662905

DP-1340

CONFINEMENT OF AIRBORNE RADIOACTIVITY

PROGRESS REPORT: JANUARY - JUNE 1973

A. G. EVANS

DU PONT
REG. U.S. PAT. OFF.

Savannah River Laboratory

Aiken, South Carolina

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION 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 22151

Price: Printed Copy $4.00; Microfiche $1.45

DP-1340
General Reactor Technology
(TID-4500-R60, UC-80)

CONFINEMENT OF AIRBORNE RADIOACTIVITY

PROGRESS REPORT: JANUARY - JUNE 1973

by

A. G. Evans

Approved by

J. M. Boswell, Research Manager
Reactor Engineering Division

Publication Date: October 1973

E. I. du Pont de Nemours & Co.
Savannah River Laboratory
Aiken, S. C. 29801

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

ABSTRACT

Radiolytic desorption tests of a series of carbon samples containing KI_3, KI, triethylenediamine (TEDA), or combinations of TEDA and KI showed that a 10 x 16 mesh coconut shell carbon containing 1% TEDA, 2% KI,and a proprietary flame retardant retained iodine better than all products tested except those containing higher TEDA concentrations.

Ignition temperature measurements on the same series of samples showed that the 1% TEDA - 2% KI product has an ignition temperature of about 330°C under SRP flow conditions (the same as the KI_3 carbon). The 1% TEDA - 2% KI carbon has been selected to replace the unimpregnated carbon currently used in the Savannah River reactor confinement system.

CONTENTS

LIST OF TABLES AND FIGURES

INTRODUCTION

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

Previous confinement system studies at Savannah River have shown that elemental iodine retention on activated carbon is most strongly influenced by the operating temperature of the carbon beds, the moisture content of the air passing through the beds, the length of time the carbon has been in service, and the radiation exposure to which the beds are subjected after iodine loading.[2-11] Radiation exposure in the carbon beds would result from adsorption and subsequent decay of radioiodine following a reactor accident. Radiolytic desorption of iodine results from free radical reactions in which organic iodides are formed and desorbed from the carbon. The controlling parameters are radiation intensity, radiation duration, carbon type, carbon service history, composition and relative humidity of the purge gas stream,[9,10] and particle size distribution of carbon granules.[11]

Impregnated carbons are more effective in retaining iodine in a radiation environment than the unimpregnated carbon currently used in the confinement system. Carbons impregnated with triethylenediamine (TEDA) or coimpregnated with TEDA-KI were shown in previous studies to be the most effective adsorbers for the radiolytically formed iodine species.[12] Carbons containing 2 to 5% TEDA were found, however, to have reduced ignition temperatures because of the low flash point and relatively high heat of combustion of the TEDA.[12]

RADIOLYTIC DESORPTION TESTS

Previous studies of the radiolytic desorption phenomenon showed that unimpregnated carbons, such as Type 416* carbon currently used in the Savannah River confinement systems, do not retain iodine as well as impregnated carbons when exposed to an

* Product of Barnebey-Cheney Company, Columbus, Ohio

intense gamma radiation field.[9-12] As part of the continuing program to evaluate and improve the capabilities of the confinement system, pilot quantities of two types of triethylenediamine-impregnated carbons were installed in 2 compartments of the confinement systems, and samples were removed after 6 and 9 months weathering (service aging). Radiolytic desorption tests performed on these samples show that a TEDA-impregnated carbon retains iodine better after 9 months service than a new unimpregnated carbon as shown in Table I.

TABLE I

Effect of Service Aging on a TEDA Carbon

Carbon Type	*Service Age,[a] months*	*Iodine Penetration,[b] %*
416 (Control)	0	0.37
TEDA[c]	0	0.034
TEDA[c]	6	0.052
TEDA[c]	9	0.120

a. Weathered in C Area Compartment 2.
b. Penetration during a "standard" 5-hour test (1 hour loading, 4 hours desorption) at 80°C, 100% relative humidity in a radiation field of 3×10^7 rad/hr.
c. Carbon coimpregnated with 2% TEDA, 2% KI.

Similar aging data for Type 416 carbon are shown in Table II. The aging data are compared graphically in Figure 1.

TABLE II

Effect of Service Aging on Unimpregnated Carbon

Service Age, months	*Iodine Penetration,[e] %*
0	0.37
21[a]	0.61
33[b]	1.04
35[c]	3.61
46[d]	4.38

a. Service aged in Compartment C-2.
b. Service aged in Compartment P-2.
c. Service aged in Compartment P-3.
d. Service aged in Compartment K-3.
e. See Table I, note b.

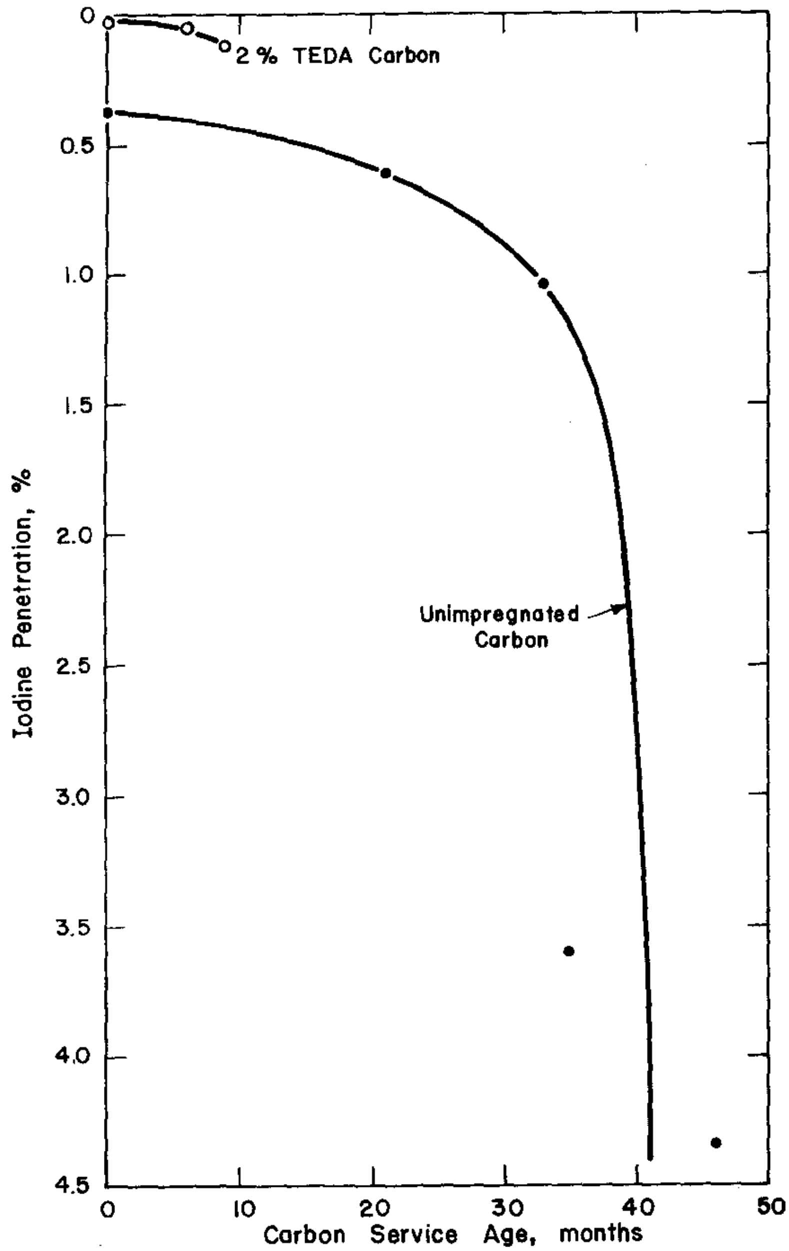

FIGURE 1. Effect of Service Aging on Two Carbons

Radiation Tests included 1-hr loading and 4-hr desorption at 80°C and 100% relative humidity in a radiation field of 3×10^7 rad/hr.

The relatively high initial iodine penetration of Type 416 carbon and the rapid rate of increase in the penetration rate beyond about 2 years service indicate that system performance might be enhanced by installation of an impregnated carbon in the Savannah River confinement system. Although the TEDA carbons were shown to be the most effective iodine sorbers in earlier tests,[9-11] potentially reduced ignition temperatures caused by TEDA combustion[12] has delayed full-scale replacement.

The earlier studies on the effect of TEDA impregnation on carbon ignition temperatures suggested that lower TEDA content carbons could be made which would reduce the ignition hazard.[12] Very little difference in iodine retention efficiency was noted in the earlier studies when the TEDA content was lowered from 5% to 2%.[12]

As a result of these earlier findings, several carbons with lower TEDA content were prepared to determine the optimum impregnation level for the Savannah River system. Two series of special carbons were prepared by North American Carbon Company.

The first series consisted of seven different impregnation combinations (plus a blank) prepared from the same lot of 8 x 16 mesh coconut shell carbon. Three samples contained TEDA (2%, 1%, and 0.5% by weight) in combination with a proprietary flame retardant. Three additional samples contained TEDA (again 2%, 1%, and 0.5% by weight) in combination with 2% KI and the flame retardant. The seventh sample contained KI and flame retardant only.

The second series consisted of three samples (plus a blank) prepared from the same lot of a narrower particle size range (10 x 16 mesh) coconut shell carbon containing TEDA (2%, 1% and 0.5% by weight) and 2% KI (plus the flame retardant). One additional sample, a 5% KI_3 carbon (10 x 16 mesh) prepared from a higher surface area coconut carbon was also included in the second series.

Replicate samples of each type of carbon were subjected to the standard 5-hour radiation test (1 hour loading and 4 hours desorption at 80°C and 100% relative humidity at an absorbed dose rate of $\sim 3 \times 10^7$ rad/hr) in the iodine radiolysis facility (Table III).

TABLE III

Iodine Penetration of Special Carbons

	Iodine Penetration, %[a]			
TEDA, wt %	*8 x 16 Mesh No KI*[b]	*8 x 16 Mesh + 2% KI*[b]	*10 x 16 Mesh*	*10 x 16 Mesh + 2% KI*[b]
0.0 (Controls)	0.27	-	0.29(No KI)	-
0.0	-	0.035	0.025(5% KI_3)	-
0.5	0.034	0.025	-	0.019
1.0	0.030	0.020	-	0.017
2.0	0.025	0.017	-	0.015

a. See Table I, note b.
b. All samples except controls and KI_3 carbon also contained a proprietary flame retardant.

The iodine penetration data in Table III indicate that carbons coimpregnated with TEDA-KI perform better than carbons containing either TEDA or KI alone. Also, TEDA-KI coimpregnation is better than KI_3 impregnation, and the 10 x 16 mesh particle size distribution is better than the 8 x 16 mesh distribution. The results show a consistent pattern of increasing iodine retention with increasing TEDA content.

IGNITION TEMPERATURE TESTS

All the special carbon samples were tested for ignition temperature in both the stainless steel apparatus (described in a previous report[12]) and in a standard quartz apparatus* (Table IV). Ignition temperatures were measured at two face velocities, 55 ft/min and 10 ft/min. The higher face velocity (55 ft/min) is the normal operating face velocity of carbon beds in the Savannah River confinement system. The lower velocity (10 ft/min) represents an extreme in minimum flow under conditions of severe HEPA filter blinding (either from smoke blinding or degradation of water repellency).

* Standard quartz apparatus as defined in the proposed ASTM D-28/IV Standard method.

TABLE IV

TEDA Carbon Ignition Temperatures[a]

TEDA Content, %	*8 x 16 Mesh Carbon*		*10 x 16 Mesh Carbon*	
	Stainless Steel,[b] °C	*Quartz,[c] °C*	*Stainless Steel, °C*	*Quartz,[c] °C*
55 ft/min face velocity				
0.0 (5% KI_3 carbon)	-	-	-	333
0.0 (Control)	340	320	330	325
0.5	335	350	320	340
1.0	330	335	315	330
2.0	300	310	300	320
10 ft/min face velocity				
0.0 (5% KI_3 carbon)	-	-	-	312
0.0 (Control)	330	315	320	300
0.5	320	340	310	325
1.0	300	320	300	315
2.0	265	280	260	270

a. Heating rate = 5°C/min.
b. Stainless steel apparatus is described in Reference 12.
c. Standard quartz apparatus is defined in the proposed ASTM D-28/IV Standard method.

Consistent patterns of lower ignition temperatures with increasing TEDA content are observed in the stainless steel apparatus. In the quartz apparatus, the unimpregnated base carbon ignites at a consistently lower temperature than impregnated carbons containing 0.5% and 1% TEDA probably because of the presence of the proprietary flame retardant in the impregnated carbon. The additional heat generation in the 2% TEDA carbons is apparently sufficient to overcome the suppressive effect of the flame retardant. In the stainless steel apparatus, combustion (particularly of impregnated carbons) is apparently partially catalyzed by metal oxides present on the steel, thus accounting for slightly lower ignition temperatures when compared to the quartz apparatus.

SELECTION OF A NEW CARBON FOR THE CONFINEMENT SYSTEM

Selection of a TEDA-impregnated replacement carbon for the Savannah River confinement system requires a compromise between iodine retention and minimum ignition potential. The iodine penetration data in Table III show that satisfactory iodine retention can be obtained with 1% TEDA and 2% KI impregnation on 10 x 16 mesh coconut carbon. The data show this carbon to be better than carbons containing either KI alone or KI_3 alone. The ignition temperature data in Table IV show that the 1% TEDA-2% KI carbon (10 x 16 mesh) is consistently better than carbons containing 2% TEDA (has a higher ignition temperature) and is about equal to the 5% KI_3 carbon. Thus, installation of the 1% TEDA product involves no greater ignition risk than installation of the 5% KI_3 product and offers slightly better iodine retention.

The iodine adsorber selected for future use in the Savannah River confinement system is a product containing 1% TEDA and 2% KI (with a flame retardant) on a 10 x 16 mesh coconut shell carbon. (The product has a North American Carbon Company designation GX-176.) Replacement of Type 416 carbon with GX-176 is scheduled to begin in the fall of 1973.

Service aging characteristics of the new carbon will be determined by frequent sampling of the confinement system carbon. Service aging data on the replacement carbon and other potentially usable carbons (0.5% TEDA, 2% TEDA, and 5% KI_3) will also be obtained from samples to be installed in the Carbon Weathering Facility (see description in Reference 10).

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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (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, S. C. (1972).

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