DP-1271

AEC RESEARCH AND DEVELOPMENT REPORT

IODINE RETENTION STUDIES

PROGRESS REPORT

JULY - DECEMBER 1970

A. G. EVANS
L. R. JONES

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Savannah River Laboratory

Aiken, South Carolina

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Reactor Technology
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IODINE RETENTION STUDIES

PROGRESS REPORT: JULY - DECEMBER 1970

by

A. G. Evans

L. R. Jones

Approved by

J. M. Boswell, Research Manager
Reactor Engineering Division

July 1971

E. I. DU PONT DE NEMOURS & COMPANY
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CONTRACT AT(07-2)-1 WITH THE
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ABSTRACT

Deterioration of iodine retention efficiency of containment system carbon can be correlated with exposure time and nitrogen dioxide concentrations in the confinement system air stream. Attempts to duplicate the natural aging process in the laboratory with NO_2-air mixtures have been partially successful under carefully controlled conditions. Carbon moisture content, NO_2 exposure concentration, and the time lapse between artificial aging and carbon testing all influence the test results.

In continued methyl iodide radiolysis tests, a gamma radiation field of $\sim 1.7 \times 10^7$ rads/hr did not significantly extend the effectiveness of carbon for methyl iodide adsorption.

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LIST OF TABLES AND FIGURES

INTRODUCTION

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

Previous reports summarize the progress from January 1965 to June 1970.[1-5] This report summarizes studies at the Savannah River Laboratory from July to December 1970.

SUMMARY

Measured average concentration of nitrogen dioxide of 0.03 ppm confirms earlier measurements. Nonuniform mixing of reactor room air with other building air before the air enters the confinement system causes slightly higher NO_2 concentrations in some compartments with subsequent faster deterioration of iodine removal efficiencies for carbon in those compartments.

An attempt to develop a laboratory-scale accelerated aging method for carbon was partially successful with dry carbon and 300 ppm NO_2 in dry air. Laboratory-aged carbon shows decreased iodine retention as a function of NO_2 exposure, as predicted from extrapolated service aging curves; however, the accelerated aging effects are temporary and decrease with time after NO_2 exposure. Laboratory exposures of used carbon to NO_2 indicated increased sensitivity to NO_2 after service in the confinement system.

Methyl iodide penetration of type 416 unimpregnated carbon in a gamma radiation field of $\sim 1.7 \times 10^7$ rads/hr is independent of temperature in the range 40-80°C at constant relative humidity. Methyl iodide penetration increases with increasing service life of the carbon.

DISCUSSION

NITROGEN DIOXIDE IN REACTOR BUILDING AIR

Iodine retention efficiency of carbon in the Savannah River confinement system decreases with increasing service.[1-6] Analyses of used carbon samples suggest that nitrogen dioxide (NO_2) is the principal contaminant in reactor building air which causes deterioration of iodine retention efficiency.[3-6] Sulfur dioxide, ethylene, and trichloroethylene are also present on the used carbon.[3,6] Fossil fuel residues are present on other confinement system components.

Nitrogen dioxide is formed near the reactor as a result of radiolysis of air and is also present in varying concentrations in supply air for the building. The other contaminants result primarily from degreasing operations in the building or from combustion of fossil fuels for steam and power generation.

The concentration of NO_2 in confinement system air was reported earlier[3] as $\sim$0.03 ppm. More recent extensive surveys of NO_2 concentrations in and around the reactor buildings confirm this value, as shown in Table I. Reactor room air (the primary source of NO_2 in the building) enters the exhaust plenum nearer Compartments 2 and 3 while clearer air from other building areas enters the plenum nearer Compartment 6 (Figure 1).

TABLE I

Nitrogen Dioxide in Reactor Building Air

	Incoming Air	Reactor Room	Upstream of Carbon Filters	Exhaust Stack
Concentration Range, ppm	0.003-0.03	0.02-0.08	0.01-0.08	0.009-0.014
Average Concentration, ppm	0.01	0.03	0.03	0.01

TABLE II

NO_2 Concentrations in Individual Filter Compartments

Compartment number	2	3	4	5	6
Average Concentration, ppm	0.030	0.032	0.019	0.026	0.022

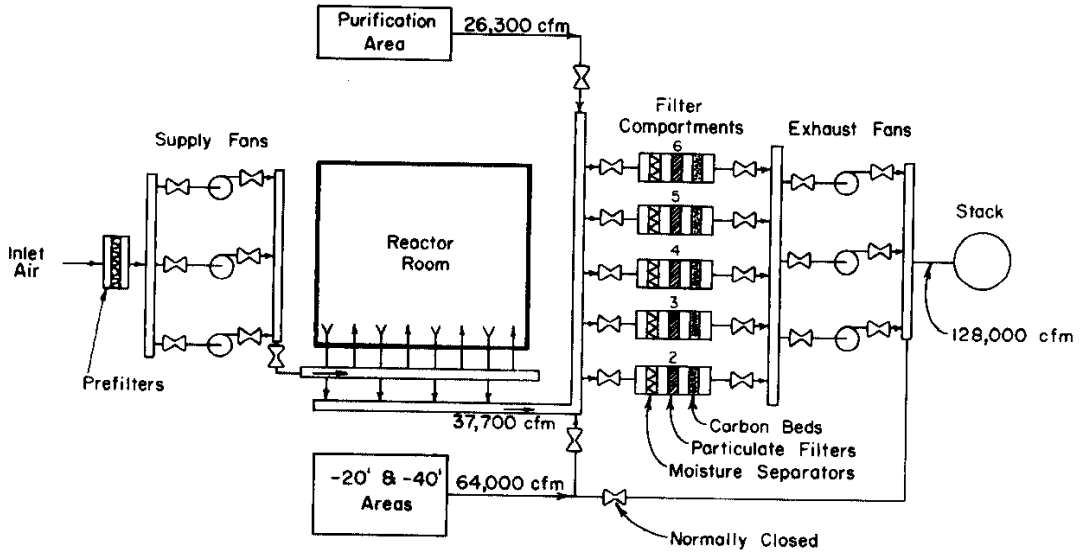

FIG. 1 FLOW DIAGRAM OF REACTOR VENTILATION SYSTEM

If NO_2 is the primary cause of deteriorating iodine retention efficiencies, the higher concentrations in Compartments 2 and 3 (Table II) should result in lower retention efficiencies in these compartments for equal service times. Comparison of iodine retention efficiency with service does show that most carbon samples taken from Compartments 2 and 3 have lower efficiencies than carbon from Compartments 4, 5, and 6 (Figure 2). The curve of efficiency versus service in Figure 2 is a least-squares fit of all test data.[5,7]

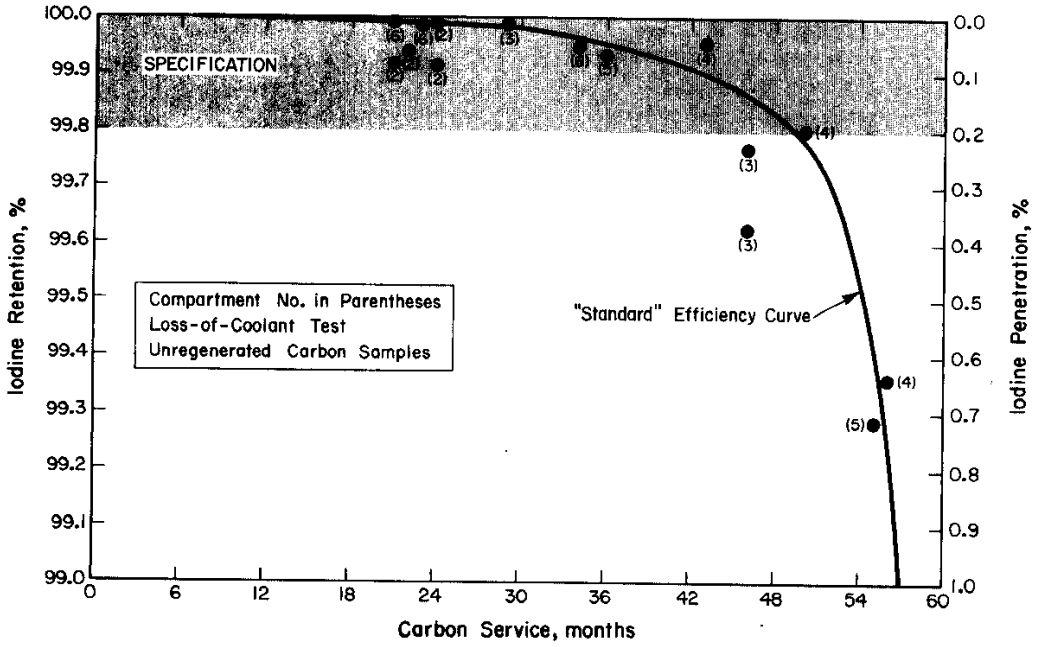

FIG. 2 IODINE RETENTION AS A FUNCTION OF SERVICE AND COMPARTMENT NUMBER

LABORATORY AGING WITH NO_2

A limited number of tests were reported earlier[3,6] which showed some correlation between carbon exposure to NO_2 and iodine retention efficiency. A more extensive series of experiments has been conducted during the current report period in an effort to develop an accelerated aging procedure for test purposes that can duplicate the deterioration of efficiency observed in service-aged carbons.

Unused carbon samples were exposed to NO_2 in air at several concentrations and under a variety of temperature and humidity conditions in an apparatus shown schematically in Figure 3. The samples were then tested for iodine retention efficiency by one of the two standard tests (loss-of-coolant or power surge test) described in a previous report.[5]

The accelerated aging method can, under carefully controlled conditions, reproduce transient iodine retention data similar to that obtained from service-aged carbon. A method has not yet been developed which can produce an artifically aged carbon similar in all characteristics to service-aged carbon.

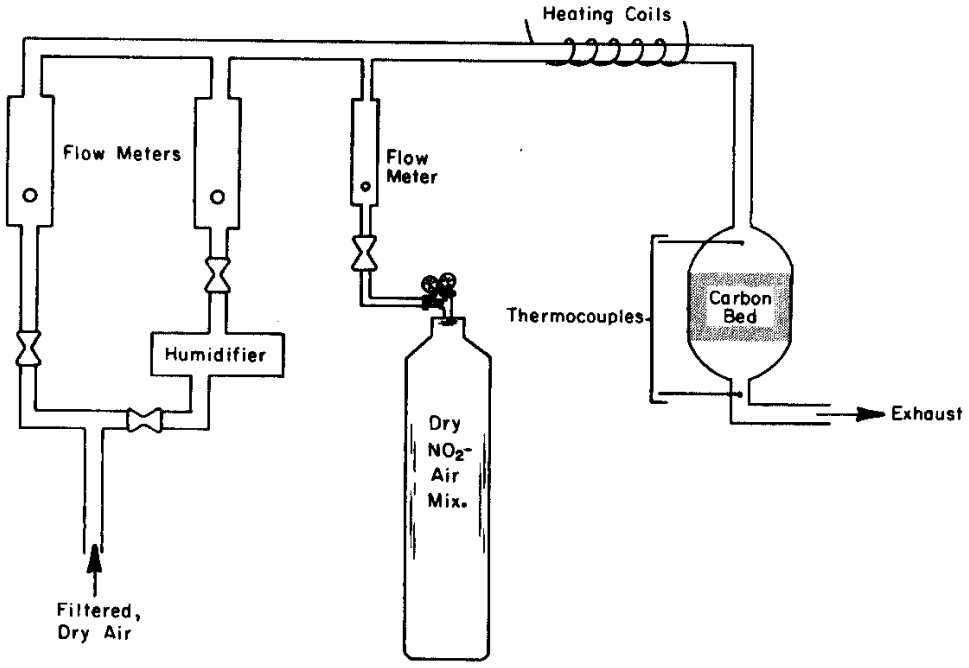

FIG. 3 NO_2 LOADING APPARATUS

In tests to develop an artificial againg method, newly purchased type 416* carbon was exposed to a dry NO_2-air mixture of 300 ppm NO_2. The 300 ppm concentration provides a concentration 10,000 times that in the confinement system and permits loading carbon with NO_2 in less than one day. Correlation of iodine penetration data with the standard efficiency was good (Figure 4). 2.53 mg NO_2/g carbon was assumed to be equivalent to one month of service (this number was calculated from air flow and carbon weight data from the confinement system with an average NO_2 concentration of 0.03 ppm).

Subsequent tests showed that changing NO_2 concentration, relative humidity of the loading air, and cure time (time lapse between NO_2 loading and iodine efficiency measurements) decreases the degree of correlation.

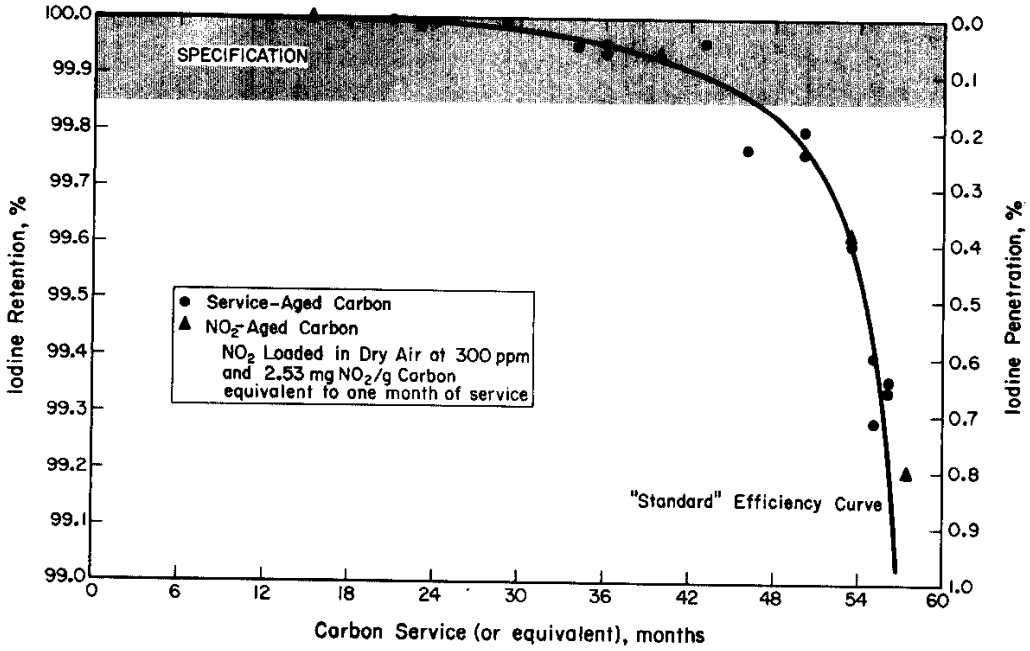

FIG. 4 NO_2 AGING OF UNUSED DRY CARBON

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

Effect of NO_2 Concentration

Reducing the loading concentration by a factor of 2 to ∿150 ppm NO_2 (with a subsequent increase in exposure time to maintain the same total mg/g exposure) resulted in iodine penetration values 3-10 times lower than predicted from the equivalent service exposure curve. When loading concentration was increased to 600 ppm NO_2, iodine penetration was 2 to 8 times greater than predicted. Typical data showing concentration effects are shown in Table III.

TABLE III

Effect of NO_2 Loading Concentration on Iodine Penetration

NO_2 Concentration, ppm	Total Exposure,[a] mg NO_2/g C	Iodine Penetration,[b] %	Predicted Penetration,[c] %
146	120	0.047	0.159
289	120	0.170	0.159
605	119	0.832	0.152

a. Total NO_2 passed through bed, not a measure of absorbed NO_2.
b. Measured ∿20 hours after NO_2 loading.
c. Based on standard efficiency curve[5] assuming 2.53 mg NO_2/g carbon equivalent to 1 month of service.

In an additional test the loading rate was held at 22 mg NO_2/min, but the face velocity was reduced to 26 ft/min from the normal velocity of 52 ft/min. The NO_2 concentration was increased to 600 ppm, from the normal 300 ppm. Iodine penetration was approximately 2 times greater than predicted from the standard curve indicating sensitivity to concentration and not loading rate.

Effect of Cure Time

Increasing the cure time (time lapse between NO_2 loading and iodine efficiency measurements) beyond 24 hours reduces iodine penetration, as shown in Table IV. In normal test sequence, the test bed and carbon are predried at 40°C for 45 minutes (inlet air temperature). The bed is then weighed and cooled in a sealed plastic bag. Once cooled, the bed is removed from the bag, loaded with NO_2, reweighed, and resealed in a plastic bag until the iodine efficiency test is run. For beds receiving low NO_2 exposures, the entire sequence, including the iodine efficiency test, is complete in a single 8-hour work period. The cure time is, thus, less than 2 hours under these conditions.

TABLE IV

Effect of Cure Time on Iodine Penetration of NO_2-Aged Carbon

Cure Time,[a] hours	Iodide Penetration, %	Predicted Penetration,[b] %	Concentration in Air, ppm NO_2	Total Loading, mg NO_2/g C
2	0.206	0.200	290	125
20	0.177	0.170	290	122
60	0.080	0.200	290	125
20	1.099	0.170	600	122
60	0.087	0.170	600	122
90	0.066	0.170	600	122

a. Time lapse between NO_2 loading and iodine penetration test.

b. Based on standard efficiency curve[5] assuming 2.53 mg NO_2/g carbon per month equivalent service.

When the test bed must be tested the next day, the cure time ranges from 16 to 24 hours. No significant efficiency variations were observed in this time span. When samples are loaded on Friday and the efficiency test run on Monday, the cure time is 60 to 70 hours. A significant decrease in iodine penetration was noted in every case where NO_2-loaded beds were stored over the weekend. With a 90 hour cure time (Table IV), the aging effects continued to diminish.

Effect of Moisture

Increasing the relative humidity of the carrier air to 50% during NO_2 loading also reduced the subsequent iodine penetration values by factors of 2 to 8 below predicted values, indicating that H_2O inhibits the reaction between NO_2 and carbon.

As stated earlier, the initial test series was performed on newly purchased, dry carbon. Attempts to obtain consistent data under the original test conditions failed after the carbon was exposed to laboratory air for a few weeks in an unsealed drum. Predrying the carbon sample with dry air at temperatures ranging from 23°C (ambient) to 60°C minimized the inconsistencies; predrying at 40°C produced the most consistent data. Weight loss measurements from the drying step indicated that moisture content variations (3.4% to 7.5%) were independent of drying temperature.

One of the aging mechanisms postulated[3,5,7] for service-aged carbon is the reaction:

$$NO_2 + C \rightarrow NO + CO$$

where each NO_2 molecule absorbed on the carbon bed destroys or occupies 3 active sorption sites (one site destroyed by oxidation and two sites occupied by the oxidation products which are sorbed on the carbon).

The mechanism in the accelerated aging method is a temporary blocking of sorption sites by the NO_2 without complete reaction with the carbon. NO_2 fumes evolved from artificially aged carbon beds while the beds were being stored in sealed plastic bags and while the beds were preheated before the power surge efficiency test. No NO_2 evolution was detected from service-aged carbon samples, even under greatly reduced pressure ($<10^{-6}$ mm Hg) and elevated temperatures (up to 300°C). Thus, it can be concluded that NO_2 reacts completely with service-aged carbon, but incompletely during the accelerated aging tests.

Effect of NO_2 on Service-Aged Carbon

Activated carbon undergoes a slow surface oxidation in a flowing air stream with the formation of incompletely oxidized carbon complexes with a general formula $C_n O_m$ (where n>m).[8] It can be hypothesized that each of these oxygen complex sites is activated for an oxidation reaction. Thus, when an NO_2 molecule is adsorbed at an activated site, the reaction

$$NO_2 + C_2O \rightarrow NO + 2\ CO$$

may take place, and a total of 5 carbon sorption sites may be removed from the surface (2 by oxidation and 3 by sorption of the oxidation products). In addition, the required activation energy would be reduced because the reaction site is partially oxidized. Carbon should thus become more sensitive to NO_2 with increasing age. Such an increased sensitivity is indicated in two tests in which service-aged carbon was treated with NO_2 in the laboratory. Results of iodine efficiency tests before and after NO_2 exposure are shown in Table V.

TABLE V

Effect of NO_2 Aging on Used Carbon

Before Artificial Aging			After Artificial Aging	
Carbon Service, months	Service Added with NO_2,[a] months	Initial Iodine Penetration, %	Expected Penetration,[b] %	Observed Penetration, %
21	12	0.004	0.032	1.176
39	12	0.039	0.255	2.497

a. Assuming 2.53 mg NO_2/g carbon per month, NO_2 loaded at 300 ppm in dry air at 52 ft/min velocity.

b. Based on standard efficiency curve for service-aged carbon.

Partial regeneration of the 21-month carbon at 60°C (as would be done during annual "Freon"* leak checking) followed by NO_2 exposure for an equivalent of 12 months service resulted in an iodine penetration of 0.034%, agreeing well with the predicted 0.032%. Partial regeneration apparently removes some of the oxide complexes and unblocks sorption sites by partially removing the oxidation products (CO and NO), thereby improving the iodine removal efficiency. Deactivation of oxide reaction sites also reduces sensitivity to NO_2. The partial regeneration of confinement compartment carbon during the annual "Freon" leak test may be beneficial, because the regeneration removes a portion of the oxide complexes and some of the oxidation products which cause deterioration of iodine removal efficiency.

* Registered trademark of Du Pont.

An Accelerated Aging Method

In summary, a method of accelerated aging of carbon was developed using NO_2 and dry air to simulate the natural aging process. Careful control of exposure conditions for the carbon sample must be maintained. The carbon must be dried before NO_2 exposure, loaded with NO_2 at ~300 ppm NO_2 in dry air, and tested for iodine efficiency within 24 hours after loading. The test carbon sample must be protected from elevated temperatures and rapidly flowing air streams to minimize desorption of NO_2 after loading.

The exposure procedure entails drying the carbon sample with air at 40°C (inlet temperature) for 45 minutes at a face velocity of 52 to 55 ft/min, and loading the NO_2 at ~300 ppm and 55 ft/min face velocity in dry air at ambient temperature. Measurement of iodine retention efficiency must then be completed within 24 hours. Results of tests under these conditions are shown in Figure 5. Total NO_2 exposure was based on the 2.53 mg NO_2/g C/month average exposure calculated for service-aged carbon.

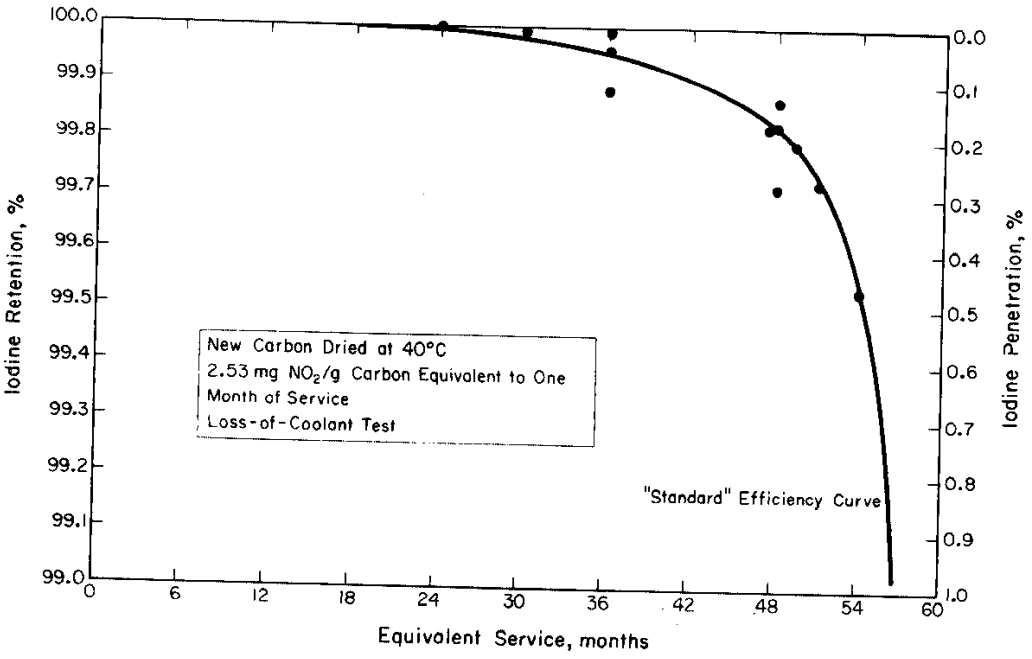

FIG. 5 NO_2 AGING OF DRIED CARBON AT 300 ppm NO_2

As stated earlier, the method described above only produces temporary aging effects. Future work will be directed toward "fixing" the aging effects. One method may be stimulation of the oxidation reaction by incorporating ozone or SO_2 in the loading stream with the NO_2 (both these contaminants are found in confinement compartment air). More careful control of loading temperature may be required with other additives, however, to remove the excess reaction heat and prevent ignition of the carbon sample.

METHYL IODIDE RADIOLYSIS TESTS

The effects of gamma radiation on the adsorption of iodine and methyl iodide on activated carbon are being measured as part of a continuing program,[1-5] in support of the reactor confinement facilities at Savannah River.

The methyl iodide radiolysis apparatus, operating techniques, and general test conditions were described previously.[5,9] Results of tests to determine the effects of face velocity, relative humidity, methyl iodide loading on the carbon, and methyl iodide concentration were also reported. This report presents results of continued tests to determine effects of temperature, carbon service age, and type of carbon on iodine retention efficiency.

Test conditions, unless otherwise specified, were:

- Relative humidity of 50%
- Face velocity of 55 ft/min
- CH_3I loading on carbon bed of 0.1 mg CH_3I/g carbon
- CH_3I concentration in steam-air mixture of 2.4×10^3 $\mu g/m^3$
- Steam-air mixture temperature of 65°C

Effect of Temperature

The effect of steam-air mixture temperature from 40 to 80°C on iodine retention is shown in Figure 6. Because the data indicated that temperature had little effect on CH_3I retention efficiency, no similar tests were run outside the gamma field. Retention of methyl iodide is affected by relative humidity[5] but not by absolute humidity or temperature within the range investigated.

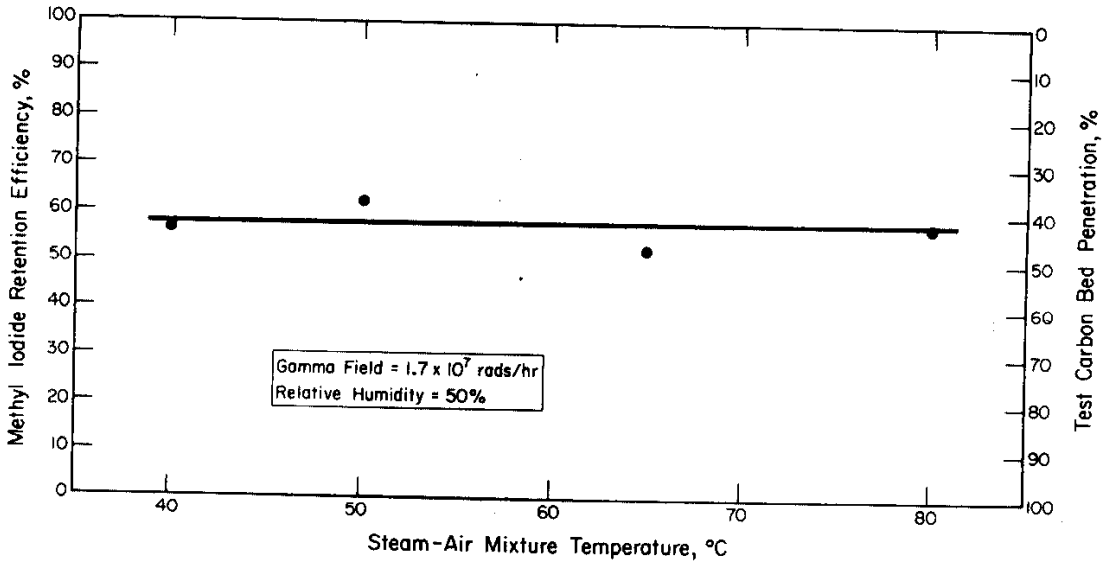

FIG. 6 CH_3I EFFICIENCY AS A FUNCTION OF STEAM-AIR TEMPERATURE

Effect of Carbon Service Age

Results of tests in the radiation field using carbon with varying service in confinement facilities are shown in Figure 7. The data indicate decreased CH_3I retention efficiency with increased carbon service age. A corresponding decrease in elemental iodide retention efficiency for used carbon measured out of the radiation field has been previously reported.[1-5]

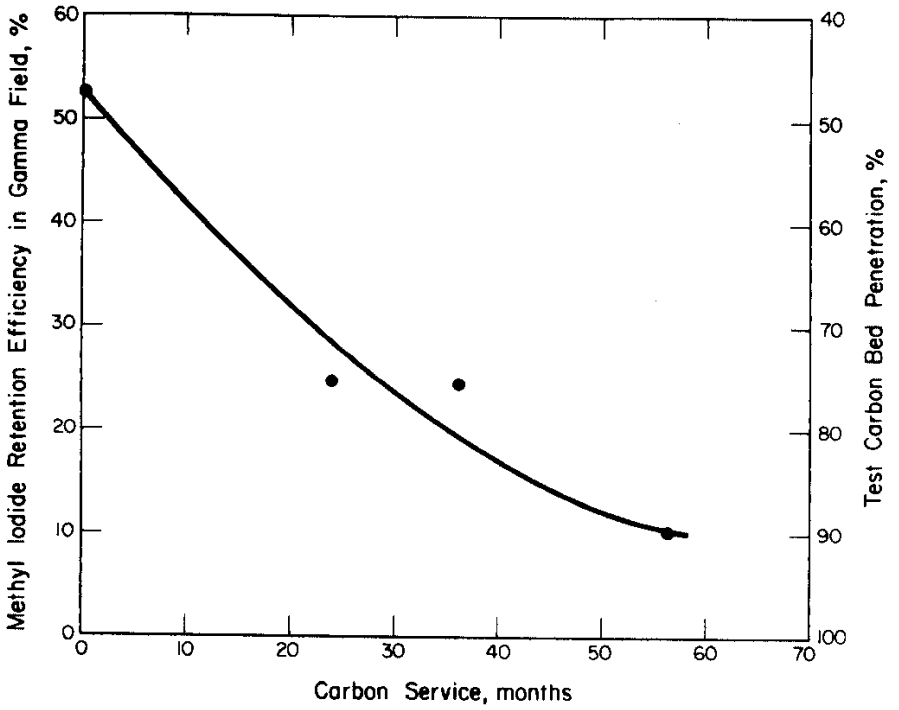

FIG. 7 CH_3I EFFICIENCY OF CARBON WITH SERVICE IN THE CONFINEMENT SYSTEM

Effect of Carbon Type

Type 727* KI-impregnated carbon was tested; results are compared with similar test results for type 416* unimpregnated carbon in Table VI. The data indicate little effect of the gamma field on the efficiency of type 727 carbon within the limits of experimental variation for the test conditions investigated. No effect of radiation on isotopic exchange between the KI impregnant and CH_3I would be expected. Additional tests with other types of carbon and other test conditions are planned.

TABLE VI

Efficiencies of KI-Impregnated and Unimpregnated Carbon

Carbon Type	Gamma Field, rads/hr	CH_3I Retention Efficiency, %
Impregnated* (BC-727)	1.7×10^7	98.41
		98.13
		97.51
	0	99.10
		98.28
Unimpregnated (BC-416)	1.7×10^7	65.75
	0	58.58

* Impregnated with KI_3.

* Products of Barnebey-Cheney Company, Columbus, Ohio

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