

AD-A014 404

LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE
AND IODINE COMPOUNDS ON ACTIVATED CARBON

R. Schwarzbach

Foreign Technology Division
Wright-Patterson Air Force Base, Ohio

31 July 1975

DISTRIBUTED BY:

NTIS

National Technical Information Service
U. S. DEPARTMENT OF COMMERCE

KEEP UP TO DATE

Between the time you ordered this report—which is only one of the hundreds of thousands in the NTIS information collection available to you—and the time you are reading this message, several *new* reports relevant to your interests probably have entered the collection.

Subscribe to the **Weekly Government Abstracts** series that will bring you summaries of new reports as soon as they are received by NTIS from the originators of the research. The WGA's are an NTIS weekly newsletter service covering the most recent research findings in 25 areas of industrial, technological, and sociological interest—invaluable information for executives and professionals who must keep up to date.

The executive and professional information service provided by NTIS in the **Weekly Government Abstracts** newsletters will give you thorough and comprehensive coverage of government-conducted or sponsored re-

search activities. And you'll get this important information within two weeks of the time it's released by originating agencies.

WGA newsletters are computer produced and electronically photocomposed to slash the time gap between the release of a report and its availability. You can learn about technical innovations immediately—and use them in the most meaningful and productive ways possible for your organization. Please request NTIS-PR-205/PCW for more information.

The weekly newsletter series will keep you current. But *learn what you have missed in the past* by ordering a computer **NTISearch** of all the research reports in your area of interest, dating as far back as 1964, if you wish. Please request NTIS-PR-186/PCN for more information.

WRITE: Managing Editor
5285 Port Royal Road
Springfield, VA 22161

Keep Up To Date With SRIM

SRIM (Selected Research in Microfiche) provides you with regular, automatic distribution of the complete texts of NTIS research reports *only* in the subject areas you select. SRIM covers almost all Government research reports by subject area and/or the originating Federal or local government agency. You may subscribe by any category or subcategory of our WGA (**Weekly Government Abstracts**) or **Government Reports Announcements and Index** categories, or to the reports issued by a particular agency such as the Department of Defense, Federal Energy Administration, or Environmental Protection Agency. Other options that will give you greater selectivity are available on request.

The cost of SRIM service is only 45¢ domestic (60¢ foreign) for each complete

microfiched report. Your SRIM service begins as soon as your order is received and processed and you will receive biweekly shipments thereafter. If you wish, your service will be backdated to furnish you microfiche of reports issued earlier.

Because of contractual arrangements with several Special Technology Groups, not all NTIS reports are distributed in the SRIM program. You will receive a notice in your microfiche shipments identifying the exceptionally priced reports not available through SRIM.

A deposit account with NTIS is required before this service can be initiated. If you have specific questions concerning this service, please call (703) 451-1558, or write NTIS, attention SRIM Product Manager.

This information product distributed by

NTIS

U.S. DEPARTMENT OF COMMERCE
National Technical Information Service
5285 Port Royal Road
Springfield, Virginia 22161

258116

①
de

FOREIGN TECHNOLOGY DIVISION

AD A014404



LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE
AND IODINE COMPOUNDS ON ACTIVATED CARBON

by

R. Schwarzbach



DDC
RECORDED
SEP 15 1975
INDEXED
D

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
US Department of Commerce
Springfield, VA. 22151

Approved for public release;
distribution unlimited.



UNCLASSIFIED

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Foreign Technology Division Air Force Systems Command U. S. Air Force	2a. REPORT SECURITY CLASSIFICATION UNCLASSIFIED 2b. GROUP
---	---

3. REPORT TITLE
LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE AND IODINE COMPOUNDS ON ACTIVATED CARBON

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)
Translation

5. AUTHOR(S) (First name, middle initial, last name)
R. Schwarzbach

6. REPORT DATE 1972	7a. TOTAL NO. OF PAGES 11	7b. NO. OF REFS 14
------------------------	------------------------------	-----------------------

8a. CONTRACT OR GRANT NO. b. PROJECT NO. c. d.	9a. ORIGINATOR'S REPORT NUMBER(S) FTD-ID(RS)I-1783-75 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
---	---

10. DISTRIBUTION STATEMENT
Approved for public release; Distribution unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Foreign Technology Division Wright-Patterson AFB, Ohio
-------------------------	---

13. ABSTRACT
07

PRICES SUBJECT TO CHANGE

EDITED TRANSLATION

FTD-ID(RS)I-1783-75 31 July 1975

FTD-75-C-002137
 LABORATORY STUDIES ON THE ADSORPTION OF RADIO-
 IODINE AND IODINE COMPOUNDS ON ACTIVATED CARBON

By: R. Schwarzbach

English pages: 11

Source: Staatliche Zentrale Fuer Strahlenschutz
 SZS-142, 1972, pp. 20-28.

Country of origin: East Germany

Translated by SCITRAN

F33657-72-D-0853

Requester: PDTR

Approved for public release; Distribution
 unlimited.

THIS TRANSLATION IS A RENDITION OF THE ORIGINAL FOREIGN TEXT WITHOUT ANY ANALYTICAL OR EDITORIAL COMMENT. STATEMENTS OR THEORIES ADVOCATED OR IMPLIED ARE THOSE OF THE SOURCE AND DO NOT NECESSARILY REFLECT THE POSITION OR OPINION OF THE FOREIGN TECHNOLOGY DIVISION.

PREPARED BY:

TRANSLATION DIVISION
 FOREIGN TECHNOLOGY DIVISION
 WP-AFB, OHIO.

LABORATORY STUDIES ON THE ADSORPTION OF RADIOIODINE AND IODINE COMPOUNDS ON ACTIVATED CARBON

R. Schwarzbach*

Activated carbons produced in the German Democratic Republic are tested with regard to their capability to adsorb radioiodine and radioactive methyl iodide. The physico-chemical properties of the available types of activated carbon are summarized. Activated carbons with most favorable properties are selected, considering different parameters. It is attempted to improve the adsorptive properties of the selected carbons by preparing them with $\text{Cu}(\text{NO}_3)_2$, AgNO_3 or KI . Furthermore, the influence of water vapor, temperature and carbon dioxide on the effectiveness of the carbon was tested. Methods and results of the tests are presented.

1. Statement of the Problem

With the establishment of many nuclear technology stations, the importance of protecting the environment from the output of radioactive materials into the atmosphere is growing. The problematic of the behavior of radioiodine, its radiotoxicity, the nature of its elimination, especially during damage situations, make it the object of many research programs and studies. [1-5]

Iodine appears in several physical-chemical forms in the atmosphere of nuclear plants: 1. bound to aerosols; 2. in the elemental form as iodine vapor; and 3. also in organic compounds (mostly as methyl iodide). As there are suitable methods and equipment for aerosol separation, here we investigate only the

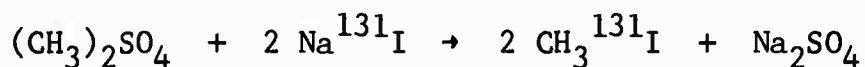
* Central Institute for Nuclear Research Rossendorf Works performed for the Federal Central Bureau for Radiation Protection, East Germany.

behavior of radioiodine in the form of iodine vapor and of organic iodine in the form of methyl iodide.

Various methods may be used for iodine removal. High decontamination factors can be attained with physical processes, such as freezing out, adsorption on solids such as silica gel, molecular sieves, Al_2O_3 , absorption in washing solutions or on materials impregnated with thiosulfate, silver nitrate, potassium iodide, tertiary amines, etc. The adsorbent most often used today in nuclear plants for separating radioiodine from the exhaust gas is activated carbon [6, 7, 8]. The activated carbons provided for iodine testing were products of the German Democratic Republic (Peoples Synthesis Plant, Schwarzheide: WDG 010, WDK 14, WDK 14F; and the "Friedrich Engels" synthetic fiber plant at Premnitz: R3, R4 and AS). The activated carbons were evaluated by measurements of the filtration effectiveness, the filtering capacity, the strength of the carbon, the flow resistance, the flammability, preparation with various chemicals to improve the adsorption properties, the desorption behavior, etc.

2. Experimental

The apparatus sketched (Figure 1) was used for testing activated carbon with elemental iodine as well as with methyl iodide. The flow of carrier gas (air) moved from the steel cylinder through a rotameter into the vessel in which iodine or methyl iodide was developed. Iodine was made by reaction of $\text{Na}_2\text{Cr}_2\text{O}_7$ with carrier-free Na^{131}I , and passed into the attached cooling vessel. The methyl iodide was generated according to the equation:



and was likewise deposited in the cooling vessel. By thermostatic control of the cooling vessel, an amount of $^{131}\text{I}_2$ or $\text{CH}_3^{131}\text{I}$

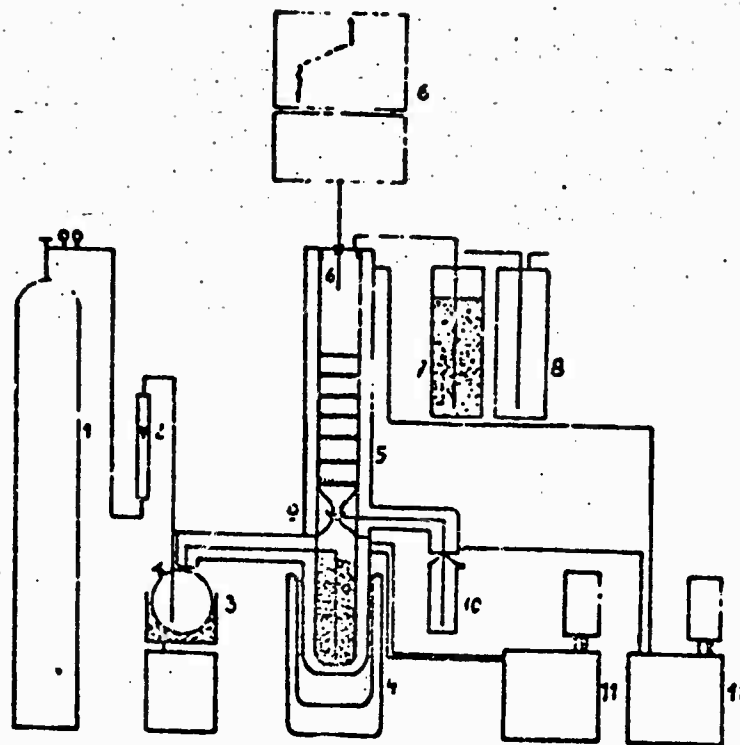


Figure 1. Apparatus for determining the adsorption of $^{131}\text{I}_2$ or $\text{CH}_3^{131}\text{I}$ on activated carbon.

1. Steel cylinder.
2. Rotameter.
3. Iodine generation vessel and control transformer.
4. Cooling vessel.
5. Filter being measured.
6. Gas flow counter tube with measuring point and recorder.
7. Safety filter.
8. Wash bottle.
9. Three-way cock.
10. Sampling bottle.
11. Thermostat.

corresponding to the temperature was vaporized out of the cooling vessel and led by the carrier gas flow through the activated carbon test bed. In order to check the activity during the experiment, a gas flow counting tube is connected following the filter, with a pulse rate meter and a recorder. The following counting tube proved to be good, because any filter leaks could be noted immediately because of the rapid rise in activity. After the activated carbon bed there was a safety filter, consisting of a washing bottle with thiosulfate solution, for complete removal of radioiodine or methyl iodide. The cooling vessel and the activated carbon filter could be heated separately with 2 thermostats. A three-way cock ahead of the activated carbon filter allowed the gas flow to be taken off for separate checking. No iodine activity could be demonstrated in the thiosulfate washing bottle after all the experiments. The actual evaluation of the activated carbon samples was done with a NaI scintillation crystal at 0.36 MeV, outside the apparatus shown.

3. Experimental Results and Discussion

3.1 Physical-chemical Parameters of the Activated Carbon

The surface areas of the activated carbons were determined with N_2 by the BET method. The results are shown in Table 1. Table 2 shows the water content of the commercial product, which was determined by weight loss after drying at $110^\circ C$. The AS carbon had the lowest water content. The residue after ignition, determined according to TGL 9493, is also shown in Table 2.

The Type WDG 010 activated carbon had more than a fourth of its weight as uncombustible residue. Determination of the carbon dusting showed that the WDG 010 activated carbon had low strength, and 50% carbon dust was found in WDG 010 when the particle size distribution was checked by sieve analysis, while the R3, R4, and AS contained less than 0.25% carbon dust. If one considers

TABLE 1. DETERMINATION OF THE SURFACE AREA BY THE BET METHOD

Carbon Type	Surface (m ² /g)
AS	1265
R3	1130
RA	710
WDK 14 P	810
WDK 14	765
WDG 010	620

TABLE 2. DETERMINATION OF THE WATER CONTENT AND RESIDUE ON IGNITION

Carbon Type	Water Content (%)	Residue on ignition (mg/g carbon)
S	4,8	81,0
3	15,8	40,6
4	12,3	76,4
DK 14 P	50,7	18,0
DK 14	42,7	41,7
DC 010	49,8	264,7

the results of these studies, it appears that the AS activated carbon has a desirable behavior with respect to the dusting resistance, water content, and residue on ignition, and has the greatest surface.

2. Measurements of the Adsorption Behavior of the Activated Carbon With Respect to Macro-Amounts of Iodine or Methyl Iodide

In order to determine the adsorption capacity of the activated carbons under static conditions, 1 g portions of activated carbon and excess iodine or methyl iodide were placed in a sealable vessel at a constant temperature of 80°C or 23°C, respectively. The course of adsorption was followed gravimetrically to constant weight. Table 3 shows the amounts of iodine or methyl iodide adsorbed, expressed in grams of iodine or methyl iodide per gram of carbon. Vogt and Meringdal [6] report similar values of 2 to 4 grams of iodine per gram of carbon.

The adsorption capacity under dynamic conditions was measured at 80°C in the apparatus sketched in Figure 1, with 1g of carbon using an incident flow velocity of 2.1m/minute for iodine. Methyl iodine adsorption was determined by means of the retention time, using gas chromatography (see Table 4). Due to the high adsorption capacity of the activated carbon, inactive iodine was used for testing. The amounts of iodine adsorbed on the activated carbons are shown in Table 5. Under the test conditions selected, the amounts adsorbed are about 1 g iodine per gram of carbon. These values are not to be considered in practice, though, because a filter cannot be used to zero efficiency, especially in nuclear plants. The actually usable filter capacity could be considerably below 0.1 g iodine per gram of carbon. From the view point of the nearly carrier-free occurrence of radioiodine in nuclear plants, this gives enormous useful lives. These filter lives are reduced by the presence of still other adsorbable foreign gases or even by the inactive iodine content of 10^{-7} g/m³ [9] in the atmosphere. With respect to activity, the filter can be loaded to about 1 Ci/g carbon. Desorption phenomena are still negligible at this activity concentration [7].

TABLE 3. STATIC ADSORPTION OF IODINE ON ACTIVATED CARBON:
AT 80°C, 1 g DRIED CARBON, DRY ATMOSPHERE

Carbon Type	Amount of Iodine Adsorbed (g iodine/g carbon)
AS	2,06
R3	1,82
RA	1,46
WIK 14 P	1,22
WIK 14	1,16
WDG 010	0,88
AS + AgNO ₃ Impregnation	1,98
R3 + AgNO ₃ "	1,74
RA + AgNO ₃ "	1,42
WIK 14 P + AgNO ₃ "	1,20
WIK 14 + AgNO ₃ "	1,12
WDG 010 + AgNO ₃ "	1,06

STATIC ADSORPTION OF METHYL IODIDE ON ACTIVE CARBON:
AT 23°C, 1 g DRIED CARBON, DRY ATMOSPHERE

Carbon Type	Amount of Iodine Adsorbed (g iodine/g carbon)
WDG 010	1,72
WIK 14	1,69
WIK 14 P	1,51
AS	1,26
R3	1,09
RA	0,88
AS + Cu(NO ₃) ₂ Impregnation	1,28
AS + AgNO ₃ "	1,24
AS + KI "	0,86

TABLE 4. DYNAMIC ADSORPTION OF METHYL IODIDE ON ACTIVATED CARBON:
1 g CARBON, 80°C, INCIDENT FLOW RATE 1.06 m/min.

Retention times in minutes, determined by gas chromatography							
Carbon Type	Untreated	CO ₂	AgNO ₃	Cu(NO ₃) ₂	Heat 470°C	Water Vapor	KI
WIK 14 F	39,1	35,9	42,1	40,4	28,1		2,8
WIK 14	34	30,4	30,2	34,4	32,2		2,2
AS	32	35	32,3	27,6	16,4	42,8	6
R3	24	14,9	20,2	16,5	11,8	17,4	3
WDG 010	13	10	11	11	17,1	12,4	1,6
RA	11	6,8	7,1	8,4	7,5	8,8	2,3

TABLE 5. DYNAMIC ADSORPTION OF IODINE ON ACTIVATED CARBON AT 80°C,
INCIDENT FLOW RATE 2.1 m/min, 1 g CARBON.

Amount of iodine adsorbed in g iodine/g carbon				
Carbon Type	Untreated	Impregnated with AgNO ₃	Impregnated with Cu(NO ₃) ₂	Tempered at 470°C
AS	1,98	0,26	1,12	2,13
R3	1,66	0,35	0,97	1,57
RA	1,09	0,22	1,06	1,21
WIK 14 F	0,93	0,21	0,53	1,24
WIK 14	0,90	0,29	0,52	0,97
WDG 010	0,87	0,21	0,46	0,89

3.3 Measurements of the Adsorption Behavior of Activated Carbons With Respect to Micro Amounts of Iodine and Methyl Iodide

Under the conditions selected, the tests with carrier-free ¹³¹I iodine and carrier-free methyl iodide gave high degrees of separation for both adsorbates. The filter efficiency was

determined from the ratio of the activity concentration on the measured filter to the total activity concentration:

$$\eta = \frac{A_1}{A_1 + A_2}$$

A_1 = activity concentration of the measured filter

A_2 = activity concentration of the residual filter

The residual filter was dimensioned so that there could be complete separation of the radioiodine or methyl iodide. Testing of the following wash bottle showed no activity. Contamination losses could be kept very small by using a glass apparatus and test temperatures of 80°C. This favorable degree of separation, even for methyl iodide, is characteristic of a dry atmosphere, while lower separation for methyl iodide is to be expected for air saturated with water vapor [10].

From Table 6 we can see that the activated carbons WDG 010 and AS had good behavior for carrier-free iodine and methyl iodide. Taking the physical properties of the activated carbons into consideration, the AS carbon was used for further experiments with the goal of improving adsorption by impregnating or activating. The retention times determined by gas chromatography are shown in Table 4. Some of the different preparations gave improvements in the retention times, especially for the AgNO_3 impregnation. The activated carbons from the German Democratic Republic production showed adsorption capacities and degrees of separation which were very similar to the Type H 32 activated carbon from the German Federal Republic, which is being used for removal of radioiodine from the waste gas streams of nuclear plants. As the tests for characterizing the adsorption behavior of the activated carbons from the German Democratic Republic production are being continued, no final evaluation can yet be given.

TABLE 6. ^{131}I ADSORPTION,
CARRIER-FREE AT 80°C ,
1 g CARBON, INCIDENT FLOW
VELOCITY 2.1 m/min

Carbon Type	%
AS	99,47
R3	79,93
RA	99,58
WIK 14 P	95,5
WIK 14	98,57
AS + KI	99,33
AS + AgNO_3	99,65
Ag + $\text{Cu}(\text{NO}_3)_2$	99,77

$\text{CH}_2^{131}\text{I}$ ADSORPTION,
CARRIER-FREE AT 80°C ,
10 g CARBON, INCIDENT FLOW
VELOCITY 0.24 m/min

Carbon Type	%
AS	99,66
R3	99,98
RA	99,08
WIK 14 P	99,96
WIK 14	97,81
AS + KI	99,84
AS + AgNO_3	99,93
Ag + $\text{Cu}(\text{NO}_3)_2$	99,68

Although most nuclear plants use activated carbons because of their high adsorption capacity, their flammability at high temperature cannot be overlooked. Various authors were able to increase the ignition temperature to 500°C with special types [11]. Along with improvement of the adsorption properties of activated carbon by impregnation and activation, there are recent literature reports on the search for other adsorber materials suitable for separating radioiodine from nuclear plants [12, 13, 14].

REFERENCES

- /1/ MISHIMAT, J.
Methyl iodide behavior in systems containing airborne radiiodine
Nuclear Safety 9 (1968) 1, 3. 35-42
- /2/ CHAMBERLAIN, A.C. u.s.a.
Physical chemistry of iodine and removal of iodine from gas streams
J. Nucl. Energy Parts A/B 17 (1963) 519-550
- /3/ DOUGLAS, K. u.s.a.
Effect of iodine concentration on the efficiency of activated charcoal adsorbers
Health Physics 19 (1970) 3. 223-233

- /4/ **CEJHAR, P.**
Chování par metyljodidu v systémech obsahujících páry radiojodu
Jaderná energie 16 (1970) 2, S. 42-47
- /5/ **KACHUTIN, J.E. n.a.**
Adsorbce parov radioaktivního jodu iz vzduchu
Atomnaja energija 26 (1969) 4, S. 390-391
- /6/ **VOGT, K.J.; MERINGDAL, J.**
Messung der Adsorptionseigenschaften verschiedener Filter für gasförmiges Jod
JUL-63 (1966)
- /7/ **STRAUSS, H.J.; WINTER, K.**
Radiojodabscheidung aus der Abluft von Kernreaktoren
in: Reaktortagung
Berlin: Dt. Atomforum 1970. 715 S.
- /8/ **WILHELM, J.**
Adsorbentmaterialien zur Abscheidung von Spaltjod aus den Abgasen kerntechnischer
Installationen
in: Reaktortagung
Berlin: Dt. Atomforum 1970. 715 S.
- /9/ **BALOGH, L.**
Acta Physiologica 14 (1958) 7
- /10/ **WINTER, K.**
Probleme der Abscheidung von radioaktivem Joddampf
Atomwirtschaft 2 (1967) 7/8, S. 440-442
- /11/ **TAYLOR, R.**
The trapping of methyl iodide at high pressure and high temperature
in: Congress International on the Diffusion of Fission Products
Montrouge: Soc. Franc. Radioprotection 1970. 549 S.
- /12/ **DONNER, Chr.; TAMBERG, T.**
Untersuchungen zur Darstellung und Prüfung einiger anorganischer Jodfiltermaterialien
Kerntechnik 14 (1972) 1, S. 22-28
- /13/ **SCHÜTTELKOPF, H.; WILHELM, J.**
KFK-1158 (1970) S. 85-99
- /14/ **HLADKÝ, E.; PIETRIK, J.; KUBIK, J.**
Průzkum možnosti použití anorganických sorbentů pro záchyt radiojodu na jaderných
elektrárnách
Jaderná energie 18 (1972) 1, S. 18-22