663800

DP-1497

CONFINEMENT OF AIRBORNE RADIOACTIVITY

PROGRESS REPORT: JANUARY - DECEMBER 1977

A. G. EVANS

E. I. DU PONT DE NEMOURS AND COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA 29801

PREPARED FOR THE U.S. DEPARTMENT OF ENERGY UNDER CONTRACT AT(07-2)-1

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 $4.50; Microfiche $3.00

663800

DP-1497
Distribution Category: UC-80

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by

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Publication Date: January 1979

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ABSTRACT

Iodine penetration and ignition temperature data obtained from carbon that was removed from the Savannah River Plant (SRP) confinement system show that Type GX-176 carbon has a useful life of at least 30 months. The 30-month service limit used for all SRP carbon is based on the most rapid rate of deterioration observed in any confinement compartment.

Development studies on new halogen adsorbers showed that coal- and petroleum-base carbons can be treated in the laboratory to retain methyl iodide as well as, or better than, commercial coconut-base carbons. These studies showed that hexamethylenetetramine (HMTA) in combination with iodine and phosphate salts are effective impregnation agents for methyl iodide retention on the non-coconut-base carbons. The studies also indicated that triethylenediamine (TEDA) is more effective for CH_3I trapping than HMTA.

Radiation stability tests for iodine-loaded, silver-exchanged zeolite (AgX) showed that some internal migration of iodine occurs in the AgX under dynamic flow conditions in a high intensity radiation environment. The tests also showed that the distribution of iodine in the test bed is more affected by the inlet air concentration than by exposure to gamma radiation.

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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 moisture droplets, HEPA filters to remove particulate radioactivity, and beds of activated carbon to remove 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. Results of previous studies are summarized in References 2 through 17. This report summarizes (1) work during 1977 to evaluate the performance of Type GX-176* carbon installed in the confinement system, (2) research efforts to develop improved carbon types to further enhance iodine retention capabilities and (3) a series of tests on iodine loaded, silver exchanged zeolite (AgX) conducted at the Department of Energy's (DOE) request to evaluate the stability of AgX under radiation stress and dynamic flow conditions.

CARBON PERFORMANCE STUDIES

Background

The air-flow pattern for each of the SRP reactors included routing of reactor room air through a single exhaust header which, in turn, discharged into one end of the confinement compartment supply plenum.[1] The distribution of particulate radioactivity on the

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

air filtration components following the K Area source-rod meltdown incident showed that this air flow pattern caused preferential loading of activity on confinement compartments 2 and 3 (the first two compartments downstream of the reactor room header discharge point).[9] SRL studies on carbon aging mechanisms indicated that NO_2 present in reactor room air was causing a more rapid deterioration of the carbon in compartments 2 and 3.[5,8,9] As a result of these findings, a second reactor room exhaust header was reopened. This header, which had been intentionally sealed when the confinement system was installed on the reactors, discharges air into the opposite end of the compartment supply plenum.

With this modification, reactor room air enters both ends of the supply plenum and should result in a more uniform aging rate in the confinement system carbon beds. In addition, a more uniform iodine loading pattern should be obtained in the unlikely event of a reactor accident. The second exhaust header was opened in all the reactor buildings immediately prior to the changeover to Type GX-176 carbon in the confinement system. Installation of GX-176 carbon began in 1974 and was completed in 1975.

SRL service aging studies indicated that GX-176 carbon should reach the end of its useful life after about 30 months of service.[16,17] Thus, replacement of the first purchase lot of the carbon was begun in 1976. The last of the material was replaced with new GX-176 carbon in December 1977. Iodine retention tests are being run on samples of carbon removed from each of the confinement compartments at the end of the first service cycle. Intermediate age data points were also obtained from selected compartments and reported earlier.[16,17]

Radiolysis Test Data

Results of radiolysis tests performed on GX-176 carbon in service in the SRP confinement system are summarized in Table 1. The data are also presented graphically in Figure 1. Data points in the figure are identified by area and compartment number. Examination of the figure shows approximately equal penetration for compartments 3, 5 and 6 in C Area. Thus, opening the second reactor room exhaust header in C Area has been effective in equalizing the aging rates of carbon in the confinement compartments. The rate of deterioration of carbon in P Area compartments 2, 3 and 4 is still significantly faster than that observed for compartment 6, however. Carbons in K Area compartments 5 and 6 are still aging more slowly than in compartments 2 and 3 as well.

TABLE 1

Radiolysis Test Data on Service Aged GX-176 Carbon

Service Age, Months	*Confinement Compartment*	*Iodine Penetration,*[a] %
0	Control	0.028
7	P-2	0.062
9	P-2	0.050
14	P-2	0.121
19	P-2	0.228
30	P-2	0.543
27	P-3	0.359
27	P-4	0.289
12	P-6	0.055
24	P-6	0.145
30	P-6	0.165
6	K-2	0.052
12	K-2	0.060
18	K-2	0.090
24	K-2	0.176
30	K-2	0.334
30	K-3	0.263
28	K-5	0.155
29	K-6	0.146
30	C-3	0.273
25	C-5	0.242
27	C-6	0.315

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[a] Elemental iodine, 1 hour loading and 4 hours desorption @ 80°C and 95% RH in radiation field of 1.5×10^7 rad/hour.

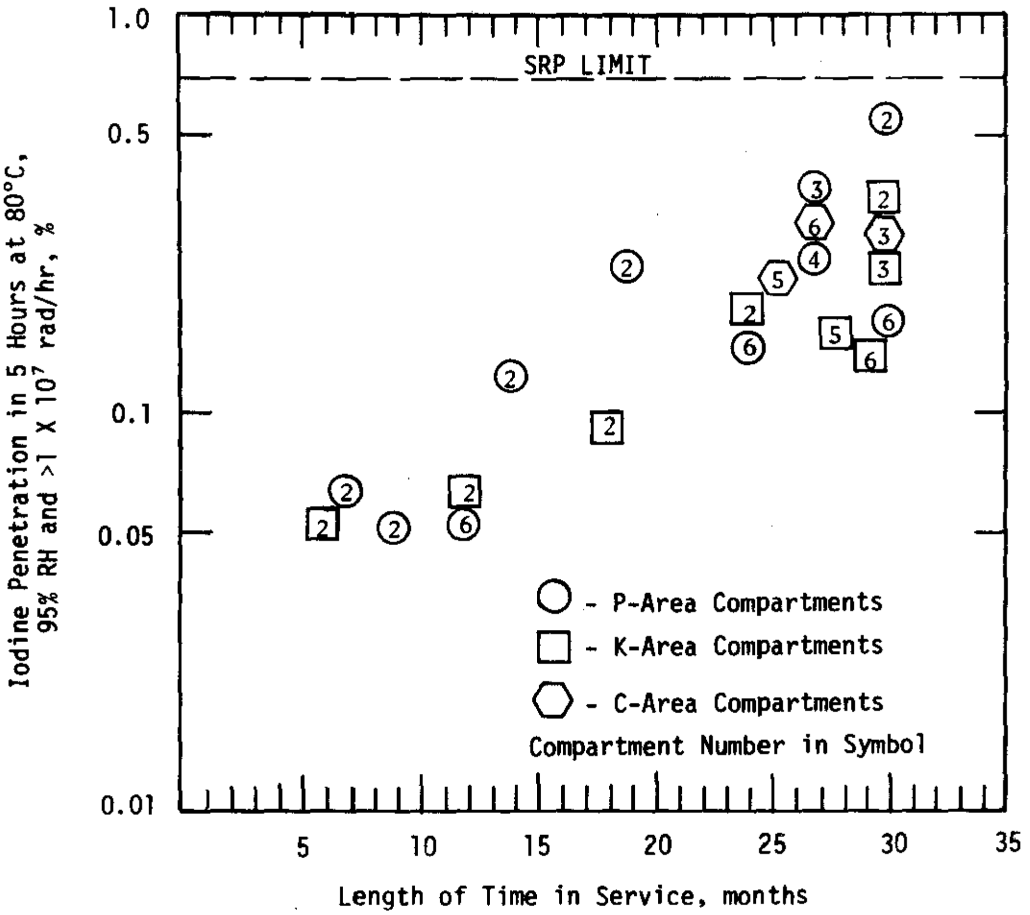

FIGURE 1. Radiolysis Test Data for Service Aged GX-176 Carbon

As indicated earlier in the background discussion, none of the GX-176 carbon was kept in service for more than 30 months. All of the carbon removed from the confinement system at the end of its useful life cycle showed iodine penetration less than the SRP limit of 0.7%. Assuming an exponential increase in iodine penetration with increasing service life for carbon used in all compartments except K-5, K-6 and P-6, a useful service life of about 38 months can be extrapolated from the data. The spread in the data reduces this value to about 33 months' service at the 95% confidence level (Figure 2). Thus, based on the iodine penetration data, extending the useful life of GX-176 beyond 30 months' service in most of the SRP confinement compartments cannot be justified.

Other Test Data

Other performance indication tests run on the confinement system carbon include the following:

(1) Iodine retention at 180°C

(2) Carbon ignition temperatures

(3) Carbon pH measurements

Iodine retention at elevated temperatures is significant because of the possibility of HEPA filter blinding under accident conditions.[14,15] If blinding occurs after the iodine is loaded onto the carbon beds, the resultant reduction in air flow will cause a rise in carbon bed temperatures. Carbon ignition temperature measurements are also made for this reason. Past studies have shown that iodine retention is strongly influenced by the pH of the carbon,[14-17] thus pH data are taken as well. Results of these three tests on service aged carbon are summarized in Table 2.

The data show a significant decrease in carbon ignition temperatures with increasing service life. While the standard ignition temperature test is not designed to give a precise measure of the temperature at which carbon will ignite under accident conditions, it does give an indication of increasing thermal instability with increasing service exposure. Since all the carbon samples tested show similar ignition temperatures after about 30 months' service (including samples from compartments K-5, K-6 and P-6), no recommendation for service use beyond 30 months in any compartment is anticipated.

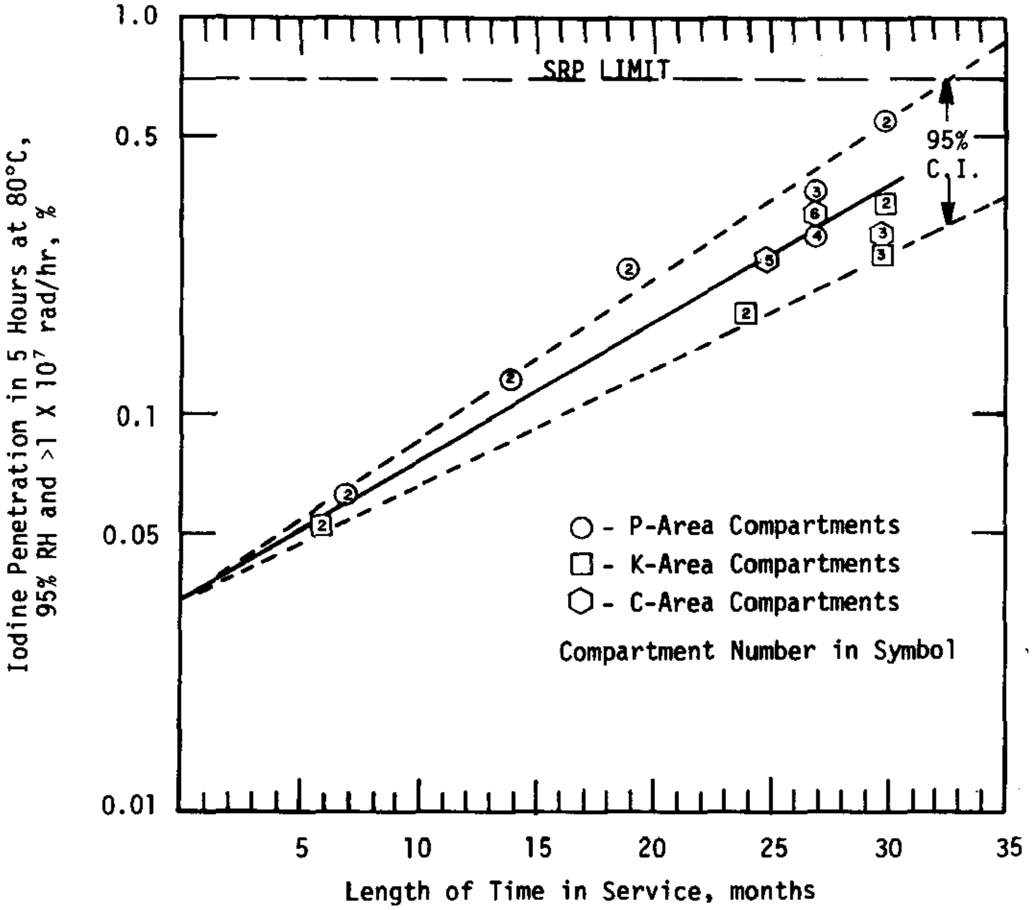

FIGURE 2. Service Life Estimates for GX-176 Carbon

TABLE 2

Other Test Data on Service Aged GX-176 Carbon

Service Age, Months	*Confinement Compartment*	*pH*[a]	*Ignition*[b] *Temp, °C*	*Iodine Penetration,*[c] *%*
0	Control	9.70	390	0.002
7	P-2	7.70	420	0.011
9	P-2	7.10	415	0.016
14	P-2	6.89	340	0.060
19	P-2	6.06	290	2.30
30	P-2	4.78	310	5.04
27	P-3	5.36	280	4.26
27	P-4	4.92	315	4.23
12	P-6	7.12	330	0.023
24	P-6	6.70	320	0.429
30	P-6	5.36	300	1.67
6	K-2	7.60	440	0.027
12	K-2	6.60	320	0.093
18	K-2	6.46	310	0.409
24	K-2	6.15	300	1.60
30	K-2	5.20	295	6.35
30	K-3	5.01	315	5.47
28	K-5	5.26	310	3.19
29	K-6	5.22	315	2.75
30	C-3	4.17	330	2.54
27	C-4	4.50	300	4.48
25	C-5	4.86	310	3.72
27	C-6	4.24	315	2.79

---

a. pH of water extract, 5 g carbon in 50 mL water, boiled, cooled to room temperature and liquid decanted.

b. ASTM D 3466 METHOD at a face velocity of 55 fpm and heating rate of 5°C/minute, reported to the nearest 5°C.

c. Elemental iodine penetration in 4 hours at 180°C.

New Test Development

The SRL radiolysis test was developed to provide a simulation of expected conditions in the SRP confinement system following a major reactor accident. Earlier experiments showed that the radiolytic desorption of iodine was independent of the dose rate above about 10^7 rad/hr.[10] Continued use of the test method for SRP carbon evaluation is thus dependent upon the availability of ^{60}Co of high enough specific activity to maintain a 10^7 rad/hr dose rate in the carbon test bed. In the absence of any planned reactor campaigns to produce or co-produce high specific radiocobalt, the radiolysis facility would soon have to be redesigned to achieve the required dose rate with the existing ^{60}Co. In lieu of an expensive redesign process to achieve temporary results (the ^{60}Co will continue to decay), an alternative test method has been devised.

The new test method will utilize ^{131}I-tagged methyl iodide in the test gas stream to simulate the mixture of organic iodides formed during the radiolytic desorption of iodine from carbon. The CH_3I injection rate can be adjusted to match the production rate of organic iodides in the test bed thus maintaining the principal desorption mechanism without relying on the intense radiation field to produce the iodine compounds. The same test apparatus can also be used to perform two of the three standard methyl iodide penetration tests specified in the RDT Standards.[18]

Construction of the new CH_3I test facility at SRL is complete, and preliminary equipment check-out is underway. The test apparatus is designed to perform over a controlled temperature range of 30°C to 80°C with controlled humidities up to 95% relative humidity.

CARBON DEVELOPMENT STUDIES

Joint efforts continued at SRL and Naval Research Laboratory (NRL) to develop new impregnant-carbon combinations for efficient trapping of methyl iodide in reactor off-gas cleanup systems. The first objective of these studies was to demonstrate that effective products could be manufactured from carbons derived from domestic raw materials. A second objective was the development of products matching the performance of triethylenediamine (TEDA) impregnated carbons without using TEDA in the impregnation formulation.

Coconut shell carbons are the preferred base materials for off-gas cleanup systems because of their unique combination of physical hardness and chemical reactivity for iodine compounds. However, excessive world demand or unfavorable international political situations could limit their availability. Advanced development of substitute carbon-impregnant combinations minimizes the risk of curtailment of reactor operations if alternative materials become necessary. Replacement of TEDA in impregnant formulations is desirable because the unfavorable combined effects of material cost, high vapor pressure and low flash point necessitate caution in both the manufacture and use of TEDA-impregnated carbons.

Earlier SRL studies showed that several domestic and imported petroleum- and coal-base carbons could be treated with combinations of iodine salts and hexamethylene-tetramine (HMTA) to produce acceptable trapping materials.[16,17] These studies also showed that incorporation of tribasic phosphate salts in the formulation was beneficial both as a flame retardant and as a buffer to maintain the high pH necessary for effective CH_3I trapping.[17]

During the current report period, efforts were directed toward finding the optimum combination of impregnants for each of the coal-base carbons. Companion samples containing TEDA were also prepared for two of the better coal carbons. Optimum impregnation formulations for four different coal-base carbons and one petroleum-base carbon are shown in Table 3. Test data for Type GX-176 carbon are shown for comparison.

Examination of the methyl iodide penetration data shows that all the laboratory prepared samples compare favorably with the commercial GX-176. TEDA impregnated samples of BPL* and 207A* show consistently lower CH_3I penetrations than their companion samples prepared with HMTA. As a result of these findings, further development of HMTA impregnated carbons was discontinued at SRL. Samples of HMTA and TEDA impregnated samples are currently being exposed to reactor building air in the SRP Carbon Test Facility (CTF) to obtain service aging data.

AgX RADIATION STABILITY TESTS

Silver exchanged zeolite (AgX) is currently used as a trapping medium for airborne radioiodine in chemical reprocessing plants and other facilities where highly oxidizing fumes in the off-gas streams prevent the use of carbon. It has also been proposed for use in reactors where high temperature gas streams could cause carbon ignition or impregnant desorption.

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* See Table 3.

TABLE 3

Optimum Impregnation Formulations for Several Base Carbons

Carbon Type[a]	Base Material	HMTA Content, wt. %	Iodine Salt Content,[b] wt %	Methyl Iodide Penetration[c], %	I_2 Penetration[d] at 180°C, %
GX-176[e]	Coconut[j]	1.0 (TEDA)	2.0 (KI)	0.45	2.0×10^{-3}
Witcarb 965[f]	Petroleum[k]	2.0	2.7	0.44	6.9×10^{-3}
MBV[g]	Coal[l]	2.0	2.7	0.44	21.0×10^{-3}
G-352[e]	Coal[j]	2.0	2.7	0.35	7.0×10^{-3}
BPL[h]	Coal[j]	1.0	2.7	0.37	0.9×10^{-3}
BPL[h]	Coal[j]	1.0 (TEDA)	2.7	0.10	1.0×10^{-3}
207A[i]	Coal[j]	2.0	2.7	0.21	1.8×10^{-3}
207A[i]	Coal[j]	1.0 (TEDA)	2.7	0.08	4.0×10^{-3}

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a. Manufacturers' designation.
b. Iodine salts = 2.18% KI and 0.56% KIO_3 unless otherwise noted. In addition to the iodine salts, 1.0% $NaH_2PO_4 \cdot H_2O$ was added with enough KOH to form the tribasic salt. The flame retardant used in GX-176 is proprietary information.
c. NRL test data. 2 in. deep beds, 25°C, 95% relative humidity and ∿0.1 mg CH_3I/g C loading.
d. SRL thermal desorption test. 1 in. deep beds, 4 hours desorption @ 180°C, 0.7 mg I_2/g C loading.
e. Products of North American Carbon Company, Columbus, Ohio.
f. Product of Witco Chemical Corp., New York, New York.
g. Product of Union Carbide Corporation, New York, New York.
h. Product of Calgon Corporation, Pittsburg, Pennsylvania.
i. Product of Sutcliff-Speakman & Co., Ltd., Leight, Lancashire, UK.
j. Sieved or ground and sieved to 10 x 16 mesh (US).
k. 8 x 16 mesh (US).
l. 6 x 14 mesh (US) material no longer supplied in this size range.

Because silver halide compounds are used as photosensitive agents in photographic and x-ray film emulsions, there are some reservations about the efficacy of AgX in these latter applications. The principal concern is the possible migration of iodine through the absorber beds under dynamic flow conditions in the high intensity radiation environment that could follow a reactor accident.

At the request of DOE, a series of tests of the radiation stability of AgX under dynamic flow conditions was conducted in the SRL radiolysis facility [10,11]. A single, long term exposure test was initially planned and executed. In this test, elemental iodine was loaded onto commercial AgX* over a 1 hour period in a high intensity radiation field. This was followed by an extended (104 hour) desorption period in the radiation field with continued air flow. Iodine desorbed from the irradiated test bed was collected on backup beds which were changed at regular intervals to evaluate specific desorption rates during the irradiation. The iodine loading on the test bed was about 6 mg I/g AgX on the 1-inch deep test bed. Air flow was maintained at 55 feet/minute face velocity and the absorbed dose rate in the test bed was $\sim 1.5 \times 10^7$ rad/hr. A steam-air mixture (80°C and ∿95% relative humidity) was employed during the loading phase and for the first 4 hour desorption period. Except for 2 four hour periods, the remainder of the test was run at 35°C and ambient humidity (ranging from 20% - 35% relative humidity at 35°C). Air at 45°C and 75% relative humidity was intentionally introduced for four hours after 45 hours exposure and 69 hours exposure to evaluate the effects of increased humidity on the desorption rate.

The specific test bed loading was chosen to be within the dynamic loading range of 1-10 mg I/g AgX found in the literature.[19-22] The one inch bed depth was dictated by the design of the radiolysis test facility, and the dose rate dictated by the specific activity of the ^{60}Co in the facility.

At the end of the radiation exposure period, the test bed was counted to determine the residual radioactive content, then carefully sectioned into ∿1/8 in. thick layers. Each layer was weighed and counted to determine the iodine distribution within the test bed.

---

* 10 x 20 mesh beads, 98^+ % exchanged. Purchased from CTI Nuclear, Inc., Denver, CO.

Results of the desorption phase of the test showed that no breakthrough of iodine occurred as a result of prolonged radiation exposure. The total penetration measured during the 105 hour test period was 0.743%. Measured desorption rates during the test are shown in Figure 3. Examination of the data shows that the specific desorption rates decreased with increasing accumulated exposure except for periods when high humidity air was intentionally introduced. The desorption rates during the last 25-30 hours of the test remained fairly constant at about 1% of the peak desorption rate observed during the initial loading phase.

Analysis of the distribution of activity in the test bed showed that about 37% of the iodine remained in the front 1/4 in. of the bed and that nearly 4% had penetrated to the last 1/8 in. segment of the test bed. The shape of the activity distribution curve (Figure 4) strongly suggests a "normal distribution" similar to that seen in ion exchange columns when an ionic species is moving through the resin beds. Numeric data are presented in Table 4.

This evidence of movement of activity in the test bed necessitated conducting additional tests in which shorter irradiation periods were employed in an attempt to determine rate constants. A bed sectioning experiment was also performed for GX-176 carbon for comparison (at a loading of 0.6 mg I/g C to stay within design loading limits).

The activity distributions in absorber beds after 5 hours exposure (1 hour loading, 4 hours desorption) are shown in Table 5 and Figure 5 for GX-176 carbon (0.6 mg/g loading), AgX at 0.6 mg/g loading and AgX at ∿6 mg/g loading. Since absorber weights and activity levels were not constant, it was necessary to normalize the data so direct comparisons could be made. The normalization was made using the function,

$$\frac{A_i/\Sigma A_i}{W_i/\Sigma W_i}$$

where: A_i = Activity in an individual segment, c/s

W_i = Weight of absorber in that segment, g

ΣA_i = Total activity in the test bed, c/s

ΣW_i = Total weight of absorber in the test bed, g

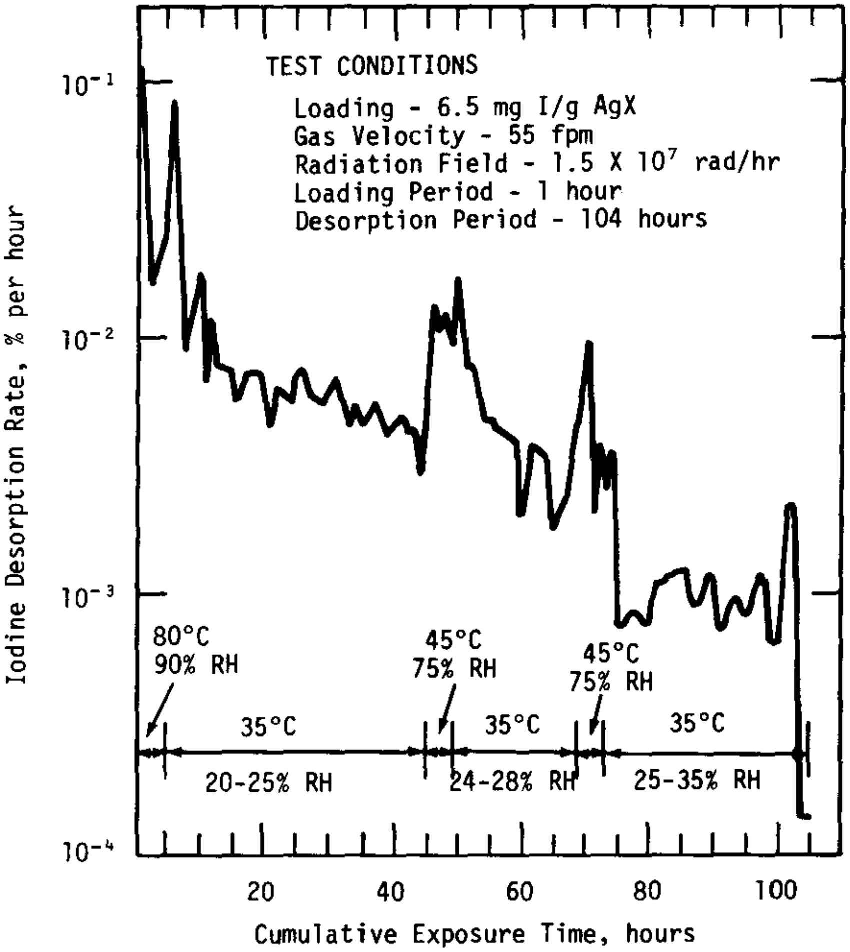

FIGURE 3. Iodine Desorption Rates From Irradiated AgX

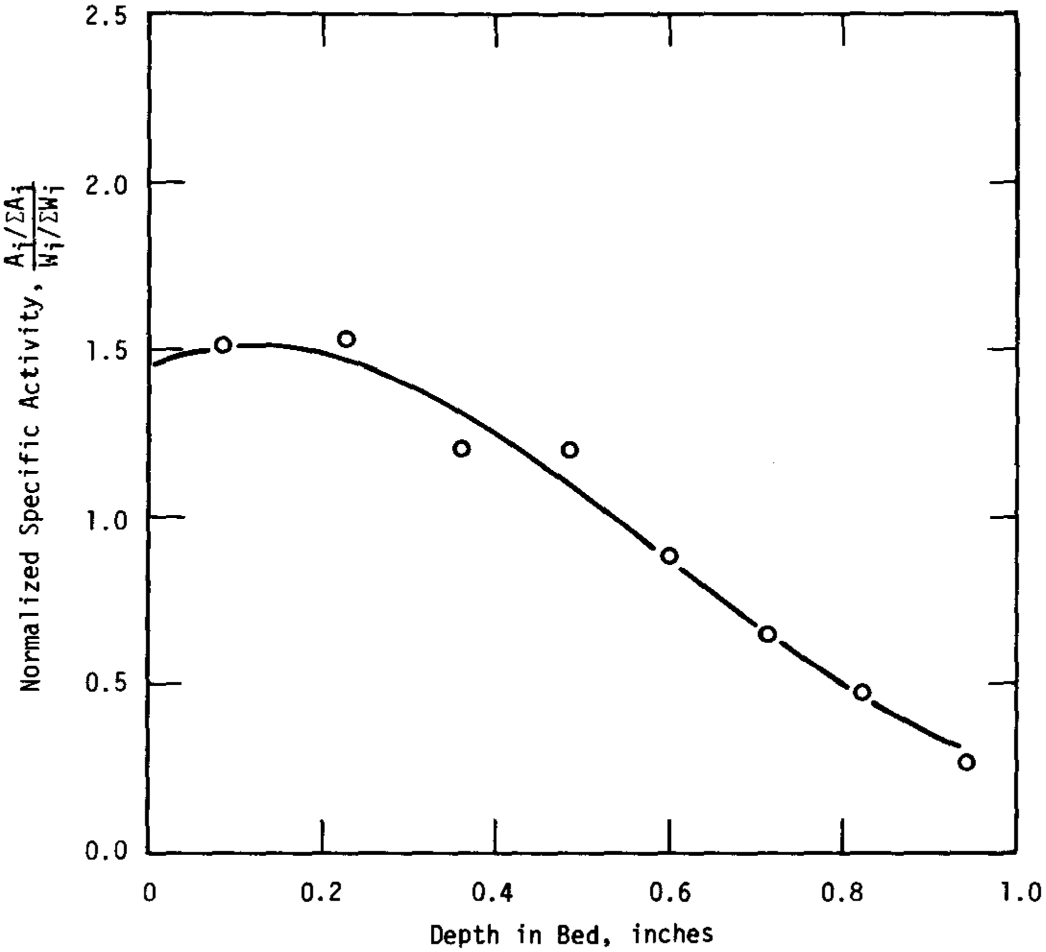

FIGURE 4. Activity Distribution in AgX Bed After 105 Hours Irradiation

TABLE 4

Radioactivity Distribution in AgX Bed After 105 Hours Exposure

Depth to Middle of Section, in.	*Normalized Specific Activity*[a], $\frac{A_i/\Sigma A_i}{W_i/\Sigma W_i}$
0.083	1.504
0.227	1.520
0.351	1.203
0.486	1.192
0.602	0.881
0.716	0.647
0.827	0.482
0.944	0.262

---

[a] Iodine Loading - 6.5 mg I/g AgX, see text for discussion of normalization function.

TABLE 5

Radioactivity Distribution in Test Beds After 5 Hours Exposure

GX-176 Carbon[a] *(0.65 mg I/g Carbon)*		*AgX*[b] *(0.68 mg I/g AgX)*		*AgX*[b] *(6.2 mg I/g AgX)*	
Depth, Inches[c]	*Normalized Activity*[d]	*Depth, Inches*[c]	*Normalized Activity*[d]	*Depth, Inches*[c]	*Normalized Activity*[d]
0.072	5.604	0.069	3.661	0.080	1.827
0.192	1.590	0.188	2.828	0.228	1.478
0.289	0.340	0.293	1.464	0.347	1.294
0.405	0.0571	0.422	0.314	0.457	1.076
0.543	0.0085	0.561	0.0364	0.589	0.711
0.671	0.0020	0.686	0.0044	0.719	0.573
0.778	0.0005	0.807	0.0007	0.826	0.467
0.914	0.0000	0.933	0.0002	0.938	0.257

---

[a] Product of North American Carbon Company, Columbus, Ohio.

[b] Type III, 10 x 20 mesh beads 98.$^{+}$% exchanged, product of CTI Nuclear Inc., Denver, Colorado.

[c] Depth to middle of bed section.

[d] $\frac{A_i/\Sigma A_i}{W_i/\Sigma W_i}$, see text for discussion.

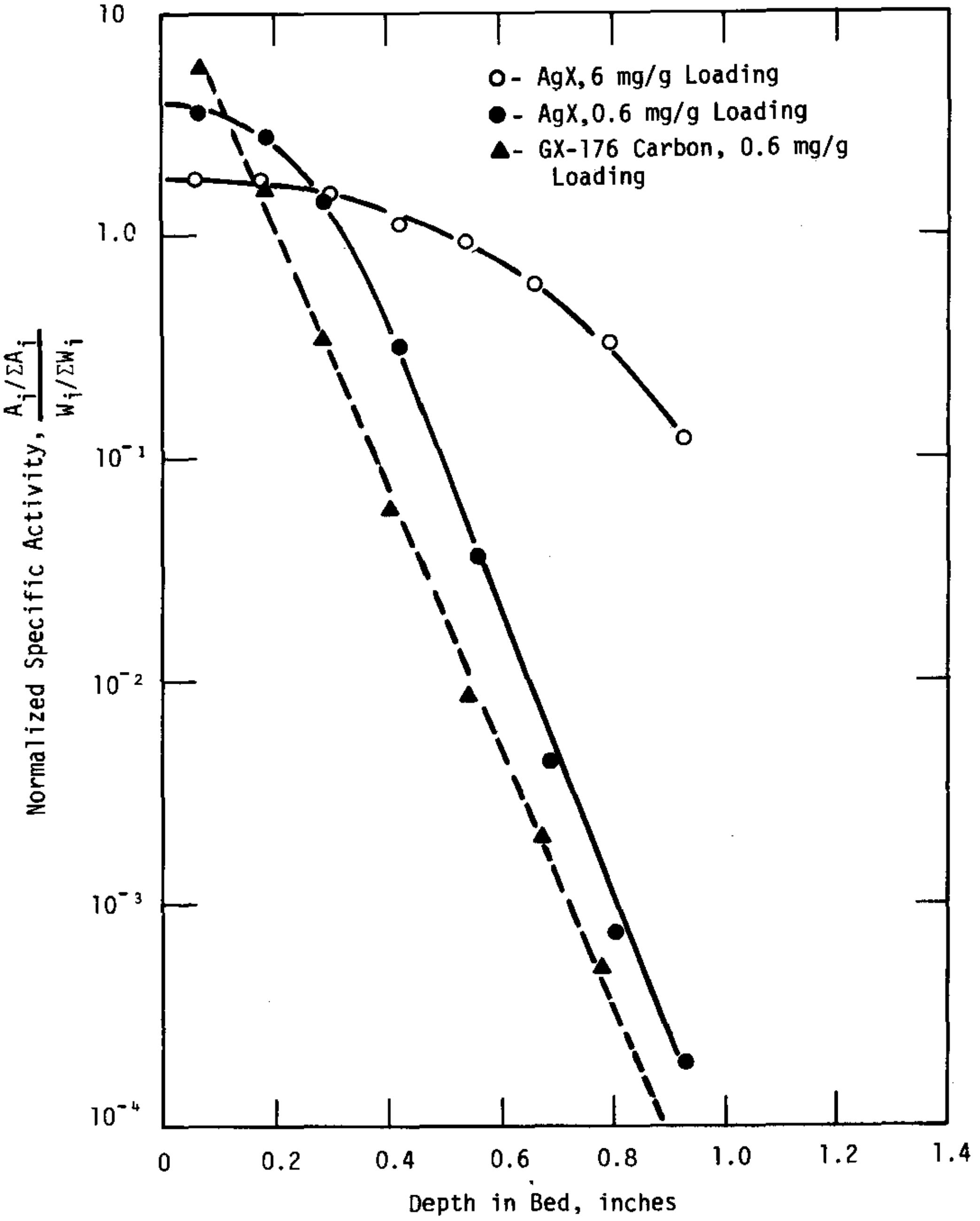

FIGURE 5. Iodine Distribution in Test Beds After 5 Hours Exposure

The data in Figure 5 show that the activity level in carbon decreases exponentially with increasing bed depth at 0.6 mg/g loading. At 0.6 mg/g loading, some penetration into the AgX bed is evident. These data suggest a somewhat slower reaction rate for airborne iodine removal in the AgX than is the case for carbon. At a loading of 6 mg/g, AgX exhibits considerable depth penetration. This deep penetration into AgX is consistent with recently published data which indicates a reaction zone of up to 5 cm depth when inlet concentrations of ∿500 mg I/m^3 air are encountered[23]. Thus, at least a portion of the activity found in the deeper layers of adsorber in the 105 hour irradiation test can be attributed to the high inlet iodine concentration and heavy loading.

In an effort to separate the heavy loading effects from the possible radiation effects, a third heavy loading experiment was performed in which the exposure time was reduced to 1 hour (the loading period only with no subsequent desorption). Data for the three heavy loading (∿6 mg/g) experiments are shown in Table 6 and Figure 6. The curves fitted to each set of data points were obtained by least squares methods assuming a normal distribution function (see Appendix A).

Examination of the curves in Figure 6 shows that some discernable movement of activity from the front toward the rear of the test beds has occurred between each of the exposure periods. The magnitude of the movement cannot be obtained directly from the data as presented because of differences in segment depths, uncertainties in the effect of slightly different loadings and the magnitude of the experimental error, and because penetration data cannot be expressed as specific activity of a bed segment.

Approximate numerical comparison can be made, however, if it is assumed that the equations obtained in the curve fitting process are reasonable approximations of the true iodine distributions. The equation for each curve can be integrated between the limits of X = 0 and X = 1 inch using the method shown in Appendix A. Corrections for activity desorbed from the bed can now be made as shown below.

$$\frac{\int_0^1 f(x)\,dx}{1 - P} = T$$

TABLE 6

Activity Distribution in Heavy Loaded AgX Beds After Irradiation

1 Hour Exposure (6.6 mg I/g AgX)		*5 Hour Exposure (6.2 mg I/g AgX)*		*105 Hour Exposure (6.5 mg I/g AgX)*	
Depth, Inches[a]	*Normalized Activity*[b]	*Depth, Inches*[a]	*Normalized Activity*[b]	*Depth, Inches*[a]	*Normalized Activity*[b]
0.061	1.831	0.080	1.827	0.083	1.504
0.179	1.797	0.228	1.478	0.227	1.520
0.303	1.524	0.347	1.294	0.361	1.203
0.426	1.103	0.457	1.076	0.486	1.192
0.540	0.911	0.589	0.711	0.602	0.881
0.665	0.592	0.719	0.573	0.716	0.647
0.797	0.326	0.826	0.467	0.827	0.482
0.932	0.119	0.938	0.257	0.944	0.262

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[a] Depth to mid-section.

[b] $\frac{A_i/\Sigma A_i}{W_i/\Sigma W_i}$, see text for discussion.

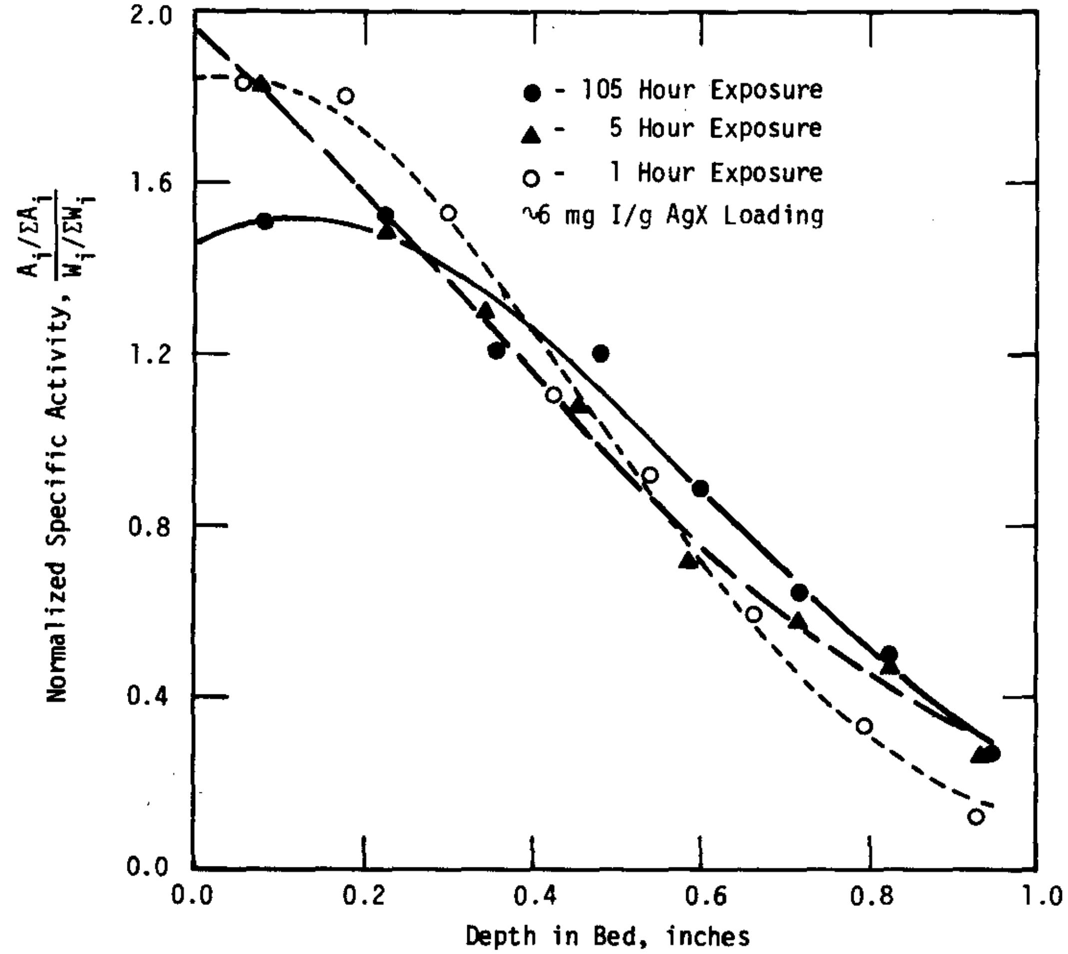

FIGURE 6. Activity Distribution in Irradiated AgX Beds

where $\int_0^1 f(x)\,dx$ = Total activity fraction on test bed

P = Total activity fraction passing through test bed (penetration)

T = Normalized total activity in test

Each equation can then be reintegrated between discrete limits and divided by T to obtain estimates of the normalized activity contained in equal depth segments of the bed. Calculated estimates of the activity contained in equal, 1/8 in. deep segments (corrected for penetration) are given in Table 7.

Numerical estimates of the movements of iodine within the test bed can be made by determining the intersection points for each pair of curves (1 hr vs. 5 hr, 1 hr vs. 105 hr, and 5 hr vs. 105 hr exposures), then integrating the area under each curve from the bed front to the intersection point. The difference in integrated area plus the difference in desorbed iodine for each pair of curves is then the estimate of the total iodine migration between each exposure time. Calculated data are shown in Table 8. The data indicate that between 1 hour and 105 hours exposure approximately 9.3% of iodine initially located in the first 0.41 in. of the bed moved to the rear of the bed. Another 0.6% of the iodine initially located in the rear of the bed moved out of the bed so that a total of about 9.9% of the total iodine migrated within or out of the 1 in. deep bed as a result of radiation exposure. Similar comparisons in the time interval from 1 hour to 5 hours exposure indicate a migration of about 5.4% of the iodine, and that approximately 6.2% of the iodine was redistributed between 5 hours and 105 hours exposure.

In the proposed reactor applications, AgX absorber beds of at least two inches depth would be used. Thus an internal redistribution of ∿10% of the iodine in the first inch of bed depth would not compromise the integrity of the absorber system.

TABLE 7

Estimated Iodine Content of Equal Test Bed Segments

Segment Depth, Inches	*Estimated Iodine Content of Each Segment, %*		
	1-Hour Exposure	*5-Hour Exposure*	*105-Hour Exposure*
0.000 - 0.125	22.98	22.86	18.44
0.125 - 0.250	21.70	19.89	18.45
0.250 - 0.375	18.57	16.61	17.13
0.375 - 0.500	14.40	13.32	14.76
0.500 - 0.625	10.12	10.25	11.81
0.625 - 0.750	6.45	7.58	8.76
0.750 - 0.875	3.73	5.38	6.04
0.875 - 1.000	1.95	3.66	3.86
Thru Bed	0.11	0.44	0.74

TABLE 8

Estimated Iodine Migration In Irradiated AgX Test Beds[a]

	Fraction of Total Iodine, %		
	Curve 1	*Curve 2*	*C(1)-C(2)*
1-Hour vs. 105-Hour			
Test Bed Iodine[b]	67.60	58.31	+9.29
Desorbed Iodine	0.11	0.74	-0.63
TOTAL MIGRATION	-	-	9.92
5-Hour vs. 105-Hour			
Test Bed Iodine[c]	45.39	39.51	+5.88
Desorbed Iodine	0.44	0.74	-0.30
TOTAL MIGRATION	-	-	6.18
1-Hour vs. 5-Hour			
Test Bed Iodine[d]	82.01	76.96	+5.05
Desorbed Iodine	0.11	0.44	-0.33
TOTAL MIGRATION	-	-	5.38

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[a] One inch deep beds loaded to ∿6 mg I/g AgX, superficial face velocity of 55 ft/min and an absorbed dose rate of ∿1.5×10^7 rad/hr.

[b] Fraction of iodine contained in the first 0.409 inches of the test bed.

[c] Fraction of iodine contained in the first 0.268 inches of the test bed.

[d] Fraction of iodine contained in the first 0.548 inches of the test bed.

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APPENDIX A

Curve Fitting Method

Depth distribution curves for the AgX test series were obtained by assuming that the general equations for the normal distribution curve best fit the experimental data

$$y = Ae^{-\frac{1}{2}\left(\frac{x-B}{C}\right)^2}$$

where y = normalized specific activity functions

x = depth in test bed, inches

A = constant representing maximum value of y

B = constant related to depth displacement in bed

C = constant related to spread in data

A computer code was written in which the observed values of x (XOBS) and y (YOBS) were used for each test. In the program, a value for A, B and C was assumed and a new value for Y (YCALC) was calculated for each XOBS value. The function

$\Delta^2 = \sum_{i=1}^{8} (YCALC_i - YOBS_i)^2$ was then evaluated. New values for B and C were then assumed and new y values calculated to obtain a new Δ^2 value. The process was repeated until the minimum value for Δ^2 was obtained for a constant value of A. The calculations were then repeated assuming a new value for A and an array of values for B and C. The equations obtained for each test represent the best fit found when values of A were incremented at intervals of 0.001 units and values of B and C were incremented at intervals of 0.0005 units. The "best fit" constants found in this manner are given in Table A.1.

TABLE A.1

Best Fit Constants for Depth Distribution Curves

Test Duration, hours	*Best Value Found For Constant*		
	A	*B*	*C*
1	1.850	0.0520	0.3970
5	2.200	-0.3030	0.6185
105	1.510	0.1260	0.4560

Integration of Curves

To obtain the area under each curve, it is necessary to evaluate the integral

$$\int dy = A \int_{0}^{1} e^{-\frac{1}{2}\left(\frac{x-B}{C}\right)^{2}} dx = \int_{0}^{1} f(x)\, dx$$

let $a = \frac{1}{2C^{2}}$ and $w = x - B$

then $dw = dx$

and $$\int_{0}^{1} f(x)\, dx = A \int_{-B}^{1-B} e^{-aw^{2}}\, dw$$

but $$\int_{0}^{Z} e^{-aw^{2}}\, dw = \frac{\sqrt{\pi}}{2\sqrt{a}} \left[\mathrm{erf}(Z\sqrt{a})\right]$$

where $\mathrm{erf}(Z\sqrt{a})$ = the error function of a number whose value is $(Z\sqrt{a})$

Therefore:

$$A \int_{-B}^{1-B} e^{-aw^{2}}\, dw = \frac{A\sqrt{\pi}}{2\sqrt{a}} \left\{\mathrm{erf}\left[(1-B)\sqrt{a}\right] + \mathrm{erf}\left[B\sqrt{a}\right]\right\}$$

substituting:

$$\int_{0}^{1} f(x)\, dx = \frac{A\sqrt{\pi}}{2\sqrt{\frac{1}{2C^{2}}}} \left\{\mathrm{erf}\left[(1-B)\sqrt{\frac{1}{2C^{2}}}\right] + \mathrm{erf}\left[B\sqrt{\frac{1}{2C^{2}}}\right]\right\}$$

(Equation 1):

$$\int_{0}^{1} f(x)\, dx = AC\sqrt{\frac{\pi}{2}} \left[\mathrm{erf}\left(\frac{1-B}{C\sqrt{2}}\right) + \mathrm{erf}\left(\frac{B}{C\sqrt{2}}\right)\right]$$

Equation 1 is the form used to integrate the curves for the 1-hr and 105-hr tests since both equations have positive (+) values for B (the apex of the curve occurs within the limits of $x = 0$ to $x = 1$). For the 5-hr test, as well as evaluation of sections of test beds between the limits of x and $x + \Delta x$, Equation 2 was used.

(Equation 2):

$$\int_{x}^{x+\Delta x} f(x) = AC \sqrt{\frac{\pi}{2}}\ \text{erf} \left[\left(\frac{x + \Delta x - B}{C\sqrt{2}} \right) - \text{erf} \left(\frac{X-B}{C\sqrt{2}} \right) \right]$$

For the 5-hr test, x = o and Δx = 1.0.

For the bed sectioning data, x = front boundary of section (0.250 inch) and Δx = depth of section (0.125 inch).

The calculated fraction of iodine in any bed segment (corrected for penetration) can be determined from Equation 3.

(Equation 3):

$$\frac{\displaystyle\int_{x}^{x+\Delta x} f(x)\ dx}{\displaystyle\int_{o}^{1} f(x)\ dx \Big/ (1 - \text{Penetration})}$$

Thus, to find the fraction of iodine in the last 1/8-in. deep segment of the test bed from the 1-hr test,

from Equation 2 and Table A.1:

$$\int_{0.875}^{1.000} f(x)\ dx = (1.850)\ (0.3970) \sqrt{\frac{\pi}{2}} \left[\text{erf} \left(\frac{1-0.0520}{0.3970\sqrt{2}} \right) - \text{erf} \left(\frac{0.875-0.052}{0.3970\ \sqrt{2}} \right) \right] = 0.9205\ [\text{erf}(1.6885)-\text{erf}(1.4659)]$$

$$= 0.01954$$

from Equation 1:

$$\int_{o}^{1} f(x)\ dx = (1.850)(0.3970) \sqrt{\frac{\pi}{2}} \left[\text{erf} \left(\frac{1-0.0520}{0.3970\sqrt{2}} \right) + \text{erf} \left(\frac{0.0520}{0.3970\sqrt{2}} \right) \right] = 0.9205\ [\text{erf}(1.6885) + \text{erf}(0.0926)]$$

$$= 1.0080$$

1 hour penetration = 0.10631%

$$\text{or } \frac{(0.01954)(1-0.00106)}{(1.0008)} = 0.01950 = 1.95\%$$

The complete list of calculated distribution values for all three tests is shown in Table 4 of the report.