particulates that might be released in the highly unlikely event of a reactor accident.[1] A continuing program is in progress at the Savannah River Laboratory to evaluate the performance of the confinement system (each reactor has an identical system) for removing airborne radioactivity under accident conditions and to develop techniques to enhance the reliability and efficiency of the system.

Previous studies at Savannah River showed that the unimpregnated Type 416 carbon (product of Barnebey-Cheney Company, Columbus, Ohio) used for many years in the system retained iodine less effectively than many types of impregnated carbons when exposed to an intense radiation field such as that which could result from the accumulation of radioiodine following a major reactor accident. A series of screening tests were conducted to aid in the selection of a suitable replacement carbon; the tests showed that coconut-shell carbons impregnated with triethylenediamine (TEDA) or a combination of TEDA and KI were the most effective adsorbers for iodine in the Savannah River confinement systems.[2,4]

Subsequent testing at Savannah River[5] confirmed the earlier observations of Adams, et al.[6] that in carbons with high TEDA content ignition temperatures are reduced significantly as the result of the fairly low (∿190°C) flash point of TEDA. Savannah River studies showed, however, that carbons with reduced TEDA content and satisfactory ignition temperature and iodine retention properties could be manufactured.[7] A 1% TEDA - 2% KI product (product of North American Carbon Company, Columbus, Ohio) is currently being installed in the Savannah River confinement system.

With the selection of a suitable replacement carbon for the confinement system,[7] research efforts were directed toward explaining the anomalous thermal desorption behavior of iodine from various commercial iodized carbons. In the earlier screening tests,[2,3] similar iodized carbons (nominally 5% KI-I_2 on

1500-m^2/g coconut carbon) exhibited highly variable results when tested for thermal desorption at 180°C (penetration values ranged from 0.023% to 12.8% in a 4-hr test). The extremes in penetration values were obtained from two different lots of carbon from the same vendor. Investigation revealed that the principal factors in retention behavior were the iodine and potassium content of the carbon (especially the atom ratio I/K) and the pH of the carbon.[8]

Previous particulate filter studies have shown that HEPA filter media deteriorates when exposed to high-intensity radiation or normal service in the confinement system.[4] In preliminary tests, the relative decrease in two media properties, water repellency and tensile strength, was measured as functions of radiation exposure time and service age. Loss of water repellency and tensile strength in irradiated media is caused by radiolytic decomposition of binder and waterproofing agents. Loss of water repellency in media exposed to normal service is caused by accumulation of hygroscopic atmospheric pollutants (soot and dust) on the media.

In this report, test filters constructed from several different types of media and sealants were tested under simulated accident conditions to determine HEPA filter degradation following radiation and service exposure. Potential effects of degraded filter performance on overall confinement system performance were evaluated to provide bases for optimum filter operation and specifications for purchase of replacement filters.

CARBON TESTING

Radiolytic Desorption Tests

DOP-Loaded Carbons

At the request of the ERDA Division of Waste Management and Transportation, several samples of carbon both exposed and unexposed to dioctylphthalate (DOP) aerosol were tested for iodine penetration in the radiolytic desorption facility. The purpose of the tests was to determine the effect of DOP aerosol penetration of HEPA filters during in-place efficiency tests on iodine retention efficiency of carbon beds downstream of the HEPA filter banks.

Two types of commercial nuclear-grade coconut carbons were exposed to a flowing air stream containing DOP aerosol at the Naval Weapons Laboratory in Dahlgren, Virginia (2 hr exposure of a 2-in.-dia. by 1-in.-deep bed to a DOP concentration of 100 μg/l at a face velocity of 40 ft/min). Measurements of the DOP concentration downstream of the carbon beds indicated a maximum carbon loading of 0.5 mg DOP / g carbon. After DOP exposure, the carbon samples and unexposed blank samples were tested for iodine retention efficiency.

Two series of samples were prepared and tested. In the first batch of samples, the iodine penetration of carbons exposed to DOP was significantly lower than that of the unexposed control samples.

Subsequent retesting of another sample of the control carbons showed that the unexposed controls were contaminated by an unidentified organic compound which caused the unusually high penetration. Purging the carbon with air during the DOP exposure period apparently desorbed the organic contaminant, thus improving the carbon performance in spite of the presence of DOP in the purge gas.

A second series of samples was then prepared using two new lots of carbon which were pretested before DOP exposure. Results of tests of the second sample series (Table 1) indicated that DOP aerosol has no effect on iodine retention efficiency.

TABLE 1

Iodine Penetration of Second DOP Sample Series[a]

DOP Exposure	*Base Carbon, %*	*TEDA-KI Carbon, %*
None (control)	0.358	0.020
0.5 mg/g[b]	0.342	0.030
None (other carbons range[c])	0.338 to 0.372	0.018 to 0.034

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[a] 1 hr loading time followed by 4 hr desorption time at 80°C and 95% relative humidity in a radiation field of $\sim 2.7 \times 10^7$ rad/hr (absorbed dose rate in carbon). Total I_2 loading of ~ 0.7 mg/g.

[b] See text for exposure conditions.

[c] Range of values obtained during other SRL test of different lots of the same carbon types which were not exposed to DOP aerosol.

The DOP concentration to which the test carbons were exposed is estimated to be at least 1000 times higher than that which would be encountered in normal operations (the required 99.9$^+$% HEPA filter efficiency would reduce the aerosol concentration by at least 1000-fold). Thus, this is a conservative test of the effect of DOP aerosol on new carbons. The long-term effects of DOP vapor on carbon (from slow volatilization of DOP trapped on upstream HEPA filters) and the effects of DOP aerosol on service-aged carbons were not tested.

Two important conclusions can be drawn from this experiment. First, DOP aerosol has no apparent effect on the iodine retention efficiency of carbon (the iodine penetration values obtained from DOP-exposed carbons were well within the range of values obtained from unexposed carbons). Second, other organic compounds can at least temporarily affect iodine retention. Thus, precautions should be taken to minimize the amount of organic vapors in the air being filtered by the carbon beds. In some cases, this may mean minimizing the use of organic solvents in the process areas of the reactor building (in Savannah River reactors, all of the reactor building air is exhausted through the carbon beds in the confinement system). Future investigations should include identification of specific organic compounds which affect iodine retention efficiency and exclusion of these compounds (if possible) from reactor building use.

Service-Aged Carbons

Service aging studies are being conducted on carbon exposed to reactor building air in full-sized beds in the confinement system and on small samples installed in the carbon test facility.[3,7] Carbons which have been service aged in the confinement system include Types 416* (unimpregnated), G-618** (5% TEDA impregnated), G-615** (2% TEDA-2% KI impregnated), and GX-176** (1% TEDA, 2% KI impregnated). All full sized beds of Types G-618 and G-615 were removed from the confinement system after only 9 months service aging because of the risk of TEDA ignition.[5] Lower TEDA-content Type GX-176 carbon was installed in the confinement system in April 1974. Changeover from Type 416 to Type GX-176 carbon was complete in 8 of the 15 compartments by the end of 1974. One bed of Type GX-176 was removed from the confinement system after 6 months service and tested for iodine retention.

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* Product of Barnebey-Cheney Company, Columbus, Ohio.

** Products of North American Carbon Company, Columbus, Ohio.

Smaller samples of 5 different types of carbon were installed in the carbon test facility (CTF) when it began operation in February 1974. Eight samples of each type of carbon (40 total samples, each containing ∿250 g of carbon) are being exposed to reactor building air in the CTF. Ten of the 40 sample positions are being exposed at superficial face velocities approximately 1.25 times the normal velocity (69 vs 55 ft/min) to obtain accelerated aging data. The remaining 30 sample positions receive air at the normal face velocity. The 5 carbon types (all 10 x 16-mesh coconut carbons) are: 1) an unimpregnated carbon (a blank to be compared with aging data obtained from Type 416 carbon), 2) a carbon impregnated with 1% TEDA and 2% KI (Type GX-176), 3) a carbon containing 0.5% TEDA and 2% KI, 4) a carbon containing 2% TEDA and 2% KI (similar to Type G-615 except for the particle size distribution), and 5) a carbon impregnated with ∿5% $KI-I_2$ (similar to the commercial iodized charcoals except for the particle size distribution). The 10x16 particle size distribution was chosen as a result of earlier tests which showed better iodine retention (in 1-in.-deep beds) for this finer mesh carbon.[7] Samples of each of the carbon types (both normal and accelerated flow) were removed after 6 months exposure.

Preliminary test data show that Type GX-176 carbon in full-size beds performs very much like Type G-615. The carbons exposed in the carbon test facility deteriorate more rapidly than compartment-aged carbons (Table 2). The reasons for the higher penetration observed in the CTF samples is currently under investigation.

TABLE 2

Radiation Tests of Service-Aged Carbons

Carbon Type	*Where Aged*[a]	*Service Age, months*	*Iodine Penetration, %*[b]
G-615	-	0	0.034
G-615	C-2	6	0.052
G-615	C-2	9	0.120
GX-176	-	0	0.022
GX-176	K-2	6	0.053
GX-176	CTF	6	0.129
GX-176	CTF	∿7.5[c]	0.249

---

a. C-2, Compartment 2 of C Reactor; and K-2, Compartment 2 of K Reactor.

b. See Table 1 for test conditions.

c. 6 months at ∿1.25 times normal flow.

Preliminary investigation indicates either that the linear face velocity in the carbon test facility is higher than in the confinement system or that the atmospheric pollutant concentration is higher at the CTF inlet plenum position (or a combination of both conditions). Earlier studies[2,9] showed that reactor room air (containing NO_2, the presumed atmospheric pollutant causing the most rapid poisoning of the confinement carbon) is not well-mixed with other building air in the confinement system supply plenum. Thus, air entering Compartments 2 and 3 contains more NO_2 than air exhausted through Compartments 4-6. The inlet plenum of the carbon test facility is in an unused blank position closer to the reactor room exhaust plenum than Compartments 2 and 3. The relatively low flow of the CTF compared to the confinement compartments (approximately 400 cfm compared to approximately 20,000 cfm for each of the confinement compartments) would cause any NO_2-free air drawn from the opposite end of the plenum to be exhausted through confinement compartments in Positions 2 and 3, and the CTF would receive air only from the reactor room end of the plenum.

Additional samples of service-aged carbons from the CTF and confinement system are scheduled for removal and testing at 6-month intervals. Data from these samples will be summarized in future progress reports.

Other Service Aging Tests

TEDA Analysis

Studies are also under way to determine the effect of service aging on the TEDA content of impregnated carbons. Because of the finite vapor pressure of TEDA (∿2 mm Hg at 40°C), some loss is anticipated as a result of air flowing through the carbon beds. Qualitative tests (consisting only of detecting the characteristic TEDA odor during ignition temperature determinations) indicate that the amine is still present on the three TEDA carbons exposed in the CTF for 6 months and on Type GX-176 exposed in the confinement system. Quantitative analytical methods are being developed to determine the TEDA content of new and used carbons.

Carbon Regeneration

Carbon ignition temperature was used as a measure of the effectiveness of several regeneration methods for reclaiming used coconut carbon. In these tests, a sample of unimpregnated Type 416 carbon service-aged for 43 months in the K-2 Compartment was first tested. Aliquots of the sample were then subjected to drying (50°C), heating (150°C), neutralization (KOH added to

∿pH 7), water leaching (to ∿pH 7), and finally to partial reactivation (∿600°C in an oxygen deficient environment). None of the treatments restored the used carbon to its original ignition temperature except the partial reactivation of the sample. Data are shown in Table 3.

TABLE 3

Effect of Regeneration Method on Used Carbon Ignition Temperatures

Treatment	*pH*[a]	*Ignition Temperature,* °*C*[b]
None[c]	3.51	280
92-hr Wash[d]	4.03	275
Dried at 125°C	2.80	275
Dried at 125°C, Neutralized, and Washed[d]	7.0	265
Dried at 200°C	3.21	285
Dired at 200°C, Neutralized, and Washed[d]	7.80	285
Partially Reactivated at 600°C	8.94	435
New Carbon (Blank)	9.50	395

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[a] pH of water extract of carbon.

[b] 55 ft/min face velocity of 5-°C/min heating rate in standard quartz apparatus.

[c] Barnebey-Cheney 416 carbon service-aged 43 months in Savannah River confinement system.

[d] Extracted with distilled water in a soxhlet extractor.

The data indicate that no simple physical or chemical treatment of used carbons can be used to reconstitute the carbon, and that full reactivation would probably be required to restore all the desirable properties.

During the determination of the ignition temperatures of the used carbon samples, a strong acidic odor resembling a mixture of short-chain organic acids and aldehydes was noted. The odor and low carbon pH indicate that the aging process probably includes partial oxidation of the carbon surface (plus accumulation of airborne acidic pollutants). Failure of the neutralization and/or leaching techniques to remove the odor-causing components indicates that they are bound to the carbon surface. Absence of the odor was noted only after heating the sample to >500°C. This again indicates that reactivation is required to restore a used carbon to a usable condition.

Water Leaching Tests

Proposals have been made by commercial power reactor vendors to provide a water spray system to cool iodine-loaded carbon beds and prevent self ignition from a combination of low air flow and radioiodine decay. In an effort to evaluate the effect of such a spray on the iodine retention properties of the carbon and to confirm basic iodine reaction mechanisms[10] (see also reaction mechanism discussion in a later section of this report), two commercial iodized coconut carbons were first loaded with ^{131}I-tagged elemental iodine and then flooded with distilled water. The flooding was carried out in two steps: first with one bed volume of water, then with an additional 9 bed volumes of water (total water = 10 bed volumes). Water collected from each step of the flooding operation (carried out without air flow) was passed through an ion exchange column, and the fraction of radioiodine removed was determined by gamma scintillation counting. In both tests, approximately 50% of the radioiodine was removed by one bed volume of water, and an additional 30 to 35% was removed by nine bed volumes of water; a total of 80 to 85% of the iodine was removed by leaching with ten bed volumes of distilled water.

Spraying the carbon beds before ignition should prevent a fire in the off-gas cleanup system by cooling the bed and by removing the heat source (radioactive iodine). Spraying the bed will, however, destroy the iodine retention capability of the carbon bed (especially for organic iodides since the nonradioactive iodine impregnant is water soluble). Moisture entrained in the flowing airstream downstream of the carbon bed would also be heavily loaded with radioiodine and a secondary cleanup system would be required before the air could be released to the atmosphere.

A bypass air stream which can provide enough flow to cool the bed would seem a more practical design. The latter method would also minimize the need for a supplementary hold tank (to store the radioiodine-loaded wash water) and a supplementary carbon filter system to remove the airborne radioiodine released as a result of operation of the fire suppression system.

The leaching tests also show that a significant portion of I_2 loaded on the carbon was converted to I^- (or another water soluble species) and indicates chemical reaction rather than physical adsorption as a basic reaction mechanism in gas phase iodine retention by carbon.

High-Temperature Desorption Tests

The high-temperature desorption test was initially designed as a screening test to aid in selecting potential replacement carbons for the Savannah River confinement system.[2] The test conditions (10-min. loading of elemental iodine at ambient temperature and humidity followed by 4-hr desorption at 180°C and <1% relative humidity) were selected to prevent damage to the *Teflon*-coated test apparatus and *Neoprene* "O"-rings used as seals in the apparatus (registered tradenames of E. I. du Pont de Nemours and Co.) The 180°C temperature is also near the upper limit of usefulness for nuclear carbons because thermal desorption of iodine becomes significant in this temperature range. HEPA filter blinding data presented earlier[9] show that this elevated temperature could be attained in a hypothetical accident. Moreover, because of the sensitivity of some commercial carbons to high temperature, the high-temperature desorption test has been proposed for inclusion in the ASTM Standards for carbon testing.

In the original screening test series,[3] 13 of the 21 candidate adsorbers were subjected to the high-temperature test and had penetration values ranging from 0.003% (5% TEDA on coconut carbon) to 12.8% (KI-I_2 on coconut carbon).[3,4] The data fall into three broad penetration categories:

1. Very low (<0.010%); coconut-base carbons either unimpregnated or impregnated with TEDA or TEDA and KI.

2. Intermediate (0.01 to 0.10%); coconut-base carbons impregnated with I_2, KI-I_2, or PbI_2.

3. Very high (>1.0%); coconut-base carbons impregnated with KI-I_2, and petroleum-base carbons impregnated with KI or TEDA.

The anomaly in the data is that four of the products originally tested were nominally identical ($\sim$5% KI-I_2 on 1500 m^2/g coconut carbon), yet the high-temperature penetration values ranged from 0.028% to 12.8%. In subsequent testing of a different manufacturing lot of the carbon showing 12.8% penetration, the penetration was only 0.023%. Preliminary investigation of the cause of the variability indicated that different methods of impregnation may have been used by the four vendors;[3] however, confirmation of the techniques could not be obtained because of the proprietary nature of the processes.

Because the two principal methods which could be employed* could result in different degrees of alteration of the chemical content of the carbon, samples of the five carbons were analyzed for K, Na, and I content by neutron activation analysis and for residual alkalinity by measuring the pH of a water extract of the carbons. The results of these analyses indicated a strong correlation between the atom ratio of iodine to potassium (I/K ratio) and the high-temperature iodine penetration test results. The sodium content of the carbons was consistently <0.2% (by weight) and was not related to penetration.

Chemical Analysis of Carbons

Results of neutron activation analyses of several commercial I_2-impregnated carbons are shown in Table 4 along with the pH and iodine penetration data. The comparison between I/K ratios and high-temperature penetration data for coconut carbons is presented graphically in Figure 1.

TABLE 4

Chemical Analysis of Commercial Iodized Coconut Carbons

Carbon Vendor	*Potassium, wt %*	*Iodine, wt %*	*Atom Ratio I/K*	*pH*[a]	*Iodine Penetration, %*[b]
A[c]	0.958	4.01	1.29	7.05	12.82
B	1.14	4.42	1.19	9.56	2.41
C	1.75	3.78	0.664	9.36	0.052
D-1[d]	1.55	3.01	0.600	9.62	0.028
A[c]	2.23	4.23	0.586	9.02	0.023
D-2	1.04	1.31	0.387	9.86	0.006
D-3	1.26	1.30	0.318	10.06	0.003

a. 5 g carbon in 20 ml distilled water. pH of water measured after 20 min soaking.

b. High-temperature desorption, see text for test description.

c. One of two manufacturing lots of the same type carbons.

d. 1, 2, and 3 represent 3 different carbon types from the same vendor.

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* The "dip" method could possibly result in a water-leached carbon; and the "spray" method could result in an unleached carbon, if the liquid addition is controlled below the saturation level of the carbon.

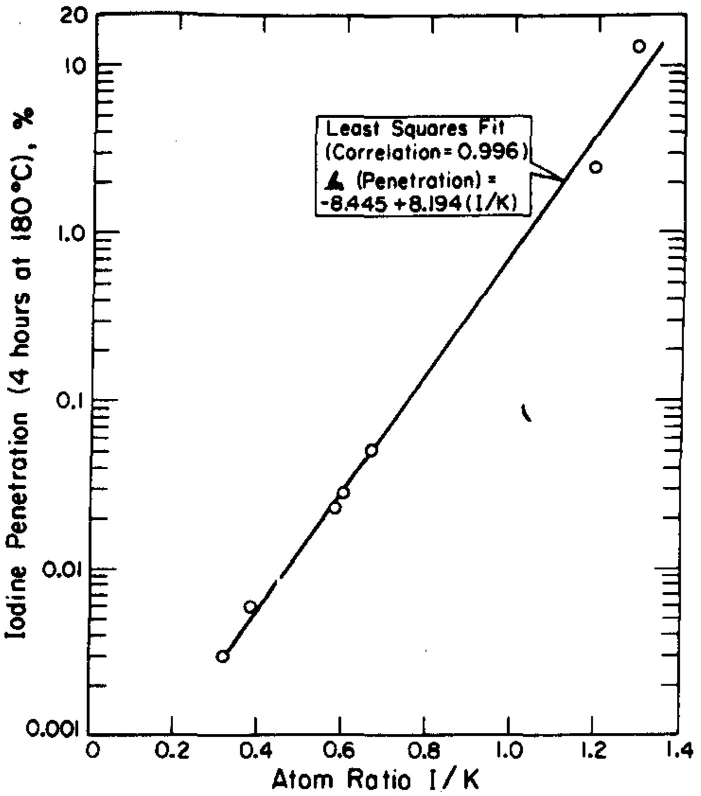

FIGURE 1. Iodine Penetration as a Function of I/K Ratio for Commercial I_2-Impregnated Carbons with No Potassium

To rule out the possibility of variations in the natural K^+ content of the base carbon (three of the four vendors use the same base carbon), representative samples of base carbon from lots manufactured over a 5-year period were also analyzed for K^+ and Na^+ content. The data show very little change from lot to lot as indicated in Table 5.

The data suggest a basic reaction mechanism for high-efficiency elemental iodine retention by activated carbon as shown in idealized form in Equation 1.

$$3I_2 + 6OH^- \rightarrow 5I^- + IO_{3-} + 3H_2O \qquad (1)$$

The potassium salts of iodine have high inherent stability, and the combination of excess K^+ and an alkaline environment best promote stabilized ionic iodine formation. Thus, coconut carbon, because of its high natural basicity and high natural K^+ content, is the best natural iodine adsorber.

TABLE 5

Chemical Content of 1500-m^2/g Coconut Carbons

Manufacture Date	*pH*[a]	*K^+, wt %*	*Na^+, wt %*
3/69	9.9	1.18	0.11
6/70	10.0	1.17	0.07
2/71	9.9	1.19	0.08
3/72	9.9	1.30	0.09
10/73	10.3	1.27	0.06

---

a. See text for test description.

Laboratory-Impregnated Carbons

In an effort to define specific reaction mechanisms, three series of experiments were undertaken: 1) vapor-phase I_2 impregnation of natural-K^+ carbon (constant K^+, varying I_2 concentration); 2) vapor-phase I_2 impregnation of a K^+-free carbon, followed by KOH impregnation (constant I_2, varying K^+ concentration); and 3) liquid KOH-I_2 impregnation of a K^+-free carbon.

To minimize the effect of a variable base carbon (base carbons have inherently different iodine capacities), a 5-gal drum of ∿1500-m^2/g coconut carbon was obtained from a carbon vendor. After preliminary analysis, a portion of the carbon was vapor-phase impregnated with I_2 at three different concentrations (nominally 2, 4, and 6 wt %). To test the Savannah River impregnation techniques, a commercial vendor also supplied samples of the same base carbon impregnated with the same concentrations of I_2.

Vapor-Phase I_2 Impregnation of Natural Potassium Carbons

Analyses of potassium and iodine contents of the I_2-impregnated, natural potassium carbons are shown in Table 6 with high-temperature iodine penetration data. The I/K ratio is also plotted in Figure 2; data from commercial carbons as a function of the logarithm of penetration are also included. A slightly better least squares correlation (0.963 vs. 0.952) was obtained when the function (I/K)/pH was plotted against the logarithm of penetration instead of I/K. This correlation indicates some dependence on pH (Figure 3).

TABLE 6

Chemical Analysis of I_2-Impregnated, Natural Potassium Carbons

Carbon[a]	*Potassium, wt %*	*Iodine, wt %*	*I/K*	*pH*	*(I/K)/ pH*	*Penetration, %*[b]
SRL-24V	1.57	2.59	0.507	10.28	0.049	0.018
SRL-25V	1.55	3.63	0.720	9.69	0.074	0.115
SRL-26V	1.40	5.45	1.203	9.50	0.127	0.339
SRL-24	1.64	1.53	0.287	9.68	0.030	0.015
SRL-25	1.52	2.67	0.541	9.30	0.058	0.057
SRL-26	1.44	5.52	1.181	8.05	0.147	0.755

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a. Arbitrary numbers: those followed by V's were vendor impregnated; the remainder were impregnated at Savannah River.

b. High-temperature desorption; see text for conditions.

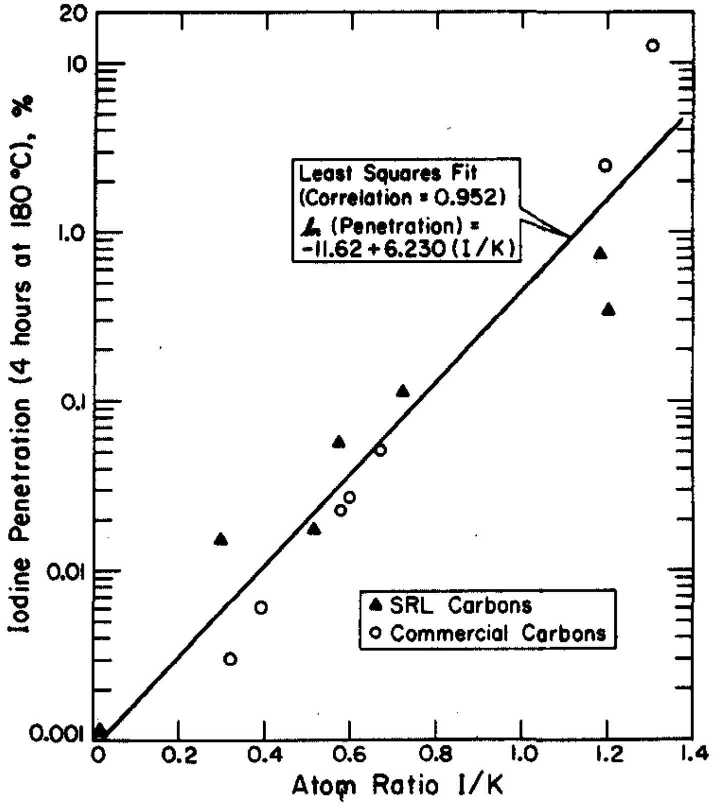

FIGURE 2. Iodine Penetration as a Function of I/K Ratio for I_2-Impregnated Carbons with Natural Potassium

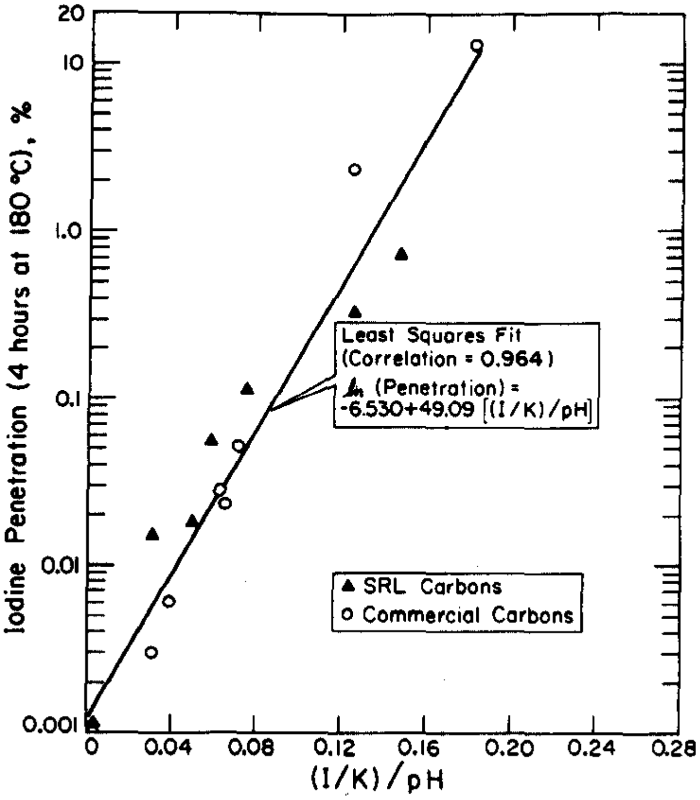

FIGURE 3. Iodine Penetration as a Function of (I/K)/pH for I_2-Impregnated Carbons with Natural Potassium

Vapor-Phase KOH-I_2 Impregnation of Potassium-Free Carbons

The second phase of the study involved removal of soluble potassium from the base carbon, vapor-phase impregnation with a constant I_2 content (∿4%), and addition of potassium (as KOH) at various concentrations. In these experiments, two methods of K^+ removal were used: 1) repeated distilled water washes (in a soxhlet extractor); and 2) dilute H_3PO_4 leaching followed by repeated distilled water washes. In all cases, the reconstituted potassium samples showed a marked deviation from a straight logarithmic increase in penetration with increasing I/K ratios [or (I/K)/pH ratios]. Samples washed with distilled water and impregnated with KOH showed consistently lower iodine penetration than commercial carbons or I_2-impregnated carbons at all (I/K)/pH ratios. The H_3PO_4-treated samples were about equivalent to natural K^+ samples at I/K ratios <1 [(I/K)/pH <0.1] and similar to water-washed samples at I/K ratios >1 [(I/K)/pH >0.1] (Figure 4). These experiments indicate that added potassium (as KOH) is more readily available for reaction than the natural potassium present in coconut carbon. The data also suggest a method for improving the high-temperature performance characteristics of noncoconut-base carbons.

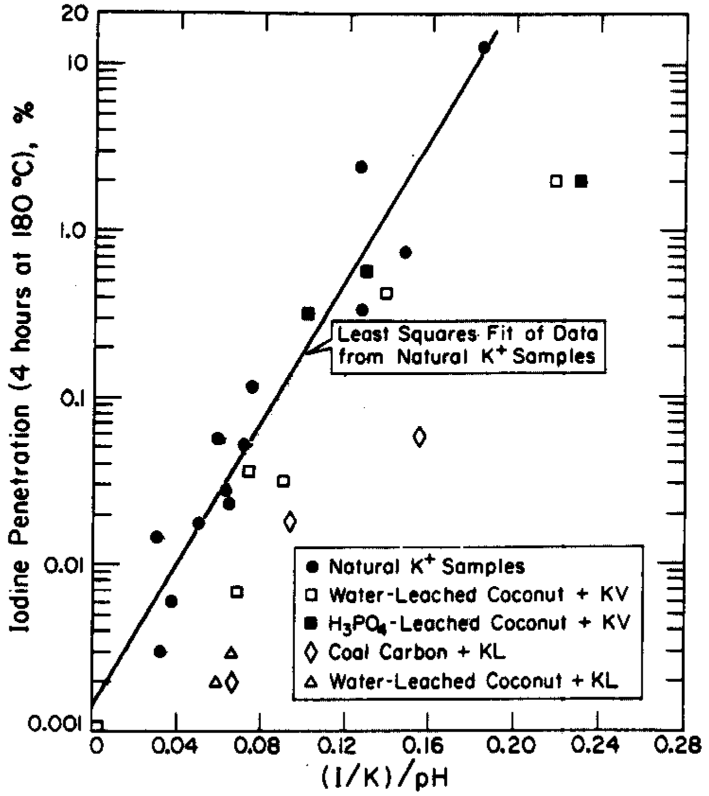

FIGURE 4. Iodine Penetration as a Function of (I/K)/pH for All Carbons

Liquid KOH-I_2 Impregnation of Potassium-Free Carbons

The third set of experiments involved co-impregnation of K^+-free carbons (coconut and noncoconut) with K^+ and I^- using a KOH-I_2 solution. Two samples were also prepared using NaOH rather than KOH to determine whether K^+ is the preferred impregnant. In the first experiments, several batches of water-washed coconut carbon were combined (the soxhlet extractor has about a 200-g capacity for coconut carbon) and were blended, and the blend was divided into two portions. The first portion was vapor-phase impregnated with I_2 (∿3.0%). One half of the iodine-impregnated carbon was then impregnated with ∿1.5% K^+ (as KOH) and designated KV for KOH + I_2 vapor. The other half was impregnated with ∿1.5% Na^+ (as NaOH) and designated NaV for NaOH + I_2 vapor. The second portion of the original blended sample was also divided into two samples. The first was impregnated with a solution of KOH-I_2 and designated KL for KOH + I_2 liquid. The second was impregnated with a solution of NaOH-I_2 and designated NaL for NaOH + I_2 liquid. The iodine/alkali metal ratio was chosen to approximately duplicate the (I/K)/pH ratio of one of the better commercial I_2-impregnated carbons.

The (I/A)/pH (where A = alkali metal) ratios are compared with high-temperature iodine penetration values in Table 7. The commercial carbon is also shown for comparison. The data show that KOH-I_2 liquid impregnation produces a superior product (sample KL) when evaluated by the high-temperature desorption test.

To investigate the applicability of the impregnation technique to other carbon types, a sample of a commercial coal-base carbon was obtained. This carbon had been screened to an 8x16 particle size distribution (the same particle size distribution as the coconut-base carbon). Both vapor-phase iodine impregnation followed by KOH impregnation and KOH-I_2 coimpregnation were used. The test data show that noncoconut carbons can be made into effective iodine adsorbers. Analytical and test data are shown in Table 8; coconut carbon sample KL is shown for comparison.

TABLE 7

Iodine Penetrations of Selected Coconut Carbons

Carbon	*(I/K)/pH*	*(I/Na)/pH*	*Iodine Penetration, %*[a]
KV[b]	0.069	--	0.007
NaV[b]	--	0.040	0.014
KL[b]	0.059	--	0.002
NaL[b]	--	0.052	0.041
Commercial	0.062	--	0.028

---

[a] High-temperature desorption; see text for conditions.

[b] See text for sample description.

TABLE 8

KOH-Impregnated Coal Carbons

Sample	*(I/K)/pH*	*Iodine Penetration, %*[a]
Coal KV[b]	0.093	0.018
Coal KL-1[c]	0.066	0.003
Coal KL-2[d]	0.066	0.002
Coconut KL[e]	0.059	0.002

---

[a] High-temperature desorption test; see text for conditions.

[b] Vapor-phase iodine (∿4%) + ∿1.2% K (as KOH).

[c] $KOH-I_2$ coimpregnation (2.8% I_2, 1.2% K by weight).

[d] $KOH-I_2$ coimpregnation (4.4% I_2, 1.9% K by weight).

[e] $KOH-I_2$ coimpregnation (2.2% I_2, 1.1% K by weight).

Other Tests of KOH-Impregnated Carbons

Impregnation of activated carbon with potassium salts lowers the ignition temperature of the carbon.[11,12] Thus, use of the $KOH-I_2$ impregnation technique involves balancing the iodine retention characteristics against the ignition characteristics of the carbon. Other factors to consider are the availability and cost of the carbon and the physical properties of the carbon that may limit its usefulness for air cleaning applications (specifically the hardness and resistance to attrition by a flowing air stream). Coconut carbon has long been used for all cleaning applications because its physical properties and iodine retention properties were satisfactory at a reasonable cost. Increasing worldwide demand for coconut carbon has caused significant price increases and accentuates the need to find alternative carbons suitable for nuclear applications.

Use of the $KOH-I_2$ impregnation technique may provide a means of improving the iodine retention properties of alternative carbons to the extent that they perform as well as or better than coconut carbon. To this end, the coal-base carbons listed in Table 8 were tested for ignition temperature, and the better sample (Coal KL-1) was tested for iodine retention in an intense radiation field (the radiolytic desorption test).[3] Data are shown in Table 9 along with comparable data for one of the better commercial coconut carbons.

TABLE 9

Comparison of Coconut and Coal Carbons

Sample	*(I/K)/pH*	*Ignition Temp., °C*[a]	*Thermal Iodine Desorption, %*[b]	*Radiolytic Iodine Desorption, %*[c]
Coal (as received)	0.006	475	0.006	∿0.3
Coal KL-1	0.066	330	0.003	0.020
Coal KL-2	0.066	310	0.002	--
Commercial Coconut Carbon	0.062	340^{+}	0.028	0.016

---

a. As measured in a standard quartz apparatus at a heating rate of 5°C/min and a superficial dry air velocity of 17 m/min (at 200°C).

b. Elemental iodine desorption in 4 hr at 180°C and <1% relative humidity.

c. Elemental iodine desorption in 5 hr at 180°C and ∿95% relative humidity and an adsorbed dose rate of 3×10^7 rad/hr.

Selection of Impregnation Salts

Having demonstrated that coal carbons can be custom impregnated to retain iodine as effectively as coconut carbons, a systematic study was initiated to find the best combination of K^+, I^-, and pH which could be used on any carbon. Solutions made with $KOH-I_2$, KI-KOH, and $KI-K_2CO_3$ were used on two different coal-base carbons and a petroleum-base carbon to optimize iodine retention. A flame retardant ($NaH_2PO_4 \cdot H_2O$) was also incorporated into some of the formulations to obtain satisfactory ignition characteristics. Various additive combinations are discussed below.

$KOH-I_2$

The most consistent results were obtained with the $KOH-I_2$ combination. A KOH solution is prepared into which I_2 crystals are dissolved. Once the I_2 has reacted (forming a colorless solution containing a complex mixture of I^-, OI^- and probably some higher oxidation states iodine salts as well), the solution is diluted with distilled water to a volume slightly less than that required to saturate the carbon being impregnated. The solution is slowly dripped onto the predried carbon in a mixing chamber (a modified stainless steel beaker with internal vanes rotated on a laboratory ball mill). The addition system is flushed with enough distilled water to rinse out the residual salts and to just saturate the carbon. The wet carbon is dried overnight in a laboratory convection oven, cooled in a dessicator, and stored in a sealed plastic bottle.

As shown by the systematic variation of I/K ratios ranging from 0.3 to 1.2 [the function (I/K)/pH ranges from 0.032 to 0.120], coal-base carbons performed consistently better than iodized coconut carbons. Comparison of the plot (Figure 5) of the function [ln (penetration) vs (I/K)/pH] indicates that the coal carbons show the same dependency on I, K, and pH as do coconut carbons (essentially the same slope), but that the penetration for the coal carbons is consistently lower by about a factor of 10. As indicated earlier, however, the ignition temperatures of these KOH-impregnated samples were consistently lower than 340°C (Table 10).

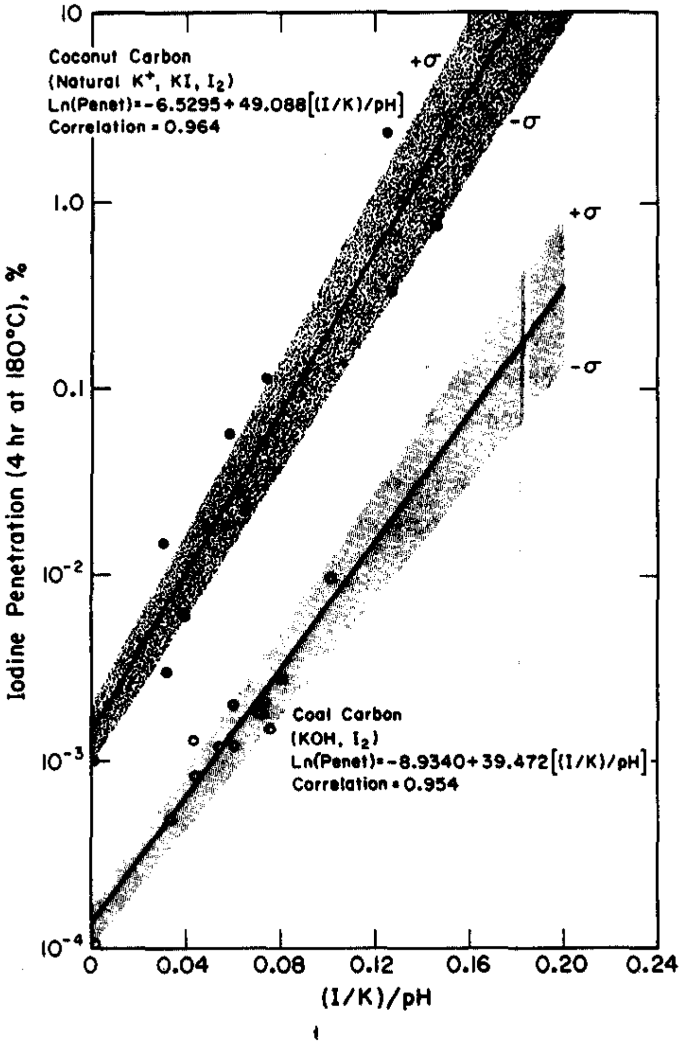

FIGURE 5. Iodine Penetration as a Function of (I/K)/pH for KOH-I_2-Impregnated Carbons

TABLE 10

Experimental Data for KOH-I_2-Impregnated Coal Carbons[a]

Impregnant Content, wt %[b] *I*	*K*	*(I/K)/pH*	*Iodine Penetration, %*[c]	*Ignition Temperature, °C*[d]
2.33	2.06	0.0332	0.00049	295
2.35	1.64	0.0429	0.0013	300
2.19	1.50	0.0431	0.00084	300
2.73	1.51	0.0537	0.0012	310
2.31	1.16	0.0586	0.0020	315
2.44	1.20	0.0603	0.0012	300
3.29	1.44	0.0697	0.0018	320
3.17	1.32	0.0720	0.0018	300
2.36	1.02	0.0728	0.0021	315
4.30	1.76	0.0759	0.0015	310
2.73	1.10	0.0790	0.0028	315
2.09	0.67	0.101	0.0097	330

---

[a] Bituminous coal-base carbon, Vendor N, ground to 8 × 16 mesh.

[b] Net impregnants added as determined by neutron activation analysis.

[c] 180° desorption test, see text for details.

[d] In standard quartz apparatus at 55 ft/min face velocity and 5°C heating rate.

Similar low penetrations were obtained from a different coal-base carbon and a petroleum-base carbon impregnated with KOH-I_2. Ignition temperatures and penetration data on these carbons are shown in Table 11.

KOH-KI

As indicated in the previous section, I_2 readily dissolves in a KOH solution forming several iodine salts so that no I_2 is actually present in the impregnation solution. The logical extension of the KOH-I_2 technique is to use iodine in the salt to avoid handling I_2. To this end, several samples were prepared with KI and KOH. As expected, the penetration data were satisfactory, but carbon ignition temperature was again lower than desired. Data are summarized in Table 12.

TABLE 11

Experimental Data for Other KOH-I_2-Impregnated Carbons

Base Carbon	*Impregnant Content, wt %*[c] *I*	*K*	*(I/K)/pH*	*Iodine Penetration, %*[c]	*Ignition Temperature, °C*[c]
Coal[a]	2.26	1.19	0.0589	0.0025	310
Coal[a]	3.20	1.48	0.0654	0.0016	320
Petroleum[b]	2.26	0.98	0.0704	0.0014	325
Petroleum[b]	3.06	1.38	0.0662	0.00066	320

---

[a] Bituminous coal-base carbon, Vendor P, sieved to 8 × 16 mesh.

[b] Petroleum-base carbon, Vendor W, 8 × 16 mesh.

[c] See Table 10 for details.

TABLE 12

Experimental Data for KOH-KI-Impregnated Carbons

Base Carbon	*Impregnant Content, wt %*[d] *I*	*K*	*(I/K)/pH*	*Iodine Penetration, %*[d]	*Ignition Temperature, °C*[d]
N[a]	2.07	1.14	0.0549	0.0017	330
N[a]	2.35	1.10	0.0668	0.0028	325
N[a]	1.82	1.18	0.0454	0.0013	295
P[b]	2.93	1.39	0.0645	0.0015	330
W[c]	2.27	1.24	0.0563	0.00066	330
W[c]	2.10	1.17	0.0538	0.0025	325

---

[a] Coal carbon, Vendor N, ground to 8 × 16 mesh.

[b] Coal carbon, Vendor P, sieved to 8 × 16 mesh.

[c] Petroleum carbon, Vendor W, 8 × 16 mesh.

[d] See Table 10 for details.

K_2CO_3-KI

As further proof that the necessary components of a successful impregnation formulation are K, I, and a pH control agent, the carbonate salt of potassium was substituted for the hydroxide salt for pH control. Without a fire retardant present, the ignition temperatures of the products were low, even though the iodine penetration values were acceptable as shown in Table 13.

TABLE 13

Experimental Data for K_2CO_3-KI-Impregnated Carbons

Base Carbon	*Impregnant Content, wt %*[d]		*(I/K)/pH*	*Iodine Penetration, %*[d]	*Ignition Temperature, °C*[d]
	I	*K*			
N[a]	2.51	1.28	0.0585	0.0010	310
P[b]	1.91	1.15	0.0513	0.0007	315
P[b]	2.51	2.07	0.0370	0.0016	285
W[c]	1.86	1.02	0.0543	0.0055	305

---

a. Coal carbon, Vendor N, ground to 8 × 16 mesh.

b. Coal carbon, Vendor P, sieved to 8 × 16 mesh.

c. Petroleum carbon, Vendor W, 8 × 16 mesh.

d. See Table 10 for details.

Flame Retardants

Three classes of compounds are commonly used as flame retardants for combustible materials: 1) halide salts, 2) borate salts, and 3) phosphate salts. Because the carbon samples are already impregnated with an iodide (KI), other halide salts were not added because F^- and Cl^- are more active chemically and would tend to release I_2 in any competing chemical reaction. Of the two remaining types of compounds, the phosphates are generally better buffering agents and have greater solubility. Buffering is important in maintaining a high pH, while solubility is important in preparing impregnation solutions. Two phosphate compounds were arbitrarily selected for initial testing, mono-basic potassium (KH_2PO_4) and mono-basic sodium (NaH_2PO_4) phosphates (or hydrated salts). Both were effective in raising the ignition temperatures of the impregnated carbons to acceptable levels (Table 14).

The data in Table 14 show that the iodine penetration values were slightly lower when the sodium salt ($NaH_2PO_4 \cdot H_2O$) is used as a flame retardant. This fact, coupled with the lower cost of the $NaH_2PO_4 \cdot H_2O$ (as compared to KH_2PO_4), makes the sodium salt the preferred flame retardant. Similar experiments using the dibasic sodium salt $Na_2HPO_4 \cdot 2H_2O$) and the dibasic potassium salt K_2HPO_4 are planned.

TABLE 14

Experimental Data for Carbons Containing Phosphate Flame Retardants

Base Carbon	*Impregnant Content, wt %*[a]			*(I/K)/pH*	*Iodine Penetration, %*[a]	*Ignition Temperature, °C*[a]
	I	*K*	*PO_4^{-3}*			
KOH-KI Carbons						
N[b]	1.87	1.06	Na[e]	0.0539	0.0024	380
N[b]	2.55	1.44	K[f]	0.554	0.0039	375
N[b]	1.88	1.33	Na[e]	0.0413	0.00096	365
P[c]	2.20	1.23	Na[e]	0.0555	0.0016	360
P[c]	2.41	1.47	K[f]	0.0370	0.0016	365
W[d]	2.07	1.08	Na[e]	0.0570	0.00053	400
W[d]	2.08	1.31	K[f]	0.0475	0.00046	395
K_2CO_3-KI Carbons						
N[b]	2.64	1.17	Na[e]	0.0706	0.0012	365
P[c]	2.65	1.31	Na[e]	0.0632	0.0032	365
W[d]	1.86	0.93	Na[e]	0.603	0.0031	375
W[d]	1.95	1.04	Na[e]	0.0592	0.0021	360

---

[a] See Table 10 for details.

[b] Coal carbon, Vendor N, ground to 8 × 16 mesh.

[c] Coal carbon, Vendor P, sieved to 8 × 16 mesh.

[d] Petroleum carbon, Vendor W, 8 × 16 mesh.

[e] $NaH_2PO_4 \cdot H_2O$ flame retardant added, 1 wt %.

[f] KH_2PO_4 flame retardant added, 1 wt %.

Other Additives

The tertiary amine additives triethylenediamine (TEDA) and triethanolamine (TEA) have also been used in a limited number of impregnations. Preliminary data (Table 15) indicate that 1% TEDA does not adversely affect the ignition temperature or high-temperature iodine penetration when a flame retardant is also present. Data on TEA-impregnated carbons are insufficient to draw conclusions (single tests of a single TEA-impregnated sample).

TABLE 15

Experimental Data for Iodized, Amine-Impregnated Petroleum Carbon[a]

Impregnant Content, wt %					*Iodine Penetration, %*[e]	*Ignition Temperature, °C*[e]
Amine	PO_4^{-3}	*I*[e]	*K*[e]	*(I/K)/pH*		
TEDA[b]	Na[d]	2.77	1.30	0.0645	0.0015	360
TEDA[b]	Na[d]	2.58	1.26	0.0636	0.0018	375
TEA[c]	Na[d]	2.67	1.44	0.0560	0.0052	370

---

a. Petroleum carbon, Vendor W, 8 × 16 mesh.

b. Triethylenediamine, 1 wt %.

c. Triethanolamine, 1 wt %.

d. $NaH_2PO_4 \cdot H_2O$, 1 wt %.

e. See Table 10 for details.

Test Program

In addition to the service aging test on confinement carbons discussed earlier in this report, radiolytic desorption tests are scheduled on the more promising of the laboratory-impregnated carbons. Arrangements are being made with a carbon vendor to impregnate larger samples of noncoconut carbons so that these samples may also be included in service aging studies in the carbon test facility. Methyl iodide penetration tests on these samples are also being arranged through a cooperative program with the Naval Research Laboratory in Washington, DC.

IODINE EVAPORATION

In a reactor accident involving fuel melting, fission products could accompany emergency coolant runoff and result in significant off-site doses. Laboratory-scale investigations have been undertaken to study the evaporation of iodine from water (a major contribution to such doses) and to develop methods to retard its release. The experimental configuration consists of a dynamic flowing air measurement which approximates the expected conditions in the environment.

Experimental results were compared with theoretical predictions available in the literature. All experiments were performed in the absence of ionizing radiation. The effects of radiation will be determined in separate experiments to be performed during the next reporting period.

Iodine Evaporation Theory

Others[13] have shown that the general mass transfer equations of Sherwood and Pigford[14] can be used to yield an equation that gives the concentration (C) of aqueous iodine in solution as a function of time(t):

$$\frac{C}{C_o} = \exp\left(\frac{-0.664\ N_{Re}^{-1/2}\ G_L A t}{PV(\mu/\rho D)^{2/3}}\right) \qquad (1)$$

where the symbols have the following definitions and values in the present experiments:

C_o = Initial aqueous concentration of iodine (mol/ℓ)

N_{Re} = Reynolds number ≃7,900

G_L = Linear velocity of air ≡ 7,200 ft/hr (an estimate)

A = Area of liquid surface

V = Volume of liquid = 3 ℓ, (A/V = 2.90/ft)

μ = Viscosity of air = 4.356×10^{-2} lb/(ft-hr)

ρ = Density of air = 0.0808 lb/ft^3

D = Diffusivity of iodine in air = 0.3875 ft^2/hr (an estimate)

t = Time, (hr)

$$P = \text{Partition coefficient} = \frac{\text{Concentration of iodine in solution}}{\text{Concentration of iodine in air at liquid-air interface}}$$

Substitution yields:

$$C = C_o \exp\ (-2.085t/P) \qquad (2)$$

where t is in minutes.

The above equation is of the same form as the first-order reaction equation that applies for simple chemical reactions (such as evaporation), and which is usually written as:

$$C = C_o \exp\ (-kt) \qquad (3)$$

Therefore, $k = 2.085/P$ (min^{-1}), and the amount of aqueous iodine evaporated at time t is given by the following equation:

$$\text{Amount of Iodine Evaporated} = (C_o - C)V = (C_o - C_o e^{-kt})V \qquad (4)$$

$$= C_o V(1-e^{-kt}) = C_o V\left[1-e^{\left(\frac{-2.085t}{P}\right)}\right] \qquad (4a)$$

Theoretical values of the partition coefficient have been estimated by Eggleton[15] for aqueous iodine solutions. Eggleton has shown that the solution of iodine in water results in the formation of ionic iodine via three principal reactions (see Equations 2-4 of the Appendix) and that at the same time iodine is lost from solution by vaporization. Only iodine in the elemental form is volatile. Eggleton treats these four competing reactions mathematically to obtain the equilibrium partition coefficient. The resulting expression* gives the partition coefficient as a function of aqueous iodide concentration, pH, and the equilibrium constants of the four competing reactions. A graph of partition coefficient vs aqueous iodide concentration is given in Figure 6 for several values of pH.

Partition coefficients of 100-125 are predicted from Figure 6 for iodine concentrations of 2×10^{-5} to 6×10^{-5} mol/ℓ; for pH's of the iodine-distilled water system of 6.2 to 6.5, and for pH's of the iodine-river water system of 6.8 to 7.2. Thus, substituting

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* A rederivation of the equation was made to correct an error that occurs in his paper.

in Equation 4a yields:

$$\text{Amount of Iodine Evaporated} \simeq 1.2 \times 10^{-4}(1-e^{-0.018t}), \text{ (mol)} \qquad (4b)$$

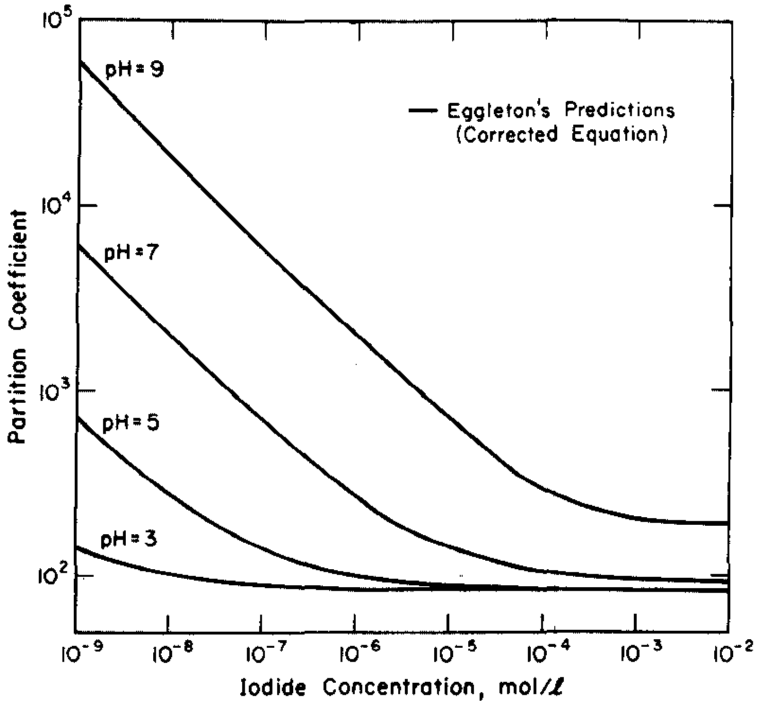

FIGURE 6. Partition Coefficient of Iodine - Water System as a Function of Iodine Concentration

Experimental Investigation

The experimental apparatus is shown schematically in Figure 7. The equipment employed consists of a 9-ℓ glass bottle that contains 3 ℓ of water and ∿3 to 30 mg of elemental iodine that is tagged with ∿2 mCi of ^{131}I. An air flow of 2 cfm is maintained through the bottle and is filtered at the input end by 6-in. of activated charcoal and a high-efficiency particulate air (HEPA) filter. The output from the bottle is passed through charcoal beds that adsorb iodine carried by the air stream. The ^{131}I activity of the carbon beds is determined with a scintillation counter (as a function of time) over the 5-hr test period. Before each test, the ^{131}I activity of both the charcoal beds and the water in the bottle is determined, and a material balance is made.

Data are reduced with a computer code modified to fit test conditions and equipment. Glass and *Teflon* are used to minimize surface adsorption of iodine.

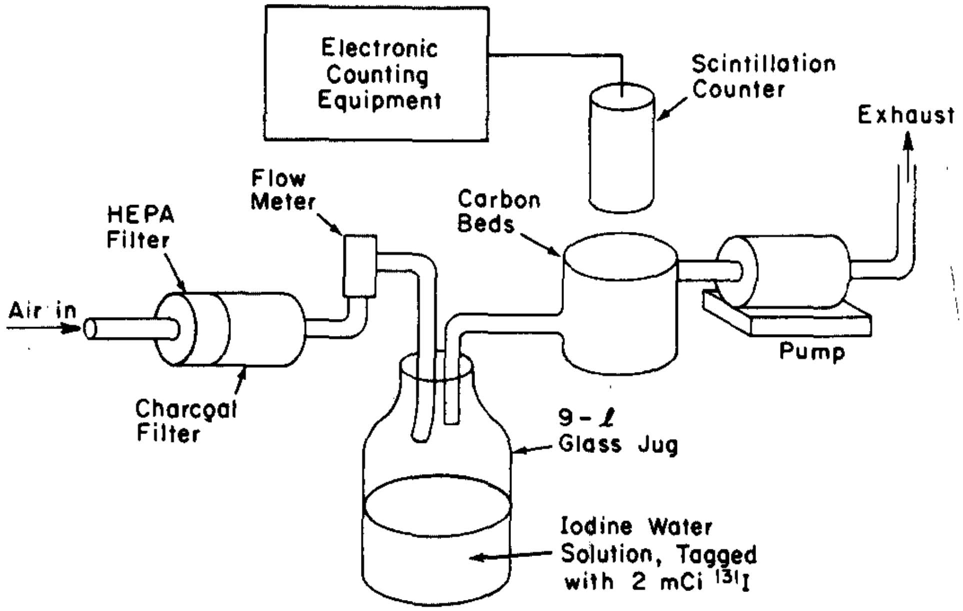

FIGURE 7. Iodine Evaporation Equipment

The iodine is tagged and put into solution at the beginning of each test by rinsing the iodine crystals through a porous glass frit and into the test bottle by means of a tube extending below the liquid surface. The initial sweep of air through the bottle is passed through a separate set of charcoal beds to remove any airborne iodine released directly into the air (during the loading), so that it will not be confused with evaporated iodine.

Experimental results for the evaporation of iodine from aqueous solutions of iodine in distilled water and river water, respectively, are shown in Figures 8 and 9 in terms of ^{131}I buildup on the carbon bed as a function of time. A nuclear poison *Polybor* (Registered tradename of U. S. Borax for $Na_2B_8O_{13} \cdot 4H_2O$) was also added (Figure 9) for one test. *Polybor* is not expected to react with iodine and hence should yield a result like any iodine-water test without additives, which it does.

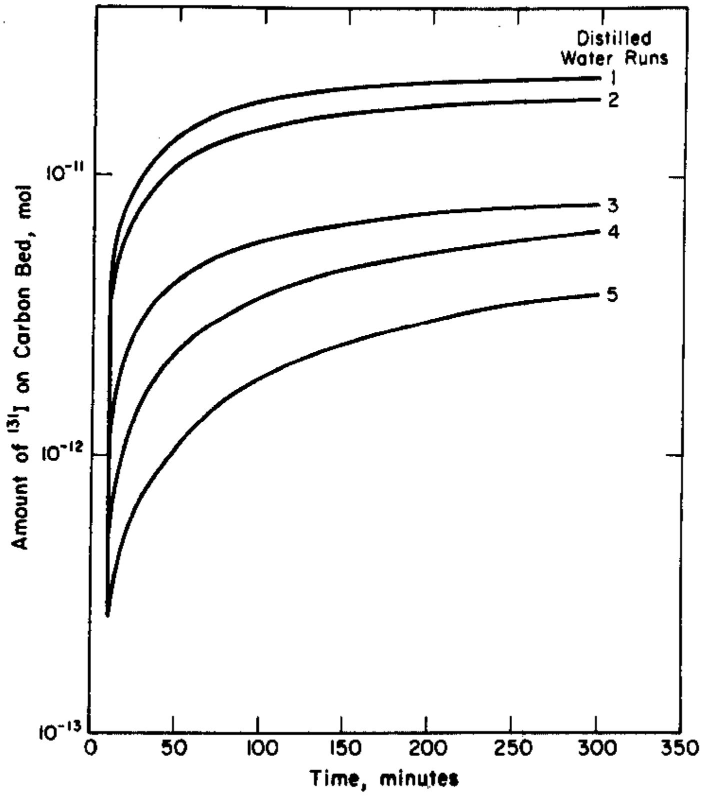

FIGURE 8. Amount of ^{131}I on Carbon Bed as a Function of Time in the Distilled Water Test

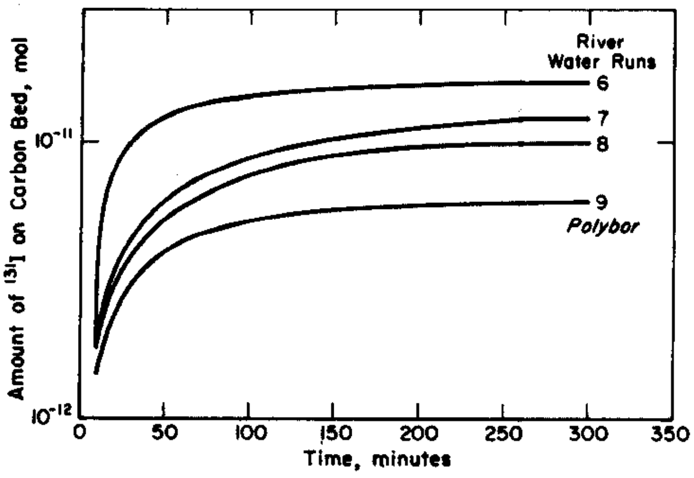

FIGURE 9. Amount of ^{131}I on Carbon Bed as a Function of Time in the River Water Test

Curve fitting was used to determine the approximate values of iodine evaporated, $C_0'V$ (for ^{131}I), and k (=2.085/P) for the tests given in Figures 8 and 9. In each case, although the total amount of ^{131}I in solution did not evaporate, $C_0'V$ for ^{131}I was a significant fraction of the original amount of ^{131}I in solution. The total iodine released as a function of initial concentration is shown in Table 16 assuming that the same fraction of inert (nonradioactive) iodine evaporated as did ^{131}I.

A comparison of Columns 4 and 5 of Table 16 with the theoretical prediction of Equation 4b shows: a) that the amount of iodine evaporated, $C_0'V$, is 16 to 82% of that expected, and b) that there is a considerable spread in individual k and P values. The lack of consistency between individual test results may indicate the presence of small quantities of interfering impurities in the water used. Other investigators[16] have also seen large variations in these quantities.

TABLE 16

Results of Iodine-Water Evaporation Tests

	C_0V *(initial), mol*	$C_0'V$ *(evaporated), mol*	*% Iodine Evaporated*	*k, min⁻¹*	*P, calc.*
Distilled Water					
1	8.10×10^{-5}	5.37×10^{-5}	66	0.0158	132
2	1.48×10^{-4}	8.73×10^{-5}	59	0.0167	125
3	9.42×10^{-5}	7.74×10^{-5}	82	0.0124	336[a]
4	1.40×10^{-5}	9.87×10^{-6}	70	0.0081	259
5	7.89×10^{-5}	4.20×10^{-5}	53	0.0049	286[a]
River Water					
6	1.98×10^{-4}	5.01×10^{-5}	25	0.0392	53
7	1.47×10^{-4}	3.51×10^{-5}	24	0.0220	95
8	1.12×10^{-4}	3.87×10^{-5}	34	0.0232	90
9	1.38×10^{-4}	2.28×10^{-5}	16	0.0234	89

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a. Because the solution depth was different in these two tests, P = 4.17/k for Test 3 and P = 1.39/k for Test 5.

To reduce iodine evaporation, two additives were examined which promote the formation of the ionic (nonvaporizable) form of iodine: ascorbic acid and sodium thiosulfate. In addition, the effect of a physical barrier (an oil film) between the solution and the air was examined.

The amount of ascorbic acid and thiosulfate added was such as to give a 10:1 mole ratio of additive to total iodine in solution. In the oil film test, 1/16-in.-thick layer of vacuum pump oil was spread on the water surface, and the iodine was injected below the oil film. The results are given in Figures 10 and 11 in the same form as the previous tests. The solid curve for the thiosulfate test in Figure 10 is only an approximate fit to the data. Due to the very large reduction in evaporation, the ^{131}I activity on the carbon bed was very small, and the counting statistics were poor. The incremental rises in ^{131}I collected partially reflect the poor counting statistics. The curve as drawn applies to approximately five hours of data, and the final saturation value, while indeterminate, is greater than the curve would indicate.

A search of the literature fails to reveal a theoretical treatment of partition coefficient vs. iodine concentration for systems with ascorbic acid and thiosulfates as additives, so that k values calculated cannot be related to partition coefficients. The significance of Table 17, however, is that both ascorbic acid and thiosulfate significantly reduce the amount of iodine evaporated over the 5-hr test period.

Conclusions and Future Program

At 25°C, 16 to 34% of the total iodine in river water evaporated in the first 5 hr. Partition coefficients for the iodine river water system differ considerably from that predicted for initial iodine concentrations of 0.47×10^{-5} to 6.6×10^{-5} mol/ℓ. Additions of ascorbic acid or thiosulfate, in the ratio of 10 mol additive to 1 mol iodine, reduce iodine evaporation by 0.012% and 0.00066%, respectively, during the first five hours of evaporation. An oil film is ineffective in retarding iodine evaporation although it slows the rate of evaporation initially.

The potentialities of thiosulfate warrant further evaluation, and an evaluation of the radiation stability of the iodine-water-thiosulfate system is planned in the ^{60}Co irradiation facility.

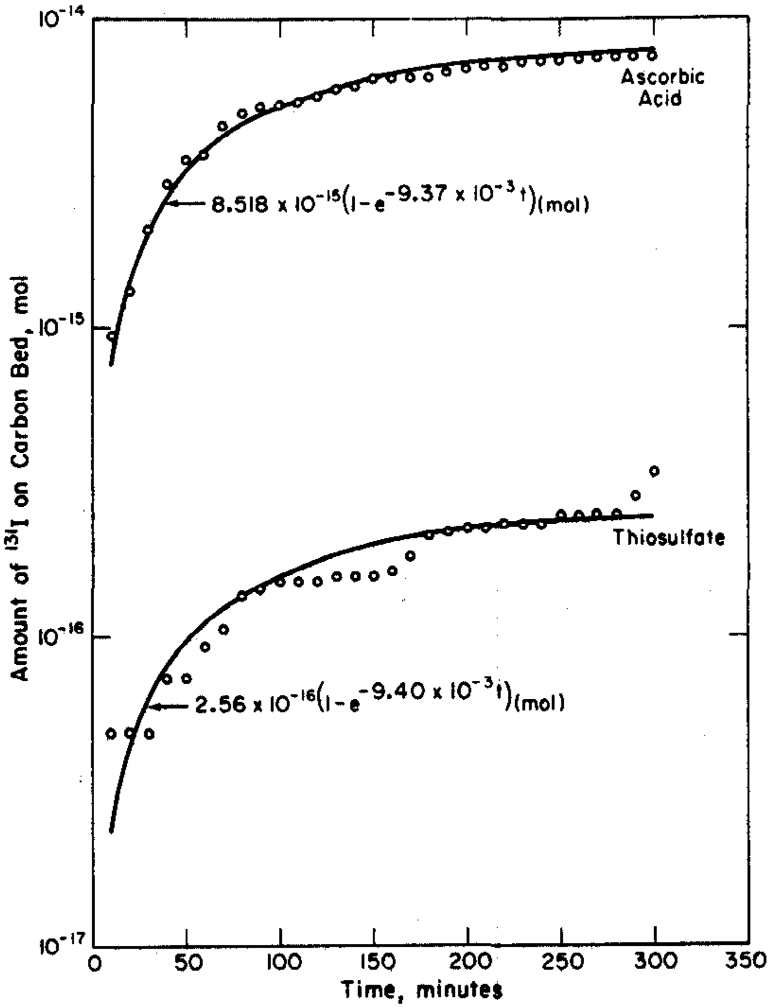

FIGURE 10. Amount of ^{131}I on Carbon Bed as a Function of Time for the Additive Test

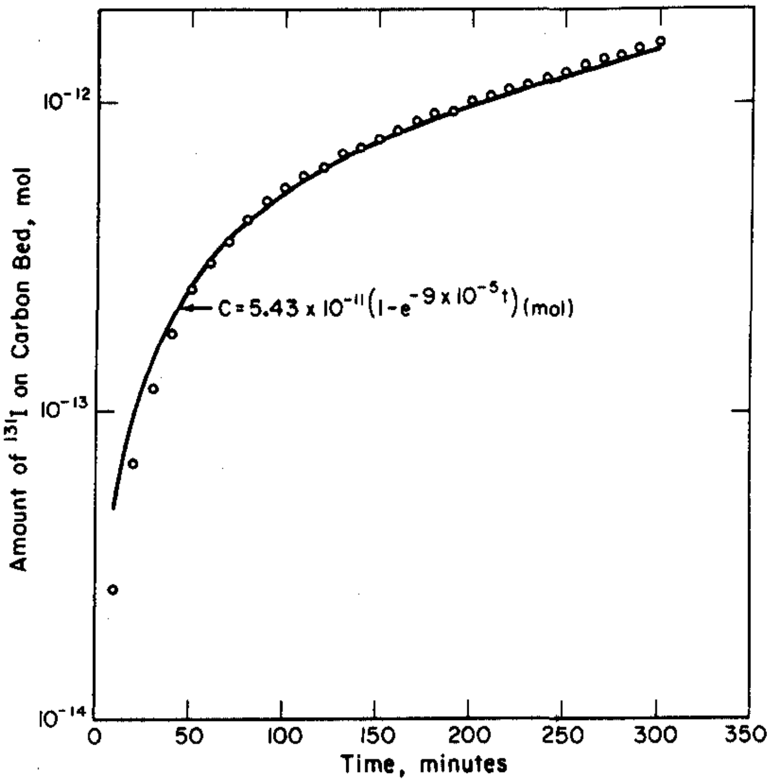

FIGURE 11. Amount of ^{131}I on Carbon Bed as a Function of Time for the Oil Film Test

TABLE 17

Results of Additive Tests

Additive	C_oV *(at start), mol*	$C_o'V$ *(evaporated), mol*	*% Iodine Evaporated*	*k, min*$^{-1}$
Ascorbic acid	2.65×10^{-4}	3.15×10^{-8}	0.012	0.00937
Sodium thiosulfate	1.64×10^{-4}	1.08×10^{-9}	0.00066	0.00940
Pump oil	2.33×10^{-4}	2.04×10^{-4}[a]	∿88%[a]	0.00009

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[a] Actually 2.8% had evaporated at the end of 5 hr; however, extrapolation of the equation on Figure 11 to saturation (t ≃ 46 days) indicates that ultimately ∿88% will evaporate.

Tests similar to those described in this report will also be performed to evaluate the performance of potassium iodide as an additive. KI is of interest because it can form KI_3, which is relatively nonvolatile. Also, the possibility of isotopic exchange with the iodine of methyl iodide needs to be examined. The effectiveness of KI in this role will be examined through the use of methyl iodide tagged with ^{131}I.*

HEPA FILTER TESTING

Exposure of small test HEPA filters to simulated Savannah River reactor accident conditions were expanded to measure performance of filters made from different types of filter media and sealants following gamma irradiation and/or service exposure. The most-severe accident test sequence (discussed in previous reports as Test 1[7,8]) and a new test intended to rupture the filters were used to characterize filter performance. Test conditions, test filters, and filter pretreatment are reviewed in the following sections. Test facilities were described in previous reports.[7,8,11]

Test Conditions

Accident Simulation Test

Temporary pressurization of the reactor room by rapid release of steam from an inadequately cooled reactor was calculated to produce a flow surge through the filters that would peak at a maximum of ∿8.5 times normal flow and would last about 25 sec. Following this flow surge, the flow would decrease to a value limited by the total pressure developed by the exhaust fans (∿6-in. H_2O) and the total flow resistance of the confinement system. Steam in the reactor room would be diluted with building supply air flow and would cause the temperature of the steam and air mixture flowing through the confinement filters to decrease from ∿90 to ∿30°C in about 5 hr. Test conditions simulating these conditions are shown in Table 18. Following the flow surge, the ΔP was limited to that which could be produced by the exhaust fans in the confinement system. Test filter ΔP and flow were monitored throughout every test to determine moisture pluggage as a function of test exposure time.

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* The release of fission-product iodine could be accompanied by some formation of methyl iodide as a result of the presence of organics in the coolant water. Methyl iodide is very volatile, and isotopic exchange may be a means for minimizing the release of iodine.

TABLE 18

HEPA Filter Accident Simulation Test Conditions

Time	*Multiple of Normal Flow Rate*	*Temperature of Steam-Air Mixture, °C*	*HEPA Filter ΔP, in. H_2O*
0 to 25 sec	8.5	92	No Limit
25 sec to 4 min	<1.0	92	<5
4 min to 14 min	<1.0	80	<5
14 min to 20 min	<1.0	70	<5
20 min to 30 min	<1.0	60	<5
30 min to 40 min	<1.0	50	<5
40 min to 2 hr	<1.0	40	<5
2 hr to 3 hr	<1.0	35	<5
3 hr to 5 hr	<1.0	30	<5

Filter Rupture Test

Although calculations indicate the most severe steam-air flow surge through the confinement filters would not last longer than ∿25 sec, a more severe test, intended to rupture the test filter, was developed to characterize more completely the different filters being tested. This rupture test exposes the filter to a steam-air mixture at 80°C and three times the normal flow rate until gross rupture occurs. A steam-air mixture temperature of 80°C was chosen because the greatest liquid water entrainment occurs when steam is mixed with air at 25°C to give a mixture temperature of 80°C. Maximizing this liquid entrainment produces the most severe condition for the test filter.

Test Filters

Smaller filters with internal construction identical to full-size filters (filter media, separator arrangement, and spacing) were fabricated by manufacturers of full-size HEPA filters using filter media designated Type F and Type E in Reference 17. These test filters (10 to 15 ft^3/min rated flow) are shown in Figure 12.

Because the internal spacing of media and separators is identical to that in a full-size filter, moisture pluggage and media rupture in these filters is considered representative of the performance of a full-size filter. However, performance of small separator-less filters or of adhesives in small test filters cannot be considered completely representative of a full-size filter. This is particularly true following radiation exposure because of geometric differences between full-size and small test filters, and the resulting stresses applied to the filter pack can be significantly different. Gross failure of a filter by blowing the filtration media completely out of the casing would probably occur at a lower ΔP for the full-size filter than for the smaller filter because the center of the full-size filter pack is not as strongly supported. In 1962, Savannah River tests of full-size filters with separators indicated the filter media would rupture, thereby reducing the filter ΔP, before the filter pack could be blown out of the casing.[18]

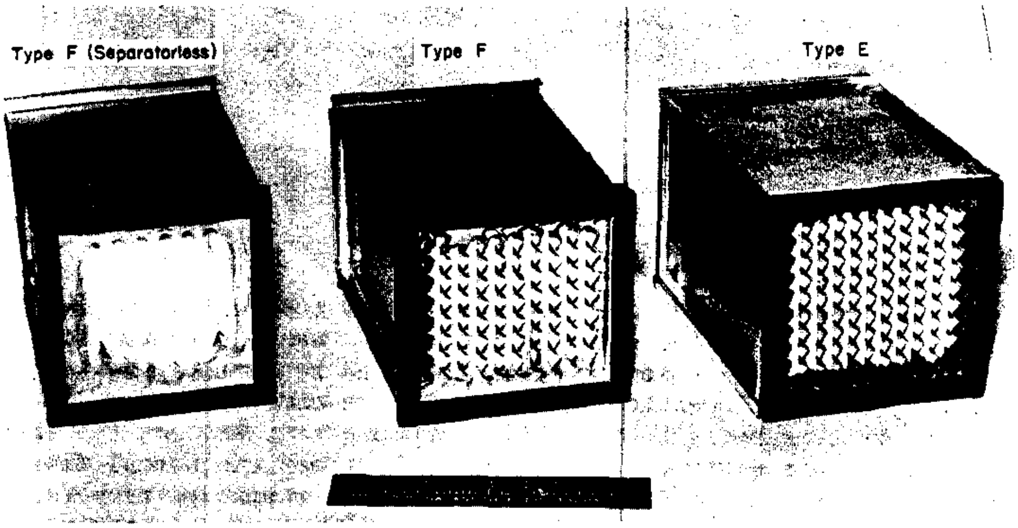


FIGURE 12. Test HEPA Filters

Type F Filters

Six groups of test filters were fabricated by combining either all-glass fiber or glass-asbestos fiber filter media (Media F in Reference 17) and a foam or a rubber-base adhesive (Adhesives A and B, respectively, in Reference 17) into filters with and without aluminum separators. For purposes of identification, the following abbreviations have been combined to designate each filter group:

G : all-glass media
GA: glass-asbestos media
S : aluminum separators
NS: no separators
RB: rubber-base adhesive
FA: foam adhesive

For example, G-NS-RB identifies a filter with all-glass media, no separators, and a rubber-base adhesive. All Type F filters have chromized steel casings.

Type E Filters

Test filters were also fabricated from materials used in Savannah River confinement filters by the manufacturer of the full-size filters. The filter media (Type E in Reference 17) is a glass-asbestos fiber media enclosed in cadmium-plated steel frames with aluminum separators.

Type E Filters with Service Exposure

Type F and Type E test filters are currently being exposed to confinement system air flows before performance testing. Four of these filters have been removed after 4- and 6-months service exposure. To indicate the expected performance of service-exposed filters, test filters were fabricated in the laboratory from media removed from full-size filters following service in the confinement system. Type E filter casings were used for the laboratory fabricated filters following rupture testing with the original filter pack.

Facilities

The ^{60}Co irradiation facility[17] was used for irradiation of test HEPA filters and other materials at a dose rate of $\sim 3.5 \times 10^7$ rad/hr. Dry air flowed through each test filter during irradiation to maintain an air temperature of $\sim 60°C$ and to remove gasses from radiolysis of air and decomposition of materials.

The carbon test facility was used to measure the effects of service exposure *in situ* and is located adjacent to the confinement filter compartments in one reactor area. Air flow upstream of the confinement filters is drawn through a full-size moisture separator, through 36 HEPA test filters, and finally through 40 test carbon beds. Test HEPA filters exposed in this facility were used to confirm results of tests with laboratory-fabricated, service-exposed filters.

Results

Filter Rupture Test

Type F Filters. Exposure of Type F filters to $\sim$3 times the normal flow of steam and air at 80°C caused moisture pluggage, which is shown as increasing filter ΔP with increased exposure time in Figure 13. Filters exposed to 2×10^8 rads in the ^{60}Co facility before testing became plugged and ruptured in from 100 to 250 minutes. Unirradiated filters plugged so slowly that the tests were terminated after 120 min. because rupture in a reasonable period of time appeared unlikely.

Exposure to flowing steam-air mixtures continued until a rupture in the filter was visible, even though in one test the filter ΔP stabilized indicating a possible rupture before visible rupture. Rupture ΔP varied widely. It depended partially on whether the filter media ruptured initially at a fold around the end of a separator, or between pleats in a separator. Partial collapse of folds occurred in some irradiated filters without separators. Several typical ruptured filters are shown in Figure 14.

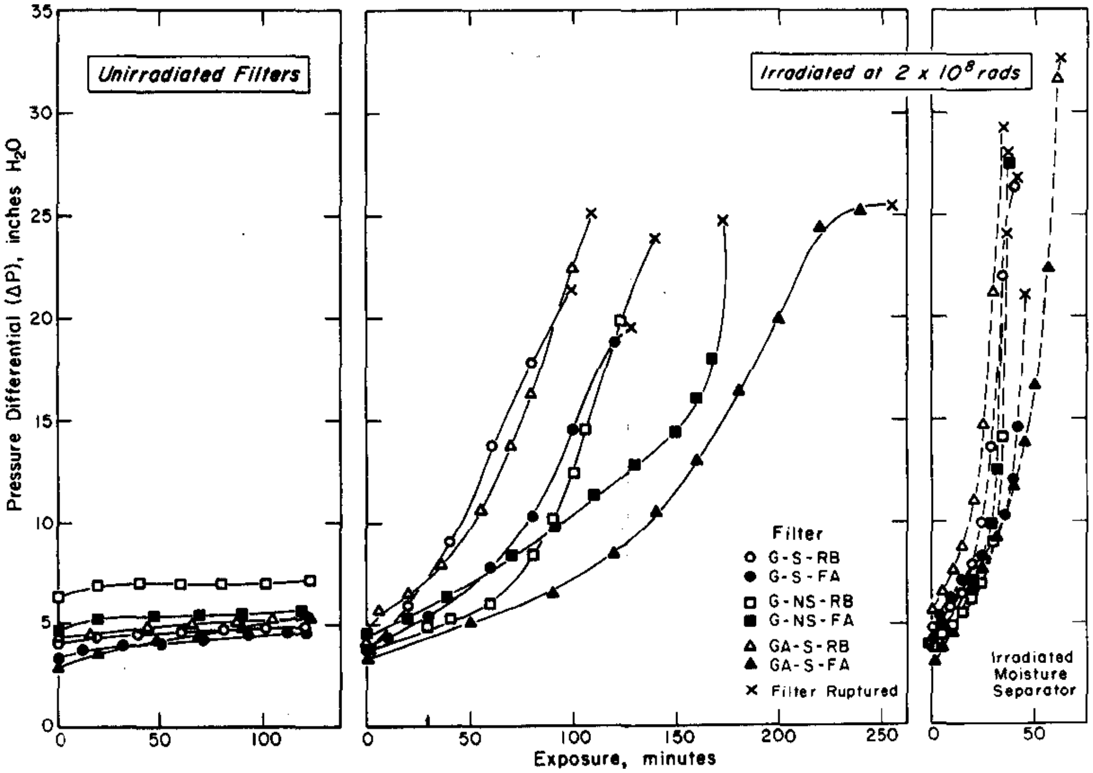
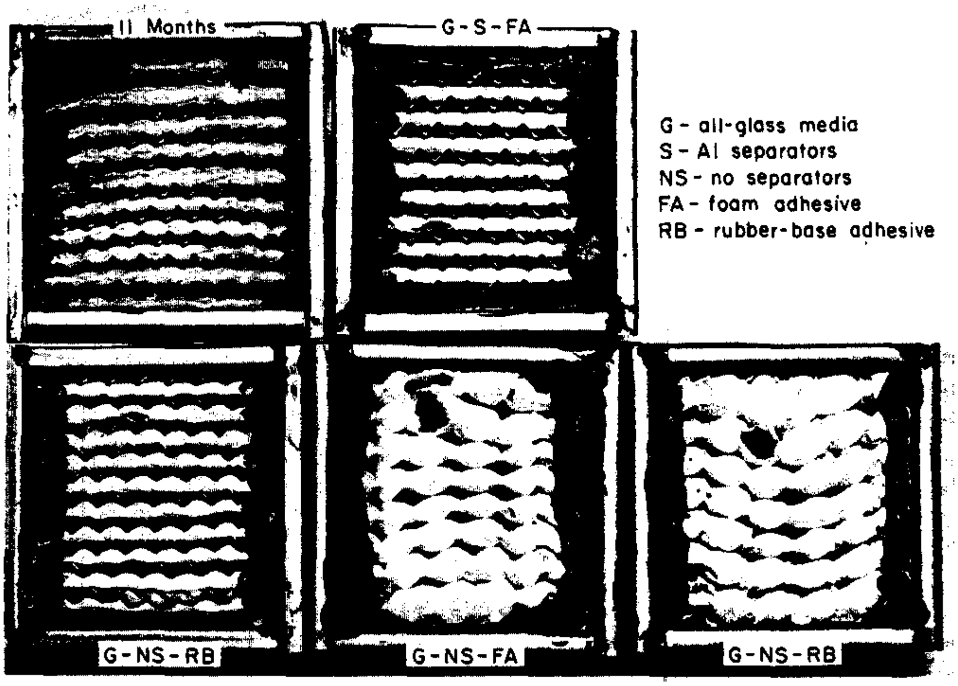
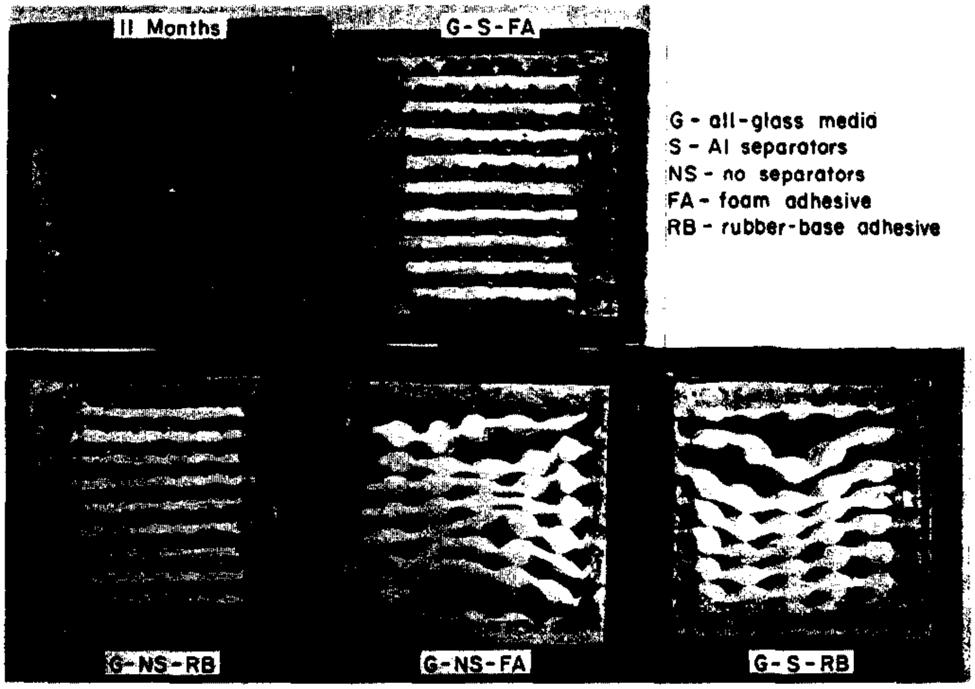


FIGURE 13. Rupture of Type F HEPA Filters



a. Downstream Face



b. Upstream Face

FIGURE 14. Ruptured Type F HEPA Filters

Replacement of test facility moisture separator units with units that had been exposed to ∿8 × 10^8 rads in the ^{60}Co facility caused more-rapid filter pluggage and rupture. The *Teflon* fibers in the irradiated moisture separators were washed completely from the stainless steel wire mesh in the first two minutes of steam-air exposure, allowing entrained moisture to pass through the separator and collect on the test filter.

Filters constructed with the foam adhesive appeared to plug more slowly than filters with the rubber-base adhesive. Such behavior would not be expected unless flow bypassed the filter media by leaking through the foam adhesive. During tests, however, no leakage through either adhesive was observed. Post-test examination revealed the rubber-base adhesive curled away from the metal filter casing after irradiation and exposure to steam-air flow, and, infrequently, loss of bonding between the foam adhesive and the metal casing was observed.

Type E Filters. Several Type E filters were exposed to rupture test conditions following irradiation to 1 × 10^8, 2 × 10^8, and 6 × 10^8 rads.

Comparison with a new, unirradiated Type E filter is shown in Figure 15. After 120 min. of exposure, rupture of the unirradiated filter appeared unlikely, while pluggage and rupture of irradiated filters occurred more rapidly with increased radiation exposure. When an irradiated (8 × 10^8 rads) moisture separator was substituted in the test facility, moisture pluggage and rupture was more rapid.

Although filters with the greater radiation exposure ruptured more rapidly, they ruptured at higher pressure differentials, apparently contradicting previous data which showed decreased wet strength of filter media with increased radiation exposure.[17] This behavior could result from less penetration of moisture into the media during the shorter exposure time before rupture or from random differences in individual filter construction.

The filter adhesive of a ruptured Type E filter curled away from the casing following irradiation and steam-air exposure; this behavior was similar to that of the rubber-base adhesive on Type F filters.

Type E Filters with Service Exposure. Test filters fabricated with media from full-size filters with 4-, 7-, 11-, and 13-months service in the confinement system were exposed to rupture test conditions. Filters with 4- and 7-months service plugged with moisture but did not rupture in 400 min of exposure, but filters with greater service ruptured rapidly. When an irradiated (8 × 10^8 rads) moisture separator was substituted, pluggage and rupture of downstream filters was even more rapid (Figure 16). Filters with 7- and 11-months service were exposed to 2×10^8 rads, and then were exposed to rupture test conditions with a new, unirradiated moisture separator in the test facility. Both filters plugged more slowly initially than unirradiated 7- and 11-months service filters, but they did rupture.

Moisture pluggage of filter media having confinement system service was more rapid because the hygroscopicity of the soot layer collected on the media during continuous air flow. This soot layer absorbed moisture from flowing steam-air mixtures much more readily than did clean, nonhygroscopic, glass-fiber filter media, even when the moisture repellency of the clean filter media was completely destroyed by radiation exposure. The presence of irradiated moisture separators aggravated the problem. The reduced removal of entrained moisture by the irradiated moisture separator exposed the hygroscopic soot on the filters to even higher moisture levels. However, the data (Figure 16) indicate irradiation reduces the hygroscopicity of the service-accumulated soot layer and also reduces the residual moisture repellency of the filter media.

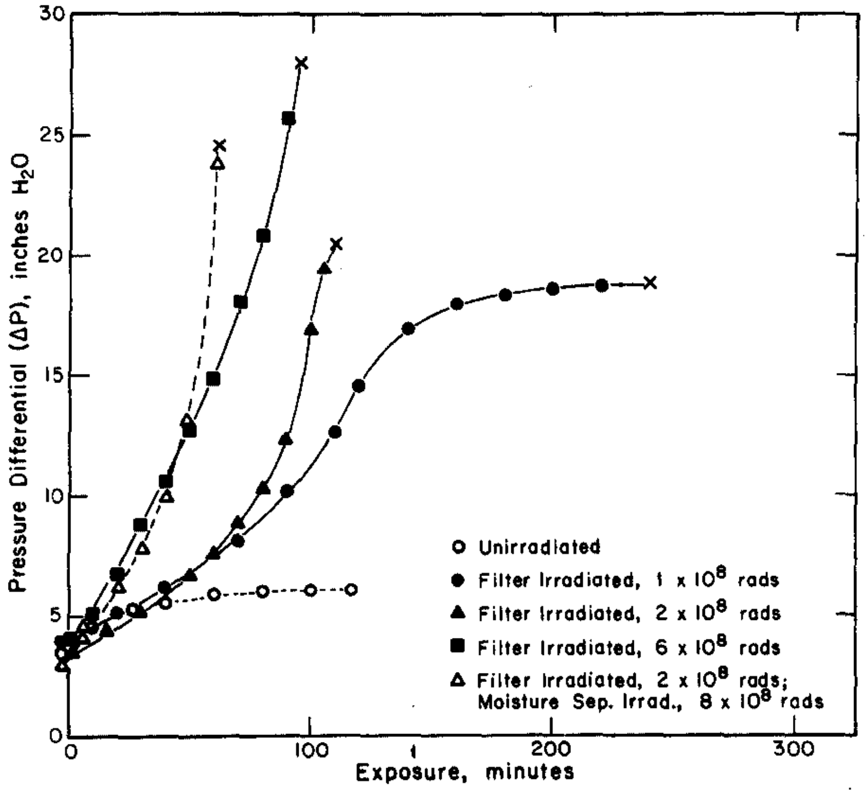


FIGURE 15. Rupture of Type E HEPA Filters

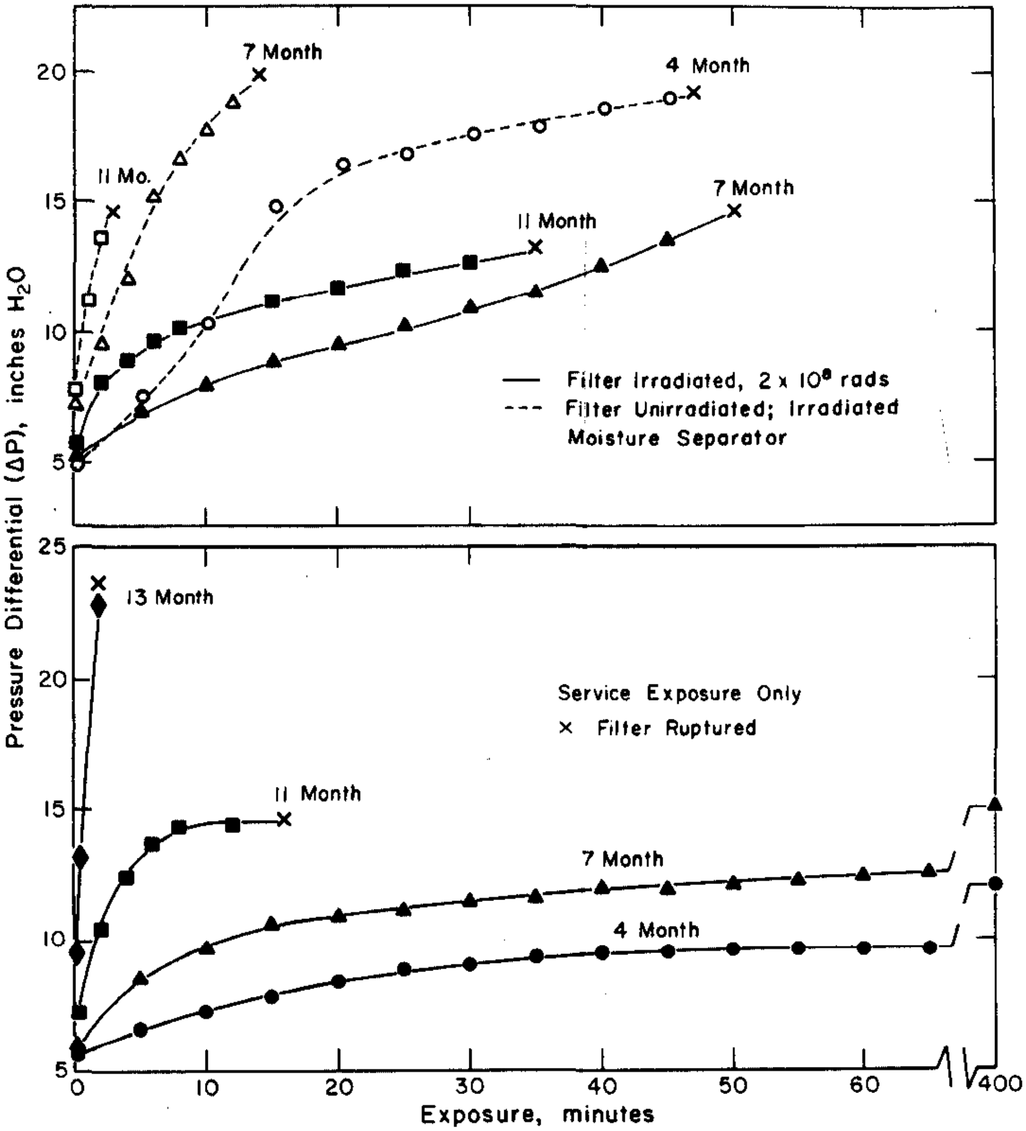


FIGURE 16. Rupture of Type E HEPA Filters with Service Exposure

Accident Simulation Test

Type F Filters. Type F filters (both unirradiated and following exposure to 2×10^8 rads) were exposed to simulated accident test conditions (Table 18). Filter flow and ΔP were monitored during each test to determine moisture pluggage.

Moisture pluggage can be expressed as the measured ΔP across the filter at a selected flow rate of steam-air mixture divided by the measured (or calculated) ΔP at that same flow rate of clean, dry air adjusted for fluid density differences. This ΔP ratio was determined for each test filter at various times throughout each five-hour test.

Calculated confinement system flow is expressed in percent of calculated normal flow with clean, dry HEPA filters (Figure 17) for each group of Type F filters (except group G-S-FA which is shown in Figure 18 for comparison with Type E filters). With the exception of group G-NS-RB filters, the initial calculated flow reduction from pluggage of Type F filters was less than 10%. As will be discussed in a later section, this initial flow reduction is of more importance than similar flow reductions at later times. The characteristic dip found during the first 25 min. exposure is caused by maximum liquid entrainment occurring in the flowing steam-air mixture at a temperature of 80°C (period from 14 to 20 min. in test sequence, Table 18). Liquid entrainment decreased at lower temperatures.

With the exception of G-NS-RB filters, performance of Type F filters was only slightly affected when filters were exposed to 2×10^8 rads and when an irradiated moisture separator was substituted in the test facility. The unique behavior of the G-NS-RB filters, especially with an irradiated moisture separator, may be the result of irregular moisture repellency treatment of the media. This supposition was not confirmed.

One G-S-RB filter and one GA-S-RB filter were exposed to confinement system service in a reactor area for 6 months. Simulated accident performance of these filters was poorer than that for new irradiated filters but similar to performance of service-exposed Type E filters (Figure 19).

Type E Filters. Simulated accident performance of Type E filters was determined following exposure to 1×10^8, 2×10^8, and 6×10^8 rads. Calculated confinement system flows for each of these filters (Figure 18) were compared to that for a new, unirradiated Type E filter. Although initial flow reduction was similar to that for the Type F filters, the unirradiated Type E filter showed better flow recovery, while the irradiated filters showed slightly greater decreases in flow with continued steam-air exposures. Substitution of irradiated moisture separator resulted in only a small effect, similar to that for Type F filters.

Type E Filters with Service. Test filters fabricated with media removed from full-size filters with 4-, 7-, 11-, and 13-months service in the confinement system were tested under simulated accident conditions. Calculated confinement system flow is shown in Figure 18 as a function of filter history. More severe flow reductions occurred for filter media with greater service exposures. Further flow reductions with substitution of an irradiated moisture separator were also more severe for media having longer service. Irradiation significantly improved performance of filters with 11- and 13-months service.

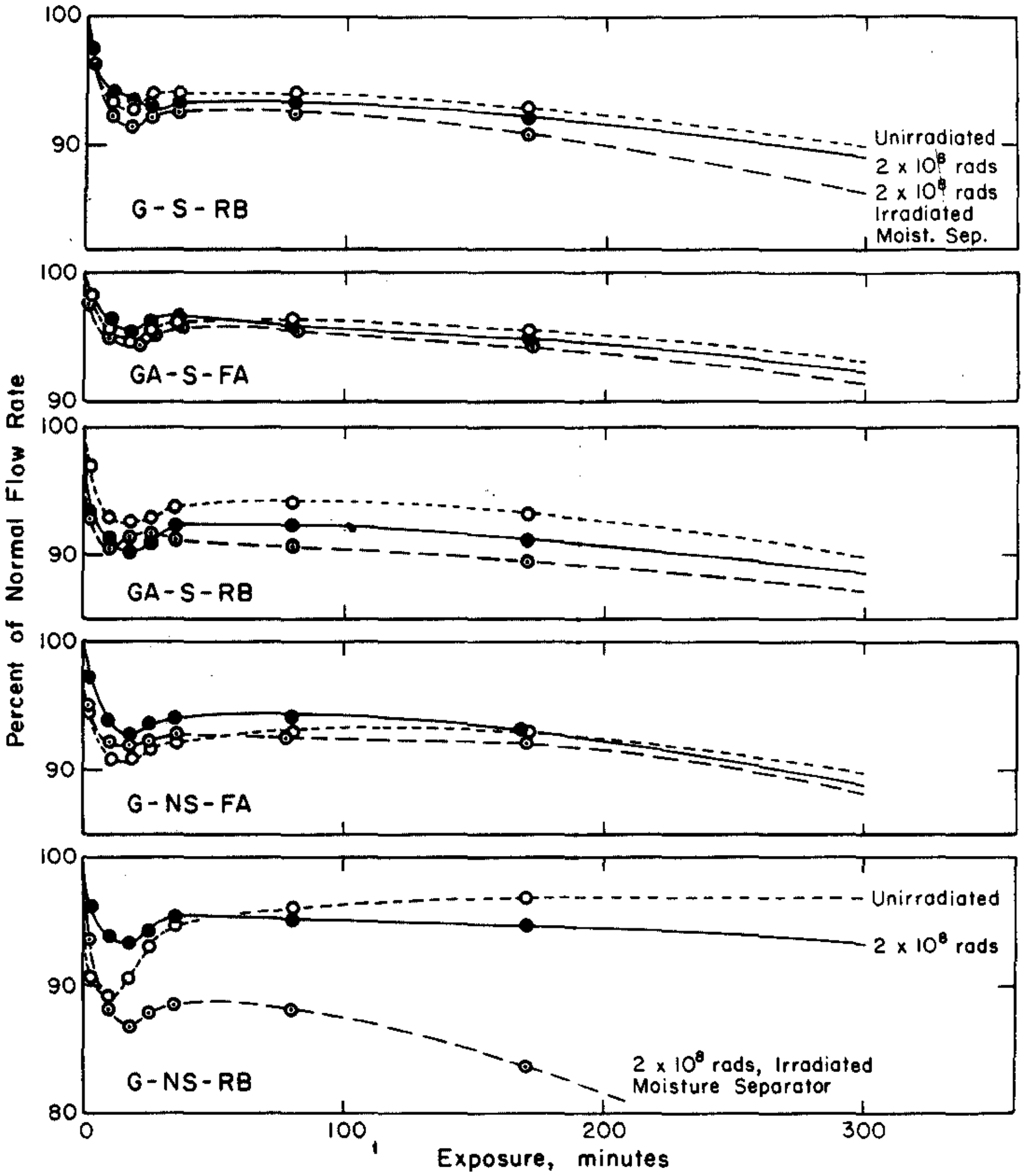


FIGURE 17. Reduced Flow through Type F HEPA Filters with Time under Simulated Reactor Exposure Conditions

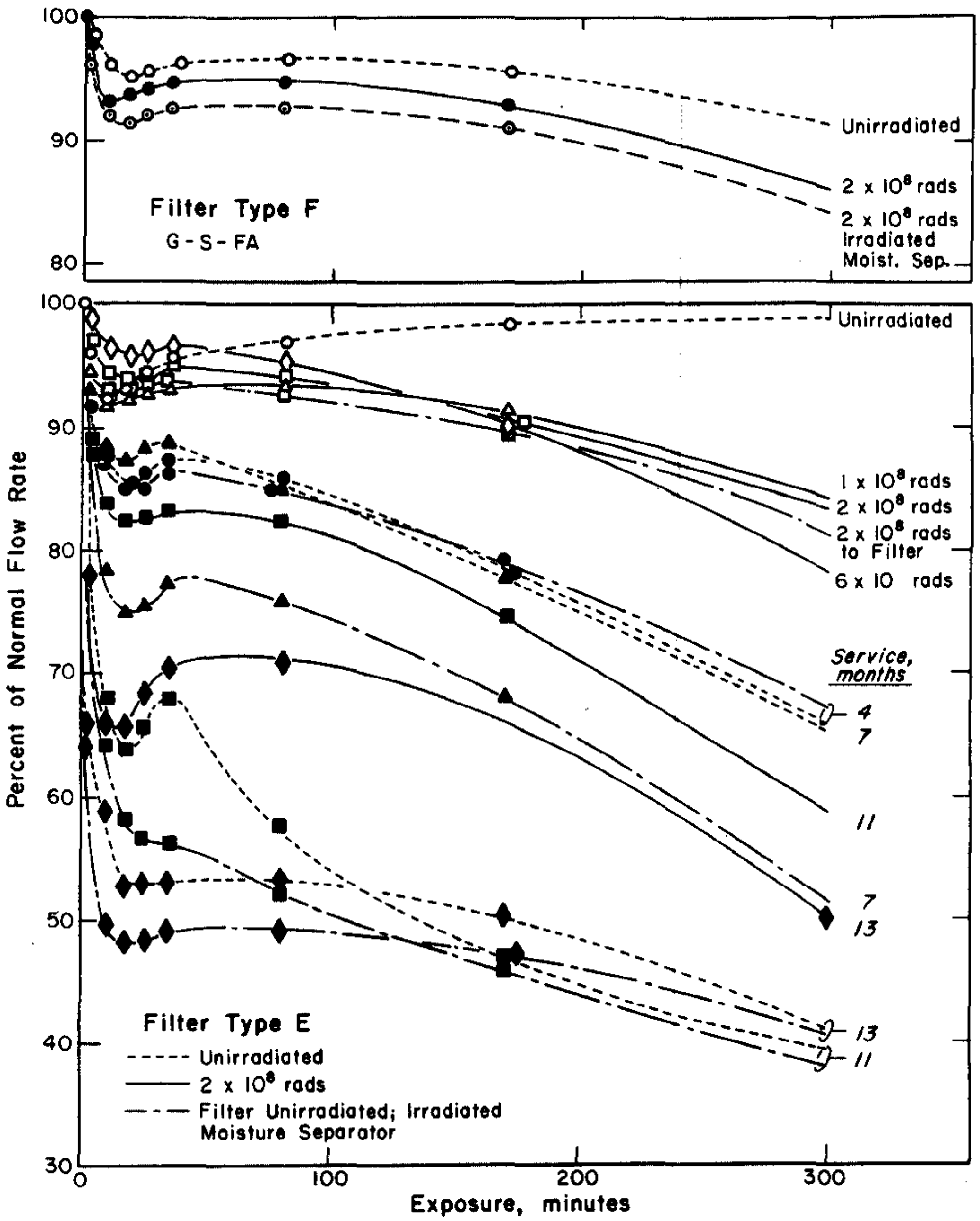


FIGURE 18. Reduced Flow through TYPE E HEPA Filters under Simulated Reactor Exposure Conditions

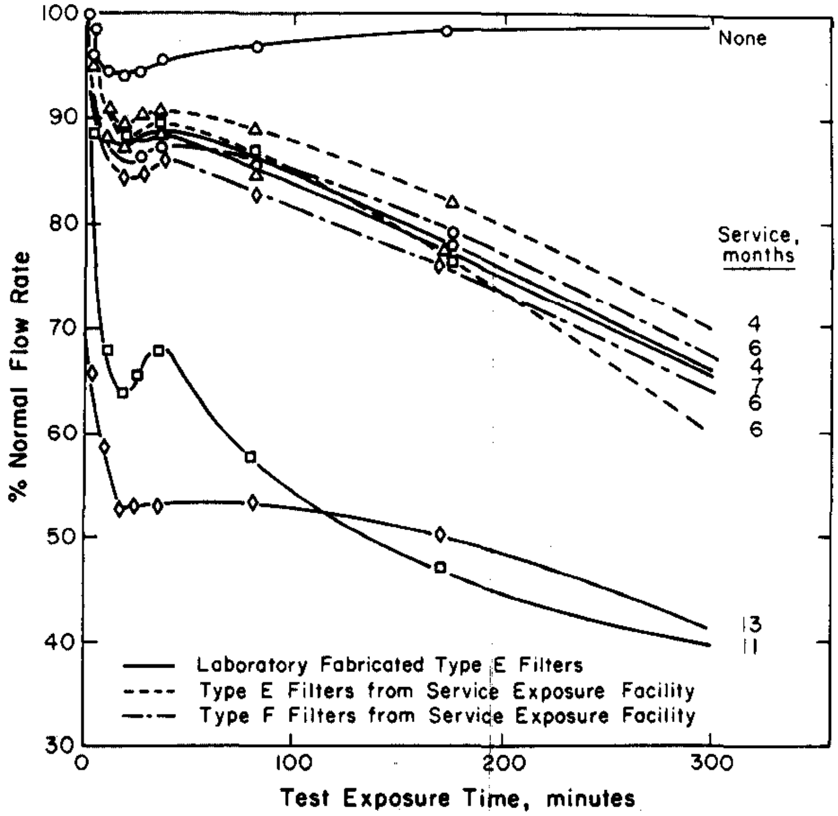


FIGURE 19. Reduced Flow through Service-Exposed HEPA Filters under Simulated Reactor Accident Conditions

One Type E filter was exposed to 4-months service and another to 6-months service in a reactor area. Simulated accident performance agrees closely with performance of laboratory fabricated filters.

Application of Results

Because the most severe flow surge postulated through the Savannah River confinement system is calculated to last no longer than 25 sec, rupture of confinement HEPA filters, even if radiation damage to moisture separators occurs, appears unlikely for filters with less than 13-months service and/or radiation exposure of less than 6×10^8 rads.

Tests in operating Savannah River reactors have shown that sufficient subatmospheric pressure following a reactor accident could be maintained at lesser air flows than have been calculated for any of the filters tested.

The reduction in confinement system flow when HEPA filters plug with moisture depends on: fixed pressure-flow characteristics of the system (pressure losses in ducts, fan head, etc.); variable operating conditions (number of exhaust fans, filter compartments on line, etc.) at the time of the postulated accident; and the increased pressure loss across the HEPA filters. Carbon adsorbers would be heated by decay of radioiodine adsorbed from the flowing air stream. Carbon temperatures following the accident would depend on the quantity of radioiodine reaching the adsorbers and several transient parameters.

The temperature of the steam-air mixture reaching the carbon adsorbers has been discussed previously (Table 18) and would be expected to decrease exponentially with time; this decrease would cause the carbon adsorber temperatures to decrease following the accident. Radioiodine decay (especially for the short-lived isotopes) would reduce the rate of heat generation and thus carbon adsorber temperatures following the accident. Reduced confinement system flow would result in slower removal of decay heat and higher carbon adsorber temperatures. Such reduced flow would also retard the transport of the radioiodine to the carbon adsorbers and allow greater decay before adsorption.

The combined effect of these transient parameters is that calculated carbon temperatures are increased by reducing system flow; however, the actual temperatures are strongly affected by the time interval between the accident and the occurrence of significant flow reduction. Temperatures calculated from performance data on a number of test filters and currently allowed confinement system operating conditions are at a maximum 20 to 30 min. after the accident occurs. The maximum calculated temperature at the carbon beds is well below the ignition temperatures of most carbons, and sufficiently below the temperature at which significant thermal desorption of iodine would be expected.

As discussed in this and previous reports, HEPA filter construction materials and complete filters have been subjected to several types of tests to determine performance characteristics following radiation and service exposure. The purpose of these tests was to correlate measured deterioration in materials and filter performance under simulated accident conditions as a basis for optimum confinement filter purchase and operating specifications.

As a result of this work, tests of media water repellency were adopted at the Government-Industry Meeting on Filters, Media, and Media Testing at Oak Ridge, Tennessee, in 1972 as a standard technique for evaluating radiation degraded filter media.[19]

The water repellency test has also been correlated with simulated Savannah River performance of service-exposed filters as shown in Table 19.

A policy of: a) initial (one time) qualification of a prospective filter supplier through laboratory-simulated accident testing of filters, and b) routine physical property tests on each roll of filter media, provides adequate quality assurance and has been adopted in Reactor Confinement HEPA Filter Purchase Specifications for the Savannah River Plant.

TABLE 19

Comparison of HEPA Filter Simulated Accident Performance and Water Column Penetration Time

Service Exposure, months	*Initial Filter Flow Reduction, %*[a]	*Water Column Penetration Time*
None	5	>48 hr
4	14	> 4 hr
7	13	> 4 hr
11	36	209 sec[b]
13	47	66 sec[b]
48	47	57 sec[b]

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a. During first 20 min of simulated accident test.

b. Average of 10 tests.

APPENDIX - CALCULATING IODINE EVAPORATION

The theoretical treatment of the partition of iodine between gas phase and water at various pH, as performed by Eggleton,[15] contains an error, as shown by the following re-examination of his mathematical treatment.

Eggleton shows that the following four reactions are significant.

$$I_2\text{(gas phase)} \overset{K_1}{\rightleftarrows} I_2\text{ (aqueous)} \qquad K_1 = \frac{[I_2]\text{ aq.}}{[I_2]\text{ gas-phase}} \qquad (1)$$

$$I_2\text{(aq.)} + I^- \overset{K_2}{\rightleftarrows} I_3^- \qquad K_2 = \frac{[I_3^-]}{[I_2]\text{ aq. }[I^-]} \qquad (2)$$

$$I_2\text{(aq.)} + H_2O \overset{K_3}{\rightleftarrows} H^+ + I^- + HIO \qquad K_3 = \frac{[H^+][I^-][HIO]}{[I_2]\text{aq.}} \qquad (3)$$

$$I_2\text{(aq.)} + H_2O \overset{K_4}{\rightleftarrows} H_2OI^+ + I^- \qquad K_4 = \frac{[H_2OI^+][I^-]}{[I_2]\text{ aq.}} \qquad (4)$$

Also, since there is no net oxidation or reduction in a hydrolysis reaction:

$$[HIO] + [H_2OI^+] = [I^-] + [I_3^-] \qquad (9)$$

The partition coefficient (P, in his nomenclature) is defined as

$$P = \frac{\text{g-mol } I_2/\ell \text{ water}}{\text{g-mol } I_2/\ell \text{ gas}}$$

$$P = \frac{[I_2]\text{aq.} + \tfrac{1}{2}\left\{[HIO] + [H_2OI^+] + [I^-] + 3[I_3^-]\right\}}{[I_2]\text{gas phase}} \qquad (10)$$

By substituting in Equation 9 expressions for $\left[I_3^-\right]$, $\left[HIO\right]$ and $\left[H_2OI^+\right]$ as obtained from Equations 2, 3, and 4 and solving for I^-, one obtains:

$$P = \frac{\left[I_2\right] \text{aq.} + \left\{1 + 2K_2 \left[I_2\right] \text{aq.}\right\} \left\{\frac{K_3}{[H^+]} + K_4\right\}^{\frac{1}{2}} \left\{\frac{1}{[I_2]\text{aq.}} + K_2\right\}^{-\frac{1}{2}}}{\left[I_2\right] \text{aq.}/K_1} \qquad (11)$$

Eggleton's Equation 11 is in error in that his denominator is given as $\left[I_2\right]$ aq. K, rather than $\left[I_2\right]$ aq./K. While this may be only a typographical error, it is also difficult to reconcile his plots of P vs. $\left[I_2\right]$ aq. with the corrected equation. We have, therefore, recalculated these curves; the results are given in Figure 1.

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