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THE IODINE COLLECTION EFFICIENCY
OF ACTIVATED CHARCOAL FROM HANFORD
REACTOR CONFINEMENT SYSTEMS:
METHYL IODIDE RETENTION
BY ACTIVATED CHARCOAL

July 1969

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By

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Radiological Sciences Department
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ABSTRACT

Activated charcoal has been used in the gas confinement system of Hanford's nuclear reactors for some time, and it was necessary that their iodine adsorption ability be precisely determined to insure operational safety. Adsorption measurements were taken while subjecting these charcoals to stress. The results were compared with those obtained from new, unused products, and an estimate was made of the loss in iodine trapping efficiency.

Adsorption tests, using both methyl iodide and iodine, were also conducted on fresh charcoal to measure the effectiveness of some of the newer commercially available products. The purpose of these tests was to aid in the selection of activated charcoal for replacement in the reactors or for use in future reactor confinement systems. The quantity of iodine emanated as methyl iodide during an accidental reactor meltdown is not accurately known; however, it is desirable to have a confinement system with sufficient capability for its removal.

A stainless-steel facility was constructed for the purpose of testing the activated charcoals of interest. The charcoal was subjected to high humidity and temperature in this facility while its adsorption characteristics were measured for various concentrations and species of iodine. Results of measurements made on charcoal from all operational and shut-down

reactors at Hanford showed severe deterioration in some. In certain cases as much as 20% of the molecular iodine was not adsorbed by a 1 in. bed of charcoal. Methyl iodide tests on unused activated charcoal illustrated that good adsorption was obtained only in those products which had been specially treated to improve their trapping ability for this species.

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INTRODUCTION

At the present time, considerable effort is being expended to determine the methyl-iodide trapping efficiency of activated charcoal used in nuclear reactor air confinement systems. (1) The concern has been over the possibility of a significant fraction of the reactor iodine inventory escaping as methyl iodide in the event of an accident. In the case of the largest water-cooled reactors, further complications could result from the release of gases having elevated temperatures and humidities. Under these more severe conditions, it is known that the charcoal trapping capacity for methyl iodide is reduced and the degree of reduction is dependent upon the severity of the conditions. (2)

Several commercial manufacturers of activated charcoal have recently incorporated special additives into their production processes in an effort to improve the trapping ability for methyl iodide. One would expect charcoal to first adsorb methyl iodide and then allow its slow removal by chromatographic elution. Charcoal additives such as KI, which exchange iodine with the organic material, provide an effective means of removing the radioiodine while treating the radiochemically-inert methyl-iodide molecule in the usual manner.

A facility was designed and constructed with features for temperature and humidity control to independently test the ability of these new activated charcoal products at Hanford.

Tests were conducted on these new charcoal products by using molecular iodine and methyl iodide under various conditions.

Nuclear reactor air-confinement cells at Hanford consist of banks of activated charcoal canisters. These canisters are 2 x 2 ft in dimension with an 8 in. depth to accommodate a larger folded area, 1 in. thick charcoal bed. Very significant results were obtained from tests on activated charcoal that had been and in the same cases, were still in use in reactor air-confinement systems. Certain charcoals were found to have remarkably retained their high efficiency for iodine collection; whereas others had deteriorated to such an extent that they were considered unqualified for continued use. Results in one instance showed the molecular iodine trapping efficiency to be less than 95% at room temperature and only 82% under humid conditions at moderate temperatures for a 1 in. charcoal bed. The deterioration of the charcoal trapping ability appears to be related to the concentration of specific impurities in the gases continuously passing through it. This factor is further complicated because certain impurities show little effect on the iodine collection ability while others are quite serious. The measured charcoal-efficiency loss in the reactor samples appeared to be a rather slow process, with perhaps months to years of continuous use constantly deteriorating the adsorption sites.

Measurements were also made of the significant radionuclides present on the charcoal in the reactor filter banks as well as the specific steady-state concentration of radioiodine.

SUMMARY

Nuclear reactor safeguard standards criteria for the performance of a gas confinement system requires that at least 95% of the iodine passing through the charcoal system be trapped. That is, no more than 5% be emitted to the environment during

accident conditions when the charcoal may be subject to hot, humid gases. Essentially, results from tests conducted under ambient conditions with molecular iodine showed that all reactor charcoals met or exceeded these standards. However, at high humidity and temperature all reactor charcoals exhibited a minimum reduction in iodine adsorption of about 2%; in a few cases, the loss was so severe as to warrant charcoal replacement. Under the more severe adsorption conditions, the iodine efficiency range for the nine reactors tested was 82 to 98%.

Experimental testing of the adsorption characteristics of new types of activated charcoal was conducted by using both iodine and methyl iodide. All the new, unused charcoals adsorbed 100% of molecular iodine in a 1 in. bed even when subjected to elevated temperatures. At high humidity the results were quite similar, with at least 99.8% iodine adsorption taking place. Ambient and elevated temperature tests showed that charcoal impregnated with KI was very effective for removing radioiodine from the methyl iodide molecule. This radioiodine was difficult to adsorb by charcoal when the high temperature gases were saturated with water vapor. Under these conditions one of the charcoal types tested exhibited greater than 95% iodine adsorption.

EXPERIMENTAL

Flow tests were conducted in which molecular iodine was passed through several plastic, metal, and glass tubes to evaluate their reactivity. Most plastics reacted with or adsorbed iodine readily and were not desirable for construction. Even teflon had a slight affinity for iodine; however, the adsorbed quantity could be slowly eluted when sufficient air flow was maintained. Glass was found to be chemically and physically inert to iodine and was used in a limited way. However, glass does not adapt itself easily to an assembly

that must be dismantled without having reactive materials for connecting joints. In our experiments, corrosion-tested stainless steel was found to adsorb iodine slightly. This surface adsorption was readily removed by continued elution. As a consequence of these tests, the facility for charcoal evaluation was constructed mainly of stainless steel by using compression type fittings (Swagelok) with a limited number of teflon-sealed pipe joints. Also, some glass was utilized where appropriate.

Major portions of the test facility are illustrated in Figures 1a and 1b. Air was drawn into the system through a flowmeter (left, Figure 1a). In tests where humidity control was unnecessary, the air may bypass all other adaptations shown in Figure 1a and go directly into the two serially arranged tube furnaces (Figure 1b). These adjust the temperature before the air enters the succeeding charcoal test chamber.

The radioactively-tagged iodine was interjected into the air stream either as methyl iodide from a pressurized stainless steel cylinder (Figure 1a) or as chemically-generated molecular iodine from a small glass flask (Figure 2). The injection point was identical in each case. Incoming air may take one of two alternate paths to the test chamber. A "T" arrangement allows this air to be either preheated or humidified or both in a stainless steel boiler.

The boiler was constructed out of 5 1/2 in. pipe with welded end plates, and stainless valves on each end allowed complete removal of the boiler from the line. A sight glass and pressure gauge were attached to the side of the boiler. The sight glass was constructed from thick glass wall tubing and the ends were stainless steel tube fittings. The lower fitting was connected to a distilled water source so that the

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FIGURE 1a. Test Facility for Measuring Methyl Iodide Adsorption
by Activated Charcoal

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FIGURE 2. Charcoal Test Facility With Molecular
Iodine Generator

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FIGURE 3a. *Charcoal Bed Test Chamber*

entrance and exit positions of the chamber. Ordinarily, only one chamber section containing a final 2 in. deep bed of activated charcoal followed the two-compartment section but a series of 2 in. deep beds could be added to the chamber by inserting additional chamber sections as just described. The sections separated by perforated screens were internally sealed by the use of O-rings mounted in grooves in the chamber section walls. Pressure was applied to the O-rings to insure a tight seal by three threaded rods holding the sections together. All threaded connections were teflon-taped for sealing purposes. The stainless steel sheathed chromel-alumel thermocouples were connected to an 8-point recorder* whose temperature range was adjusted with resistor boards to the designed range for a test series. The thermocouple readings were recorded in sequence at 30 sec intervals.

The gases in tests conducted without additional humidity adjustment were exhausted at the exit of the test chamber. High humidity experiments utilizing the boiler required the remainder of equipment shown in Figure 1b. The air stream was diverted through the glass condenser coils, which were operated in parallel, to allow the high flow rate desired. Condensed water from this air was also forced up the coil to the overflow bulb located at the top of the apparatus. Here the air-water mixture was separated and the water was channeled to the lower glass reservoir. Rather large amounts of water were collected during some tests, and up to 2.5 liters could be easily handled. The reservoir was internally calibrated for water volume, and each test could then be calibrated for humidity purposes. A stopcock allowing rapid water removal was attached at the bottom of the reservoir. For high-humidity long-term tests, the water in the boiler could be

*Honeywell Brown Electronik, Minneapolis-Honeywell Regulator Company, Brown Instrument Division, Philadelphia, Pa.

replenished and the reservoir exhausted at calibrated intervals, thus allowing continuous operation at high humidity. The cooled moist air was then led to a small tube furnace* (extreme right, Figure 1b) where it was warmed to reduce its relative humidity and was subsequently passed through a 3 in. bed of activated charcoal contained in a stainless steel sampler. The purpose of this final trap was to ensure the complete removal of all iodine used in the tests. The absence of ^{131}I activity in this trap throughout the test series provided proof of the effectiveness of the test chambers' charcoal beds. The gas flow in the entire system was maintained by pump or vacuum supplied at the exhaust end of the final trap. This connection was not illustrated.

PREPARATION OF MOLECULAR I_2 TAGGED WITH ^{131}I

Generally, the charcoal was tested in the facility with a loading of about 100 μg molecular iodine per gram of charcoal in the upstream bed. Figure 2 illustrates the iodine generation apparatus and the flowmeter through which air was drawn into the overall system. Air flow adjustments were made by downstream valve control so that the exact linear gas velocity in the charcoal bed could be maintained. Molecular iodine was prepared by the following reaction:

A measured quantity of sodium iodide was added to a 3N H_2SO_4 solution containing sufficient acid to complete the reaction as written. To this was added a quantity of ^{131}I in basic sulfite solution sufficient to tag the inert iodine and allow easy analysis of the sampler sections. About 5×10^{-3} μCi of the ^{131}I were introduced as a tracer in the test and the iodide

* Fisher Micro Combustion Furnace, Fisher Scientific Company, Pittsburgh, Pa.

was oxidized to iodine by slow addition of NaNO_2 . All iodine-containing solutions were treated with Na_2SO_3 and held slightly basic to keep the iodine in the iodide form prior to use. The rate of iodine addition to the air stream (and the resultant iodine-air concentration) was partly controlled by the NaNO_2 addition. The iodine formed in the generator was swept from the solution into the air stream by using helium gas. This maintained positive pressure in the generator which prevented air entering the system from bypassing the flowmeter.

PREPARATION OF METHYL IODIDE TAGGED WITH ^{131}I

Efficiency tests of activated charcoal required the preparation of methyl iodide having a known and useful amount of ^{131}I tracer. An experimental apparatus was set up for the preparation of the iodide which consisted of a three-neck flask connected to a water-cooled condenser (Figure 4). A thermometer was placed in one neck and a separatory funnel in the remaining neck. The flask was wrapped with heating tape which was temperature controlled by a variable transformer. A mixture of 8 g KI, premixed with about 0.01 mCi ^{131}I in the form of a NaI solution; 4.3 ml H_2O ; and 0.6 g CaCO_3 powder was placed into the flask containing a magnet stirrer. The flask was warmed to 65 °C while stirred slowly. Drops of dimethyl sulfate (4.7 ml) were added to this mixture from the attached separatory funnel. The product, formed by the reaction,

was slowly distilled from the mixture and condensed into a collecting vial containing anhydrous CaCl_2 at 0 °C. The dry methyl iodide (4 ml) was decanted into a storage vial containing a drop of mercury to prevent iodine formation. The vial was stored in a deep freezer, and aliquots of methyl iodide were taken by using precooled pipettes.

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FIGURE 4. Apparatus for Preparation of Tagged Methyl Iodide.

Figure 2 illustrates the one-liter stainless steel cylinder with sensitive pressure gauge used for dispersing the radioactive methyl iodide. After the cylinder was precooled in the deep freeze, 100 μ liter aliquots were transferred to the open cylinder. The cylinder valve was replaced, and the outlet was connected to a high-pressure argon gas source. The cylinder was pressurized to 100 psi and then allowed to stand overnight to facilitate mixing of the argon and methyl iodide. The vapor pressure of methyl iodide at room temperature was sufficiently high to ensure complete vaporization of the compound within the cylinder, with the argon gas acting as a carrier for iodide dispersal. By carefully bleeding the cylinder into the flow stream, the resultant air-iodide concentration was controlled. Air concentrations of methyl iodide were typically 10 mg/m³, and the charcoal loading for methyl iodide was about 1 to 2 mg/gram charcoal tested.

CHARCOAL TESTING

The test chamber was assembled as follows:

- About 36 g of charcoal were placed in the first 1 in. bed of the primary test section.
- The material was agitated to provide close packing.
- The separator screen and steel sleeve were inserted and aligned to allow the center thermocouple to be mounted at the time.
- Thirty six grams of charcoal were also placed in the second bed.
- The final two inch chamber section used for backup purposes was similarly prepared.
- The sections were assembled to form the test chamber.
- Three bolts that produce the pressure seal on the O-rings in each section were tightened firmly but without excessive pressure.

- A vacuum was drawn from the exhaust end of the system and the test began.

No other control was necessary for charcoal tests at ambient temperatures.

Generally, the iodine was loaded onto the charcoal in about 30 min. Air elution continued for 2 hr after test initiation in a typical experiment. When the iodine collection efficiency of the charcoal bed was measured at elevated temperatures, slight changes in operating procedures were used. The air flow was adjusted to the proper level, and the two-series tube furnaces were turned on. The temperatures of the air in the charcoal beds were measured by the three chromel-alumel thermocouples in the test facility and recorded on the 8-point recorder. Furnace temperature control was maintained by using an iron-constantan thermocouple and a relay operated controller. When the air temperature reached the desired level, iodine was introduced and the experiment proceeded. Temperature control was generally within 1 °C of the desired temperature, with small adjustments made in the controller-thermostat level.

To test the capability of activated charcoal under rather extreme operating conditions, experiments were conducted under humid conditions at elevated temperatures. The furnace used to supply heat to the boiler was turned on with the boiler sealed from the system. Air flow through the system was started and the tube furnaces turned on; all controlling and recording systems were operating. The pre-exhaust heater was turned on and set at a point which provided about 50 °C air to the exhaust trap. This greatly reduced the air humidity prior to passing through the final charcoal trap. As the water in the boiler was heated, the internal pressure rose and when it reached 30 psi, the downstream boiler valve was slowly opened and moisture entered the air stream. Approximately 1 hr was required to bring the boiler to

temperature. The upstream valve was then opened and the bypass valve closed, forcing all incoming air from the flow-meter to pass through the boiler and to become saturated. Heating and humidification were continued for at least another hour while the temperature and humidity in the charcoal beds stabilized. During this time, the condensers were in operation collecting water. When a satisfactory stable condition was noted, the water level in the collection reservoir was measured, the iodine generation was initiated, and the recorder was marked for operation. Adjustments in tube and boiler temperature were made to maintain the desired conditions. Practice in this technique was necessary to become familiar with the particular characteristics of the system. The rate of water condensation was indicative of the humidity of the operation, and it was relatively easy to maintain almost 100% humid conditions in the test chamber. The average humidity during a test was calculated from the quantity of water collected and total air flow through the system. Corrections were made for the water remaining in the exhaust air at the exhaust gas temperature. The quantity of water collected was in agreement with the theoretical amount expected in saturated air at 100 °C. Air saturated at 100 °C contains about 600g H₂O/m³; therefore, quite a large volume of water was collected in each test. At the completion of a test, the height of the water in the reservoir was noted.

After the facility was shut down and air flow was terminated, the test chamber was disassembled, and the charcoal from each bed was transferred to a polystyrene screwtop jar which fit into a well-type gamma-ray spectrometer (9 in. NaI (Tl)). Similar measurements were also made on the charcoal in the exhaust trap. The total iodine collected on each bed was calculated from the amount of ¹³¹I gamma ray detected under the 0.35 MeV photo peak. Samples of all

charcoal types tested were taken for γ -ray analysis to measure the possible interference in the ^{131}I gamma energy region. These measurements showed no unusual background interference levels for new charcoal samples; however, several radio-contaminants were found in used, reactor charcoals. The amount of this residual interference found in the region of the ^{131}I gamma spectrum was used to determine the quantity of tracer activity to be used in the experiments. In this way, interpretation of the results was simplified.

RESULTS AND DISCUSSION

RADIONUCLIDES DEPOSITED ON ACTIVATED CHARCOAL IN REACTOR AIR CONFINEMENT SYSTEMS

Before tests were conducted on the effective adsorption of reactor charcoal for iodine, the interference level of radionuclides already present on the material had to be measured. Gamma-ray spectral analysis was conducted on all reactor charcoals measured in this work in the well-type NaI (Tl) counting arrangement previously described. Data taken from charcoal in reactors inoperative for about three years are shown in Table 1.

TABLE 1. *Long-Lived Radioisotopes Collected by Charcoal Traps in Reactor Airstreams at Hanford^(a)*

<u>Isotope</u>	<u>Half-Life</u>	<u>D Reactor, pCi/3</u>	<u>H Reactor, pCi/g</u>	<u>F Reactor,^(b) pCi/g</u>
^{60}Co	5 yr	7.2	6.2	6.1
^{137}Cs	30 yr	3.4	4.5	4.0
^{125}Sb	2.7 yr	2.0	1.6	1.4
^{54}Mn	254 day	0.2	0.18	0.0

(a) *The charcoal traps were preceded in all cases by particulate filters*

(b) *F Reactor has been shut down for a longer period than the others.*

The minimal quantity of the specific radionuclides detected in the background analysis of these charcoals presented no significant interference to the ^{131}I efficiency measurements. The charcoals examined from operating reactors, however, exhibited gamma spectra from the isotope ^{131}I with smaller interferences from 9.2 hr ^{135}Xe and other radioisotopes of these elements. Isotopes such as 1.8 hr ^{41}Ar were also identified in fresh materials. Generally, the quantity of ^{131}I tracer added during an experiment could be increased to reduce this interference to a negligible level; however, in one instance where $7 \times 10^{-2} \mu\text{Ci/g}$ was found in charcoal from an operating reactor, this was not feasible. Another approach used for reactor charcoal samples containing moderate amounts of ^{131}I , was the careful mixing of charcoal before loading it into the test beds. This method insured a homogeneous distribution of the existing adsorbed nuclides. At the completion of a test, the third bed, which was originally new-charcoal, now contained only iodine from eluting the second bed during the experiment. This quantity was also indicative of the amount of iodine eluted from the first to the second bed since these charcoal beds were subjected to the same experimental conditions. From this information and the quantity of added iodine found on the beds after the test, the charcoal efficiency could be calculated. These results were consistent with those obtained by conducting tests on the charcoal after a long delayed period which allowed the ^{131}I activity to decay to an insignificant level.

IODINE EFFICIENCY TESTS ON CHARCOAL FROM HANFORD REACTORS

Activated charcoal* has been used in all Hanford reactors for periods of several years to act as gas purifiers of reactor exhaust gases. Generally, the charcoal had been in use for a continuous five year period. This charcoal, mounted in

* *Dorex air recovery, Conner Engineering Corporation, Danbury, Connecticut*

special high surface area canisters stacked in rows, forms a very large adsorption trap for gaseous radio impurities such as ^{131}I , which may accidentally be released from the reactor system. Typically, there may be seven rows of these canisters occupying a room that acts as a large duct some 20×30 ft in cross section. Samples of this charcoal taken from selected reactor canisters were tested under various conditions to measure their efficiency for trapping molecular iodine. In some instances, these samples were taken from operating reactors, while sometimes the reactors had been shut down for several years. Samples were also taken from reactors in which special studies had been conducted. Table 2 illustrates the results of these measurements, along with results from testing of the unused original activated charcoal for comparative purposes. The iodine was generated in about 30 min for all cases. The resultant specific iodine loading in all cases was about $250 \mu\text{gI}_2/\text{g}$ charcoal for the first one-inch bed. It should be re-emphasized that these are results from tests utilizing molecular iodine. Primarily, these data indicate that molecular iodine can and does escape adsorption through 1 and often 2 in. of charcoal. The extent of iodine escape in certain cases must be considered rather unexpected for the test conditions described. In all cases, reduced adsorption was evident as the severity of the test conditions were increased. As expected all but one of the charcoals tested exhibited high efficiencies for iodine under ambient or normal room conditions. Only under stress could real differences in the capabilities of the charcoals be distinguished. The primary purpose of the containment system is to provide protection under hazardous accident conditions, at which time the charcoal will probably be subjected to high temperature and humidity. From these present experimental data, the satisfactory charcoals tested can be grouped into those maintaining a minimum of 95% efficiency under the most severe conditions used in these

**TABLE 2. Efficiency for Molecular Iodine Adsorption by Activated Charcoal
Used in Nuclear Reactors**

Sample Source	Gas Flow Conditions									Linear Flow Rate, ft/min	Elution Time, min
	26 °C, 50% Humidity			100 °C, 15% Humidity			100 °C, 100% Humidity				
	First (a) Second (b) Third (c)			First (a) Second (b) Third (c)			First (a) Second (b) Third (c)				
	First (a)	Second (b)	Third (c)	First (a)	Second (b)	Third (c)	First (a)	Second (b)	Third (c)		
New Reactor											
Charcoal	100	0.0	0.0	100	0.0	0.0	99.8	0.2	0.0	65	120
H Reactor	98.8	0.9	0.3	98.2	1.7	0.1	97.4	2.3	0.3	65	120
D Reactor	99.7	0.3	0.0	97.7	2.3	0.0	96.1	3.5	0.4	65	120
DR Reactor							97.9	1.2	0.9	65	120
F Reactor	99.4	0.6	0.0	98.2	1.8	0.0	96.9	2.5	0.6	65	130
KW Reactor	99.6	0.4	0.0	91.9	5.5	2.6	90.4	8.8	0.8	65	120
KE Reactor	94.3	5.7	0.0	88.5	9.3	2.2	81.8	16.9	1.3	65	120
N Reactor	99.5	0.5	0.0	98.7	1.2	0.1	97.5	2.3	0.2	65	120
B Reactor							94.1	5.1	0.8	65	120
C Reactor -1	99.1	0.9	0.0	98.2	1.6	0.2	96.6	3.2	0.2	50	260
C Reactor -2							95.8	3.7	0.5	65	120
C Reactor -3 ^(d)							94.7	4.7	0.6	65	120
C Reactor -4 ^(e)							91.7	7.8	0.6	65	120

- (a) First: 1 in. charcoal bed
 (b) Second: 1 in. charcoal bed
 (c) Third: bed acting as absolute iodine trap, at least 2 in. deep maintained at ambient conditions (new charcoal)
 (d) Sample taken from seventh row or edge of canister banks
 (e) Sample taken from fourth row or center of canister banks

tests. The charcoal efficiencies from KE and KW Reactors, which have subsequently been replaced, were quite poor under stress. KW Reactor charcoal at first appeared to be quite good; however, its iodine adsorption capability was marginal and quickly deteriorated with stress. Several factors are believed to affect the iodine adsorption ability of charcoal. Included in these are the impurity levels of gaseous contaminants passing through the charcoal and the length of time the charcoal has been in continuous use. Nuclear reactor design has been such that varying gaseous mixtures have been utilized in different reactors; consequently, different pollutant levels may be present in certain cases. Activated charcoal, although having a large adsorption surface, still can have its adsorption quality impaired or poisoned by adsorption of certain materials. Since the K reactors use nitrogen gas as one of their coolant gases, the presence of oxides of nitrogen in the containment system is highly probable. This would reduce the iodine collection efficiency of their charcoal beds. Evidence of the presence of NO_2 on the surface of the charcoal came from the high corrosion rate of the cadmium plate on the surface of the charcoal canister containers. A marked increase in this corrosion was found on the downstream or exhaust side of the bed, indicating the pre-concentration of NO_2 in the bed followed by its delayed elution. In all probability, the gas was converted to nitric acid after combination with water on the charcoal surface. The upstream or gas entrance end of the canister had little, if any, corrosion compared to that found downstream. Thus, the impurity level itself was only slightly reactive; however, after concentration in the bed, the corrosive agent produced visible damage. The formation of NO_2 was assumed to be from NO oxidation, which itself is known to be found in nitrogen containing mixtures under severe irradiation conditions. Further evidence of the acid nature of the adsorbed charcoal gases in these two reactors came from pH measurements

of aqueous solutions used to leach contaminants from the charcoals. A comparison of these pH measurements with charcoal from reactors not using nitrogen for cooling purposes showed that acidic constituents were present on the charcoals used in reactors employing nitrogen gas.

Several samples of activated charcoal were removed from a number of locations in C Reactor. The first two results, C-Reactor -1 and -2, showed small differences which were probably more indicative of the experimental precision than the small change in flow rates or elution times. It was also apparent from C-1 that increased elution time did not significantly affect the results of these experiments. Charcoal located near the center of the containment cell exhibited a significantly lower iodine adsorption (compare results from C Reactor -3 and -4 tests). The continuous air flow at the center of the cell was much higher than at the periphery; therefore, higher charcoal contaminant levels were likely present. This accelerated the deterioration processes in the charcoal and reduced its iodine collection efficiency.

Since specific charcoals had proven satisfactory in the trace pollutant level tests, experiments were needed to determine if they would retain their collection efficiency for iodine in a reactor with a high air-pollution level. For this purpose, the charcoal filter beds in KE Reactor were replaced with charcoal from other high-efficiency reactors, and the iodine collection efficiency was determined as a function of time. After four months of operation, tests conducted at ambient and under severe conditions showed that the charcoal adsorption efficiency was as high as it had been in the previous reactor. The condition of this charcoal will be studied in the future since the deterioration process is slow. This slow deterioration was fortunate, since it is absolutely essential that a positive method of safeguards be maintained. The

data point up the need for continuous surveillance of such a trapping system to prevent continued use of deteriorating charcoal with its inherent increased environmental hazard potential.

NEW CHARCOAL TESTS

The original charcoal test facility was designed to measure the efficiency of new activated charcoal products currently available commercially. This information would then be used to select a product for future use in reactors consistent with present and anticipated reactor safeguard standards. In addition, economical justification was required for the selection made with respect to such factors as charcoal type and the necessary bed depth. Some measurements of efficiency of selected charcoals were made, and the results are shown on Table 3. The experimental data demonstrated that even under the most severe test conditions, the capability for trapping molecular iodine by new charcoal was very high. A 1 in. bed depth was quite effective in removing iodine gas for both normal and impregnated charcoals. The situation changed considerably for the methyl iodide species. Tests conducted with unimpregnated charcoals, such as Barnebey-Cheney type 592, showed a marked reduction in collection efficiency at high humidity and temperature. In contrast, a 1 in. bed of type 727 impregnated with KI removed greater than 97% of the radioiodine in the methyl iodide. The methyl iodide collection efficiency was probably similar to an unimpregnated charcoal. An efficiency of 97 percent was considered adequate in relation to reactor safeguards standards.

Experimental evidence from methyl iodide tests using longer elution times than those listed in Table 3 indicates two operative mechanisms for iodide trapping. Impregnated charcoal (KI) exchanges its inert iodine for the radioiodine in the methyl iodide molecule, thus binding it firmly in that charcoal section (barring re-exchange with additional organic

TABLE 3. The Efficiency for Iodine and Methyl Iodide Collection by Activated Charcoal

Charcoal Type	Iodine Form, %	Gas Flow Conditions									Flow Rate, ft/min	Elution Time, min
		26 °C, 50% Humidity			100 °C, 15% Humidity			100 °C, 100% Humidity				
		First (a)	Second (b)	Third (c)	First (a)	Second (b)	Third (c)	First (a)	Second (b)	Third (c)		
Dorex	I ₂	100	0.0	0.0	100	0.0	0.0	99.8	0.2	0.0	50	120
BC-727 (d)	I ₂	100	0.0	0.0	100	0.0	0.0	99.93	0.07	0.0	65	140
BC-592 (e)	CH ₃ I							91.7	6.1	2.2	65	120
BC-727	CH ₃ I	100	0.0	0.0	100	0.0	0.0	97.3	1.7	1.0	65	120
BC-763 (f)	CH ₃ I							Less than BC-727			65	120

(a) First: 1 in. charcoal section

(b) Second: 1 in. charcoal section

(c) Third: sampler section (>2 in. deep) containing charcoal (KI) maintained near ambient conditions

(d) BC-727 - Barnebey-Cheney Type 727 activated charcoal containing KI to enhance ¹³¹I collection from CH₃I.

(e) BC-592 - Barnebey-Cheney Type 592 activated charcoal containing a high temperature additive.

(f) BC-763 - Barnebey-Cheney Type 763 activated charcoal containing KI and an additive for high temperature use.

organic iodides). Continued elution of this would presumably have no effect on the entrapped iodine; however, continued elution did remove radioiodide from such a charcoal bed. The rate of removal was not as rapid as in tests conducted with unimpregnated charcoal. These results indicated that some of the radioiodide trapped on the impregnated bed was adsorbed and re-eluted chromatographically, while another part exchanged with the KI and was no longer available for elution.

Economically speaking, the impregnated charcoals represent a significantly higher investment cost for a nuclear reactor system. The need for such a special trapping material is dependent upon the iodine species of importance in a confinement system. The fraction of organic iodides present in reactor gas is not accurately known; however, work in this field is presently in progress.⁽³⁾ From estimates of this fraction and from data obtained in this study, one can properly select the type and depth of an iodine trapping bed which exceeds that required in health hazard standards for environmental safety.

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