Radioiodine canisters, cartridges, and their sorbents were tested with vapors of iodine compounds. The purposes were (1) to select a species for use in performance testing, (2) to determine how useful nonradiometric species would be for such testing, (3) to study effects of humidity and other parameters on sorbent performance, and (4) to make limited comparisons of canisters and cartridges currently available from U.S. manufacturers. An experimental apparatus was constructed to generate normal iodine and radioiodine vapors and to detect their penetrations through test beds. Methyl iodide was found to be the most penetrating vapor form. It provides an upper limit to methyl radioiodide penetration. At high humidity, methyl iodide removal deteriorates rapidly. Triethylenediamine-impregnated charcoal is the most effective charcoal tested. Larger beds give higher efficiencies.

Respirator canister testing for radioiodine*

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introduction

The fission product radioisotope iodine-131 (^{131}I) in airborne respirable forms represents a potential health hazard in nuclear environments. The toxicity of this radioisotope of high specific activity (1.24×10^5 Ci/g) and moderate half-life (8.05 d) is enhanced by the physiological process of concentrating iodine in the thyroid. In routine and emergency maintenance or repair operations it is sometimes necessary for a worker to approach a source of radioiodine, so that personal respiratory protection is required. Presently, the Nuclear Regulatory Commission (NRC) limits the use of air-purifying respirators to protection against radioactive materials present as particulate aerosols for which high-efficiency respirator filters have been shown to be effective.(1,2) While unapproved respirator canisters and cartridges are manufactured to collect iodine and radioiodine vapors, the NRC restricts their use due to (1) unknown collection efficiencies of sorbent cartridges for vapors of radioiodine compounds, (2) unknown service lives for providing adequate protection, and (3) no warning of breakthrough.

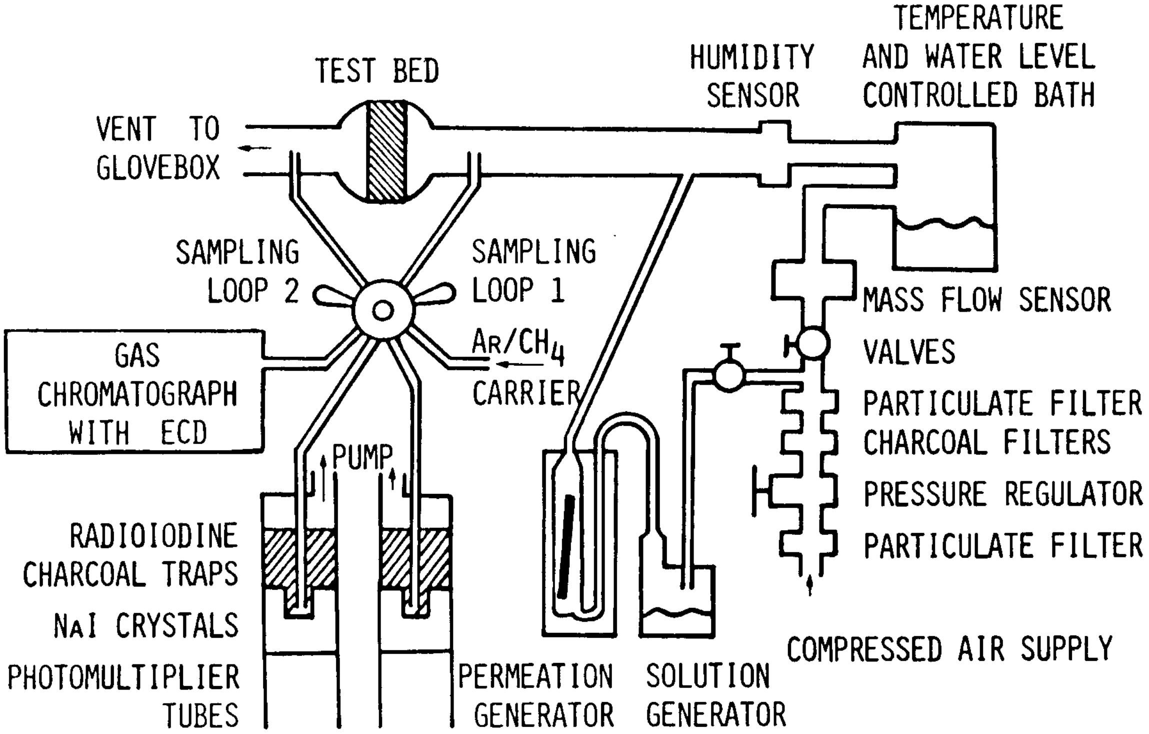

Figure 1 — Experimental apparatus for testing air-purifying respirator cartridges and canisters using radioiodine and normal iodine vapor species.

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*This work was supported by the Nuclear Regulatory Commission and performed at the Los Alamos Scientific Laboratory operated under auspices of the U. S. Department of Energy, Contract No. W-7405-ENG-36.

TABLE I
Characteristics of Canisters and Cartridges Tested

			Charcoal Bed Geometry[A]			Charcoal Impregnants[B]
Source	Type	Designation	Cross, Section (cm^2)	Depth (cm)	Volume (cm^3)	(Weight Percent)
MSA	Canister	GMR-I	110[C]	3.2	350	5% KI_3
	Canister	GMR-I (TEDA)[D]	110[C]	3.2	350	5% KI_3, 2% TEDA[E]
	Canister	GMR-S	110[C]	3.2	350	Metal and Ammonium Salts[F]
Scott	Canister	600252-75	87	3.8	330	5% TEDA
	Canister	282 OAP-R	87	3.8	330	Metal and Ammonium Salts[F]
	Cartridge	604550-75	48	1.3	62	5% TEDA
	Cartridge	604403-75	48	1.3	62	5% TEDA
Norton	Cartridge	Type I[G]	44	2.4	106	5% TEDA
	Cartridge	Type II[H]	44	2.4	106	5% TEDA

[A] Measured from opened canisters.

[B] Best information from manufacturers.

[C] Oval cross section.

[D] The GMR-I (TEDA) designation is used for GMR-I canisters manufactured after July, 1979 through at least April, 1980.

[E] TEDA = triethylenediamine.

[F] Whetlerized charcoal.

[G] Granule size 8-16 mesh.

[H] Granule size 12-20 mesh.

In the Industrial Hygiene Group of the Los Alamos Scientific Laboratory, we have accepted the responsibility of providing the NRC with information and guidance regarding the use of air-purifying respirators for radioiodine. A test procedure and approval schedule is being developed that will allow certification of respirator canisters or cartridges for this application. The Testing and Certification Branch of the National Institute for Occupational Safety and Health has indicated that it would promulgate regulations for approval, if techniques and instrumentation were developed. The NRC would authorize the use of approved units for certain applications.

Previous research on this project established the feasibility of constructing a respirator canister of adequate efficiency (protection factor = 1000).(3) A preliminary test system was built and used to measure breakthrough curves for methyl iodide vapor and potential radioiodine sorbents. Effects of environmental and sorbent bed design parameters on the retention of methyl iodide were studied. These parameters included relative humidity, flow rate, bed diameter, bed depth, and challenge concentration. Observed effects of challenge concentrations (1-1000 ppm) on canister service life were explained in terms of Freundlich adsorption isotherms.

Additional studies, reported here, of commercial radioiodine canisters and cartridges and their sorbent contents have been done to establish: (1) What iodine vapor species should be used for a test of effectiveness? (2) Can a nonradiometric test using normal ^{127}I species give results representative of effectiveness against radioiodine species? (3) What are the effects of conditions such as humidity and flow rate (work load)? (4) How well do currently available sorbents, canisters, and cartridges work?

experimental

apparatus

The apparatus used to measure the penetrations of volatile iodine and radioiodine compounds through test beds, canisters, and cartridges is diagrammed in Figure 1. It was built inside a fume hood to exhaust any toxic vapors which might have been released. Radioiodine solutions and contaminated sorbents were contained for further safety within a glove box with charcoal and HEPA exhaust filters. Vapor generation and test bed exposures were done within the glove box. Compressed air was filtered, regulated for proper flow rate, and humidified before entering the glove box. An electronic mass flow meter (Datametrics 800-L) which monitored airflow was periodically checked using a dry test meter (Singer DTM-325) at the test bed location. Humidification was accomplished by passing air through the headspace over a heated water reservoir. A humidity monitor/controller (Phys-Chemical Research Corp.) which regulated water temperature was calibrated with a dew point hygrometer (EG & G 911) at the test bed location. Water level was maintained automatically by a conductive liquid level control (Lumenite Electronic Co.).

Vapors were generated in two ways shown in Figure 1. Liquid methyl iodide and methyl radioiodide sealed in a Teflon permeation tube were released at a steady rate by permeating into a 500 cm^3/min airflow. Temperature control (25-70 °C, ±0.1 °C) of this permeation tube was by the Calibration System (Analytical Instrument Development, Inc., Model 303). Alternately, methyl radioiodide ($CH_3{}^{131}I$), elemental radioiodine ($^{131}I_2$), and hyporadioiodous acid ($HO^{131}I$) were generated from aqueous solutions. A syringe was used to inject 10 mL of solution into 100 mL of distilled water or other reagent solution in the glass container in the

TABLE II
Radioiodine Test Results for Canisters[A]

Canister Type	Test Vapor	Average Percent Instantaneous Penetrations (and Standard Deviations)[B] 0-2 h	2-4 h	4-6 h	6-8 h	8-10 h
Scott 600252-75	$CH_3{}^{131}I$	-0-	---	0.24 (0.02)	---	---

		1.07 (0.06)	-0-	0.61 (0.05)	---	---
	$^{131}I_2$	-0-	-0-	-0-	---	---
	$HO^{131}I$	---	-0-	0.10 (0.03)	0.08 (0.03)	-0-
GMR-I (TEDA)	$CH_3{}^{131}I$	0.24 (0.08)	4.43 (0.16)	6.09 (0.17)	---	---
		0.99 (0.41)	2.46 (0.73)	7.54 (1.06)	---	---
	$^{131}I_2$	0.71 (0.04)	0.10 (0.02)	0.10 (0.02)	---	---
	$HO^{131}I$	---	0.08 (0.04)	-0-	-0-	-0-
GMR-I	$CH_3{}^{131}I$	3.34 (0.52)	8.40 (0.53)	21.29 (0.65)	---	---
	$^{131}I_2$	0.17 (0.02)	0.07 (0.01)	-0-	---	---
	$HO^{131}I$	---	-0-	0.07 (0.03)	0.17 (0.07)	0.11 (0.03)
Scott 282 OAP-R	$CH_3{}^{131}I$	19 (2)	98 (4)	100 (7)	---	---
	$^{131}I_2$	-0-	0.07 (0.01)	0.18 (0.02)	---	---
	$HO^{131}I$	---	-0-	-0-	0.27 (0.04)	0.75 (0.04)
GMR-S	$^{131}I_2$	-0-	-0-	-0-	---	---
	$HO^{131}I$	-0-	-0-	-0-	---	---

[A] 64 L/min, 97 ± 3% RH.

[B] Zero value (-0-) means not significantly greater than zero at the 95% confidence level. Dash (---) means not measured.

lower center of Figure 1. The volatile iodine compounds in this stirred mixture entered the head space and were swept by 0.5 L/min of air through Teflon and glass tubing into the main airflow. Water vapor was also generated. Output of volatiles from solution dropped exponentially from the time of injection. Generator output and main airstream passed through sufficient length of 2.4-mm-i.d. glass tubing and two elbows to mix thoroughly before entering the test bed. Sections of the glass flow system and the test bed were connected with O-ring seals and clamps. Challenge air and test bed effluent air were sampled continuously through Teflon tubes connected to the glass system.

The detector for methyl iodide was a gas chromatograph (Varian 1520) with a linearized electron capture detector (Tracor Instruments). Air from upstream and downstream of the test bed was drawn (0.8 L/min) through matched Teflon sampling loops attached to a 10-port valve (Valco Instrument Co.) of Hastalloy-C (for inertness). This valve was pneumatically actuated by a digital valve sequence programmer (Valco Instrument Co.) to alternately inject the upstream and downstream air at 5-min intervals. The chromatographic column was 1.8-m × 4-mm-i.d. glass packed with 15% OV-7 on 100/120-mesh Chromosorb G. Operating conditions were 100 °C and 20 cm^3/min 19:1 $Ar:CH_4$ carrier gas. An electronic peak integrator (Spectra Physics Minigrator) quantitated the methyl iodide peaks and recorded elapsed times. Calibrations of this analytical system were made using weighed permeation tubes to generate known methyl iodide concentrations in air.

The radiometric detectors continuously collected and measured ^{131}I from the 0.8 L/min air samples passing through the gas chromatograph sampling valve. Figure 1 shows the charcoal trap and 7.6-cm-diam × 7.6-cm-thick NaI(Tl) well-type (52-mm deep × 29-mm-diam) scintillation crystal with integral photomultiplier tube (Harshaw Chemical Co.). High-efficiency charcoals were used: 5%-TEDA impregnated (Barnebey Cheney CN 2762) for $CH_3{}^{131}I$ and activated charcoal (Union Carbide ACC) for $^{131}I_2$ and $HO^{131}I$. The majority of radioiodine was collected at the bottom of the well, resulting in good detection efficiencies (~0.5) for the 0.364 MeV gamma-ray.(4) Each detector for upstream and downstream air had its own preamplifier, amplifier, single-channel analyzer, and counter (all from Ortec). They shared the power bin (Ortec), high voltage power supply (Canberra), timer (Ortec), and printer (Ortec). Linear-log rate meters (Mech-Tronics) were used for count rate monitoring. Detector counts were taken for 5-min intervals and printed together. Each detector trap and crystal was shielded by 5 cm of lead to reduce background counts. Fresh charcoal was placed in the detector traps for background counts before each new bed was tested. The detectors were calibrated almost daily for relative sensitivities by sampling the same radioiodine-containing air.

TABLE III
Radioiodine Test Results for Cartridges[A]

Cartridge Type	Test Vapor	Average Percent Instantaneous Penetrations (and Standard Deviations)[B] 0-2 h	2-4 h	4-6 h
Norton Type I	$CH_3{}^{131}I$	0.03 (0.01)	1.94 (0.06)	3.34 (0.60)
	$^{131}I_2$	-0-	-0-	-0-
Norton Type II	$^{131}I_2$	-0-	-0-	-0-
Scott 604403-75	$CH_3{}^{131}I$	1.18 (0.33)	9.27 (0.29)	11.50 (0.34)
	$^{131}I_2$	0.04 (0.01)	-0-	-0-
Scott 604550-75	$CH_3{}^{131}I$	1.98 (0.17)	10.71 (0.99)	12.87 (0.90)

[A] 32 L/min, 97 ± 3% RH.

[B] Zero value (-0-) means not significantly greater than zero at the 95% confidence level.

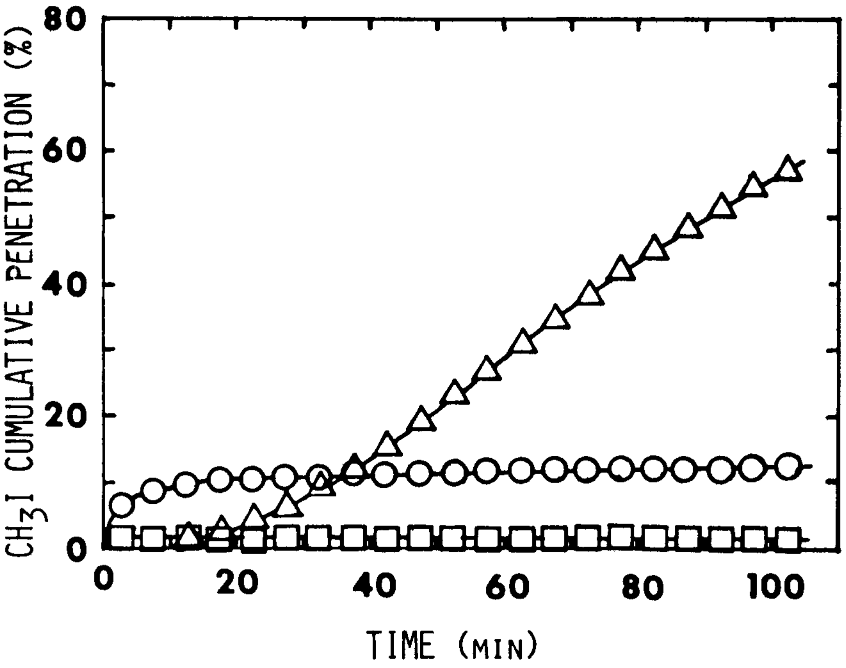

Figure 2 — Methyl iodide cumulative fractional penetrations as functions of time for charcoals from three respirator canisters: □, Scott 600252-75 (5% TEDA); ○, MSA GMR-I (5% KI_3 + 2% TEDA); △, MSA GMR-I (5% KI_3).

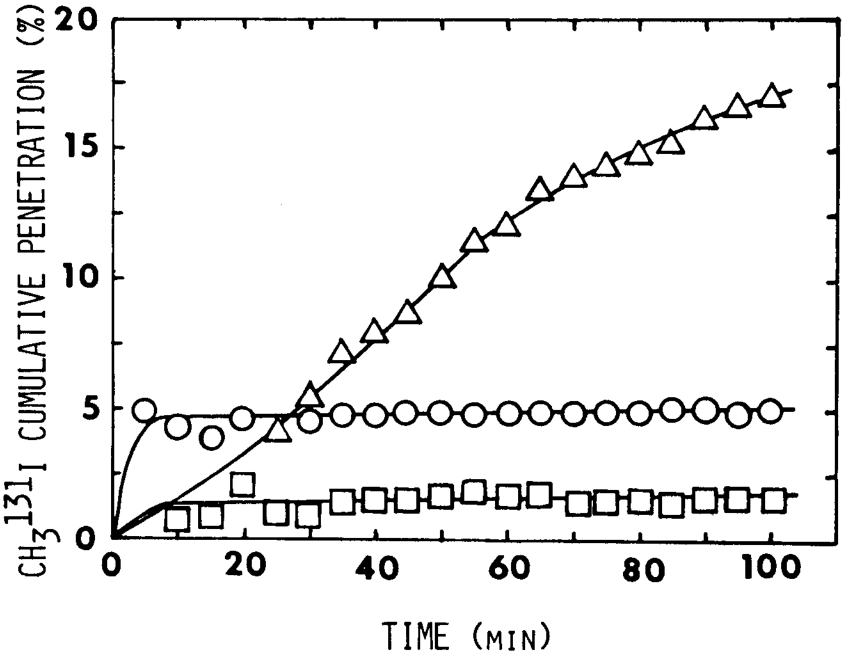

Figure 3 — Methyl radioiodide cumulative fractional penetrations as functions of time for charcoals from three respirator canisters: □, Scott 600252-75 (5% TEDA), ○, MSA GMR-I (5% KI_3 + 2% TEDA), △, MSA GMR-I (5% KI_3).

reagents

The source for radioiodine 131 was ICN Chemical and Radioisotope Division, Irvine, CA. Methyl radioiodide was ordered as 5 mCi ^{131}I in 1-3 mL of total methyl iodide. Stated purity was at least 99%. Two milliliters were used to fill a permeation tube and 1 mL was dissolved in 1 L of double distilled water. This aqueous solution (2.3 g/L or 0.016 mol/L) was used 10 mL at a time for generating as described above. Radioiodine in the form of NaI in 0.05 N NaOH was purchased in a 5 mCi amount. Stated purity was at least 99% with an $^{127}I/^{131}I$ ratio less than 10. This material of about 1 mL volume was added to 1 L H_2O containing 0.127 g of dissolved I_2 (0.127 g/L or 5×10^{-4} mol/L). Isotope exchange occurred to form $^{131}I_2$. This solution was used to generate both $^{131}I_2$ and $HO^{131}I$. For $HO^{131}I$, 10 mL of this latter solution were injected into 100 mL of 4×10^{-3} mol/L $NaIO_3$ at pH = 2 to cause the reactions:(5)

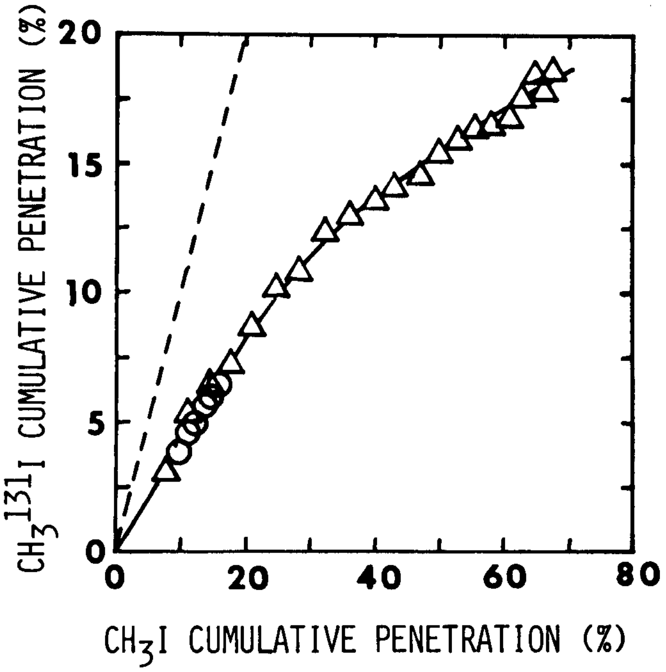

Figure 4 — Comparisons of cumulative fractional penetrations of methyl radioiodide and methyl iodide for two 5% KI_3 charcoals: Δ, GMR-I; O, GMR-I (TEDA).

$$2\,I_2 + IO_3 + 6\,H^+ + 2\,H_2O \rightleftharpoons 5\,H_2OI^+$$

$$H_2OI^+ + H_2O \rightleftharpoons HOI + H_3O^+ .$$

No attempt was made to determine the extent of HOI production, since no analytical method is known which distinguishes this unstable species from I_2.

test beds

Air-purifying respirator canisters and cartridges were obtained from three U.S. commercial sources; Mine Safety Appliances Company (MSA), Pittsburgh, PA; Norton Company, Safety Products Division, Cranston, RI; and Scott, Health/Safety Products, South Haven, MI. They each claimed, by labelling or personal manufacturer information, to be of some use for protection against iodine or radioiodine vapors. Each type contained a particulate filter followed by a sorbent bed containing a coarse grained charcoal. The distinction which is made in this paper between canisters and cartridges is that the latter are used in pairs and are physically smaller. For some experiments beds of 2.4-cm diam were prepared from charcoals taken from canisters. The term "test bed" will be used in this paper to refer to a canister, a cartridge, or an experimental bed. Table I lists characteristics of the canisters and cartridges and their charcoal contents.

results

comparison of vapor species

Penetration test results at high humidity (97 ±3%) for the three radioiodine vapor species are tabulated in Table II for five canisters (64 L/min) and in Table III for four cartridges (32 L/min). Pulses of challenge vapor were generated from solution at 2-h intervals. Two-hour average penetrations and standard deviations (given in parentheses) were determined by linear regression analysis of 5-min counts in the downstream detector vs the upstream detector. Relative sensitivity of the two radioiodine detectors determined by daily calibrations was taken into account. Any penetrations calculated to be within 95% confidence levels of zero were considered as zero and listed as -0-. In only two cases were more than one canister or cartridge of a type tested for a given radioiodine vapor. Therefore, these results cannot reflect variations within a given type. Each canister or cartridge was tested at least three successive times.

methyl iodide vs methyl radioiodine

Cumulative fractional penetrations through three types of impregnated charcoals are compared for methyl iodide (Figure 2) and for methyl radioiodide (Figure 3). The test beds, 3.75-cm deep by 2.4-cm diam consisted of charcoals taken from MSA GMR-I, GMR-I (TEDA), and Scott 600252-75 canisters. Each bed was preconditioned for 2 h at the test conditions of 3 L/min airflow and 86 ± 3% relative humidity before being challenged with 1.5 ppm (7 mg/m^3) methyl iodide tagged with ^{131}I. Cumulative fractional methyl radioiodide penetrations were calculated directly from 5-min interval counts of radioiodine trapped in the detectors. Cumulative fractional penetrations of methyl iodide were calculated by integrating instantaneous upstream and downstream concentrations determined by gas chromatography. The 5% TEDA-impregnated charcoal from the Scott canister was the most efficient, allowing nearly constant 1.5 ± 0.5% penetration of both methyl iodide and radioiodine throughout the experiment. The GMR-I (2% TEDA, 5% KI_3) was less efficient at about 10 ± 2% methyl iodide penetration and 5 ± 1% methyl radioiodide penetration after an initial equilibration period. The GMR-I (5% KI_3) charcoal was most efficient at the beginning of the experiments,

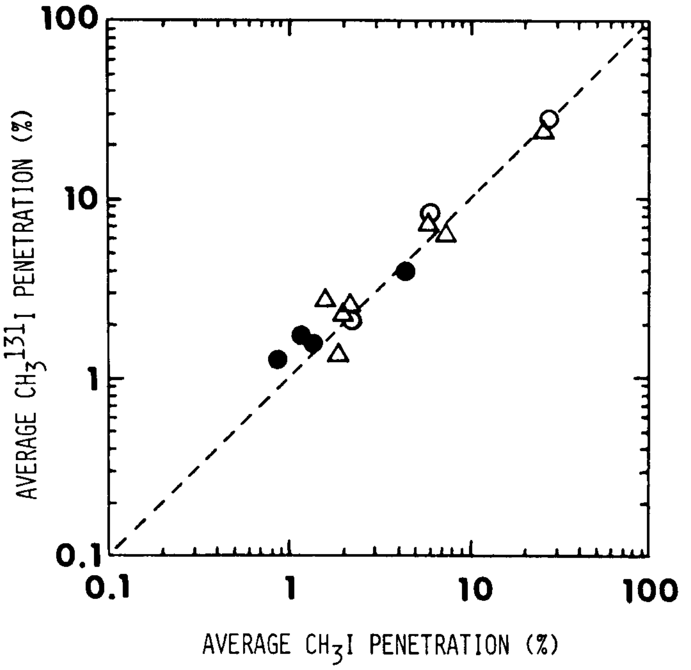

Figure 5 — Comparison of average fractional penetrations of methyl radioiodide and methyl iodide for a 5% TEDA impregnated charcoal (Scott 600252-75). Permeation tube generation: O, 97% RH and ●, 85% RH; aqueous solution generation: Δ, 97% RH.

but rapidly and steadily deteriorated to give a 60% cumulative fractional methyl iodide penetration and a 17% cumulative fractional methyl radioiodide penetration by 100 min.

Results from experiments with iodized charcoals are compared in Figure 4, which shows $CH_3{}^{131}I$ cumulative fractional penetration vs CH_3I cumulative fractional penetration. The data points all fall below the equality (dashed) line, *i.e.*, CH_3I penetration greater than $CH_3{}^{131}I$ penetration. Also, in the region of practical interest (less than 10% penetration) the difference is apparently only a small and constant factor, about two in these cases.

A more-extensive comparison of fractional penetrations for Scott (5% TEDA) beds is summarized in Figure 5. These results are from 14 experiments at two humidities, for two generation methods, and for three bed depths (1.25-3.75 cm). Each graphed point represents the average of 20 to 30 measurements for a given experiment. The penetration values all fall close to the theoretical (dashed) equality line. Therefore, for this type of sorbent (TEDA only) measurements of molecular CH_3I penetrations are direct measurements of the ^{131}I penetrations when the radioiodine challenge is in the form of $CH_3{}^{131}I$.

A fourth type of charcoal, from an MSA GMR-S canister, was tested to compare methyl iodide and radioiodide penetrations. This Whetlerized charcoal is impregnated with metal and ammonium salts, but contains no impregnants that react with methyl radioiodide. Therefore, removal of ^{131}I in $CH_3{}^{131}I$ can occur only by physical adsorption of the molecule. Cumulative fractional penetrations of methyl iodide and methyl radioiodide are compared in Figure 6 for duplicate experiments. The data points closely fit the equivalence line until the amount desorbing from the test bed equals that entering it. Then there was a slight deviation in the direction of greater radioiodine penetration than methyl iodide penetration. This deviation is explained as the result of forming volatile iodides other than methyl iodide. The radioiodine detector is not compound specific, but the gas chromatograph is. Instantaneous fractional penetrations (effluent concentration/challenge concentration) of methyl iodide increased with time and even exceeded 100% as the vapor adsorbed at the beginning was displaced into the air. Breakthrough times of methyl iodide average 33 ± 3 min at 0.1%, 49 ± 6 min at 1%, and 68 ± 8 min at 10% instantaneous penetration.

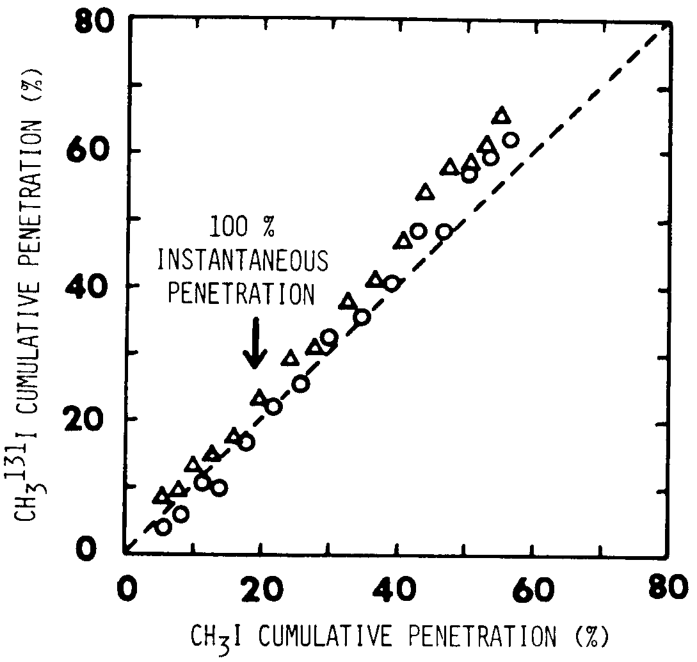

Figure 6 — Comparisons of cumulative fractional penetrations of methyl radioiodide and methyl iodide for a Whetlerized charcoal (GMR-S) for two separate experiments (Δ and O).

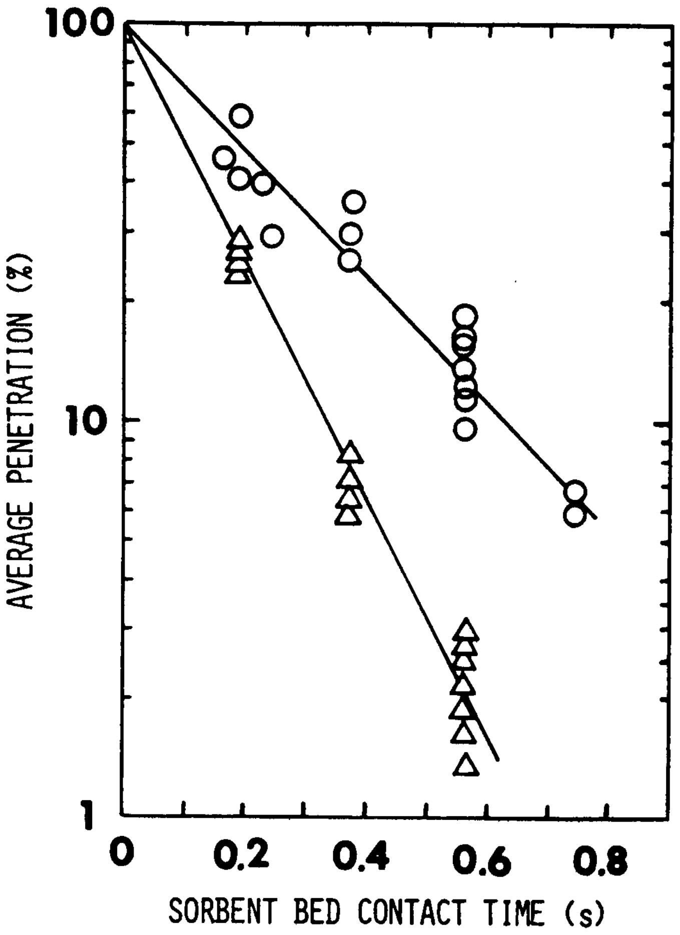

Figure 7 — Average instantaneous fractional bed penetrations as logarithmic functions of bed contact times: Δ, methyl iodide and methyl radioiodide for a 5% TEDA charcoal (Scott); O, methyl radioiodide for a 5% KI_3 charcoal (GMR-I).

removal kinetics

Another series of experiments with the TEDA and KI_3-impregnated respirator canister charcoals was done to establish the rate orders of methyl iodide and radioiodide removal. The ranges of test conditions were:

Bed depth: 1.25 - 3.75 cm
Bed diameter: 2.4 cm
Airflow rate: 1.8-4.2 L/min or 6.7-15.3 cm/s
Bed residence time: 0.16-0.75 s
Relative humidity: 86 ± 3%
Concentrations: 6-1200 nCi/m^3 ^{131}I
0.19-72 mg/m^3 CH_3I
Conditioning Period: 2 h

TABLE IV
First Order Rate Coefficients for Methyl Iodide and Radioiodide Removal[A]

Charcoal Impregnant	Charcoal Source	Rate Coefficient (s^{-1}) Total $CH_3{}^{131}I$	Total CH_3I	Isotope Exchange[B]
5% TEDA	Scott 600252-75	6.9 ± 0.5	7.1 ± 0.3	None
5% KI_3	GMR-I	3.6 ± 0.3		3.6 ± 0.3
5% KI_3 + 2% TEDA	GMR-I (TEDA)	4.9 ± 0.1	3.0 ± 0.1	1.9 ± 0.2

[A] 2-h preconditioning at 86% RH.

[B] Difference of preceeding two columns.

Seventeen tests with methyl radioiodide generated from a permeation tube were done using iodized charcoal from an MSA GMR-I canister. In each test the radioiodine penetration remained nearly constant, while methyl iodide penetration increased steadily with time until it exceeded 100%. When the logarithms of radioiodine penetration fractions were plotted against bed contact times a straight line with an intercept of 1.0 resulted (Figure 7). This indicates that the radioiodine removal reaction (isotope exchange) is simple first order in methyl radioiodide concentration. Molecular methyl iodide breakthrough times of 4.6 ± 1.6 min at 1%, 17 ± 2 min at 10%, and 42 ± 3 min at 50% were independent of challenge concentrations (0.19-3.8 mg/m^3). This indicates that the physical adsorption isotherm is linear for this molecule and sorbent.

Ten such experiments using 5% TEDA charcoal from Scott 600252-75 canisters were also done at similar conditions. Semilog plots for penetration fractions (both methyl iodide and radioiodide) vs bed contact times (Figure 7) showed that the chemisorption reaction is also by a simple first order rate. Both methyl iodide and radioiodide are removed from air at the same rate. Four experiments were also done at different bed depths (1.25-5.0 cm) using charcoal from MSA GMR-I (TEDA) canisters. Penetrations of methyl iodide and radioiodide during each run were both constant, but not equal. The difference for this mixed impregnant (2% TEDA and 5% KI_3) sorbent is due to isotope exchange. Average first-order rate coefficients calculated from the slopes of plots such as Figure 7 are listed in Table IV.

exposure effects

When canisters or cartridges were tested more than once with methyl radioiodide at 2-h intervals while continuously exposed to very high humidity (97 ± 3% RH) air, increasing penetrations were observed. This is illustrated by the results in Tables II and III. Such an effect could be due to (1) loading the test bed with methyl iodide in previous tests and/or (2) loading it with water vapor by exposure to large volumes of high humidity air. Studies were done to sort out these effects using Scott 600252-75 canisters (5% TEDA charcoal) at 64-L/min airflow rate. Methyl iodide was generated from aqueous solutions (0.23 g/100 mL) at selected times while a canister was exposed to high (97 ± 3%) or medium (50 ± 3%) relative humidity air. Penetration results vs exposure times are shown in Figure 8. Box sizes represent

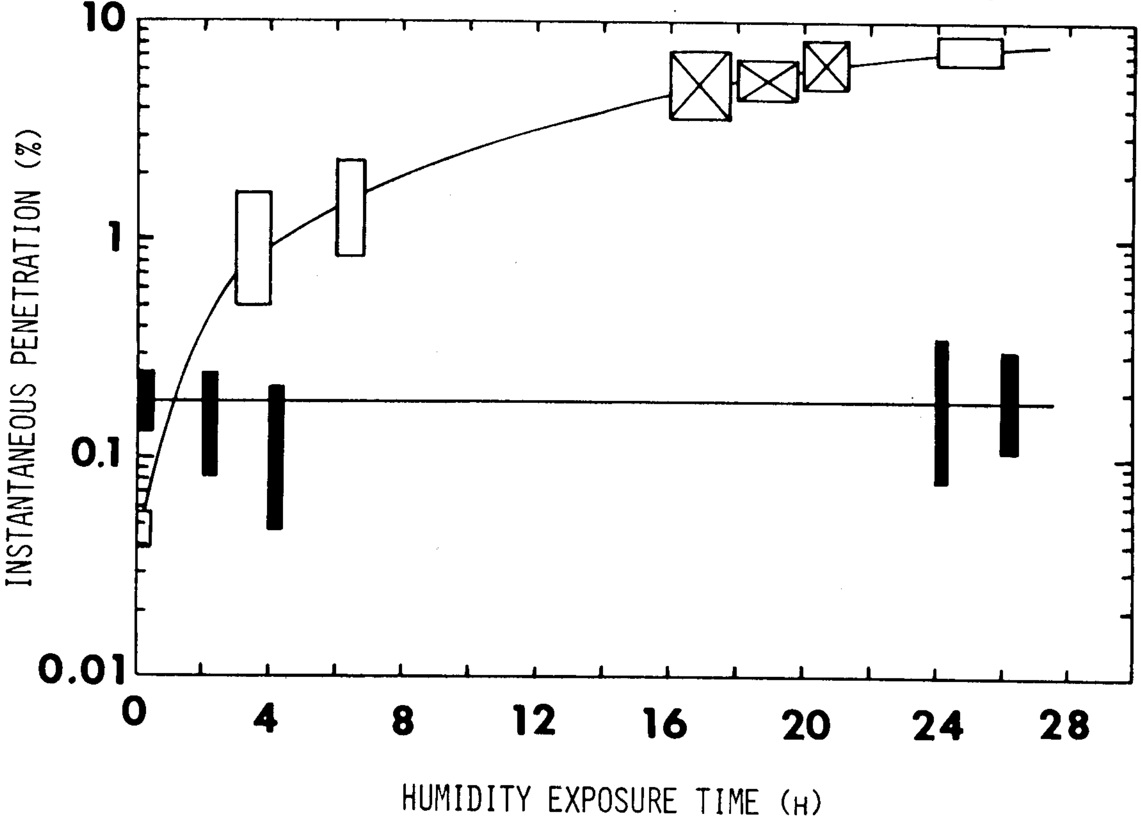

Figure 8 — Average instantaneous fractional penetrations of methyl iodide through Scott 600252-75 (5% TEDA) canisters as functions of exposure times: □, 97 ± 3% RH; ■, 50 ± 3% RH.

ranges of data obtained. In the first experiment (open rectangles) a fresh canister was tested after 0, 3, 6, and 24-h exposures to 97% RH air. Penetrations increased by over two orders of magnitude. In the second experiment another canister was exposed at this humidity for 16 h before being tested at 16, 18 and 20 h. These data (shown as rectangles with x's) fell on the same curve as those from the first experiment. In the third experiment (solid rectangles) another canister was tested at the 50% RH and 0, 2, 4, 24, and 26-h exposures. Even for the longest time and highest bed loading the penetration at 50% RH was not significantly changed from the beginning. These results clearly show that the deterioration in canister effectiveness was due to the high humidity of the air and not due to methyl iodide loading.

cartridge comparisons

Four cartridges, all containing 5% TEDA-impregnated charcoals, were tested with methyl radioiodide at 0, 2, and 4-h exposure times to 32 L/min, 97 ± 3% RH air. Methyl iodide penetrations again increased with exposure times. Maximum penetrations (humidity equilibrations) were reached in about 3-4 h. Average penetrations measured during 4-6 h by gas chromatography and by radiometric counting are listed in Table V. The values from the two methods are in good agreement. Cartridges with larger sorbent volumes (Table I) of similar sized and impregnated charcoals gave lower penetrations. That this is due simply to increased bed contact time is shown in Figure 9. This semilog plot also includes data from Table II for the Scott 600252-75 canister. The average first order rate coefficient is 17.6 s^{-1} (standard deviation = 1.3s^{-1}). This correlation should be useful for improving efficiencies by redesigning canisters and cartridges.

conclusions

A. Methyl iodide was the vapor form of radioiodine that most readily penetrated the respirator canisters and cartridges which were tested. Penetrations of $^{131}I_2$ and $HO^{131}I$ at high humidity were low (≤0.75%) and, with one exception, did not increase significantly with exposure and loading.

TABLE V
Cartridge Penetration Fractions of Methyl Iodide and Methyl Radioiodide after Equilibration at High Humidity[A]

Cartridge Type	Average Percent Instantaneous Penetrations (and Standard Deviations)		Bed Contact Time (s)	Charcoal Granule Mesh
	CH_3I	$CH_3{}^{131}I$		
Norton Type I	3.75 (0.76)	3.34 (0.06)	0.20	8-16
Norton Type II	1.50 (0.15)		0.20	12-20
Scott 604403-75	13.24 (0.59)	11.50 (0.34)	0.12	8-16
Scott 604550-75	16.52 (0.61)	12.87 (0.90)	0.12	8-16

[A] 32 L/min, 97 ± 3% RH, 4 h.

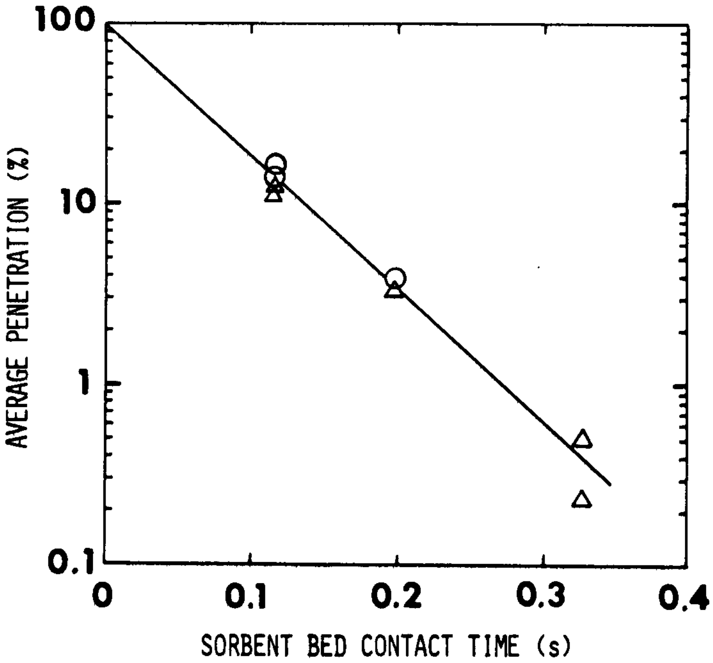

Figure 9 — Dependence of average instantaneous fractional penetrations on bed contact times for canisters and cartridges containing 5% TEDA impregnated charcoals: O, methyl iodide; Δ, methyl radioiodide.

Since methyl iodide is the most volatile organic iodide compound, other organic iodides should be retained on these canisters or cartridges with the same or higher efficiencies. Therefore, methyl iodide should be used as the test species to determine the upper limit penetration of vapors containing radioiodine.

B. Normal methyl iodide can be used to determine the upper limit of penetration to be expected for methyl radioiodide. Isotope equivalent efficiencies have been demonstrated for sorbents not impregnated with normal iodine or iodide. Normal methyl iodide tests cannot measure the removal of ^{131}I by isotope exchange on iodized charcoals and, therefore, give a high (conservative) estimate of methyl radioiodide penetration. However, there are currently no commercial radioiodine canisters or cartridges that have charcoals impregnated with iodide only. Soon MSA will also use 5% TEDA charcoal in their GMR-I canisters.(6)

C. The reaction of TEDA impregnant with methyl iodide vapor is by first order kinetics. The isotope exchange of iodide impregnant to remove the radioiodine from methyl radioiodide is also by first order kinetics. Effluent vapor concentrations decreased exponentially with bed depth. These results confirm that removal efficiency was independent of vapor concentrations within the bed. This is an important conclusion, since the radioiodine concentrations to be encountered in nuclear environments are many orders of magnitude lower than the ppm concentrations required for a nonradiometric test. The first order kinetics also implied that contact time of vapor within the sorbent bed is critical. Contact time is determined by canister geometry and airflow rate (*i.e.*, work load). A high flow rate should be chosen for a canister test to approach the upper limit of average vapor penetration. The arbitrary test standard is 64 L/min for canisters and 32 L/min for individual cartridges used in pairs.(7)

D. When the challenge of a vapor to a test bed is at a high enough concentration and continuous, the bed will become loaded and will decrease in efficiency of vapor removal. The resulting increase in penetration with time is called a breakthrough curve. Breakthrough times for selected penetration fractions are often dependent on the vapor challenge concentration.(3,8) At high relative humidities charcoal beds become loaded with water vapor, also increasing penetration of test vapor with time. It was shown that for efficient sorbents at low challenge concentrations or loadings, the relative humidity effect may be the most significant. Therefore, the time of exposure of a canister or cartridge to high humidity air is an important parameter in a test procedure or a usage protocol.

E. Canister charcoals containing 5% TEDA impregnant were more effective for methyl iodide removal than those containing 5% KI_3. Lower TEDA loading (2%) resulted in lower efficiencies, as expected from simple kinetics. Charcoals with these impregnants are more efficient for methyl radioiodide removal than those without, except for short periods with fresh canisters.

F. The larger canisters (used alone) were more effective for methyl iodide removal than cartridges (used in pairs) even though the flow rate through each cartridge was half as much. Also, the cartridges deteriorated in efficiency more rapidly due to high humidity. Magnitudes of efficiencies can be correlated with volumes of charcoal and bed contact times.

G. Insufficient data are available to rate cartridges and canisters for radioiodine removal. Variations within brands and types have not been established. Also, their contents are subject to change by the manufacturers. These unknowns emphasize the need for an ongoing certification program. Such a program to be carried out in the NIOSH Testing and Certification Branch, is an ultimate goal of this project.

acknowledgment

The assistance of Frank Valdez in performing some of the respirator canister and cartridge tests is gratefully acknowledged.

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