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REMOVAL OF ELEMENTAL RADIOIODINE FROM FLOWING HUMID AIR BY IODIZED CHARCOALS

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ABSTRACT

Certain types of iodized charcoal have been found to be useful for trapping radioactive methyl iodide. The question has been raised, however, as to whether or not the iodizing process reduces the capacity of the charcoal for elemental iodine (I_2) to an unacceptable level. Investigation concerning this point has been made for the conditions of room temperature and of very high relative humidity, 98% and 100%. In the case of the 100% relative humidity condition, the charcoal was caused to contain bulk phase liquid water, i.e., it was water-logged. According to the results obtained, the iodized charcoals that were tested possess ample capacity at 25°C for I_2 when the relative humidity was 98%. The water-logged iodized charcoals exhibited erratic I_2 removal performance which ranged from good to fair. This somewhat unpredictable behavior is probably associated with sweep gas channeling in the charcoal bed rather than overloading of the charcoal with I_2. Obviously, operational conditions that might result in water-logging should be avoided if efficient trapping of I_2 is required.

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INTRODUCTION

Certain specially iodized (impregnated) charcoals have been observed to have the capability of removing radioactive methyl iodide ($CH_3{}^{131}I$) from flowing humid air and steam-air under a wide variety of conditions (1-6). The efficacy of these charcoals is, in part, due to an isotopic exchange reaction, in addition to the sorption processes peculiar to the charcoal. Because iodized charcoal itself is impregnated with one or more iodine-containing substances, during manufacture, the question has been raised as to whether or not the capacity of such charcoal for elemental iodine (I_2) has been reduced to a low and thereby unacceptable level. Previous study of the trapping of elemental iodine by charcoal had been concerned only with the non-iodized variety of charcoal and no attention was given to the ultimate capacity of the charcoal for gas-phase iodine under dynamic conditions (7). Accordingly, an investigation was initiated to remedy this lack of information, and a significant quantity of data has been obtained at room temperature for four varieties of iodized charcoal under conditions of extremely high loadings of I_2 and of high relative humidity.

Conceivably, isotopic exchange may be a factor in elemental radioiodine trapping by iodized charcoals. If so, the effect would be beneficial provided the impregnant had not previously been exposed to radioiodine or, if it had, provided the specific activity of the impregnant iodine was less than that of the I_2 entering the adsorber. In what follows, the retention of the ^{131}I of I_2 by iodized charcoal will be treated as though it is entirely due to sorption of I_2. In any event, as in radioactive methyl iodide trapping, the important consideration is whether or not the

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radioactivity is stopped on the charcoal, and, in this work, the primary attention was directed toward ascertaining the extent to which this was accomplished.

IODIZED CHARCOALS TESTED

The types of commercial iodized charcoal that were tested in this study are listed in Table 1. Details regarding the procedures for iodine impregnations are not given because of proprietary considerations. MSA 85851 and BC-727 are more-or-less equivalent and contain about 5%, by weight, of the impregnated iodine form(s); BC-272 is similar to BC-727 but contains only about 2%, by weight, of the impregnated iodine form(s); BC-117 is a new type of iodized charcoal and details on its impregnation are lacking.

Table 1. Identification of the Iodized Charcoals

Charcoal	Lot No.	Mesh Size	Manufacturer or Supplier
MSA 85851	93066[a]	8-14 Tyler	Mine Safety Appliances
BC-727	01345	8-14 Tyler	Barnebey-Cheney
BC-272	4767[a]	8-14 Tyler	Barnebey-Cheney
BC-117	53167[a]	10-14 Tyler	Barnebey-Cheney

a. ORNL-assigned number.

EXPERIMENTAL PROCEDURES

A total of ten experiments were performed; in each experiment there were three separate 2-inch depths of charcoal tested, in effect, providing a total of thirty tests. Also, each 2-inch depth of charcoal under test consisted of two 1-inch depths, in series, so that I_2 efficiencies corresponding to both 1- and 2-inch depths of charcoal could be determined.

The apparatus in which the tests were conducted is schematically illustrated in Fig. 1 and pictured in Fig. 2. Each set of collection beds (three sets per experiment) included a 1-inch depth of unimpregnated charcoal and two 1-inch depths of iodized charcoal; high-efficiency filter media were also included in some of the later tests. The amount of I_2 injected was determined by weighing the U-tube before and after the experiment; due allowances were made for any deposition upstream of the test beds. The I_2 removal efficiencies of each charcoal bed under test were calculated from the results of a quantitative determination (by gamma spectrometry) of the quantity of ^{131}I in each section of the apparatus that had been exposed to the tagged iodine vapor.

The first four experiments, Nos. 1 through 4, were similar with respect to the experimental conditions involved with the exception that the I_2 loading, very high in all four cases, was varied over a factor of about two; the prevailing relative humidity (at the middle of the 2-inch depths of charcoal being tested) was calculated to have been 98%. This value is based upon measurements of pressure differential in the system starting from a point where the air stream was saturated and at room temperature and proceeding downstream.

In experiments Nos. 5 and 6, the charcoal was preequilibrated with respect to moisture content as was done in Nos. 1-4, but, shortly before I_2 injection, water was poured over two of the 2-inch depths of charcoal (Beds A and C) being tested and, also, they were maintained slightly below room temperature during the period of I_2 injection and during a part of the subsequent exposure to flowing air. That is, they were water-logged during this period and the relative humidity was, therefore, 100%. The third of the three 2-inch depths of charcoal under test (Bed B), however,

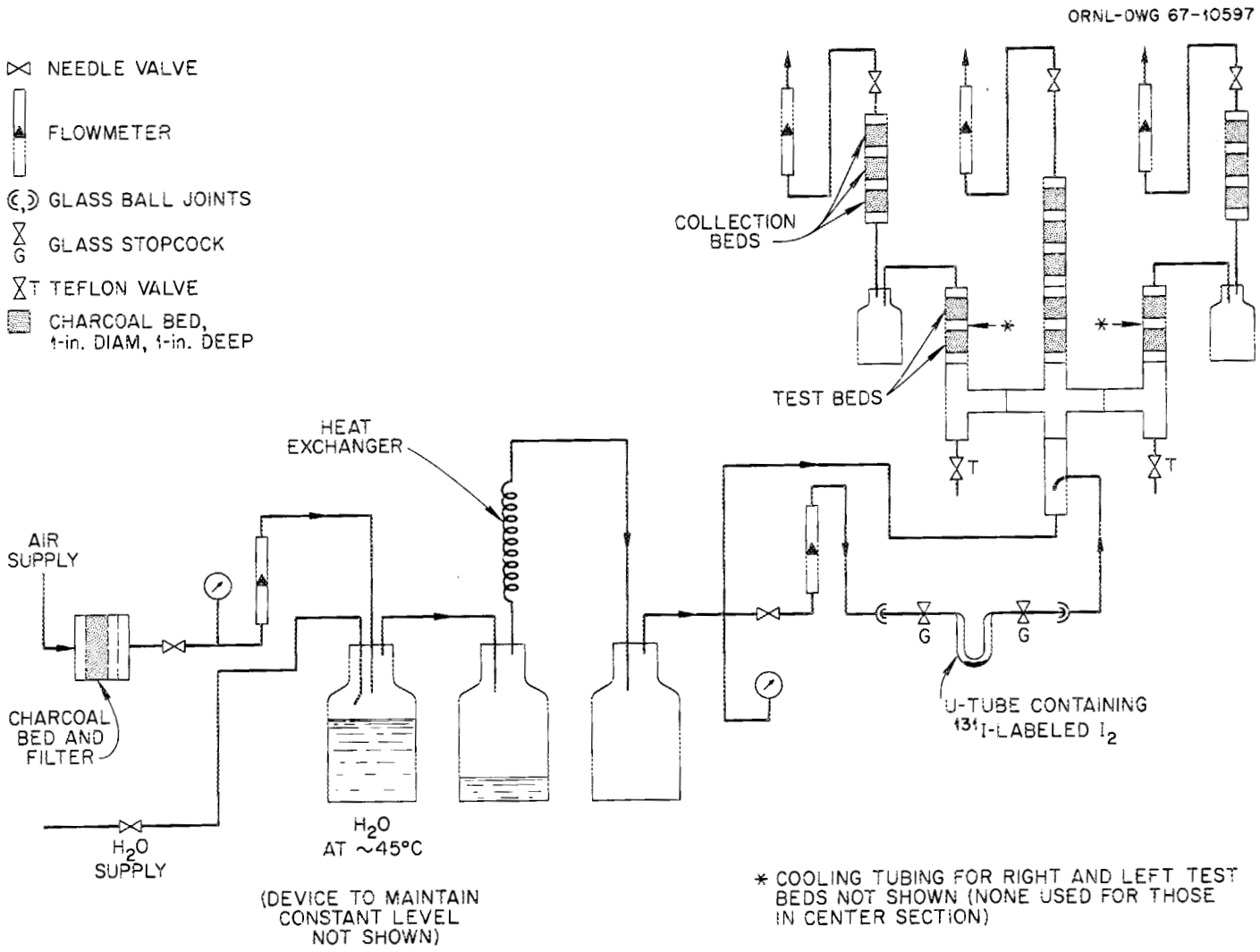

Fig. 1. Schematic of Apparatus Used to Investigate Removal of I_2 from Flowing Air by Iodized Charcoals at Room Temperature.

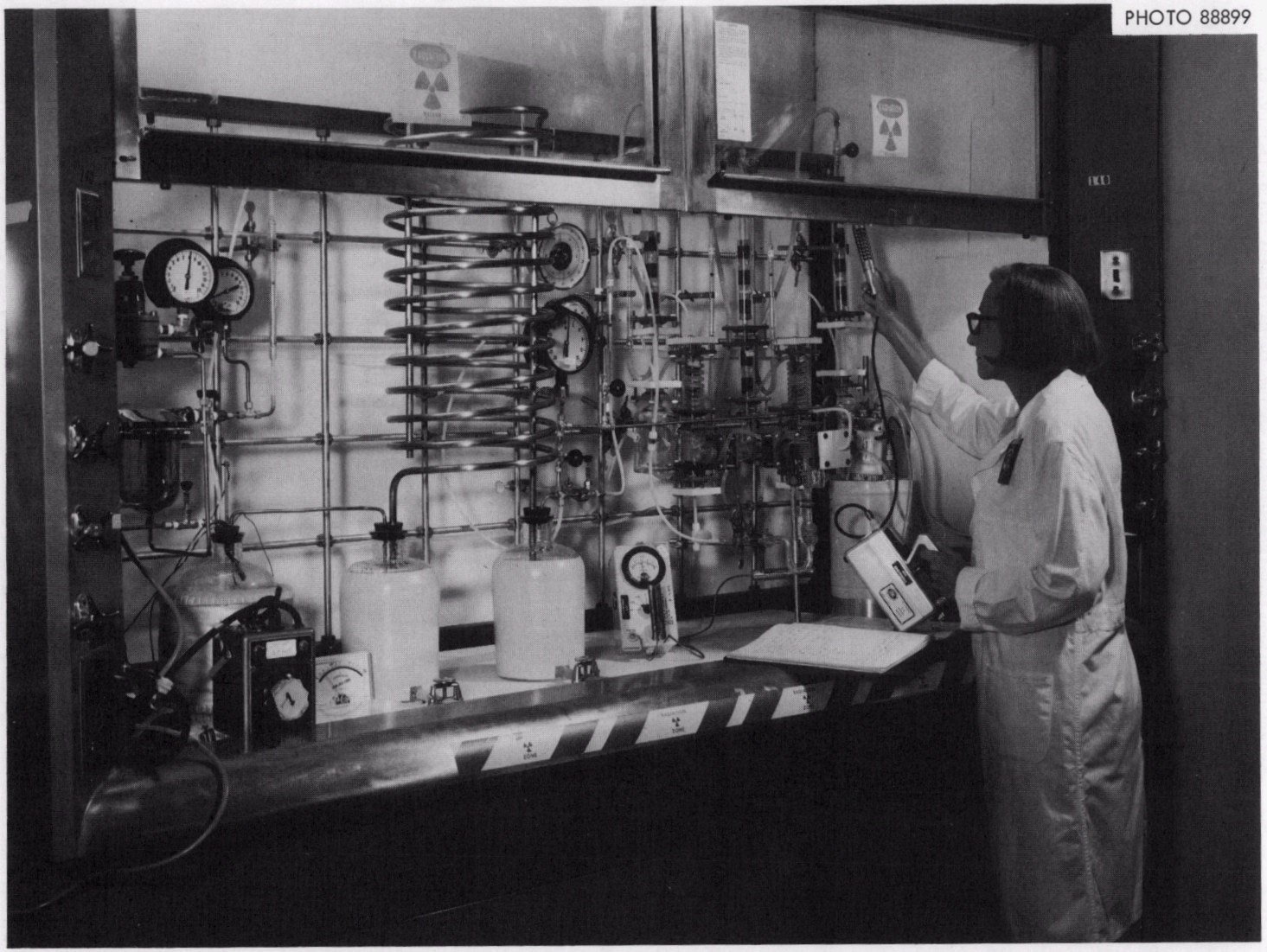

Fig. 2. Photograph of Experimental Setup.

was not so treated and the calculated relative humidity for it was, as before, 98%. Experiments 7 through 10 were performed similarly to experiments 5 and 6 except that precooling of the humidified air entering the water-logged test beds was applied; this variation produced some mist formation slightly upstream of the wetted beds. In order to avoid the problems associated with sampling ^{131}I-bearing charcoal dripping with liquid water, the short period of air sweep subsequent to iodine injection (and while the charcoal was visually wet) was followed by a much longer sweep during which the prevailing relative humidity in the charcoal test beds was slowly decreased from 98% or 100% (whichever applied) to a value of around 50%. The effect on removal efficiency of this additional sweep was negligible since the iodine had already become affixed in the charcoal. No experimental evidence of elution of iodine radioactivity during this purge was noted.

RESULTS AND DISCUSSION

The results, together with the more pertinent of the experimental conditions employed, are given in Tables 2, 3, and 4. Data for experiments Nos. 1-4 are in Table 2, data for the non-wetted test beds (the B group) of experiments Nos. 5-10 are in Table 3, while data for the wetted test beds (the A and C groups) of experiments Nos. 5-10 are in Table 4. Examination of the distribution of radioactivity in the charcoal collection beds downstream of the test bed did not reveal any information of significance regarding the form or character of the iodine penetrating the test bed. In some of the later runs high-efficiency filters were placed downstream of the first 1-inch section of the collection bed in an effort to determine if the radioactivity penetrating this depth of charcoal was

Table 2. Efficiency of Iodized Charcoals for Removing Elemental Radioiodine at Very High Loadings from Flowing Humid Air at Temperatures Around 25°C

Air velocity (superficial): 40 fpm
Relative humidity (R.H.): 98%
System pressure: approximately 1 atm

Experiment Number	Charcoal Under Test[a]	I_2 Injection Time (hr)	Duration of Additional Air Sweep at 98% R.H. (hr)	I_2 Loading[b] for 2-in. Depth of Charcoal (mg/g)	Inlet I_2 Conc. (mg/m^3)	I_2 Removal Efficiency	
						For 1-in. Depth(%)	For 2-in. Depth(%)
1A	MSA 85851	15	10	47	93	99.91	99.947
1B	BC-727	15	10	47	93	99.87	99.89
1C	BC-272	15	10	47	93	99.86	99.90
2A	MSA 85851	10	15	66	196	99.90	99.92
2B	BC-727	10	15	64	190	99.94	99.960
2C	BC-272	10	15	60	178	99.965	99.976
3A	MSA 85851	8	16	34	126	99.985	99.987
3B	BC-727	8	16	34	126	99.968	99.970
3C	BC-272	8	16	34	126	99.973	99.982
4A	MSA 85851	15	9	50	99	99.90	99.93
4B	BC-727	15	9	51	101	99.93	99.948
4C	BC-272	15	9	50	99	99.91	99.955

a. Lot numbers are given in Table 1.
b. This refers to I_2 injected during test and does not refer to impregnant.

Table 3. Efficiency of Iodized Charcoals for Removing Elemental Radioiodine at Fairly High Loadings from Flowing Humid Air at Temperatures Around 25°C

Air velocity (superficial): 40 fpm
Relative humidity (R.H.): 98%
System pressure: approximately 1 atm

Experiment Number	Charcoal Under Test[a]	I_2 Injection Time (hr)	Duration of Additional Air Sweeps		I_2 Loading[b] for 2-in. Depth of Charcoal (mg/g)	Inlet I_2 Conc. (mg/m^3)	I_2 Removal Efficiency	
			At 98% R.H. (hr)	At 98→50%R.H. (hr)			For 1-in. Depth(%)	For 2-in. Depth(%)
5B	BC-727	4	3	65	11	80	99.88	99.93
6B	BC-727	4.25	3	64	16	111	99.957	99.979
7B	BC-727	4	3	64	6	42	99.987	99.993
8B	BC-117	5	2	65	4	34	99.990	99.998
9B	BC-117	5	2	65	6	48	99.995	99.998
10B	BC-727	5	2	64	11	64	99.92	99.998

a. Lot numbers are given in Table 1.

b. This refers to I_2 injected during test and does not refer to impregnant.

Table 4. Efficiency of Water-Logged Iodized Charcoals for Removing Elemental Radioiodine at Fairly High Loadings from Flowing Humid Air at Temperatures Around 20°C

Air velocity (superficial): 40 fpm
Relative humidity (R.H.): 100%
System pressure: approximately 1 atm

Experiment Number	Charcoal Under Test[a]	I_2 Injection Time (hr)	Duration of Additional Air Sweeps		I_2 Loading[b] for 2-inch Depth of Charcoal (mg/g)	Inlet I_2 Conc. (mg/m^3)	I_2 Removal Efficiency	
			At 100% R.H. (hr)	At 100→50% R.H. (hr)			For 1-in. Depth(%)	For 2-in. Depth(%)
5A	BC-727	4	3	65	11	80	99.33	99.993
5C	BC-272	4	3	65	10	77	99.73	99.982
6A	BC-727	4.25	3	64	17	118	96.63	99.89
6C	BC-272	4.25	3	64	15	104	99.57	99.948
7A	BC-727	4	3	64	5	39	98.43	99.90
7C	BC-272	4	3	64	5	36	99.65	99.991
8A	BC-117	5	2	65	4	32	95.96	99.91
8C	MSA 85851	5	2	65	5	30	95.55	99.49
9A	BC-117	5	2	65	6	48	98.64	99.967
9C	MSA 85851	5	2	65	8	48	97.49	99.84
10A	BC-727	5	2	64	10	61	99.33	99.970
10C	BC-272	5	2	64	11	64	99.62	99.982

a. Lot numbers are given in Table 1.

b. This refers to I_2 injected during test and does not refer to impregnant.

associated with a particulate form of iodine or particulate material; no evidence of particulate iodine was found. Actually, the quantity of radioactive material entering the collection beds was so small that very little could be done with it in the effort to identify its form.

The results in Table 2 do not show any pronounced trends. The iodine loadings were deliberately caused to be exceedingly high (much higher than any loading proposed for a full-scale charcoal adsorber system) in order to make immediately apparent any overloading of the remaining capacity of the impregnated charcoals for elemental iodine. As may be judged from the high efficiencies that were obtained, these iodized charcoals under the stated laboratory conditions are effective up to the highest I_2 loading that was studied. It is rather difficult to determine an exact number for the I_2 loading that is applicable throughout the depth of the charcoal bed. Actually, essentially all of the iodine mass is stopped by the first inch of charcoal and, within this inch of depth, the loading varies quite widely from front to back. Since most installed or proposed charcoal adsorber systems in the large power reactors contain a 2-inch depth of iodized charcoal for control of methyl iodide, the I_2 loading will be expressed on the basis of mass of iodine injected divided by the mass of charcoal contained in a 2-inch depth of charcoal under the geometry of our test beds; i.e., 1-inch diameter right cylinder, 2 inches long. If one wishes to observe the loading per 1-inch depth of charcoal, then the loading values in Tables 2-4 should be doubled. The situation with respect to loading at higher temperatures will, more

than likely, not be as favorable, and one of the extensions of this work will be to investigate this point. If the performance of the three charcoals is compared on the basis of the Table 2 results, no differences that can be deemed truly significant are evident; the averages of the 1-inch efficiencies (four) for the individual types varies from 99.924% to 99.927% while, for the 2-inch efficiencies, the analogous range is 99.942 % to 99.953%.

In Table 3, the I_2 loadings are still high, but considerably lower than those of the preceding table. This may be the explanation for the 1- and 2-inch efficiencies for BC-727 in Table 3 being, on the average, somewhat higher than the respective values in Table 2. The data for type BC-117 charcoal, while limited in number, indicate that it is highly effective for trapping I_2 under these conditions.

Considering now the results for water-logged charcoal in Table 4, the variation in performance imposed by this condition appears to obscure any small variations due to other factors, as is illustrated most dramatically by the 1-inch efficiencies. For instance, the 1-inch value for experiment 7A is only 98.43% while that for 10A is 99.33% and there is no evidence to suggest that the controllable experimental conditions of 7A were more severe than those of 10A. The probable explanation is that the degree of sweep gas channeling was higher in the test producing the lower efficiency. Provided this explanation is correct, good I_2 removal efficiency can be obtained with a 1-inch depth of iodized charcoal even when water-logged and excellent removal efficiency for I_2 can be obtained with a 2-inch depth if sweep gas channeling can be limited. Of course, control of channeling would be almost impossible to achieve

and it would be preferable to avoid altogether having the iodized charcoal in a water-logged condition, not only from the standpoint of trapping elemental radioiodine, but also with regard to the trapping of radioactive methyl iodide, as has been discussed elsewhere (4-6).

The apparent I_2 removal efficiency of charcoal has, in certain studies, been observed to be relatively low when the inlet iodine concentration was low (8). This has been explained on the basis of a greater proportion of the I_2 being converted to more penetrating forms such as methyl iodide and/or iodine-bearing particulates. Consequently, another planned extension of the present work is to investigate this concentration effect as it applies to iodized charcoals.

CONCLUSIONS

1. Certain types of iodized charcoal (four have been tested) are highly effective at around 25°C for trapping elemental radioiodine from flowing air at high relative humidity (98%) and at extremely high I_2 loadings on the charcoal.
2. Iodized charcoal when it is water-logged (100% relative humidity plus liquid water in the bed) may exhibit either fairly poor or good I_2 removal efficiency (at around 20°C). This erratic performance is probably associated with sweep gas channeling, and whether or not channeling occurs appears to be on a somewhat random basis.
3. Further investigation of I_2 trapping by iodized charcoals is needed and should include effects of elevated temperature and of low I_2 concentration in air on removal efficiency.

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390. General Atomic Library (Attn: A. J. Goodjohn)
391-405. Division of Technical Information Extension (DTIE)
406-407. Reactor Division ORO
408. Laboratory and University Division, ORO