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DP-1053

AEC RESEARCH AND DEVELOPMENT REPORT

NONDESTRUCTIVE TEST OF CARBON BEDS FOR REACTOR CONFINEMENT APPLICATIONS

PROGRESS REPORT
OCTOBER 1964 - JANUARY 1966

D. R. MUHLBAIER

DU PONT
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Savannah River Laboratory

Aiken, South Carolina

Printed in USA. Price $2.00
Available from the Clearinghouse for Federal Scientific
and Technical Information, National Bureau of Standards,
U. S. Department of Commerce, Springfield, Virginia

664111

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Engineering and Equipment
(TID-4500)

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by

David R. Muhlbaier

Approved by

E. C. Nelson, Research Manager
Reactor Engineering Division

Issue Date: June 1966

E. I. DU PONT DE NEMOURS & COMPANY
SAVANNAH RIVER LABORATORY
AIKEN, SOUTH CAROLINA

CONTRACT AT(07-2)-1 WITH THE
UNITED STATES ATOMIC ENERGY COMMISSION

ABSTRACT

A nondestructive in-place test for detecting leaks in a bank of installed carbon beds with "Freon-112" tracer was developed by the Savannah River Laboratory and demonstrated in the reactor confinement facilities at the Savannah River Plant. Leakage flow (flow that bypasses the carbon) of 0.01% of the total air flow through the beds is detected readily. Installed carbon beds with about 2 years exposure to exhaust air flow in the confinement system were evaluated successfully with the test. Standardization of the test is in progress for general application to in-place testing of carbon beds.

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NONDESTRUCTIVE TEST OF CARBON BEDS FOR REACTOR CONFINEMENT APPLICATIONS

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INTRODUCTION

Halogen vapor (principally ^{131}I) that might be released accidentally into the buildings of Savannah River Plant (SRP) reactors would be removed by normal, routine passage of the exhaust ventilation air through carbon beds. New carbon beds are tested for absence of leaks by a nondestructive technique in which "Freon-12"* is used as a tracer. Development of this technique by the Savannah River Laboratory (SRL) is discussed in progress report DP-870(1). The "F-12" technique is limited to testing new carbon beds with air at a maximum velocity of 20 ft/min and with carbon containing no more than 5% sorbed H_2O. In addition, the test is unsatisfactory for used carbon because adsorbed impurities cause the "F-12" to desorb too quickly (in less than one minute) for a definitive measurement of the leak path.

Because of these limitations, work was undertaken to develop a standardized nondestructive test that would be generally applicable for in-place leak testing of installed, used carbon beds. The limitations imposed on air velocity and sorbed H_2O were reduced significantly by the use of "F-112", a halogenated hydrocarbon less volatile than "F-12" (DP-910(2), 920(3), and 950(4)). This report discusses work (October 1964 - January 1966) to develop an in-place leak test of used carbon beds installed in the SRP confinement system. A final report will be issued when the test is standardized for general application to in-place leak testing of installed carbon beds.

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* "Freon" and combinations of "Freon-" and "F-" with numerals are Du Pont's registered trademark for its fluorinated hydrocarbons.

SUMMARY

An in-place leak test for a complete bank of installed carbon beds was developed by SRL and demonstrated in the SRP reactor confinement system. In-place "F-112" tests of used carbon beds in five SRP filter compartments showed that the average leakage flow (flow that bypasses the carbon) was <0.03% of the total air flow through the beds. This leakage flow also corresponds to an iodine adsorption efficiency of ⩾99.96% for all banks of carbon beds, because small-scale tests (in which there were no detectable leak paths) showed that the iodine adsorption efficiency of representative samples of the carbon was >99.99%. The beds were exposed for ~2 years to exhaust air flow in the confinement system prior to the "F-112" and iodine tests.

The in-place "F-112" test is relatively easy to conduct. The precise values of air flow, "F-112" injection rate, and calibration of the "F-112" detectors are not required. Only the relative calibration of two detectors for "F-112" is necessary. Mixed air-"F-112" samples from upstream and downstream of the carbon beds were supplied to the "F-112" detectors.

New carbon is "F-112" leak tested at air velocities to 70 ft/min and with H_2O sorbed to ~20%. The air velocity and sorbed H_2O are reduced to 10 to 20 ft/min and to ~4% for a satisfactory in-place test of used carbon (exposed to air flow with trace contaminants). Water is desorbed from the carbon prior to the test by heating the inlet air (at 5 to 10 ft/min velocity) to reduce its relative humidity to <25%.

Exposure of the carbon beds to normal exhaust air flow desorbs over 95% of the "F-112" in about 3 weeks, but "F-112" sorbed on the carbon does not affect its iodine adsorption efficiency.

Standardization of the "F-112" test for general application to in-place testing of carbon beds is in progress, and will be reported at a later date.

DISCUSSION

LEAK TEST DEVELOPMENT

Leaks in or around carbon beds can be measured by injecting a known concentration of adsorbable tracer upstream of the carbon bed and measuring the downstream tracer concentration. If the carbon removes all of the tracer that passes through it, the ratio of the downstream tracer concentration to the upstream concentration represents the fraction of the total flow that bypasses the carbon (leaks). This principle is used to measure leaks in carbon beds by the "F-12" technique(1) and the in-place "F-112" leak test discussed in this report. However, "Freon" is physically adsorbed and, consequently, will desorb with time. Very rapid desorption of the "Freon" is undesirable, because of the difficulty in distinguishing between "Freon" penetration caused by desorption and penetration caused by leaks. Therefore, the tests must be conducted at conditions that prevent rapid "Freon" desorption.

Before the development of the in-place leak test of installed carbon beds, preliminary tests were conducted on small-scale carbon beds to evaluate the performance of different "Freon" compounds for improved adsorption characteristics on new carbon.(2,3,4) Based on this work, "F-112" was selected for primary evaluation as the tracer in the development of an in-place leak test, with "F-113" as a backup tracer. The principal properties of the "Freon" compounds mentioned in this report are listed in Appendix A. The proprietary materials that were used were selected because of convenient availability in standard, reliable quality. Presumably other materials might have been used. Studies continued with small-scale carbon beds of used carbon (carbon with several months service in the reactor exhaust ventilation system) before testing was begun on full-size carbon beds. The small-scale tests showed that "F-112" was satisfactory for used carbon, so further work with "F-113" was suspended. Only "F-112" was used for tests of full-size carbon beds; these tests led to the development of the in-place leak test described in this report.

"Freon" Tests of Small-Scale Carbon Beds

Small-scale carbon beds, described in DP-910(2), were used in the preliminary evaluation of the adsorption of "F-112" and "F-113" on used carbon. The beds were packed with representative samples of carbon removed from full-size carbon beds that had been subjected to various service times in an SRP reactor ventilation system.

Tests were conducted at air velocities to 70 ft/min and with carbon containing up to 25% sorbed H_2O. The results for new and used carbon are summarized in Table I. The results show that the adsorption characteristics of "F-112" differ significantly between new and used carbon. Tests with "F-112" on new carbon showed acceptable results for leak testing at velocities to 70 ft/min and up to ~25% sorbed H_2O. However, "F-112" tests of used carbon with 15 months service in the reactor exhaust ventilation system did not produce acceptable results at 70 ft/min and with only 3.5% sorbed H_2O. These results showed that both velocity and H_2O content must be reduced for an acceptable test of used carbon. A moderate reduction in velocity did not produce acceptable results. The adsorption efficiency at 50 ft/min and 6 to 10% sorbed H_2O was not satisfactory. A typical adsorption efficiency curve of an unsatisfactory test of a small-scale bed packed with used carbon is shown in Figure 1. Further tests were conducted on full-size carbon beds to accurately define the limits for an acceptable test of used carbon.

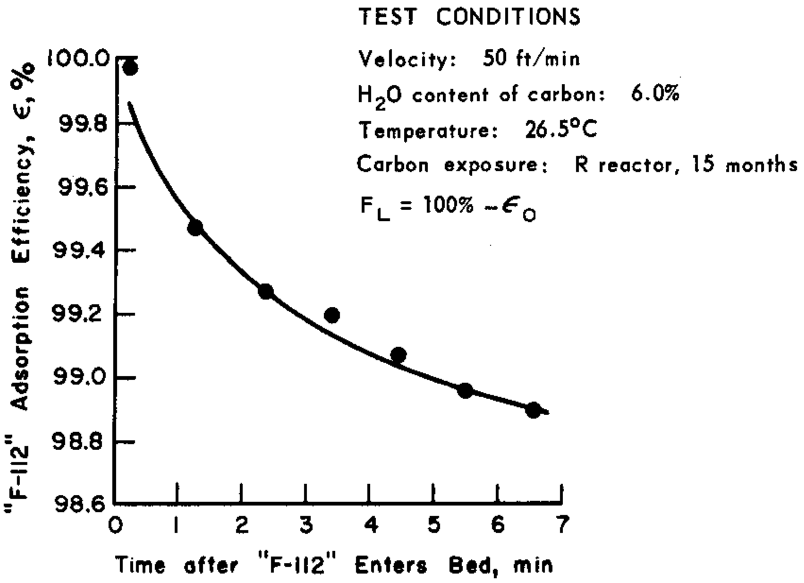
Subsequent tests of "F-112" on used carbon showed that the conditions for an acceptable test are similar to those for "F-12" on new carbon.(1) Similar tests on two used carbon beds showed the superior adsorption characteristics of "F-112." The beds were packed with identical carbon which was exposed in the P-reactor ventilation system for 14 months. The carbon was heated in air flowing at 20 ft/min and ~40°C for about 2 hours to remove sorbed H_2O. Both carbon beds were tested in the same manner except one test used "F-12" and the other "F-112" as tracer. The results (Figure 2) confirmed that "F-12" is unsatisfactory for testing used carbon, because the efficiency was reduced to 99.9% in <1 minute, which is insufficient time to evaluate the performance of installed beds. Under the same conditions, "F-112" showed no penetration (<0.01%).

TABLE I

"Freon" Efficiency of Small-Scale Carbon Beds

	Test Conditions			"Freon"		
Carbon	Air Velocity, ft/min	Sorbed H_2O, %	Temp, °C	Type	Upstream Conc, ppm	Efficiency at 5 min, %[a]
New	70	27	20	"F-112"	10	99.98
	70	25	25.5	"F-113"	5	99.9
R reactor (15 months)	70	16	22	"F-112"	3	99.0
	70	3.5	25	"F-112"	3	99.5
	70	1.8	28	"F-113"	5	99.75
	50 (repeat)	6.0	26.5	"F-112"	3	99.0
L reactor (2 years)	50	10.4	25	"F-112"	3	99.5
	50	7.0	23.5	"F-112"	3	99.7

(a) An acceptable test efficiency is 99.9+% at 3 to 5 minutes after "Freon" injection.



TEST CONDITIONS

Velocity: 50 ft/min

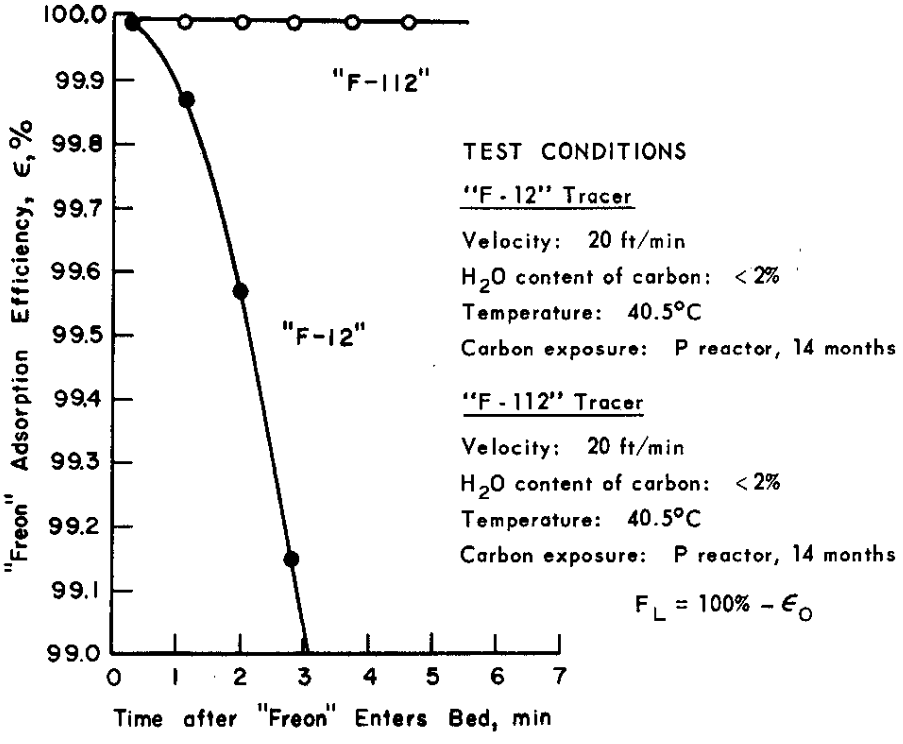
H_2O content of carbon: 6.0%

Temperature: 26.5°C

Carbon exposure: R reactor, 15 months

$F_L = 100\% - \epsilon_O$

FIG. 1 TYPICAL "F-112" EFFICIENCY OF SMALL-SCALE USED CARBON BED



TEST CONDITIONS

"F - 12" Tracer

Velocity: 20 ft/min

H_2O content of carbon: < 2%

Temperature: 40.5°C

Carbon exposure: P reactor, 14 months

"F - 112" Tracer

Velocity: 20 ft/min

H_2O content of carbon: < 2%

Temperature: 40.5°C

Carbon exposure: P reactor, 14 months

$F_L = 100\% - \epsilon_O$

FIG. 2 EFFICIENCY OF USED CARBON FOR "F-12" AND "F-112"

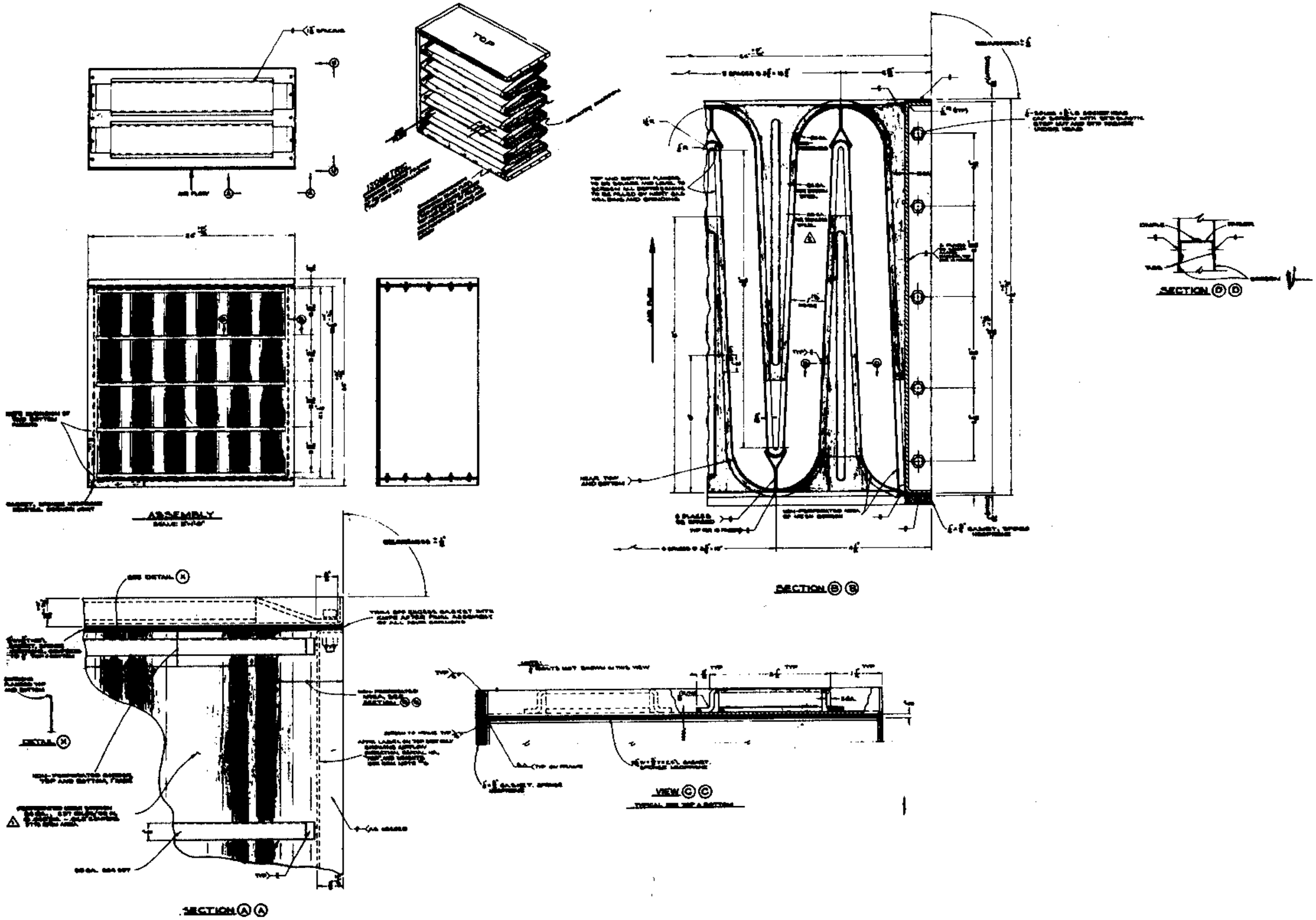


FIG. 3 FULL-SIZE SRP CARBON BED

"Freon" Tests of Full-Size Carbon Beds

Full-size SRP carbon beds, as shown in Figure 3, were used to evaluate the performance of "F-112" on new and used carbon. The used beds were production units that had been in service in the reactor exhaust ventilation system for several months before testing.

"F-112" tests of a new full-size carbon bed are shown in Figure 4. Although the "F-112" efficiency decreased to 99.9% in ~3 minutes, the tests were satisfactory, because any leakage flow could be determined with confidence due to the shape of the curve. The leakage flow that bypassed the carbon is calculated from the following equation:

$$F_L = 100\% - \epsilon_0 \qquad (1)$$

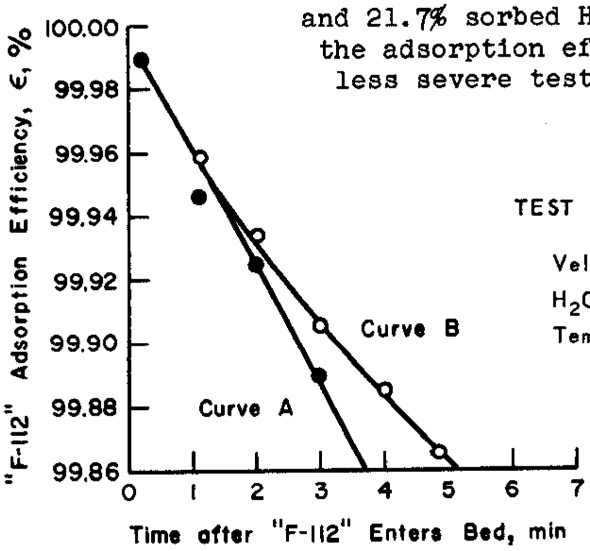
where

F_L = leakage flow, % of total flow through bed(s)

ϵ_0 = "F-112" adsorption efficiency extrapolated to time zero from a plot of the "F-112" efficiency versus time after "F-112" enters the carbon bed(s).

In Figure 4, the efficiency at time zero is >99.99%; hence, the leakage flow is <0.01%.

The first test shown on Figure 4 (Curve A) was made at a face velocity of 70 ft/min and with 22.4% sorbed H_2O in the carbon. After the test, "F-112" was desorbed from the carbon, and a repeat test was made. The repeat test (Curve B) was at 60 ft/min velocity and 21.7% sorbed H_2O. A slight improvement in the adsorption efficiency occurred because of less severe test conditions.



TEST CONDITIONS	Curve A	Curve B (repeat test)
Velocity:	70 ft/min	60 ft/min
H_2O content of carbon:	22.4%	21.7%
Temperature:	23°C	21°C

$$F_L = 100\% - \epsilon_0$$

FIG. 4 "F-112" EFFICIENCY OF FULL-SIZE NEW CARBON BED

The "F-112" adsorption efficiency of carbon is gradually reduced with service in the SRP reactor ventilation system. This effect is caused by the loss of adsorption sites due to the adsorption of trace impurities in the ventilation system air. Thus, used carbon must be tested at a lower H_2O content (when compared with new carbon) to compensate for the loss in active sites. Reduction in face velocity also provides more time for "Freon" to adsorb. The effect of the carbon's service on the "F-112" adsorption characteristics is summarized in Table II. By comparing the test conditions and efficiencies of new carbon and carbon with 7 and 15 months service in C reactor, it is apparent that less severe test conditions (lower velocity and/or lower H_2O content) are required for an acceptable test of older carbon. Loss in "F-112" adsorption efficiency will probably continue with increased carbon service as indicated by the "F-112" test results of K-reactor carbon with 2 years service.

TABLE II

"F-112" Efficiency of Full-Size Carbon Beds

	Test Conditions			Desorbed Before Test[a]	Upstream "Freon" Conc, ppm	Efficiency at 4 min, %[b]
Carbon	Air Velocity, ft/min	Sorbed H_2O, %	Temp, °C			
New	70	22.4	23	-	10	99.85
P reactor (15 months)	30	16.6	21	No	10	99.98
	35	15.9	23.5	No	10	99.92
C reactor (7 months)	25	9.1	28	Yes	10	99.87
	20	5.6	33	Yes	10	99.99+
	25	6.9	36	Yes	10	99.99
	25	2.0	46	Yes	10	99.99+
C reactor (15 months)	20	20.7	31	No	20	99.5
	20	4.3	31	No	10	99.94
	20	2.0	40	No	20	99.99+
K reactor (2 years)	20	4.0	32	Yes	10	99.5
	15	4.7	32	Yes	20	99.68
	15	2.0	43.5	Yes	20	99.98

(a) Desorption was performed by purging carbon with air at ~90°C for at least 20 hours.

(b) An acceptable test efficiency is 99.9+% at 3 to 5 minutes after "Freon" injection.

Tests have shown that under humid conditions heating the ambient air before it passes through used carbon beds improves "Freon" adsorption. This occurs because heating the air reduces the relative humidity which in turn reduces the H_2O content of the carbon. Air with a dew point of $<20°C$ can be heated to 50°C and passed through carbon to reduce the H_2O content from up to ~30% to <4%. This method was used to successfully test used carbon with as much as two years service (Table II). The method, when applied to testing installed SRP carbon beds, was implemented by installing heaters upstream of the carbon beds and reducing the air flow. Figure 5 shows the time required to desorb a new SRP carbon bed with up to 22% H_2O to <4% H_2O with air at ~50°C and flowing at ~4.5 ft/min velocity. Within 20 hours, the H_2O content could be reduced to <4%. The desorption time can be decreased by increasing the air flow and/or temperature.

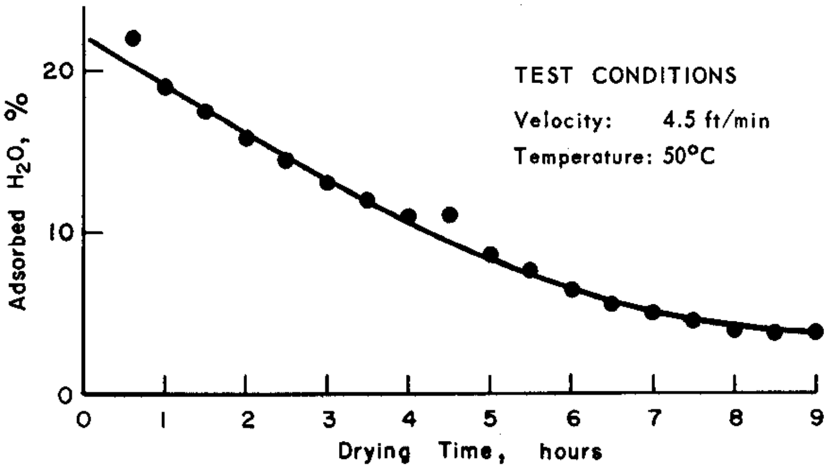


FIG. 5 DRYING TIME FOR CARBON BED AT REDUCED AIR FLOW

The ability of the "F-112" technique to detect a known leak path was successfully demonstrated by simulating a leak in a used carbon bed, as shown in Figure 6. Extrapolation of the curve (Figure 6) to zero time confirms the expected efficiency of 99.95% for the known leakage flow of 0.05%.

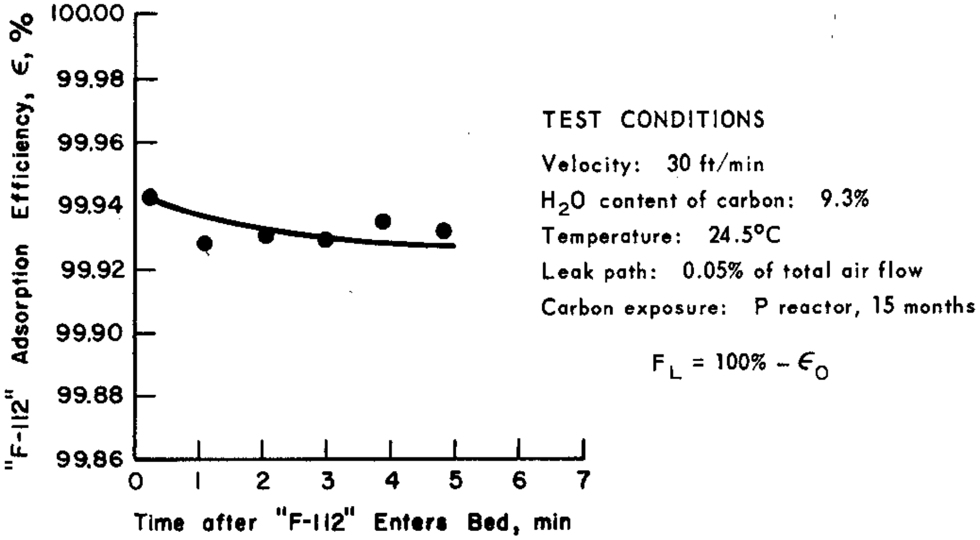
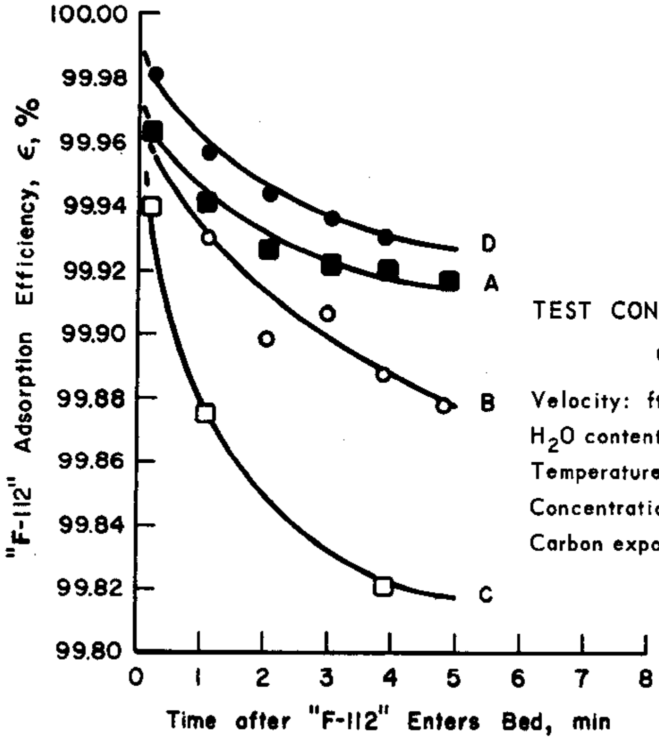


FIG. 6 "F-112" EFFICIENCY OF USED CARBON BED WITH LEAK PATH

Tests also showed that the upstream "Freon" concentration significantly affected the "Freon" adsorption efficiency of used carbon beds. Higher concentrations increase the "Freon" loading on the carbon but reduce the effect of adsorbed impurities, as shown by Curves A through D of Figure 7. The four tests were made with one used carbon bed, which was desorbed of all significant residual "F-112" prior to each test. The tests were conducted with inlet "F-112" concentrations of 2, 10, and 20 ppm by volume. The best adsorption efficiency was obtained at the highest "F-112" upstream concentration as explained below. In adsorption, the capacity of the carbon for most gases increases as the relative pressure (concentration) increases. Activated carbon contains many adsorption sites with various energy levels. Adsorption occurs first on the most active sites and proceeds to the less active sites as the relative pressure is increased. When the carbon contains small amounts of adsorbed impurities (as does used SRP carbon) the most active sites are occupied by the impurities. Therefore, at very low "Freon" concentrations the capacity of the carbon for "Freon" is reduced, and the "Freon" penetrates the bed more quickly with time. At higher inlet concentrations, at least the same absolute amount of "Freon" penetrates the bed, but the percent penetration is less and therefore the indicated efficiency is higher.



TEST CONDITIONS

Curve	A	B	C	D
Velocity: ft/min	35	35	35	35
H_2O content of carbon: %	15.9	15.4	16.8	15.5
Temperature: °C	23.5	25	24.5	24.5
Concentration: ppm	10	10	2	20
Carbon exposure:	P reactor, 15 months			

$F_L = 100\% - \epsilon_0$

FIG. 7 EFFECT OF UPSTREAM CONCENTRATION ON "F-112" EFFICIENCY OF USED CARBON BEDS

Carbon removed from the SRP reactor ventilation system after several months service contains several impurities that cause loss in the "Freon" and iodine adsorption efficiency. Mass spectrometric analyses of gas (vacuum extracted from carbon at ~100°C) showed as much as 7 mol % NO.(8) Qualitative analysis also showed the presence of trichloroethylene (which is used in the reactor building to degrease fuel elements) and two other unidentified substances. The analysis was performed with a gas chromatograph and electron capture detector, so the unknown substances were probably high boiling point (>50°C) halide compounds with concentrations in the low ppb range.

Iodine Tests of Small-Scale Carbon Beds

Tests showed that the in-place "F-112" test of installed carbon beds would not significantly affect the iodine adsorption efficiency of the carbon. The iodine adsorption efficiency of new carbon containing 3.4 mg "F-112" per gram of carbon and tested with an iodine-air mixture was 99.9+%. The "F-112" loading represented at least ten consecutive "F-112" leak tests, each with a duration of ~5 minutes, at an inlet concentration of 20 ppm by volume. Carbon with 15 months exposure in the C-reactor ventilation system and containing up to 0.33 mg of sorbed "F-112" per gram of carbon (equivalent to at least one "Freon" test) was evaluated. Iodine tests (with air or steam-air mixture) showed that the removal efficiency for elemental iodine was 99.9+%. The carbon was not desorbed prior to the tests, and the results are comparable with those reported(8) for used SRP carbon with no "F-112" loading on the carbon. Desorption of the more volatile impurities (e.g. NO) from the used carbon prior to the iodine test improved the efficiency for iodine removal (99.99+%) even with carbon containing up to 0.73 mg "F-112" per gram of carbon (equivalent to at least two "F-112" tests). The carbon was desorbed with air at 80 to 95°C for 2 hours. Table III summarizes the iodine tests of used carbon containing "F-112".

TABLE III

Iodine Tests of Used Carbon Exposed to "F-112"(a)

"F-112" Loading, mg "F-112"/g carbon	Carbon Desorbed(b)	Iodine Carrier	Iodine Removal Efficiency, %
0.18	No	Air	99.92
0.18	No	Steam-air	99.98
0.33	No	Air	99.94
0.73	Yes	Air	99.99+
0.73	Yes	Steam-air	99.99+
0.73	Yes	Steam-air	99.99

(a) The carbon was removed from C reacter after 15 months exposure in the ventilation system.

(b) Desorption of the carbon was performed by exposure to flowing air at 80 to 95°C for 2 hours.

"Freon" Detector

The detectors used for measurement of the "Freon" concentrations were model 680 Aerograph Pestilizer gas chromatographs with an electron capture detector. A gas sampling valve was also attached for ease in sampling the air stream. The instruments are made by Varian-Aerograph (formerly Wilkens Instrument & Research, Inc.), Walnut Creek, California. A photograph of the detector is shown in Figure 8. The operating principles of the instrument were discussed in DP-870.(1)

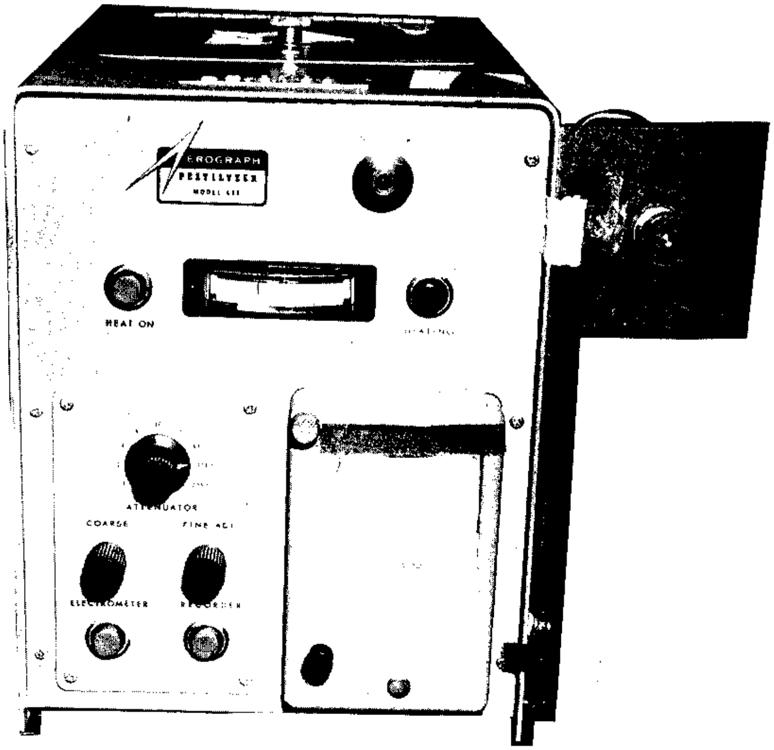


FIG. 8 "F-112" DETECTOR

The chromatograph column used during the tests to separate the "F-112" from the air sample for detection by the electron capture cell was a 5 ft long x 3/32 inch ID stainless steel column. The column, which was originally packed with 30% SF-96 on 45/60 "Chromosorb"* P, was modified by baking in an oven at 350°C for 16 hours to remove most of the stationary phase (SF-96) from the "Chromosorb". Nitrogen at a flow of ~75 cc/min was purged through the column during the baking operation. This column produced sharp peaks and good resolution of O_2 and "F-112"; "F-112"-air samples could be analyzed every 50 seconds.

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* "Chromosorb" is a registered trademark of Johns-Manville Company.

The sensitivity of the electron capture detector is affected by the sample size and operating conditions such as the cell temperature, N_2 flow, and cell voltage. However, these factors are relatively unimportant for the "F-112" leak test, because the detectors are calibrated prior to each test. The sensitivity of the detectors for "F-112" is ~1 ppb by volume. Therefore, leaks of <0.01% can be detected with a "F-112" concentration of 20 ppm by volume upstream of the carbon beds.

"Freon-112" Desorption

"Freon-112" is physically adsorbed and therefore can be desorbed from activated carbon. Desorption of "F-112" from SRP-type activated carbon occurs very slowly at the conditions recommended for in-place leak testing. Thus, a repeat test of carbon beds can be accomplished without desorbing the previously adsorbed "F-112". Several satisfactory repeat tests have been conducted on new and used carbon beds without desorbing the previously adsorbed "F-112".

Desorption of "F-112" from activated carbon is enhanced by high temperature and high H_2O content as shown by Table IV. These desorption times and conditions are applicable to carbon beds with loadings of ~0.4 to 0.8 mg "F-112" per gram of carbon. The "F-112" was adsorbed on the front of the carbon bed and desorbed by driving the "F-112" through the bed and out the back, except where noted. Desorption of a given amount of "Freon" becomes easier with increased "Freon" loading. The air velocity is also a factor affecting "F-112" desorption time, but its influence was not studied.

TABLE IV

Desorption of "F-112" From New Carbon(a)

	Air Conditions			
"F-112" Desorbed, %	Velocity,(b) ft/min	Relative Humidity, %	Temp, °C	Desorption Time
>95	70	<3	100	3 weeks
>95	70	75+	30	3 weeks
~90	70	<3	130+	5 days
~70	70	55 to 65	30	10 days
~1	70	<40	30	1 day
>95	70(c)	<3	95	3 days

(a) Data for new small-scale carbon beds with ~0.4 to 0.8 mg "F-112"/g carbon at starting conditions.

(b) The "F-112" was desorbed from the opposite side to which it was first adsorbed except where noted.

(c) The air flow was reversed through the carbon bed to desorb the "F-112" from the same side of the bed on which it was first adsorbed.

IN-PLACE LEAK TEST OF INSTALLED CARBON BEDS

Technique

As previously discussed, leaks in a carbon bed can be determined by measuring the concentration of an adsorbable tracer both upstream and downstream of the bed and calculating the concentration ratio. Previous tests of SRP carbon beds had shown that this method was satisfactory in measuring leakage flow under specified conditions. Details are given in section, "Freon" Leak Test of Full-Size Carbon Beds.

The maximum acceptable leakage flow for SRP carbon beds is 0.1% of the total air flow. However, the in-place test was designed to detect at least a 0.01% leak (one-tenth of the maximum acceptable leak) by injecting at least 10,000 units of tracer upstream of the test carbon bed with the capability to detect 1 unit downstream of the carbon bed.

Figure 9 shows a diagram of the equipment for testing installed carbon beds at SRP. Only three connections to the filter compartment are required. One connection at the top of the compartment is used for the "F-112" injection. The other two connections are required for the sampling system; air samples are pumped from upstream and downstream of the carbon beds and conveyed to two detectors for measurement of the "F-112" concentrations.

The "Freon" injection apparatus consists of a pressurized "F-112" reservoir containing an immersion heater to preheat the "F-112" (freezing point = 23.8°C) to ~40°C and thin-walled stainless steel tubing. The tubing (3/32-inch OD with 0.010-inch wall by 45 ft long) is attached to the reservoir and isolated from the liquid "F-112" by a valve. The stainless steel tubing is used as a resistance heater to vaporize the "F-112" before injection in the filter compartment. The tubing is insulated electrically from the "Freon" reservoir and filter compartment. During operation, the reservoir is pressurized with nitrogen to a maximum of 75 psig and about 15 amperes (at 115 volts) is passed through the stainless steel tubing. The reservoir pressure forces liquid "F-112" into the heated tubing where it is vaporized and superheated before it enters the filter compartment. Tests demonstrated that the injection apparatus provides a uniform and hydrodynamically stable flow of superheated "F-112" to the filter compartment, and no detectable "F-112" condensation occurred when the superheated "F-112" was mixed with air at the end of the tube. Reservoir pressures above 75 psig caused saturated "F-112" vapor at the tube effluent.

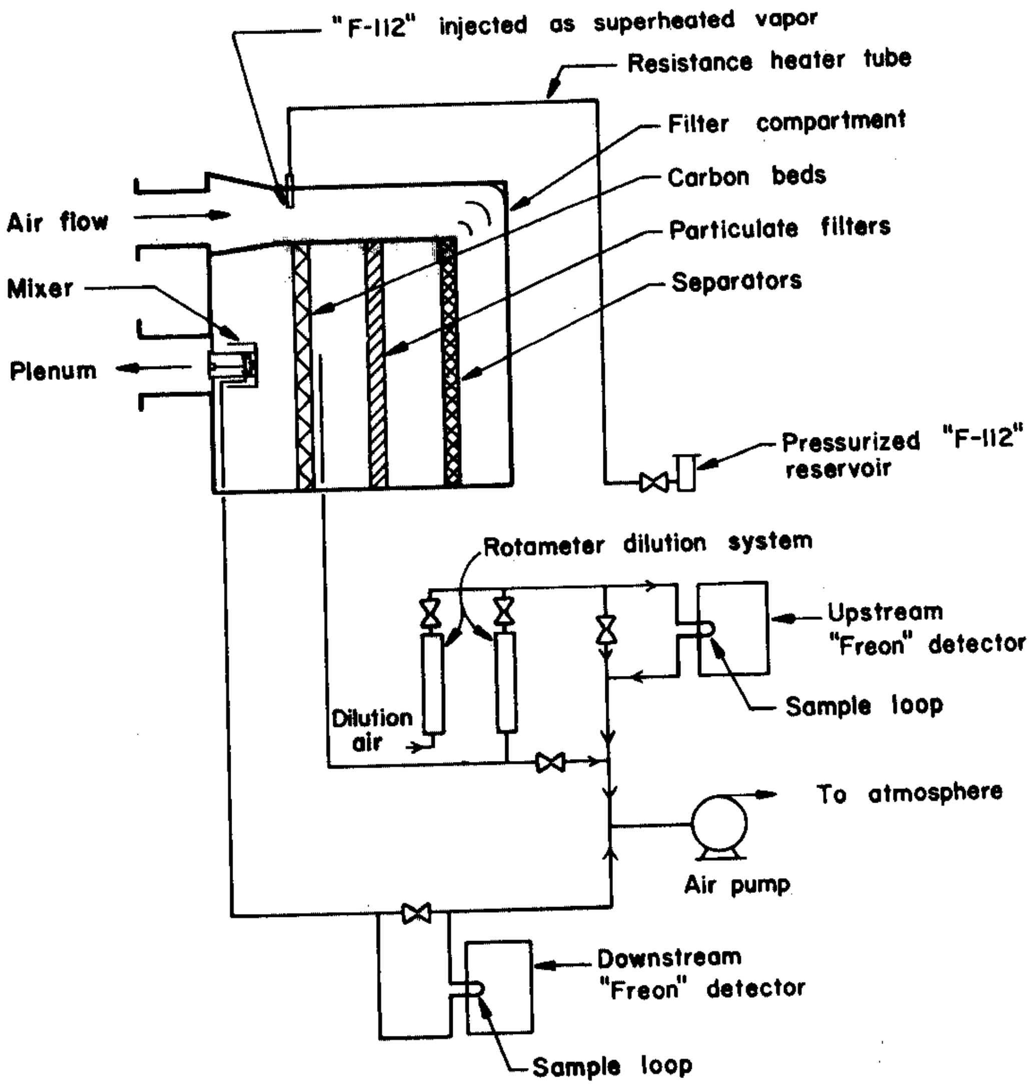

FIG. 9 DIAGRAM OF EQUIPMENT FOR TESTING INSTALLED SRP CARBON BEDS

The sampling system consists of a pump to draw air samples from both the upstream and downstream sides of the test carbon beds. Two identical gas chromatographs with electron capture detectors are used to measure the "Freon" concentrations; each has a linear range for detection of "F-112" of about 1 to 100 ppb by volume. This detection range is ideal for the downstream "Freon" concentration, but the upstream concentration exceeds the linearity range of the instrument. The upstream sample is diluted with a known quantity of air to reduce the concentration to within the linear range before measurement by the detector. Calibrated rotameters are used in the dilution system to permit determination of the dilution factor.

As discussed previously, the leakage flow (Equation 1) is determined from a plot of the "F-112" adsorption efficiency versus time. The "F-112" adsorption efficiency as a function of time after "F-112" enters the carbon bed is calculated from the following equation:

$$\epsilon = 100 \left[1 - \left(\frac{R_d}{R_u} \frac{1}{f_d} \frac{C_u}{C_d} f_c\right)\right] \qquad (2)$$

where

ϵ = "F-112" adsorption efficiency, %

R_d = downstream detector response for "F-112", recorder units

R_u = upstream detector response for "F-112", recorder units

f_d = dilution factor of sample from upstream carbon bed to upstream detector, dimensionless constant

C_u = upstream detector response to standard calibration solution, recorder units per microliter

C_d = downstream detector response to standard calibration solution, recorder units per microliter

f_c = calibration conversion factor, dimensionless constant

During the "F-112" tests, it is not necessary to know the absolute values of the following: tracer concentrations; air flow through the carbon beds; or calibration of the "Freon" detectors. However, an estimate of the air flow is necessary to set the upstream tracer concentration at 10,000 to 20,000 times the minimum downstream detector response so that leaks of 0.01% of the total flow can be determined.

Relative calibration of the detector is accomplished by injecting a solution of "F-112" in hexane onto the chromatograph column.* The solution contained a given mass of "F-112" per unit volume (e.g., 10^{-11} g "F-112"/µl of hexane). The response of both detectors to the same "F-112"-hexane solution is determined in recorder units per microliter (RU/µl).

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* This is a standard chromatographic technique. The hexane is used to dilute the "F-112" and enable injection of very small quantities of "F-112". The hexane does not interfere with the detection of "F-112" because the chromatograph column separates the sample components. One sample can be analyzed every 50 seconds.

Equation (2) contains two constants. The dilution factor (f_d) converts the relative "F-112" concentration measured by the upstream detector to the relative value upstream of the test carbon beds. f_d is equal to the dilution ratio of the rotameter dilution system (e.g., if dilution ratio = 500:1, f_d = 500).

The second constant of equation (2) is the calibration conversion factor (f_c), which converts the relative "F-112" calibration determined from the "F-112"-hexane mixture to the relative "F-112" calibration in an "F-112"-air mixture. This factor is needed because "F-112" is detected in air during the leak test and not in hexane. Although the detectors respond equally to equal masses, whether in air or hexane, the ratio of mass injected to the detectors by each method is not necessarily equal. For the "F-112"-hexane method, the ratio of mass injected to each detector was set equal to one (1 μl for upstream detector per 1 μl for downstream detector). For the "F-112"-air calibration, the volume of sample injected to each detector is fixed and depends on the measured volume of the sample loop (Figure 9). For example, the sample volume may equal 0.60 ml on the upstream detector and 0.50 ml for the downstream detector. The calibration conversion factor (f_c) would then equal 0.60/0.50 or 1.20. The calibration conversion factor is not necessary, if the detectors are calibrated with an "F-112"-air mixture. However, such calibrations are inconvenient for most in-place tests, because a small quantity of "F-112" must be vaporized and stored in a relatively large cylinder of air.

Equation (2) contains two quantities, R_u and R_d, which are the upstream and downstream detector response to "F-112" only. If the test system contains an impurity that appears at the same point on the detector chromatogram as "F-112", then the impurity provides a background signal that must be subtracted from the "F-112" signal for the correct quantities of R_u and R_d. This condition occurred in the leak test of SRP carbon beds when trichloroethylene appeared in the confinement system air. A background signal would also exist if "F-112" desorbed from the carbon at the time of a repeat test.

Work is in progress to standardize the leak test for general applications. The leak test apparatus is being modified to make it more portable. Upon completion of this work, a final report will be issued on details of the equipment, technique, and procedure. However, a general outline of the leak test procedure is presented in Appendix B.

Mixing

For a definitive measurement of leakage flow through carbon beds, the upstream "Freon" concentration must be nearly uniform across the face of the carbon beds and the sample removed downstream of the beds for "Freon" analysis must be representative of a nearly uniform mixture. Otherwise, if a rarified or concentrated mixture occurred upstream or downstream of the location of a leak path, the indicated amount of the leakage flow would vary by the same percent variation in the local "Freon" concentration from the mixed mean concentration. Hence, a significant leak might not be detected if the mixing was inadequate.

Mixing tests were conducted in a SRP filter compartment to determine (1) the "F-112" upstream mixing performance, and (2) a method of obtaining a downstream sample representative of a mixed sample. The tests showed that the upstream mixing was satisfactory when "F-112" was injected at the point shown on Figure 9 and without any other device to promote mixing. The upstream mixing was within ±7% of the calculated mixed mean concentration.

Sampling for measurement of the downstream "Freon" concentration had to be done in the short distance (~9 feet) between the carbon beds and the plenum. Sampling could not be done in the plenum, because the "Freon" concentration would be diluted by the air flow from other systems. Downstream mixing tests showed that a completely mixed air sample could not be obtained with a single point sampler and a single mixing orifice installed in the compartment effluent nozzle.

Figure 10 shows the location of the mixing and sampling devices that were developed to obtain a mixed sample from downstream of the carbon beds. The mixer consists of two coaxial pipes; it is 36 inches long by 35 inches in diameter. Mixing is promoted by flow in the annular section of the mixer and by 35 holes (3.5 inches in diameter) at the inlet end of the inner pipe. The mixer is concentric with a 23-inch-diameter hole in the center of a cover plate that is installed in the effluent nozzle of the filter compartment. A multipoint sampler is installed downstream of the mixer. The sampler consists of a hub with eight radial spokes and two circumferential arcs between each spoke. The sampler was made of 1/2-inch-diameter stainless steel tubing, and 96 holes (0.021-inch diameter) are located at ~2-inch intervals in the spokes and arcs. A sample flow of about 0.7 cfm is removed from the hub with an air pump.

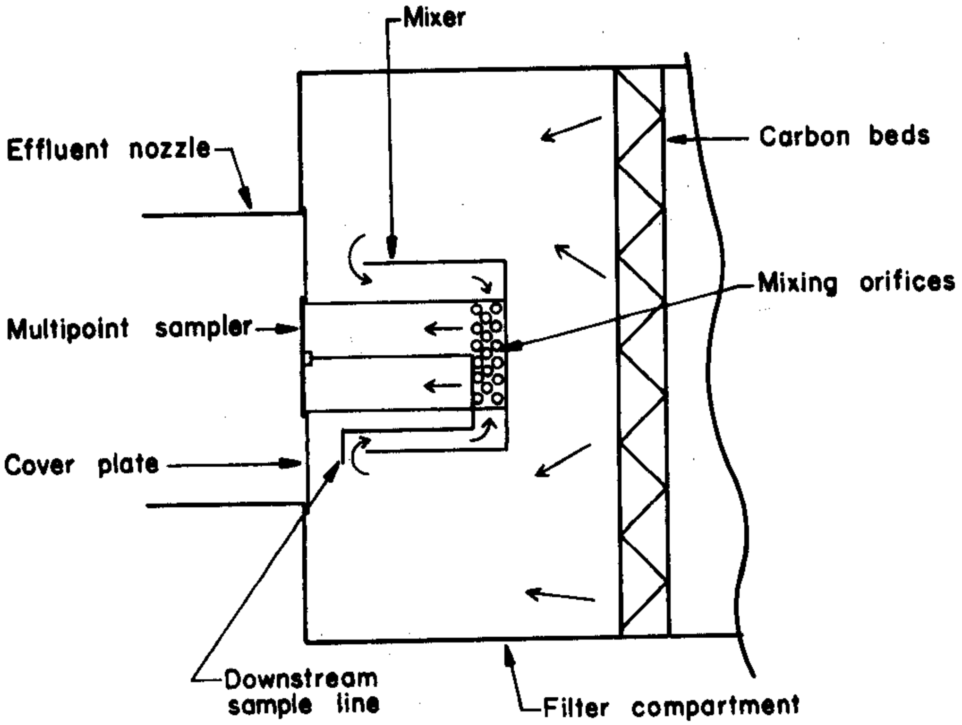


FIG. 10 "F-112" AIR MIXING DEVICE

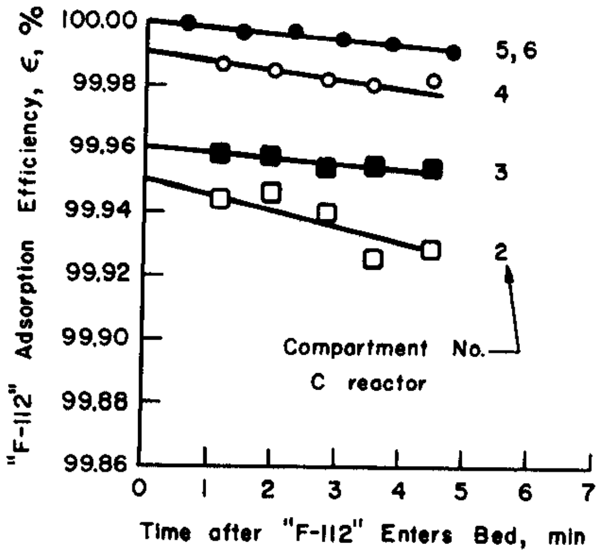
(Section Through Effluent Nozzle)

Tests of the mixer and multipoint sampler showed a maximum variation of ±10% from the calculated mixed mean "F-112" concentration downstream of the carbon beds. The tests were made by injecting "F-112" at a known rate and at several different positions downstream of the carbon beds.

Measurement of the percentage leakage flow during in-place leak tests of SRP carbon beds can be predicted with a standard deviation of ±9% when all errors in measurements of the "F-112" concentrations are combined. The errors result from incomplete mixing, both upstream and downstream, and variation in calibration of the "Freon" detectors and the dilution rotameters.

Test Results

The technique and equipment previously described were successfully demonstrated on banks of carbon beds installed in C reactor at SRP. The "F-112" adsorption efficiency for the five compartments is shown in Figure 11, and the leakage flow is summarized in Table V. Each compartment passed the leak test, and the average leakage flow for the five compartments was <0.03% of the total air flow. The leakage flow for each bank of carbon beds is obtained by subtracting the "F-112" adsorption efficiency at time zero (Figure 11) from 100%.



$F_L = 100\% - \epsilon_0$

FIG. 11 "F-112" EFFICIENCY OF BANKS OF INSTALLED SRP CARBON BEDS

TABLE V

Measured Leaks in Carbon Beds Installed in C Reactor Confinement System

Filter Compartment	Leakage Flow, %
2	0.05
3	0.04
4	0.01
5	<0.01
6	<0.01
Average	<0.03

Compartment 3 required two tests because the first test showed leaks of ~0.2%. Inspection of the carbon beds and mountings showed that several mounting bolts were loose. After the bolts were tightened, the leakage flow was reduced to 0.04%, well within the acceptable range. Apparently, the bolts were not tightened adequately during installation of the carbon beds, because this condition was not experienced in any of the other compartments.

The leakage at normal air flow is probably less than the value measured by the "F-112" leak test at reduced flow. The leak tests were conducted at 10 to 25% of rated flow. The normal operating flow varies from ~70 to 100% of rated flow. If turbulent flow exists in the leak path, the actual leakage flow at 70% of rated flow is ~30% less than that measured during the leak test, because the pressure drop across the carbon beds varies as the 1.3 power of the flow.

For a definitive measurement of leaks, the H_2O content of used SRP carbon beds had to be less than ~4%, as previously discussed. The H_2O content of the carbon varies from 10 to 25% by wt (in equilibrium with air at 50 to 80% relative humidity) so the carbon was dried in place before the test. Four heaters (total capacity 72 kw) were installed at the separator bank near the bottom of the filter compartment. The heaters increased the temperature and consequently reduced the relative humidity of the air to <25% at the carbon beds. Carbon in equilibrium with air at <25% relative humidity will contain <4% H_2O. The carbon was desorbed at approximately 5 ft/min air velocity for ~1 day prior to the "F-112" leak test. This drying method was satisfactory in reducing the H_2O content below 4% and, secondly, increased the elemental iodine efficiency of the carbon by desorbing NO(6).

Trichloroethylene was detected downstream of the carbon beds during the "F-112" leak tests. Because of detector operating conditions and similarities in the compounds of "F-112" and trichloroethylene, the detector does not distinguish between the two compounds. However, tests showed that trichloroethylene (in the low ppb concentration range observed during the test or in concentrations at least equal to the "F-112" concentration) has no significant effect on the sensitivity of the electron capture detector for "F-112". Thus, any background signal from trichloroethylene was subtracted from the total signal to obtain the actual "F-112" concentration.

Application

The "F-112" leak test was developed to measure leakage flow through carbon beds installed in reactor confinement systems. The test was successfully demonstrated at SRP. The same technique can be used to evaluate other carbon beds of different design and different carbons. However the maximum test conditions (maximum face velocity and moisture content of carbon) may vary for different systems. The bed design and service experience will probably be the most important factors governing the test conditions. Test conditions for carbon beds in standby confinement systems should be similar to those of new SRP carbon beds if of similar design. Carbon beds in systems that are continuously on-line and exposed to impurities in the air should require test conditions similar to those used at SRP. The maximum permissible test conditions should be determined experimentally for each system. However, the best "F-112" adsorption will occur at the lowest face velocity and H_2O content that can be maintained in the system during the leak test.

Figure 12 shows a diagram for the application of the "F-112" leak test to a typical carbon bed installation. "F-112" is injected upstream of the test carbon beds, preferably before several elbows or far enough away so that thorough mixing of air and "F-112" will occur before reaching the carbon beds. Downstream of the test carbon beds, a mixed sample must be obtained.

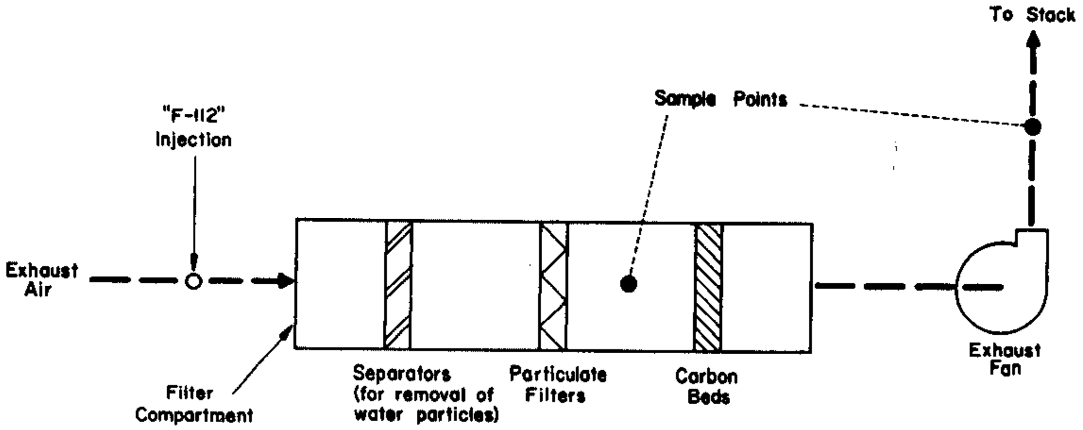


FIG. 12 DIAGRAM OF EQUIPMENT FOR "F-112" LEAK TESTING OF TYPICAL CARBON BED INSTALLATIONS

This can be accomplished by installing a mixing device or by removing a sample downstream of the exhaust fan. However, if the "F-112" must travel a significant distance before reaching the sample point, the system should be checked to make certain that no significant "F-112" is adsorbed by the duct and that a representative sample reaches the sample point. Otherwise, an error could be introduced in measuring a leak. In the tests at SRP, there was no significant "F-112" adsorption on the duct between the carbon beds and the sample point. Adsorption of "F-112" on surfaces upstream of the carbon beds was of no concern, because the "F-112" concentration was measured immediately upstream of the carbon beds.

Significant "F-112" adsorption on the sample lines must be avoided to ensure that a representative sample reaches the detectors. Both the line material and transit time are important factors. Tubing materials such as stainless steel, "Teflon"*, and glass are satisfactory for conveying "F-112"-air samples to the detector. Sample transit times limited to several seconds are also satisfactory. In the tests of SRP installed carbon beds, flexible sample lines of "Teflon"* were used. The lines were 50 ft long by 1/4-inch ID and conveyed a purge flow of ~0.7 cfm.

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* "Teflon" is Du Pont's registered trademark for its fluorocarbon resins.

APPENDIX A

PROPERTIES OF HALOGENATED HYDROCARBON MATERIALS

Trademark	Chemical Formula	Boiling Point, °C at 1 atm.
"F-12"	CCl_2F_2	-29.8
"F-112"	CCl_2F-CCl_2F	92.8
"F-113"	$CCl_2F-CClF_2$	47.6

APPENDIX B

GENERAL PROCEDURE FOR "F-112" LEAK TESTING

1. Set up test apparatus in area convenient to carbon beds to be tested.

2. Establish that all equipment is functioning properly.

3. Dry carbon beds with air at a relative humidity <25% so that the H_2O content of the carbon will not cause rapid "F-112" penetration of the beds.

4. Calibrate each detector with "F-112"-hexane solution.

5. Establish test air flow through the carbon beds at a velocity that will not cause rapid "F-112" penetration of the beds.

6. Inject "F-112" upstream of the test carbon beds.

7. Sample air and analyze for "F-112" upstream and downstream of the carbon beds. The first downstream sample should be analyzed as soon as equilibrium conditions are established at the detector.

8. Continue "F-112" injection and sampling for 4 to 5 minutes and then stop the injection.

9. Analyze test data.

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