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NONDESTRUCTIVE TEST OF CARBON BEDS FOR REACTOR CONTAINMENT APPLICATIONS

PROGRESS REPORT
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Savannah River Laboratory

Aiken, South Carolina

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NONDESTRUCTIVE TEST OF CARBON BEDS FOR
REACTOR CONTAINMENT APPLICATIONS
Progress Report: June 1962 - December 1963

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ABSTRACT

A nondestructive technique was developed by the Savannah River Laboratory for evaluating carbon beds used in reactor containment applications. "Freon-12" was used as a tracer to detect leak paths that would reduce the efficiency of carbon beds for removal of radioactive I_2 vapor. Leaks greater than 0.006% of the total flow were detected with an electron-capture-type instrument. The current method is suitable for testing dry carbon beds prior to field installation and at air velocities up to 20 ft/min.

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INTRODUCTION

Fission products that might be released accidentally into ventilating air of Savannah River Plant (SRP) reactor buildings are removed by passing the air through filters to remove particulates and through carbon beds to remove iodine vapor. Development of the air treatment system by the Savannah River Laboratory (SRL) has been discussed⁽¹⁻³⁾. The need for tests to evaluate the performance of these air treatment units is well recognized, and standardized tests for evaluating particulate filters are available (dioctylphthalate-aerosol penetration tests)⁽⁴⁻⁶⁾. This report describes a technique developed by SRL for evaluating the behavior of activated carbon beds to be installed in SRP reactor buildings. Subsequent reports (to be issued at intervals of 2 to 4 months) will discuss the progress and results of new work to develop a standardized nondestructive test for carbon beds after their installation in filtration systems.

SUMMARY

A nondestructive technique was developed by SRL for detecting leak paths in carbon beds. Leak paths, caused by settling or voids in the beds or by leaks around gasket seals, would reduce the halogen adsorption efficiency of the beds. "Freon-12"* was used as a tracer to detect any leak paths. "Freon-12" was readily desorbed from the carbon by normal exposure to the ventilating air stream; however, carbon beds almost saturated with "Freon-12" had no loss in efficiency for iodine adsorption.

"Freon-12" was injected into the air upstream of the bed to give an inlet concentration of 500 ppm, and the "Freon-12" downstream of the bed was measured by an electron-capture-type detector. The usable sensitivity of the detector during routine testing was 0.03 ppm for "Freon-12"; therefore, leaks greater than 0.006% of the total air flow were detected readily. The detector is extremely sensitive to halides and practically insensitive to hydrocarbons and other common interferences.

*Du Pont's trademark for dichlorodifluoromethane.

The current technique is suitable for evaluating new activated carbon beds of the type described in DP-778⁽¹⁾, under the following conditions:

- Maximum air velocity of 20 ft/min (based on carbon bed face area)
- Maximum H₂O content in carbon about 5% by weight
- Maximum air temperature of 35°C

The technique was used successfully to evaluate over 800 carbon beds prior to their installation in the containment facilities of the SRP reactor buildings.

Development of a standard test for installed carbon beds has been started at SRL because of (a) leak paths that can develop in gasket seals during field installation, (b) limitations of water vapor content and air velocity in the current "Freon-12" technique, and (c) general lack of a standard for testing carbon beds. The first part of the program, evaluation of other "Freon" compounds that may reduce current restrictions on the test conditions, is underway.

DISCUSSION

TEST EQUIPMENT

The facility for evaluating carbon beds (Figure 1) was a modification of the facility for testing full-size separators and particulate filters described in detail in DP-812⁽²⁾. Air was drawn through the particulate filter rack and into the blower where it was discharged into a 16-inch-OD pipe. "Freon-12", which was used as a tracer, was injected near the blower on the downstream side at a concentration of 500 ppm by volume. The "Freon-12" and air are mixed thoroughly in the upstream piping before they reach the carbon bed to be evaluated.

Carbon beds were housed in a duct about 2 ft x 2 ft. Expansion of air from the pipe to the duct occurred in a tapered section designed to produce a uniform velocity distribution. A distorted velocity profile may cause a high local velocity that could cause rapid penetration of the carbon bed by "Freon-12".

FIG. 1 FACILITY FOR EVALUATING FULL-SIZE CARBON BEDS

After the air passed through the carbon bed, it was mixed by an orifice and then exposed to a sampling nozzle located in the jet-stream downstream of the orifice. Thorough mixing was essential to prevent possible channeling of any "Freon-12" retained in the air along the walls of the duct. The effectiveness of the mixing orifice was demonstrated by injecting a known concentration of "Freon-12" at the edge of the duct downstream of a carbon bed to mock up a leak path at the outermost edge of the bed. Sampling of the air stream showed that thorough mixing had occurred and that a similar leak path in an actual carbon bed would be detected readily. The air flow was measured by a calibrated orifice located at the end of the duct.

During the test, a purge flow of 5000 cc/min was supplied continuously to the sampling system. A side sample stream of about 30 cc/min was drawn from the purge flow and supplied to a 6-way sampling valve of the "Freon" detector. A flow diagram of the sampling system is shown in Figure 2, and is described in greater detail under "Freon" Detector. Complete details of the test procedure for evaluating carbon beds are described in the Appendix.

FIG. 2 SAMPLING AND DETECTION SYSTEM

"FREON" DETECTOR

The instrument used for detection of "Freon-12" was a Model A-600-B gas chromatograph with an electron capture detector, made by Wilkens Instrument & Research, Inc. A Leeds & Northrup Co. strip-chart recorder was used to record the response of the detector. The automatic sampling system was controlled by a timer made by Zenith Electric Co.

Of primary importance to the detection system was the electron capture cell. The cell is extremely sensitive to all halides, including the "Freon-12" (CCl_2F_2) used in the technique described here, and relatively insensitive to pure hydrocarbons. The theory and operating techniques of the electron capture detector as well as precautions and source of faulty indications are described in the literature⁽⁷⁻¹¹⁾. A schematic diagram of the cell is shown in Figure 3. The cell is an ionization chamber which was attached to an electrical potential; an electrometer was used to detect small changes in current flow through the cell. The cell is made of a metal cylinder cathode with a tritiated titanium foil insert (250-mc radioactive source that emits beta particles). A metal tube is inserted in one end of the cylinder; the tube, which is insulated from the cathode by glass, serves as an anode.

FIG. 3 ELECTRON CAPTURE DETECTOR CELL

Dry N_2 gas was constantly purged through the cell as a carrier during normal operation. The N_2 was passed through a molecular sieve column to remove vapor impurities from the N_2 upstream of the instrument. Beta radiation from the tritium source ionized N_2 molecules and formed electrons that produced a steady current; which is called the standing current. If a sample containing electron-absorbing molecules (such as "Freon-12" or O_2) is injected into the N_2 carrier gas, the current is reduced by an amount proportional to the sample concentration. The current flow through the cell was recorded, and changes in the current flow for known "Freon-12" concentrations were used to indicate the unknown sample concentration. A variable resistor (installed in one of the electrical leads from the cell as shown in Figure 3) was used to adjust the standing current of the cell for maximum "Freon-12" sensitivity. Calibration of the cell for known "Freon-12" concentrations in air is shown in Figure 4.

FIG. 4 "FREON-12" CALIBRATION OF ELECTRON CAPTURE DETECTOR

Because the carbon beds were located in an air stream, "Freon-12" had to be detected in an air sample. Furthermore, air contains about 20% O_2 , and the simultaneous admission of O_2 and "Freon-12" in an air sample to the cell would result in the O_2 response masking the relatively small response of "Freon-12" at concentrations <1 ppm. Consequently, "Freon-12" and O_2 were separated before entering the detector cell. This separation was accomplished by a chromatograph column used in conjunction with the 6-way sampling valve discussed below. The chromatograph column consisted of stainless steel tubing, 10 ft x 3/32 in. ID, that was packed with 30% SF-96 on 45/60-mesh "Chromosorb" P made by Johns-Manville. An interval of 1 minute between samples was required with the column at 50°C and with a N_2 flow of 50 cc/min to obtain adequate resolution of the O_2 and "Freon-12".

The 2-position, 6-way sampling valve (Figure 2) was operated by a solenoid connected to the automatic timer. Before injection of a sample to the detector, the normal sample flow passed (a) through one chamber of the sample valve, (b) through a sample loop (volume of 1/4 ml), and (c) through a second valve chamber to a rotameter that exhausted to the atmosphere. The normal N_2 flow passed through the third valve chamber and then through the chromatograph column and the electron capture cell. When sampling was started, the automatic timer actuated the solenoid for a period of 5 seconds and at 1-minute intervals. The solenoid repositioned the 6-way sampling valve, and the N_2 flow was diverted as shown in Figure 2. In the sampling position, the sample flow was shut off momentarily and the N_2 flow swept out the 1/4-ml sample; the N_2 sample mixture then flowed to the chromatograph column.

The chromatograph column separated the O_2 and "Freon-12"; O_2 was eluted first, followed by "Freon-12". At the end of the column, the sample entered the electron capture cell. A typical recorder trace of the O_2 and "Freon-12" response of the detector is shown in Figure 5. The drift in the trace with time was apparently caused by accumulation of small quantities of O_2 that was not eluted completely from the column during the sampling interval; this slight drift had no significant effect on the "Freon-12" sensitivity or the test procedure.

During production testing, the usable sensitivity of the detection system for "Freon-12" was about 0.030 ppm (1 recorder unit deflection as shown by the calibration, Figure 4). With a "Freon-12" concentration of 500 ppm upstream of a carbon bed, leaks equivalent to or greater than 0.006% of the total bed flow were detected readily.

FIG. 5 TYPICAL RESPONSE OF ELECTRON CAPTURE DETECTOR FOR OXYGEN AND "FREON-12"

The upper limit for detection of "Freon-12" (consistent with linear response) was not determined, but the manufacturer reported that it would be a concentration equivalent to 30% of the standing current (or about 1 ppm for the conditions shown in Figure 4). The inlet "Freon-12" concentration was determined in the test from measured flows of "Freon-12" and air (indicated by a calibrated rotameter and orifice meter, respectively).

The "Freon-12" calibration of a given electron capture cell was reproducible at the operating conditions shown on Figure 4. No significant variation in the calibration was observed during repeated checks of any one cell, although variations may exist between different cells and at different operating conditions. After 1 to 3 months of service, the standing current of the cell decreased due to adsorption of vapor impurities on the tritium source. These impurities were probably volatile organics eluted from the chromatograph column, but impurities in the air samples could cause the same effect. To restore the standing current and the sensitivity for "Freon-12", the cell was cleaned in a 5% KOH solution.

RESULTS

A typical recorder trace from a "Freon-12" test of a carbon bed is shown in Figure 6. Initially no "Freon-12"

FIG. 6 RECORDER TRACE OF TYPICAL "FREON-12" EVALUATION OF CARBON BED

was detected downstream of the bed. After a few minutes, desorption or penetration began and the "Freon-12" concentration in the exit stream increased. Typical curves of the "Freon-12" adsorption efficiency of acceptable and unacceptable beds are shown in Figure 7. Curve A represents an acceptable bed as discussed above. Curve B represents an unacceptable bed that had settled and reduced the bed thickness about 50% at the boundaries of the bed casing. Curve C represents an unacceptable bed in which 0.12% of the total flow bypassed the carbon through leak paths.

Qualitative tests showed that the adsorption-desorption characteristics of "Freon-12" on carbon beds depended principally on air velocity and H_2O content in the carbon. Inlet "Freon-12" concentration and air² temperature had a lesser effect. As the magnitude of any one of these conditions increased, "Freon-12" desorbed or penetrated the carbon bed more quickly with time, and the general shape of the "Freon-12" efficiency curve was similar to that shown by Curve B, Figure 7; consequently, it was not possible to determine if significant leak paths or bed settling existed in carbon beds if the conditions were changed. The effect of trace impurities adsorbed on carbon during extended exposure to atmospheric air flow is also not known at the present time. These effects were eliminated in evaluating SRP carbon beds for leaks by exposing individual beds to

FIG. 7 "FREON-12" ADSORPTION EFFICIENCY OF ACCEPTABLE AND UNACCEPTABLE CARBON BEDS

conditions that were demonstrated to have no significant effect on the results of the technique. These conditions were:

- New carbon beds of the type described in DP-778⁽¹⁾
- Maximum air velocity of 20 ft/min (based on carbon bed face area)
- Maximum air temperature of 35°C

Some of the production carbon beds contained excessive amounts of H₂O (because of packing and/or storage during humid summer conditions), and these beds showed excessive "Freon-12" penetration. Qualitative examinations showed that the H₂O content in the carbon was about 5% or greater by weight. Subsequently, the beds were desorbed of H₂O and "Freon-12". Acceptable adsorption efficiencies were obtained when the dry beds were re-evaluated with "Freon-12".

Small-scale carbon beds with a "Freon-12" removal efficiency of 99.99+% also removed 99.99+% of I¹²⁷-I¹³¹ vapor mixtures in air flowing at 70 ft/min and at 30°C. The small-scale beds, techniques, and equipment for the destructive I₂ tests are described in DP-778⁽¹⁾. A known leak with a calculated leakage flow of about 0.04% of the total flow through

a carbon bed was confirmed by the "Freon-12" evaluation. Tubing was installed in the bed, as shown in Figure 8, so that part of the flow bypassed the carbon. A subsequent destructive test of this bed showed an I_2 adsorption efficiency of 99.99%. The I_2 penetration of the bed was only $\frac{1}{4}$ of that indicated by the "Freon-12" evaluation; the lower I_2 penetration was apparently caused by I_2 adsorption on the tubing wall and/or I_2 adsorption on small carbon particles in the tubing.

FIG. 8. SECTION OF CARBON BED WITH KNOWN LEAK PATH

"Freon-12" is readily desorbed from carbon beds by exposure to exhaust air flow of reactor ventilating systems for about 1 day. Exposure of carbon beds to air flowing at 70 ft/min and at 100°C desorbed all detectable "Freon-12" in about 2 hours. Small-scale beds almost saturated with "Freon-12" (exposure to "Freon-12" at a concentration of 500 ppm in air for $\frac{1}{2}$ hr) continued to adsorb 99.99% of I_2 vapor when tested as discussed above. Consequently, it was not necessary to desorb "Freon-12" prior to installation of carbon beds.

The "Freon-12" technique was used successfully to evaluate 840 carbon beds to be installed in SRP reactor buildings. Detailed results of nondestructive and destructive evaluations of these beds will be discussed in a later report.

The current "Freon-12" technique, in conjunction with destructive I_2 tests of typical carbon samples, provides a satisfactory method of evaluating carbon beds prior to field installation. The technique would not be effective for detecting leak paths in gasket seals of carbon beds after field installation because of the limitations discussed previously. Development of a standard test for installed carbon beds has been started at SRL. Initial work will be to reduce restrictions on the current method. Preliminary analysis indicates that other "Freon" compounds with higher molecular weight and boiling point will not be affected by air velocity and adsorbed H_2O to as great an extent as "Freon-12". No loss in sensitivity of detection is expected, because the sensitivity of the electron capture detector apparently increases with increasing number of halogen molecules in the compound. Evaluation of these "Freon" compounds has been started.

APPENDIX - PROCEDURE FOR "FREON-12" LEAK EVALUATION OF CARBON
BEDS TO BE INSTALLED IN PRODUCTION FACILITIES

1. Energize electron capture instrument in accordance with manufacturer's instructions and establish the following conditions:

Electrometer settings

Input impedance - 10^7 ohms

Output sensitivity - 1X

Attenuator - 1X

Detector standing current - 200 recorder units

Oven temperature - 50°C

N₂ carrier gas flow - 50 cc/min

2. Start air blower and sampling system to verify that no "Freon-12" was left in system from previous evaluation.
3. If no "Freon-12" is detected, stop blower and sampling. Inspect carbon unit for gasket defects (repair if necessary) and install in facility.
4. Start blower and adjust air flow at 250 cfm (approximately 20 fpm face velocity). Maximum allowable air temperature is 35°C. Start sampling system.
5. Analyze two air samples and then inject "Freon-12" at a concentration of 500 ppm in the air upstream of carbon unit. The injection begins 20 seconds after completion of the last air sample to allow for residence time in the duct and sampling system.
6. Analyze three samples taken downstream of carbon unit for "Freon-12". After the third sample is taken, "Freon-12" injection, sampling, and the blower are stopped.
7. The heights of the first and second "Freon-12" peaks are indicative of the carbon bed efficiency. If the "Freon-12" concentration in any of the three samples exceeds 0.400 ppm (99.92% bed efficiency), the bed is set aside for additional evaluation.

8. If the carbon bed does not meet the above requirements, the bed is exposed to air flowing at 1000 cfm (70 fpm face velocity) and at 100°C for 2 hours to desorb the "Freon-12". This exposure removes all detectable "Freon-12" as well as H₂O adsorbed on the carbon. The bed is then evaluated to determine if the unit was defective or if an excessive amount of H₂O was adsorbed on the carbon.
9. After completion of the "Freon-12" evaluation, the pressure drop across the bed is measured at the rated air flow of 1000 cfm.

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