DP-1542

CONFINEMENT OF AIRBORNE RADIOACTIVITY: FINAL PROGRESS REPORT JANUARY/DECEMBER 1978

A. G. EVANS

E. I. du Pont de Nemours & Co. (Inc.)
Savannah River Laboratory
Aiken, South Carolina 29801

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

Printed in the United States of America

Available from

National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy A03; Microfiche A01

726245

DP-1542
Distribution Category: UC-80

CONFINEMENT OF AIRBORNE RADIOACTIVITY: FINAL PROGRESS REPORT JANUARY/DECEMBER 1978

by

A. G. Evans

Approved by:

G. F. Merz, Research Manager

Nuclear Engineering Division

Publication Date: February 1980

E. I. du Pont de Nemours & Co. (Inc.)
Savannah River Plant and Laboratory
Aiken, South Carolina 29801

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY UNDER CONTRACT DE-AC09-76SR00001

ABSTRACT

A new test method has been developed at the Savannah River Laboratory for evaluating the iodine retention capabilities of carbon used in the airborne-activity confinement system. Methyl iodide tagged with I-131 is injected into a test gas stream continuously for 5 hours with test conditions of 80°C temperature, 95% relative humidity, and 55 feet per minute linear flow velocity. Results show that the CH_3I retention efficiency is independent of the inlet CH_3I concentration over the range of at least 0.9 to 200 $\mu g/m^3$ in the test gas stream. Comparative data show excellent correlation between CH_3I penetration and radiolytic iodine penetration (the iodine radiolysis test previously used to evaluate carbon performance) over at least 2/3 of the useful life of carbon in the Savannah River Plant confinement system. The method was also used to evaluate the effects of paint fumes on in-service carbons and showed that solvent exposure reduced carbon service life by 5 to 7 months. Experimental carbons both before and after service exposure in the SRP carbon test facility were also evaluated.

CONTENTS

LIST OF FIGURES

LIST OF TABLES

CONFINEMENT OF AIRBORNE RADIOACTIVITY: FINAL PROGRESS REPORT - JANUARY-DECEMBER 1978

INTRODUCTION

The airborne activity confinement system for each of the Savannah River Plant (SRP) production reactors is a continuously online, off-gas cleanup system designed to collect halogens and particulates that could be released in the highly unlikely event of a reactor accident.[1] Active components in the system include moisture separators to remove entrained water droplets, HEPA filters to remove particulate radioactivity, and beds of activated carbon to remove volatile halogens. All air from the process areas of the reactor buildings passes through the confinement system before being exhausted to the atmosphere.

The Savannah River Laboratory (SRL) has conducted a continuing program to characterize and improve the performance of the confinement system components under contract with the DOE Division of Waste Management and Transportation.[2-18] This report summarizes research activities during the calendar year 1978 and is a final progress report. Included in the report is a discussion of methyl iodide test results for new and service-aged samples of Type GX-176* carbon (the type of carbon used in the SRP confinement system) as well as test results for experimental carbons receiving service exposure in the P-Area Carbon Test Facility.

CONFINEMENT SYSTEM STUDIES

Background

The SRP confinement systems were placed in service in 1964 with unimpregnated Type 416** carbon installed in the beds to contain the airborne halogens that could be released in the unlikely event of an accident.[1] Radiolytic desorption mechanisms were discovered in 1971 which could result in the release of some of the radioiodine trapped on the carbon beds.[9,10] These earlier studies showed that, in the presence of a high-intensity radiation field, some of the iodine is converted from inorganic salts to organic iodide compounds (primarily methyl iodide) which are poorly

* Product of North American Carbon Company, Columbus, Ohio.
** Product of Barneby-Cheney Company, Columbus, Ohio.

retained by unimpregnated types of carbon. Changeover from unimpregnated Type 416 to impregnated Type GX-176 carbon was initiated in 1974 to reduce the potential release rate associated with the radiolytic desorption mechanisms. The SRL radiolysis test was adopted as the standard test method for carbon in the SRP confinement system and has been used in all subsequent service-aging studies.[11,18]

In the radiolysis test, elemental iodine tagged with I-131 is loaded onto a test bed of carbon for 1 hour in a steam-air environment (80°C, 95% relative humidity) in the presence of a high-intensity gamma radiation field (>1.0 x 10^7 rad/hr absorbed dose rate in the carbon bed). The loading period is followed by a 4-hour desorption period during which the temperature, humidity and radiation field strength are maintained at the same level as during the loading period. Airflow during the test is maintained at a linear face velocity of 55 feet per minute (the nominal face velocity for the SRP carbon beds).

Earlier studies showed that above a dose rate of about 10^7 rad/hr, radiolytic desorption does not increase with increasing radiation intensity.[9,11] Below 10^7 rad/hr, however, the reaction is dose rate dependent.[9] The dose rate attainable in the SRL radiolysis facility is now approaching 10^7 rad/hr so that a major equipment redesign would be required to raise the field intensity into the range where reliable test results can be obtained. Thus a program was initiated to determine whether alternative methods could be used to evaluate the iodine retention capabilities of the SRP confinement system carbon.

Since the radiolytic desorption phenomenon has been shown to be primarily a reaction in which organic iodides are formed and subsequently desorbed from the carbon, development of a test method employing direct injection of organic iodides into the test gas stream was undertaken. Methyl iodide was chosen as the test compound, both because it is the predominant species identified in radiolysis tests and because comparative data on methyl iodide retention efficiencies were available from other sites.

Comparison of results from radiolysis and methyl iodide tests was made by performing CH_3I retention tests on carbon samples previously evaluated using the radiolysis test method.

Test Equipment

A new test apparatus was designed and constructed at SRL to inject a wide range of CH_3I concentrations into the test gas

stream (Figure 1). The apparatus can also control the temperature, humidity, and flow rate in the system over a wide enough range to facilitate comparative testing with other sites.

The injection system consists of a calibrated pressure bomb assembly, a micro-metering valve, and a calibrated flow meter. A known amount of radioactive CH_3I is pipetted into the filling port and the system is sealed. Hydrocarbon-free, compressed air is then added to the assembly to a known pressure. The CH_3I-laden air is then metered into the test gas stream at a predetermined rate to achieve the desired concentration. The bomb assembly is also equipped with appropriate safety pressure relief valves (not shown), and a carbon safety bed to permit depressurization without releasing radioactive gases into the fume hood.

A constant temperature oven is used to house the test bed assembly, a steam-air mixing chamber, and a dew-point hygrometer. Prefiltered, metered air can be routed through a low-temperature humidifier assembly (not shown) or routed directly into the mixing chamber through a preheated line. Filtered steam is added to the chamber for the higher temperature tests to achieve the desired humidity. A small portion of the air downstream of the mixing chamber is pulled through the hygrometer (equipped with a precision platinum resistance thermometer) to determine the dew point of the test gas stream. A second precision platinum resistance thermometer is used to measure the temperature of the gas to obtain the relative humidity. The CH_3I-air mixture from the bomb assembly enters the test gas stream downstream of the hygrometer, but upstream of the test bed. The flow-rate from the bomb assembly is maintained at <1/300 of the total sample flow so that the flow rate's effect on the relative humidity is negligible.

The CH_3I-laden air is then routed through the test bed assembly, out of the oven through a water-cooled condenser and through the backup bed assembly. Heated air is added to the gas stream downstream of the condenser to improve the backup bed efficiency by lowering the humidity. Methyl iodide collected in the condensate is accumulated on an ion exchange column to improve the counting geometry.

Activity on the test beds, backup beds, and ion column is determined using a calibrated gamma scintillation counter and single-channel pulse-height analyzer. Activity decay corrections, pressure and material balance calculations, and preparation of a data summary report are accomplished using a computer code written for the new test method.

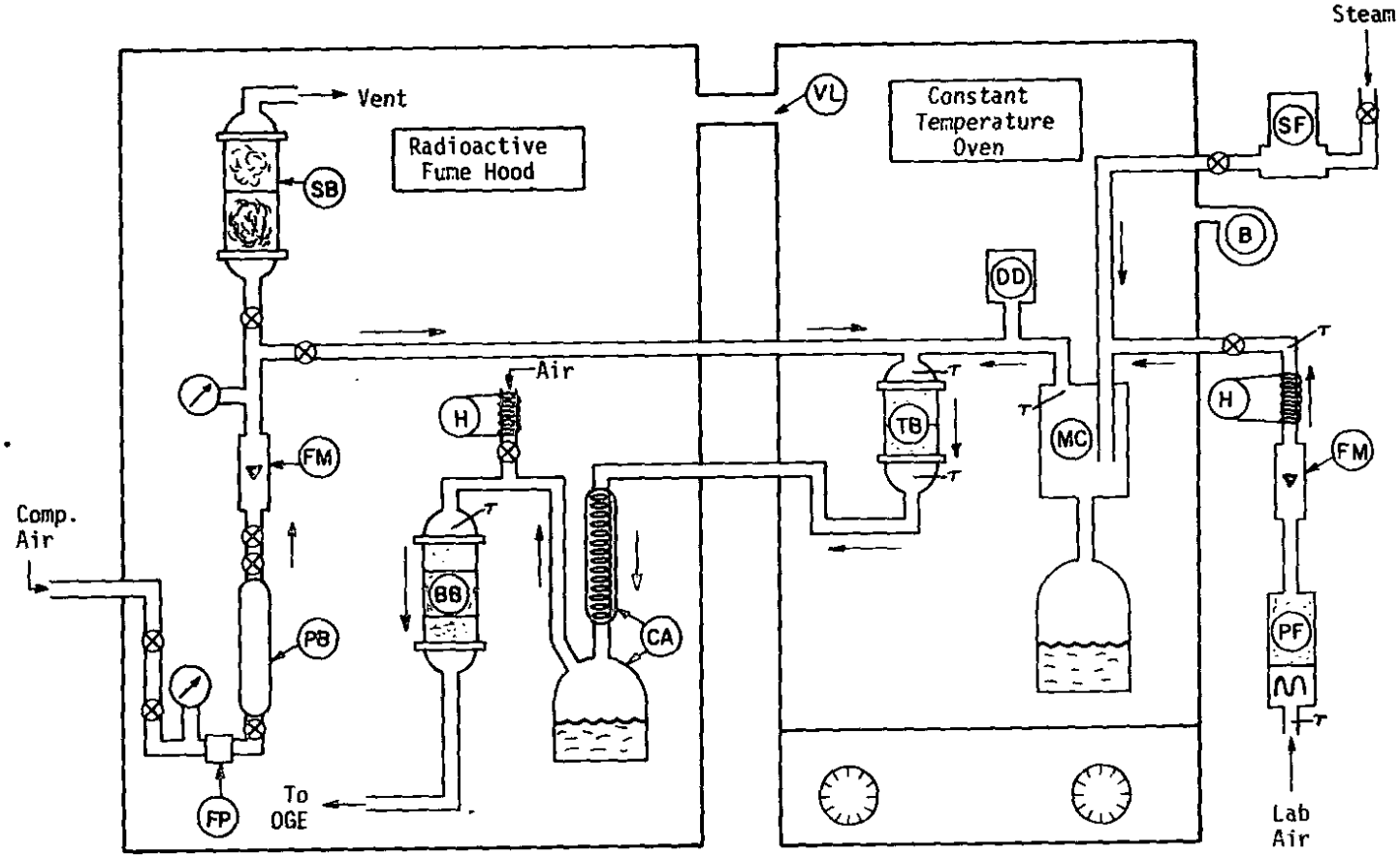

FIGURE 1. Schematic Diagram of the Methyl Iodide Test Apparatus

Methyl Iodide Test Results

New Carbon

The initial tests with the new apparatus were made with unused carbons under various flow, humidity, and inlet concentration conditions. The primary purpose of these tests was to establish baseline data for comparisons with service-aged carbons. A secondary purpose was to determine the test sensitivity to the more important control parameters for routine equipment operation. Three types of carbon were used in these tests: 1) unimpregnated Type 416, 2) impregnated Type GX-176, and 3) impregnated Type G-618.* Type 416 carbon was chosen because of its low retention efficiency for methyl iodide (typical of unimpregnated carbons). Type GX-176 (co-impregnated with 1% TEDA** and 2% KI)[13] is the carbon used in the SRP confinement system. Type G-618 carbon (impregnated with 5% TEDA) was chosen as a reference carbon because it normally exhibits a very high methyl iodide retention efficiency.

All tests were performed at a test bed temperature of 80°C (the temperature used in the radiolysis test), and most were performed at 95% relative humidity (RH). Some tests were made at 90% RH for comparison. Linear face velocities employed were 55 feet per minute (fpm), which is the nominal face velocity for confinement system carbon beds, and 83 fpm (about 1.5 times the nominal face velocity).

The test procedure includes 1) a preheating period to bring the test carbon bed to 80°C with no gas flow through the system (a step necessary to prevent thermal regeneration of used carbons in later tests); 2) a 30 minute pre-equilibration period with steam-air flow (necessary to establish thermal equilibrium between the test carbon and test gas stream); and 3) a 5-hour methyl iodide injection period in the steam-air environment (corresponding to the 5-hour test period used in the radiolysis test). Methyl iodide gas flow is stopped 5 minutes before the end of the test to purge the upstream sample lines for contamination control purposes.

Test data are summarized in Table 1. The data show that a 1-in.-deep bed of new GX-176 retained an average of 93.63% $\pm$ 1.40% of the methyl iodide incident at the front face (80°C, 95% RH and at a face velocity of 55 fpm).† By comparison, new G-618 carbon had a retention efficiency of 89.19% $\pm$ 1.98%, and new 416 carbon retained only 5.39% $\pm$ 0.56% under the same test conditions.

---

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

** Triethylenediamine, $C_6H_{12}N_2$.

† The $\pm$ value is the standard error of the replicate determinations.

TABLE I

Methyl Iodide Test Results for New Carbons
(All tests run at 80°C)

Carbon Type	Face Velocity, fpm	Relative Humidity, %	Inlet CH_3I Concentration, g/m^3	Methyl Iodide Retention Efficiency, %
GX-176 (Summary)				
	83	95	102.2	66.61
	83	90	4.6 - 28.5	83.83 ± 1.60
	55	95	0.9 - 194	93.63 ± 1.40
G-618 (Summary)				
	83	95	21.0	65.64
	83	90	5.2 - 27.2	80.37 ± 0.27
	55	95	2.9 - 189.6	89.19 ± 1.98
416 (Summary)				
	83	95	11.3 - 29.2	3.16 ± 0.24
	55	95	16.4 - 18.7	5.39 ± 0.56
GX-176 (Individual)				
	83	90	17.9	81.61
	83	90	10.1	85.38
	83	90	28.5	83.93
	83	90	4.6*	84.41*
	55	95	21.8	94.32
	55	95	19.0	95.22
	55	95	0.9*	92.78*
	55	95	24.8	94.67
	55	95	1.3*	92.28*
	55	95	20.6	93.08
	55	95	138.6	91.03
	55	95	194.0	94.69
	55	95	10.3*	94.63*
G-618 (Individual)				
	83	90	27.2	80.08
	83	90	26.5	80.43
	83	90	5.2*	80.61*
	55	95	36.4	86.71
	55	95	31.2	90.61
	55	95	2.9*	89.91*
	55	95	25.2	89.39
	55	95	1.7*	89.60*
	55	95	23.6	88.26
	55	95	189.6	88.09
	55	95	182.8	88.46
	55	95	21.1*	87.67
416 (Individual)				
	83	95	11.3	3.43
	83	95	29.2	3.24
	83	95	25.3	3.12
	83	95	24.5*	2.85*
	55	95	18.7	4.76
	55	95	17.4	5.58
	55	95	16.4*	5.84*

* Denotes the second 1-in.-deep test bed in series (see text).
All efficiency values given are for 1-in.-deep test beds.
± values listed are standard deviations for the average values.

The data also show that the retention efficiency is independent of the incident methyl iodide concentration over at least a 200-fold concentration range (from 0.9 to 194 $\mu g/m^3$ for GX-176). Some of the wide ranges of inlet concentrations were obtained by using two 1-in.-deep test beds in series. The efficiencies and inlet concentration values were calculated for the second bed from efficiency and concentration data for the first bed. In Table 1, the first bed in each of the paired data sets is listed first, and the second beds are marked with an asterisk. Examination of the data shows that there is no statistically significant difference between first and second bed efficiencies obtained in this manner. A two-bed sample train supplies duplicate analytical data from a single test. A wide variation in efficiency between the two test beds can also be used to indicate flooding of the first bed and thus flag invalid tests.

Both the flow rate through the bed and the relative humidity had a very significant effect on retention efficiency for impregnated carbons. Increasing the linear face velocity by 50% (from 55 fpm to 83 fpm) decreased the efficiency by about 33% for both GX-176 and G-618 (from 93.63% to 66.61% for GX-176, and from 89.16% to 65.64% for G-618). At 83 fpm face velocity, decreasing the relative humidity from 95% to 90% increased the efficiency by about 25% (from 66.61% to 83.83% for GX-176, and from 65.64% to 80.37% for G-618). These data indicate that careful control of both flow rate and relative humidity is required to obtain reproducible results. However, the fact that the standard deviation obtained from the GX-176 is only $\pm$1.4% indicates that adequate control can be obtained with the instrumentation available.

One surprising feature of the test series on new carbons was the consistently poorer performance of G-618 compared to GX-176. G-618 was chosen as a reference carbon because test data from other sites showed this generic type of carbon (5% TEDA on coconut charcoal) to be the most efficient collector of methyl iodide of the commercial carbons available. The most probable explanation for the phenomenon can be found by comparing test conditions used to evaluate retention efficiency.

Most of the data that has been previously reported are based on standardized tests[19] specified in RDT M16-1T (a predecessor to the proposed ASTM standard). In the RDT tests (whether run at 25 or 80°C), CH_3I is injected over a shorter period of time (120 minutes) and at a higher concentration (1750 g/m_3), then desorbed (without further CH_3I addition) for another 2 hours. Methyl iodide reacting with the TEDA in the charcoal tends to form the thermally stabilized [$(CH_3I)_2$:TEDA] complex, which has a melting point of 284°C, while the unreacted TEDA has a boiling point of 174°C. Any TEDA volatilized during the desorption phase of the RDT test will not result in an apparent decrease in efficiency since no

no more methyl iodide is being injected. In the more conservative SRL test, continued CH_3I injection into a decreasing inventory of residual TEDA results in an increasing penetration rate with increasing time into the test. Thus, using a lower methyl iodide loading for a carbon containing only TEDA as a CH_3I trapping agent, the SRL test will give a lower efficiency than the RDT test. Co-impregnated GX-176 is less affected by the TEDA loss since KI is also present to react with the CH_3I. If the SRL test were performed at 25°C, the G-618 carbon would probably exhibit a much higher efficiency, since the TEDA volatilization would be much lower (TEDA has a 22 mm Hg vapor pressure at 80°C and a 0.2 mm Hg vapor pressure at 25°C).

Testing at 80°C for several hours is probably conservative (at least for an SRP reactor), since steaming would not continue for several hours after an SRP design basis accident. Prolonged exposure of the carbon to methyl iodide is not conservative, however. Radiolysis reactions will continue to produce organic iodides internally for several days in radioiodine-loaded carbon beds. Thus the SRL test conditions simulate expected accident conditions more accurately than RDT test conditions for the SRP carbon beds.

Service Aged Carbon

As expected from earlier Naval Research Laboratory (NRL) data, methyl iodide retention by service-aged carbons decreases rapidly with increasing time in service.[17] In addition, the SRP and NRL data are in good agreement when bed depth corrections are applied to the NRL data. A summary of all data is given in Table 2 and Figure 2. Examination of the data shows that a 1- in.-deep bed of new GX-176 retains about 93% of the incident methyl iodide. After 18 months of service, the retention decreases to about 36%, and some samples show only 7 to 8% retention after 27 months of service.

A fairly uniform decrease in efficiency is observed in all samples for the first 15 months of service. Beyond 15 months of service, however, the data become more scattered, which is consistent with earlier radiolysis test results showing variation in aging rates as a function of the confinement compartment in which the carbon was exposed.

The methyl iodide efficiency data can be fitted to the mathematical model shown in Equation 1.

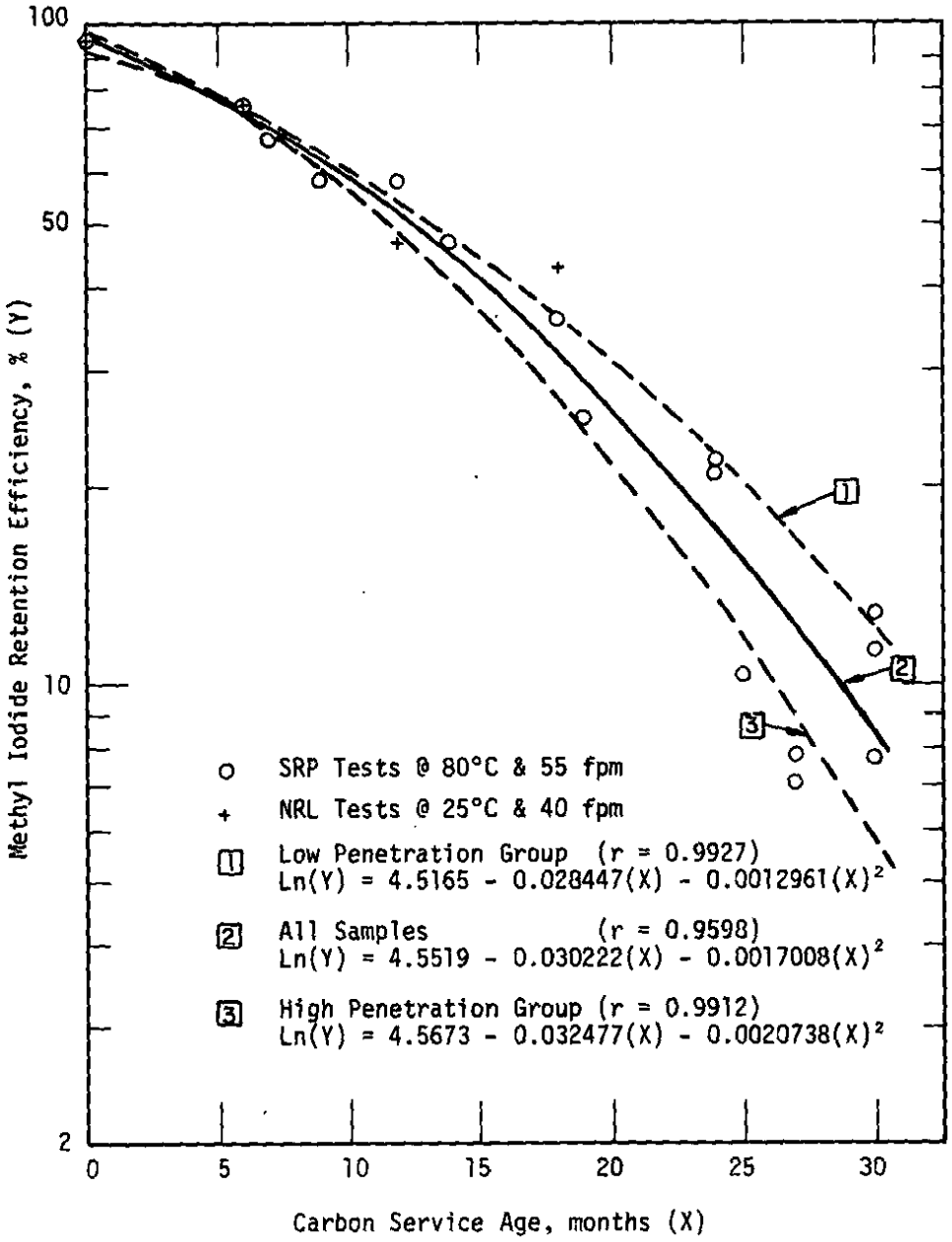

FIGURE 2. Methyl Iodide Retention as a Function of Carbon Service Age

TABLE 2

Methyl Iodide Test Results for Used GX-176 Carbon

Data Source	Service Age, mo	Compartment Numbers	Methyl Iodide Retention Efficiency, %*	Standard Deviation, %
NRL	0	-	93.292	N.A.**
SRL	0	-	93.632	±1.405
NRL	6	K-2	75.875	N.A.**
SRL	6	K-2	75.430	±1.510
SRL	7	P-2	67.427	±1.981
SRL	9	P-2	58.531	±4.376
NRL	12	K-2	46.615	N.A.**
SRL	12	K-2	57.998	±1.490
SRL	14	P-2	47.121	±2.418
NRL	18	K-2	43.343	N.A.**
SRL	18	K-2	35.799	±3.438
SRL	19	P-2	25.430	±4.836
SRL	24	K-2	21.793	±8.211
SRL	24	P-6	21.122	±5.459
SRL	25	C-2	10.299	±5.073
SRL	27	P-3	7.823	±3.113
SRL	27	C-4	7.064	±3.990
SRL	30	P-6	12.779	±5.357
SRL	30	K-2	11.337	±4.933
SRL	30	P-2	7.745	±2.722

* Efficiency of a 1-in.-deep test bed. SRL tests run 5 hours at 80°C, 95% relative humidity, and 55 fpm face velocity. NRL tests run 4 hours at 25°C, 95% relative humidity, and 40 fpm.

** Standard deviation for NRL data not available.

$$\ln(Y) = A + BX + CX^2 \quad (1)$$

Where: Y = Methyl iodide retention efficiency, percent
A, B, and C = Constants
X = Carbon service age, months

When all the test data are considered together, an average aging curve (shown as the solid line in Figure 2) can be obtained. However, the least squares correlation coefficient for this data grouping is 0.9598, indicating only a moderately good fit to the model. When the data are grouped into two categories (a low and a high penetration group as was necessary in earlier analysis test data), a much better fit is obtained. The high penetration group (the lower dashed curve in Figure 2) is composed of those samples showing the maximum penetration in the earlier radiolysis tests. The remainder of the samples were lumped together into the low penetration group (the upper dashed curve in Figure 2). Values obtained for the constants A, B, and C as well as the correlation coefficients for the curve fits are given in Table 3.

TABLE 3

Least Squares Constants for Methyl Iodide Efficiency Curves

Sample Group	Constant A	Constant B	Constant C	Correlation Coefficient
All	4.5519	0.030222	0.0017008	0.95982
High Penetration Group	4.5673	0.032477	0.0020738	0.99124
Low Penetration Group	4.5165	0.028447	0.0012961	0.99268

Correlation with Radiolysis Test Data

A detailed discussion of the effect of service on radiolytic iodine penetration is contained in Reference 18. Data from this publication are summarized in Figure 3. Data points falling on or near the upper boundary of the upper shaded area represent the most conservative estimate available for the rate of increase in radiolytic iodine penetration with increasing service life. When data points from this high penetration group are considered separately, they fit the mathematical model shown in Equation 2 with a correlation coefficient of 0.9936.

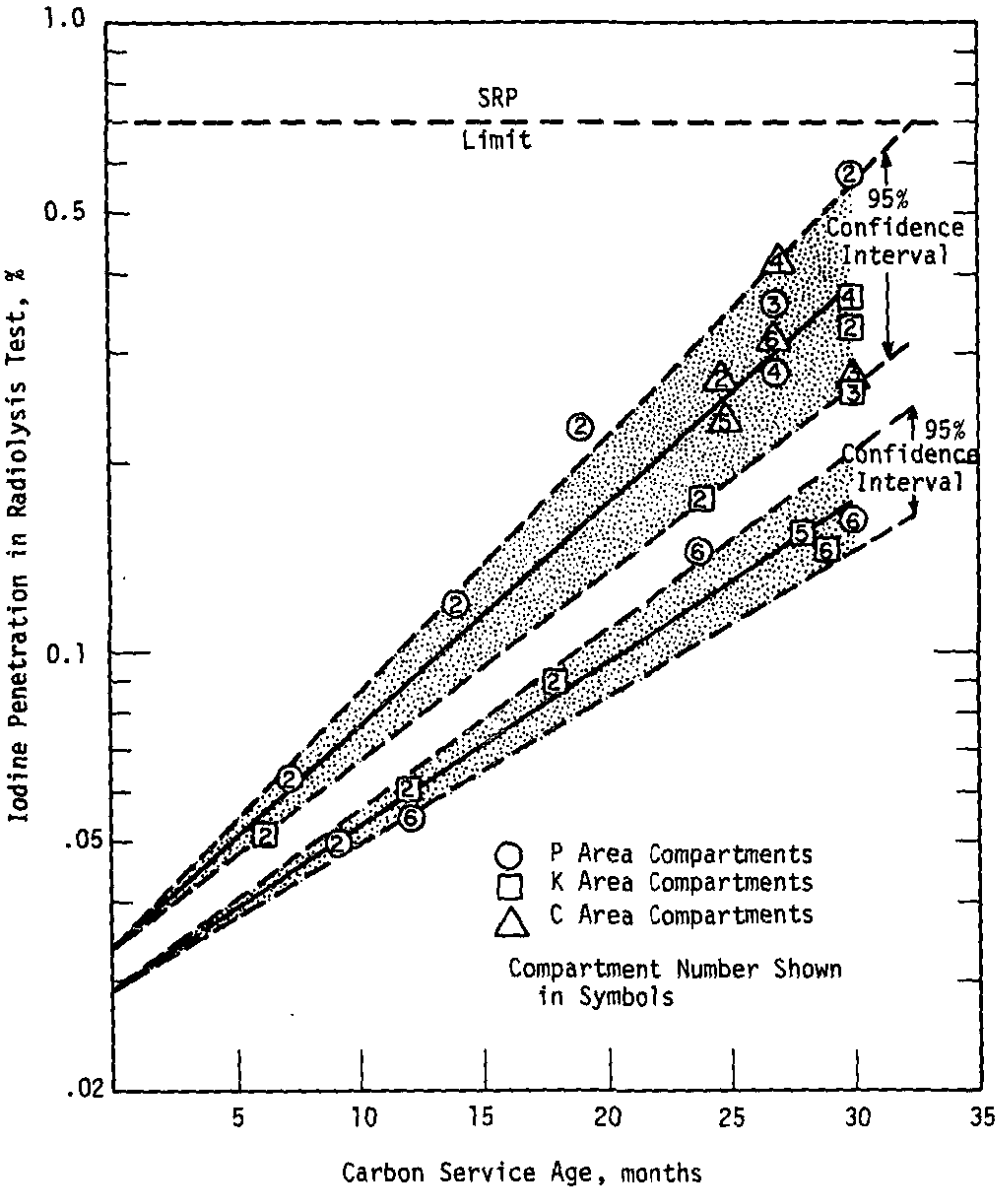

FIGURE 3. Radiolytic Penetration as a Function of Service Age

$$\ln(Y) = A + BX \qquad (2)$$

where: Y = Radiolytic iodine penetration, percent
A = Zero service intercept (-3.5014, or 0.0302% penetration)
B = Curve slope (0.095765/month)
X = Carbon service age, months

If methyl iodide penetration is compared to radiolytic penetration instead of age for the same carbon samples, a fairly good correlation is also obtained (correlation coefficient of 0.9898) with the following regression coefficients:

Y = Radiolytic iodine penetration, percent
A = Zero service intercept (-3.7506 or 0.0235%)
B = Curve slope (0.030663/% CH_3I penetration)
X = Methyl iodide penetration, percent

Data points for the correlations are given in Table 4 and shown in Figure 4.

Even though the spread in the data is greater than would be desired as the carbon approaches the end of its useful service life, the model is an excellent fit for the data for the first 19 months of service (correlation coefficient of 0.9990). More complex mathematical models provide slight improvements in correlation coefficients, but with poorer statistical accuracy (lower 'F' values and higher standard deviations).

Thus, Equation 2 represents the best fit for the correlation between radiolytic iodine penetration and methyl iodide penetration. The excellent fit of the data over nearly 2/3 of the useful carbon life means that the methyl iodide test can be used with reasonable confidence to evaluate the condition of the carbon in the SRP confinement system. The test method can detect the variations in aging rates encountered in different confinement compartments, and can identify carbons which were both intentionally and unintentionally exposed to organic pollutants.

As can be seen from the standard deviations listed in Table 2, a wider spread in data was encountered with used carbons than with new carbons, particularly as the carbon accumulates more service exposure. At least part of this variation is attributable to sampling and compositing techniques. Carbon from the front (inlet) face of the bed is more severely degraded than that from the back face of the bed. When the carbon is removed from the confinement system beds, an effort is made to obtain a blended composite. Any non-homogeniety in the sample tested can cause variation in the test results.

TABLE 4

Methyl Iodide Penetration and Radiolytic Iodine Penetration in High Penetration Group

Data Source*	Service Age, mo.	Compartment Number	Methyl Iodide Penetration, %**	Radiolytic Iodine Penetration, %†
NRL	0	-	6.708	0.028
SRL	0	-	6.368	0.028
NRL	6	K-2	24.125	0.052
SRL	6	K-2	24.570	0.052
SRL	7	P-2	32.573	0.062
SRL	14	P-2	52.879	0.121
SRL	19	P-2	74.570	0.228
SRL	25	C-2	89.701	0.270
SRL	27	P-3	92.177	0.359
SRL	27	C-4	92.937	0.414
SRL	30	P-2	92.255	0.577

* Data source for methyl iodide data only.

** Efficiency of a 1-in.-deep test bed. SRL tests run 5 hours at 80°C, 95% relative humidity, and 55 fpm face velocity. NRL tests run 4 hours at 250°C, 95% relative humidity, and 40 fpm.

† Iodine desorbed in 5 hours at 80°C, 95% relative humidity, and absorbed dose rate of $>1.5 \times 10^7$ rad/hr in the carbon.

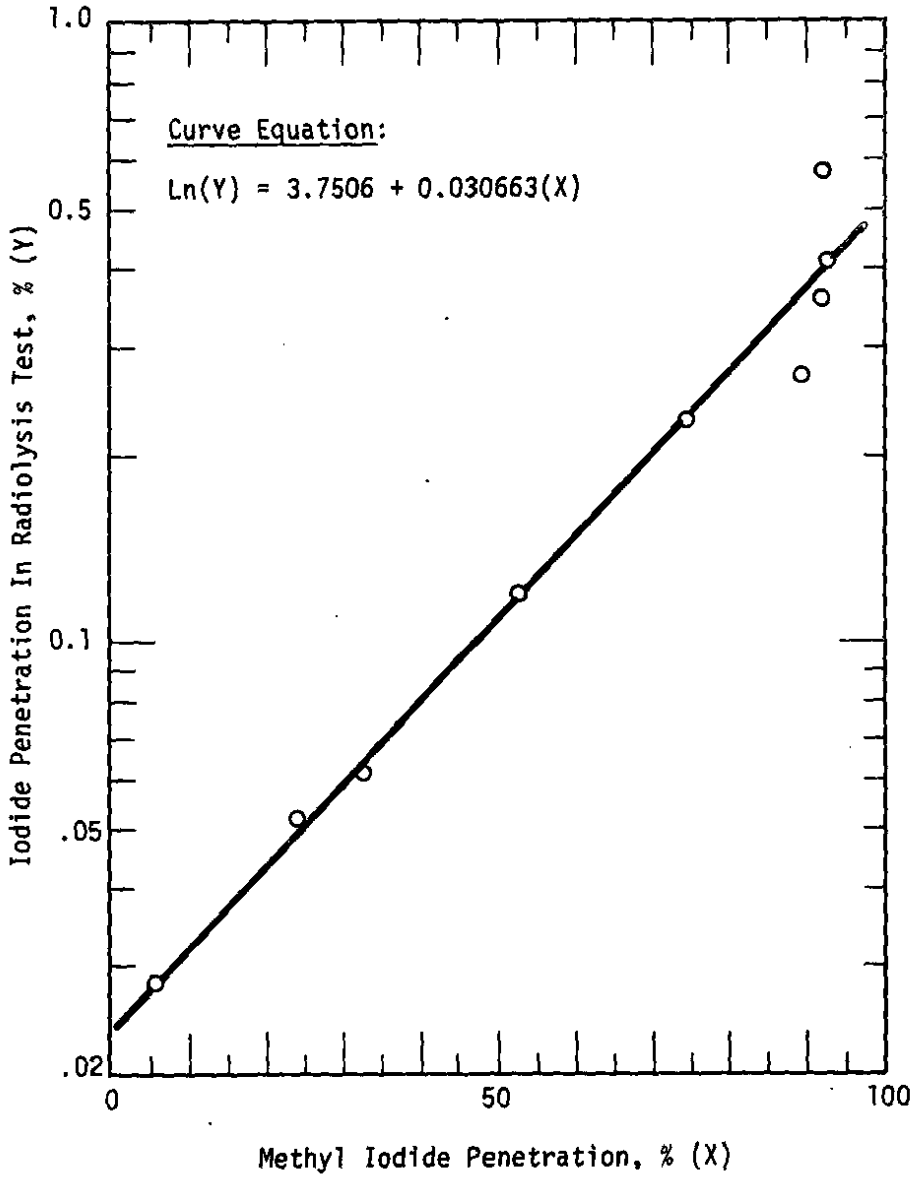

FIGURE 4. Radiolytic Penetration vs. CH_3I Penetration - Maximum Penetration Group

Another cause of variation is the difference in inlet air temperatures between the first and second bed. Heat liberated during moisture adsorption by the first bed can raise the inlet temperature of the second bed by as much as 20°C during the pre-equilibration period. While the exit temperature from the second bed comes to within 1°C of the inlet temperature of the first bed within 30 minutes, some thermal regeneration of the second bed occurs causing slightly better efficiency. This improved efficiency in the second bed for used carbons causes poorer agreement between the two beds.

Accepting only the test results from the first test bed in the series is an overly conservative approach. NRL data[20] show that carbon at the front face of a segmented test bed exhibits up to 10 times higher methyl iodide penetration than does the carbon at the rear face of the same bed. Mixing the poor front face carbon with the better rear face carbon (as is necessary when sampling the SRP beds) results in a poorer retention than would be obtained if the carbon could be tested in an undisturbed condition. The improved efficiency of the second bed compensates for the slightly poorer efficiency of a composited sample. Thus, the average efficiency obtained from two composited beds in series gives a reasonable approximation of the true efficiency of an undisturbed bed.

Effect of Paint Fumes on Carbon

Two paint fume exposures occurred during the report period which showed the adverse effect of organic solvents on the ability of carbon to retain radioiodine. The first incident was an accidental exposure which resulted from the use of an enamel paint in the personnel areas of the K Area reactor building. Fumes from the painting operations were pulled through cracks around doorways into the process area (which is maintained at subatmospheric pressure) and subsequently accumulated on the confinement system carbon.

The carbon poisoning incident was detected during the annual leak testing of the confinement system operations a few weeks after the painting was complete. Successful Freon® (Du Pont) tests could not be obtained for two of the confinement compartments due to the presence of multiple peaks in the leak detector readout equipment. A bed of carbon was removed from one of the compartments (K-2) and sent to SRL for analysis. Repeated heating of the compartment (to about 40°C above ambient temperature) with a low-flow air purge resulted in an eventual successful leak test without the interfering peaks. Interference in a second compartment (K-3) was so severe that the carbon was replaced.

A sample of the K-2 compartment carbon was purged with hot (180°C) nitrogen, and the desorbed gases were collected in a liquid nitrogen cold trap. Gas chromatographic analysis of the vapor revealed the presence of Freon® 112 (residual from leak testing operations), toluene, xylene, and three cyclohexane compounds. Analyses of the same type of enamel used in the reactor building revealed the presence of all of the same compounds except the Freon® 112.

Subsequent methyl iodide testing of the K-2 carbon showed the sample to have a retention efficiency of 30.2% ± 3.2% after 15 months of service (average efficiency for a 1-in.-deep test bed). Earlier testing of carbon from the K-2 compartment[9] (low penetration location) showed an expected retention efficiency of approximately 45% after 15 months of service (curve 1, Figure 2). Thus the paint fume exposure incident reduced the methyl iodide retention efficiency by an equivalent of approximately 5 months of normal service, and early replacement of the K-2 carbon has been recommended.

The second incident was a planned exposure resulting from repainting operations in the reactor room. The painting was scheduled during a long shutdown in the area when no fuel was in the reactor tank. All exhaust air from the building was intentionally routed through a single filter compartment (C-3) during the painting period and for approximately a week afterward (to allow for complete paint drying). The remaining four compartments were then returned to service, and the "sacrificial" carbon in C-3 was replaced.

Carbon samples were obtained from the C-3 compartment both before and after the planned exposure; methyl iodide testing of the samples showed a retention efficiency of 35.0% ± 3.7% with 15 months of service before paint fume exposure (as expected from a "high penetration group" compartment) and 16.9% ± 8.1% after organic solvent poisoning. Thus the paint fumes reduced the efficiency by an amount equivalent to about 7-1/2 months of normal service.

Administrative controls are being instituted which will limit all future painting operations in the reactor buildings to periods when the fumes can be exhausted through sacrificial carbon beds. Additional restrictions will also be placed on other uses of hydrocarbon and chlorinated hydrocarbon solvents in the personnel areas of the buildings (procedural controls already restrict the quantity of these materials allowed in the process areas).

CARBON TEST FACILITY STUDIES

The carbon test facility (CTF) is a specially designed weathering station in P Area which has been used to obtain service-aging data on small carbon samples and scale model HEPA filters.[15] Air from the confinement system supply header is drawn through a moisture separator, an array of small HEPA filters, and an array of carbon test beds, then returned to the supply header. Because the air passing through the CTF is refiltered in the confinement system before discharge to the atmosphere, carbons with unknown performance characteristics can be evaluated without compromising the integrity of the main off-gas cleanup system.

Samples of confinement system carbon (GX-176) are simultaneously exposed in the facility with vendor-supplied carbons and special experimental carbons to obtain comparative data on weathering rates. Thus, the CTF can be used to evaluate a variety of candidate replacement carbons while maintaining the requisite high level of performance for the main system. Results of some of the vendor qualification tests and experimental carbon tests are summarized in the following sections.

Vendor Qualification Tests

A sample of Type 509-1 carbon* (the Barneby-Cheney version of GX-176) was supplied to SRL to be evaluated as a potential replacement for the Type GX-176 carbon now in service in the confinement system. Comparative results obtained for the two types of carbon are shown in Table 5.

Because of the consistently poorer performance of Type 509-1 when compared to Type GX-176 under the same exposure conditions, the 509-1 carbon was rejected as a potential replacement for GX-176.

Experimental Carbon Tests

Cooperative studies between SRL and NRL have been under way for the past 3 years to develop improved impregnated carbons for trapping various forms of airborne radioiodine. Laboratory impregnations of coal base carbons were made which demonstrated that hexamethylenetetraamine (HMTA) in combination with phosphate-buffered iodine salts could be added to the base carbon to produce an efficient trapping medium for both organic and inorganic radioiodine compounds.[16,18]

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

TABLE 5

Comparative Performance of Carbon Types GX-176 and 509-1 in the CTF

Carbon Type	CTF Exposure Time, months	Approximate Equivalent Confinement Service, months*	Radiolytic Iodine Penetration, %**
GX-176	0	0	0.028
509-1	0	0	0.042
GX-176	6	7.5	0.129
509-1	6	7.5	0.208
GX-176	18	22.5	0.375
509-1	18	22.5	0.434

* CTF exposed samples deteriorate about 1.25 times faster than confinement system exposed samples because of differences in the linear face velocity of air passing through the carbon beds.

** The SRL radiolysis test; see text for test description.

The final phase of the studies was preparation with pilot-scale quantities of the experimental carbons followed by weathering experiments to determine how well the products perform after service exposure. NRL contracted with a commercial carbon impregnation firm to prepare 55-pound pilot lots of 5 different base carbons. Samples of each of the experimental carbons were exposed in the CFT for 6 months, removed, and the iodine retention properties determined utilizing the SRL methyl iodide test described earlier in this report. Results of tests run at SRL are summarized in Table 6. NRL methyl iodide test data for unexposed samples of the same carbon are included for comparison. Portions of the CTF-weathered carbon were also sent to NRL for further testing.

CTF-aged GX-176 samples were not tested for CH_3I penetration for direct comparison. However, GX-176 exposed 7 months in compartment P-2 (adjacent to CTF) showed an average methyl iodide penetration of 10.62% (2-in.-deep bed). Since the CTF is operated at a linear face velocity of about 65 fpm, 6 months of exposure in the CTF is approximately equivalent to 7 months in compartment P-2. Thus, the NRL experimental samples on 207-A base carbon* and BPL base carbon* appear to age at about the same rate as the coconut base GX-176 in this short exposure.

* See Table 6.

TABLE 6

Test Data for New and Service-Aged Experimental Carbons

Sample No.[a]	New Carbon			Used Carbon[e]	
		Methyl Iodide Penetration, %			
	pH[b]	NRL[c]	SRL[d]	pH[b]	CH_3I Penetration, %[d]
4314C[f]	9.19	1.54	1.77	6.33	14.88
4315C[g]	9.02	0.26	0.13	6.96	7.48
4316C[h]	9.64	0.90	0.38	6.78	11.57
4317C[i]	9.29	0.46	0.47	6.69	13.52
4318C[j]	9.14	0.72	4.46	6.75	27.85
GX-176[k]	9.70	0.45	0.41	7.70	10.62 $\pm$ 0.74

a. NRL sample designation for impregnated carbon; see base carbon types listed below.

b. 5 grams carbon brought to a boil in 50 ml distilled H_2O, cooled in a sealed flask to room temperature. pH measured on decanted supernate.

c. CH_3I penetration of a 2-in.-deep bed at 25°C, 95% RH, and a superficial face velocity of 40 fpm.

d. CH_3I penetration of a 2-in.-deep bed at 80°C, 95% RH, and a superficial face velocity of 55 fpm.

e. 6 months of exposure of 1-in.-deep beds in the CTF. Samples composited and repacked into two 1-in.-deep beds in series before CH_3I test.

f. 8 x 16 mesh, type ACC base carbon supplied by Union Carbide Corporation, New York, NY.

g. 10 x 16 mesh, type 207A base carbon supplied by Sutcliff-Speakman & Co., Ltd, Lancashire, UK.

h. 8 x 20 mesh, type BPL base carbon supplied by Calgon Corporation, Pittsburg, PA.

i. 10 x 16 mesh, type WVH base carbon supplied by Westvaco, Covington, VA.

j. 8 x 16 mesh, type 915 base carbon supplied by Witco Chemical Corporation, New York, NY.

k. GX-176 Carbon with 7 months of exposure in compartment P-2.

REFERENCES

1. W. S. Durant, R. C. Milham, D. R. Muhlbaier, and A. H. Peters. **Activity Confinement System of the Savannah River Plant Reactors.** USAEC Report DP-1071, E. I. du Pont de Nemours and Co, Savannah River Laboratory, Aiken, SC (1966).

2. W. S. Durant. **Performance of Activated Carbon Beds in SRP Reactor Confinement Facilities - Progress Report: September 1961 - September 1965.** USAEC Report DP-1028, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1966).

3. 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, SC (1966).

4. 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, SC (1969).

5. 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, SC (1969).

6. R. C. Milham and L. R. Jones. **Iodine Retention Studies - Progress Report: July 1969 - December 1969.** USAEC Report DP-1234, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1970).

7. A. G. Evans and L. R. Jones. **Iodine Retention Studies - Progress Report: January 1970 - June 1970.** USAEC Report DP-1259, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1971).

8. A. G. Evans and L. R. Jones. **Iodine Retention Studies - Progress Report: July 1970 - December 1970.** USAEC Report DP-1271, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1971).

9. A. G. Evans and L. R. Jones. **Confinement of Airborne Radioactivity - Progress Report: January 1971 - June 1971.** USAEC Report DP-1280, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1971).

10. A. G. Evans and L. R. Jones. **Confinement of Airborne Radioactivity Progress Report: July 1971 - December 1971.** USAEC Report DP-1298, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1972).

11. A. G. Evans and L. R. Jones. **Confinement of Airborne Radioactivity - Progress Report January-June 1972.** USAEC Report DP-1316, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1973).

12. A. G. Evans. **Confinement of Airborne Radioactivity - Progress Report: July 1972 - December 1972.** USAEC Report DP-1329, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1973).

13. A. G. Evans and L. R. Jones. **Confinement of Airborne Radioactivity - Progress Report: January 1973 - June 1973.** USAEC Report DP-1340, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1973).

14. A. G. Evans and L. R. Jones. **Confinement of Airborne Radioactivity Progress Report: July 1973 - December 1973.** USAEC Report DP-1355, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1974).

15. A. H. Dexter, A. G. Evans, and L. R. Jones. **Confinement of Airborne Radioactivity - Progress Report: January - December 1974.** USERDA Report DP-1390, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1975).

16. A. G. Evans and A. H. Dexter. **Confinement of Airborne Radioactivity - Progress Report: January - December 1975.** USERDA Report DP-1430, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1976).

17. A. G. Evans. **Confinement of Airborne Radioactivity - Progress Report: January-December 1976.** USERDA Report DP-1463, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1977).

18. A. G. Evans. **Confinement of Airborne Radioactivity - Progress Report: January-December 1977.** USDOE Report DP-1497, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, SC (1979).

19. **RDT Standard, Gas-Phase Adsorbents for Trapping Radioactive Iodine Compounds.** USDOE Report RDT M-16-1T, Division of Reactor Research and Development, U.S. Department of Energy (1977).

20. R. R. Bellamy and V. R. Deitz. **Confirmatory Research Program - Effects of Atmospheric Contaminants on Commercial Charcoals.** Paper presented at the 15th DOE Air Cleaning Conference, Boston, Mass., August 10, 1978 (to be published in Conference Proceedings).