SESSION IX

MONITORING

Thursday, August 15, 1974
CHAIRMAN: Dr. A.P. Malinauskas

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CONTINUOUS MONITORING OF RADIOACTIVE IODINE EMISSIONS

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Abstract

Two important reasons for the continuous monitoring of airborne ^{131}I in nuclear installations are:

1. Requirement of immediate startup of standby iodine filter systems for off-gas cleaning at the moment of inadmissible iodine release to the environment.
2. Control of the level of airborne radioactive iodine inside a nuclear installation to avoid contamination of and incorporation by the operating personnel.

The detection of ^{131}I in atmospheric concentrations down to $2x10^{-9}$ Ci/m^3 and even less, which may be necessary in health physics, calls for a specific measurement technique. A monitor has been developed for the continuous measurement of the ^{131}I-content of air in which iodine is accumulated on a special filter cartridge. The ^{131}I-activity is measured continuously by detection during the accumulation process and displayed on a ratemeter. An alarm may be given if a certain level of the total accumulated iodine activity (A) is reached in the filter cartridge (integral method) or a preset ratio of dA/dt is exceeded (differential method).

Elemental iodine and methyl iodide can be measured with the same detection sensitivity even in air of extremely high humidity. The influence of rare gas activity on the measurement is low due to the use of an inorganic iodine sorption material.

The construction principles of the continuous iodine monitor will be discussed. Data will be given on the length of the sampling period preceding the alarm, for two different versions of the monitor, the detection limit and the selectivity of ^{131}I-measurement in off-gas which might also contain radioactive rare gases.

I. Introduction

Detection of the most crucial iodine isotope for safety purposes, ^{131}I, in the range of the minimum airborne concentration requires a special method of measurement. Direct measurement of ^{131}I in air is not feasible in the presence of other gaseous airborne contaminants, e.g., noble gas isotopes or if the iodine measurement is disturbed by an increased local dose due to other radioactive sources. These conditions may be expected to be present under conditions of monitoring the recirculation room air and exhaust air of nuclear power stations.

For the reasons cited above it is necessary to enrich ^{131}I in a suitable filter cartridge for measurement purposes. The iodine activity may be continuously measured by a detector and indicated on a ratemeter during the very enrichment procedure. If a preset pulse rate is reached, a signal indicating the airborne contamination can be actuated after a certain collection time which is practically inversely proportional to the activity concentration of iodine in the air because of the ratio between measurement time and halflife of ^{131}I.

II. Design Criteria

A number of aspects rule the design and construction of the setup referred to above:

- The larger the volume flow of air passing through the filter cartridge, the more quickly airborne contamination will be detected. Large volume flows require large filter cartridges because a certain minimum stay time of the air in the iodine adsorbing material is required for complete iodine separation.
- The sensitive detector volume should be kept as small as possible in the interest of a low background count rate. This also applies to the total volume of the detector and the filter cartridge because there must be a shielding enclosing both the detector and the filter cartridge.
- Adsorption of noble gases onto the surface of the iodine sorption material of the filter cartridge should be as low as possible so that low iodine concentrations can be detected in an air additionally contaminated by noble gases.
- Gaseous iodine may be present in various forms, mainly as elemental iodine (I_2) and methyl iodide (CH_3I). The removal efficiency of iodine sorption materials differs for the different forms of gaseous iodine. Accordingly, there will be a different activity distribution of the iodine components in the filter cartridge (e.g. steep removal profile for I_2, flatter removal profile for CH_3I).
- The removal of CH_3I is influenced very strongly by the relative humidity of the intake air. As the humidity of the air increases the removal efficiency of the filter cartridge will decrease.

- Iodine, especially elemental iodine, is deposited on surfaces. The effect on reading of a contamination of the intake paths and the detector should be minimized by the choice of material and the shape of the unit.

- Particulate radioactive material which may upset the measurement of iodine should not be allowed to be removed in the filter cartridge. If the air is prefiltered by means of an aerosol filter, no gaseous iodine should be removed in the filter.

- The iodine activity should be measured as specifically as possible, e.g., by means of a single channel analyzer through the photopeak.

III. Measuring Setup

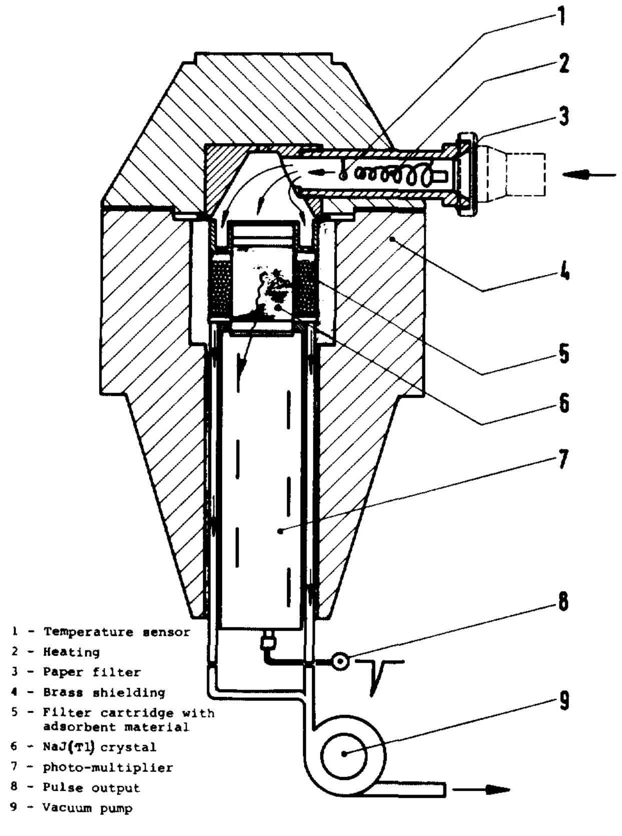

Fig. 1 Detector of the iodine monitor with collection device

Fig. 1 shows an arrangement of the detector and the iodine filter cartridge in which the points mentioned above have largely been taken into account. The same setup was chosen for the iodine monitor developed jointly by the Karlsruhe Nuclear Research Center and Herfurth GmbH, Hamburg. A filter cartridge encloses a cylindrical NaI(Tl) scintillation crystal. The detector and the filter cartridge are surrounded by a shielding. Contamination of the interior of the air intake and distribution ducts due to iodine plateout is largely avoided by the shape of the unit and the use of teflon as the wall material. The intake air of the filter cartridge is preheated to reduce the relative humidity of the air. Heating by 7 °C is sufficient to reduce the relative humidity, which above all affects the removal behavior of methyl iodide, from 100% to $\leq$ 70% at air temperatures of $\leq$ 40 °C. In this way, the volume flow of air through the filter cartridge can be greatly increased (reduction of the required stay time) and iodine sorption material on an inorganic basis can be used whose application will greatly reduce noble gas adsorption in the filter cartridge.

Experiments were performed on a prototype iodine monitor in order to dimension the filter cartridge(1). In Table I, only the results of a few measurements will be given which were performed with the present filter cartridge of the iodine monitor in which an Ag-impregnated inorganic sorption material (AC-6120) is used for iodine trapping. The efficiency of the iodine detector in these experiments was 3.4%, which corresponds to a sensitivity of detection of 1.25 cps/nCi ^{131}I. Background was around 0.8 cps. The removal efficiency of the filter cartridge can be enhanced by variation of the grain size and of the impregnation of AC-6120, the rest of the design remaining unchanged.

IV. Influences of Noble Gases upon the Measurement

Radioactive noble gas isotopes of xenon and krypton occurring in the exhaust air of reactors and fuel reprocessing plants can greatly affect the continuous measurement of iodine. This applies in particular to ^{135}Xe whose activity concentration in the exhaust air of reactors may exceed that of ^{131}I by several orders of magnitude.

A series of measurements were performed to determine the relative adsorption behavior of ^{133}Xe on various iodine sorption materials. After the adsorption-desorption equilibrium of xenon had been established between the flowing gas phase (air) and the surface of the iodine sorption material, the ^{133}Xe activity was measured in the sorption bed (Table II). The count rates measured at the same ^{133}Xe concentration in the air and with otherwise unchanged boundary conditions in sorption beds made of impregnated activated charcoal Norit CG 1, molecular sieve type LMS 13X-Ag and AC-6120 showed a ratio of 184:1.42 :1. Hence, if the iodine sorption material mentioned last is used in the filter cartridge instead of activated charcoal, an increase in the background count rate due to Xe activity present in the air may be expected which is more than 2 orders of magnitude smaller.

For a direct measurement of the influence of ^{135}Xe upon the background count rate of the iodine monitor the shortlived ^{135}Xe ($T_{1/2}$ = 9.5 h) was produced by activation of ^{134}Xe in the thermal column of a reactor. With a width of the window of 20% and setting to the photopeak of ^{131}I of the single channel analyzer connected to the detector, an increase in the background count rate from 0.8 cps (background) to 6.8 cps was found for an activity concentration of the intake air of 160 μCi ^{135}Xe/m^3. The sensitivity of the iodine monitor is around 1.25 cps/nCi ^{131}I. A measurement effect corresponding to this iodine activity would be caused by a ^{135}Xe activity concentration of the intake air of 33 μCi/m^3.

While iodine is continuously removed in the filter cartridge so that increasing count rates are registered as the collection time advances, there is no longer any increase in the count rate after the adsorption-desorption equilibrium of the noble gases has been established in the filter cartridge. Accordingly, the disturbance of the iodine measurement by very high noble gas activity in the intake air continues to lose significance as collection time goes on.

V. Prefiltering of Radioactive Aerosols

The removal of radioactive aerosols by filters from the intake air of the iodine monitor may be necessary to prevent any falsification of the count rate for gaseous iodine due to spurious activity partly removed in the filter cartridges as aerosols. This removal by filters is permissible only if the aerosol filter does not retain any major fractions of gaseous iodine. Since elemental iodine is particularly reactive and moreover is strongly adsorbed onto surfaces, its removal by an aerosol filter (Luwa Gelb) was measured in the intake air of the filter cartridge. The data are given in Table III.

In experiments with carrierfree ^{131}I a fraction of $\leq$ 0.8% was retained by the aerosol filter. In the (high) range of concentration between 1.6 and 2700 μg I_2/m^3 of air the maximum iodine removal efficiency of a Luwa Gelb filter was 3.7%. Whether it is meaningful to have a prefiltration of aerosols (radioactive iodine may be bound to aerosol particles) depends upon the merits of each specific case. If necessary, the aerosol filter can be incorporated in the adsorber section of the filter cartridge to allow also iodine bound to particles to be measured. The possible increase in the background count rate due to the removal of other radioactive aerosols in the filter cartridge would then have to be tolerated.

VI. Sensitivity of Detection and Collection Time

The sensitivity of iodine detection and the time until a given alarm threshold is reached are a function of the efficiency η of the detector, the iodine activity concentration c, the intake volume flow of air v, and the background count level R_o. If a low airborne iodine concentration is assumed, the actuation of an alarm will be determined by the collection time needed for removal of a sufficient level of iodine activity in the filter cartridge. By contrast, the time constant of the ratemeter measurement can be neglected. The alarm threshold can be set to a measurement rate which is given by x times the background count rate or, if the background count rate is high, rather by x times the standard deviation of the background count rate.

If an ^{131}I load on the filter cartridge stemming from a previous collection time is present at the beginning of the new collection time, the alarm threshold must be shifted to a higher level in the simple design of an iodine monitor without a digital differential calculator. The collection time necessary for the alarm threshold to be reached will then be extended.

If the alarm threshold is set the way outlined above, the collection time t is determined as follows:

$$t = \frac{(x-1) \cdot R_o \cdot 100}{c \cdot k \cdot \eta \cdot v} \text{ (h) and} \qquad (1)$$

$$t = \frac{x \cdot \sqrt{R_o} \cdot 100}{c \cdot k \cdot \eta \cdot v} \text{ (h)} \qquad (2)$$

with R_o indicated in cps, c in Ci/m^3, v in m^3/h and η in %; k (conversion factor) = 3.7×10^{10}; η was determined to be 3.4% for the iodine monitor.

The necessary collection time for the detection of an ^{131}I concentration in the air of 2×10^{-9} Ci/m^3 is only 3.8 min for $R_o = 0.8$ cps and $v = 5$ m^3/h, if $2 R_o$ is assumed to be the lower limit of detection (calculated by Eq. (2)).

Measurements with an iodine monitor designed in the light of the aspects listed above in collaboration between the Karlsruhe Nuclear Research Center and Herfurth GmbH, Hamburg, resulted in the operating times listed in Table IV up to the point of alarm actuation as a function of the ^{131}I concentration in the intake air.

The measurements were performed in the controlled area in the presence of an increased local dose rate caused by hard γ-radiation. The background was 9.5 cps instead of the level of 0.8 cps measured outside the controlled area. The intake path of the air containing CH_3I was 1.65 m long and consisted of a multiply bent and flanged glass tube of an inside diameter of 25 mm. The linear air velocity in the glass tube was 2.7 m/sec. For actuation of the alarm a level was preset which corresponded to an increase in the background count rate R_o by a value of $4\sqrt{R_o}$. As was to be expected, the adsorption of $CH_3{}^{131}I$ in the line (concentration ≤ 3 μg CH_3I/m^3 of air) and the air intake and the time constant of the ratemeter (10 sec was the value selected) resulted in times to alarm actuation which were a few minutes (3.1 to 6.4 minutes) longer than had been calculated according to (2).

VII. Direct Indication of Airborne ^{131}I Concentration

If the concentration of ^{131}I in the air is constant, a monotonically increasing pulse rate is obtained because the halflife of ^{131}I (8.05 d) is long enough to prevent a saturation equilibrium of the activity removed to be reached even approximately during the measuring time. If a direct indication of the concentration of ^{131}I in the air is wanted, the rate of increase of the pulse rate must be determined.

A digital ratemeter additionally installed in the iodine monitor cyclically, corresponding to the set measuring period, e.g. every 3000 seconds, supplies a new measured result of the mean pulse rate to a digital differential calculator which then indicates the result of the calculation

$$R_n - R_a = R_d \qquad (3)$$

where R_n is the "new mean pulse rate," R_a the "old mean pulse rate," and R_d the pulse rate difference over the measuring period considered.

The concentration of ^{131}I in the air (in nCi/m^3) results from R_d (in cps) and the period of measurement (in sec) to be

$$c = \frac{k \cdot R_d}{t} \qquad (4)$$

where k is a constant; for a sensitivity of detection of the iodine monitor of 1.25 cps per nCi of ^{131}I and a flow of 5 m^3/h (corresponding to 1.39×10^{-3} m^3/sec) of air through the filter cartridge, k = 576 [nCi sec^2/m^3 count].

Here is one example: R_a = 10 cps, R_n = 11.2 cps, R_d = 1.2 cps, measuring time = 3000 sec.

$$c = \frac{576 \times 1.2}{3000} = 0.23 \pm 0.031 \text{ nCi/m}^3.$$

R_a = 10 cps means that approximately 8 nCi of ^{131}I had already been removed by the filter cartridge.

Table V is a compilation of a few values of R_d as a function of the ^{131}I concentration in air and the length of the measuring period preset on the ratemeter.

The measuring period should be chosen on the basis of the pulse rate R_o existing at the beginning of the measurement (from the ^{131}I load existing on the filter cartridge and the background count rate) because the statistical error of R_d at low airborne ^{131}I concentrations is determined largely by the statistical error of R_o.

In Table VI this fraction (ΔR_d) of the overall statistical error of R_d is given for a few examples as a function of pre-loading of the filter and the length of the measuring period. The data in Tables V and VI indicate the measuring periods that should be preset for the measurement of minute airborne activities if the filter cartridge is already preloaded at the beginning of the measurement, so that values of R_d are obtained with a sufficiently small statistical error. Table VI under t_g also indicates the overall collection time of the iodine monitor which results in the buildup of the pulse rate R_o at certain ^{131}I concentrations in air.

If the digital ratemeter indicates a pulse rate, e.g., of 10 cps before the beginning of the measuring period (corresponding to 8 nCi of ^{131}I in case of a negligible background count rate), a measuring period of approximately 1000 sec should be set if an airborne activity of 2 nCi/m^3 is to be detected clearly.

It should be pointed out that the warning thresholds installed will actuate an alarm independent of any setting of the measuring period if there is a sudden occurrence of higher concentrations. This is achieved by having the digital difference calculator continuously generate the respective difference. However, the difference calculated is stored in the display only at the end of a measuring period.

Reference

(1) Wilhelm, J.G., "Entwicklung eines Prototyp-Jodmonitors", Report of the Karlsruhe Nuclear Research Center, KFK 1365, p. 105-109 (1970)

Appendix

Tab. I Removal efficiencies for I-131 of the filter cartridge of the iodine monitor

Iodine sorption material: AC 6120, grain size: 1 - 2 mm
Impregnation: 8.8 % Ag
Air flow: room air of 20°C and 70 % R.H.
Air heated to 47°C prior to entering the filter cartridge. Volume flow 4.7 m^3/h

Experiment I : CH_3I-air mixture, concentration 70 μg CH_3I-127/m^3, activity concentration 8.3 nCi CH_3I-131/m^3

Experiment II : I_2-air mixture, concentration 40 μg I_2-127/m^3, activity concentration 135 nCi I-131/m^3

Depth of bed (cm)	Stay time (s)	Removal efficiencies in %	
		Experiment I	Experiment II
1.8	0.05	76.1	- +
3.6	0.10	96.0	99.91
5.4	0.15	99.1	99.95

\+ not measured

Tab. II Xe-133 equilibrium activity in various iodine sorption materials

Carrier gas: laboratory air, 1 atm, room temperature, linear air velocity 17 cm/sec

Xe-133 activity: approx. 50 μCi Xe-133/1 of air

Time to establishment of the adsorption-desorption equilibrium in the sorption bed: 3 - 6 min (controlled in several long time tests extending up to 6 h)

Sorption bed: depth 10 cm, diameter 2.5 cm, volume 49 cm^3

Sorption material	Specific surface according to BET + m^2/g	Xe-133 equilibrium activity in the adsorber bed in μCi ++
Empty tube exposed to Xe-133-air mixture	-	0.0175 (in tube volume)
Activated charcoal (Norit CG I)	1000	1.67
Molecular sieve (LMS 13 X changed to Ag form)	490	0.0129
$AgNO_3$ impregnated inorganic material AC 6120	125	0.00909

\+ Sorption material before impregnation and ion exchange, respectively

++ Error of single values referred to calibration ± 5 %. The activity of the Xe-133 source was known within ± 30 %

Tab. III Removal of I-131 (loaded in the form of elemental iodine) on Luwa Gelb type paper filters

Iodine concentration in air + ($\mu g/m^3$)	Desorption time (min)	Removal efficiencies of paper filters (%)	
		Filter 1	Filter 2
Carrierfree sample	5	0.69	0.80
Carrierfree sample	30	0.53	0.49
Carrierfree sample	60	0.32	0.34
1.6	5	3.5	3.7
3.5	5	3.0	3.0
65	45	0.61	0.40
84	60	1.5	0.34
100	5	3.0	3.2
470	5	1.9	1.7
550	5	0.92	0.61
580	5	2.4	2.4
850	15	1.8	1.2
1100	30	0.24	0.39
2700	5	0.36	0.39

+ averaged over an exposure time of 5 minutes

Tab. IV Time to alarm for various airborne activity concentrations of CH_3I-131 (iodine monitor without a differential calculator)

I-131 concentration in air in nCi/m^3	2	5	10	50
Operating time, measured, in min	49.5	24.3	14.5	4.9
Collection time as calculated according to (2), in min	44.6	17.9	8.9	1.8

Tab. V Pulse rate difference R_d as a function of the I-131 concentration and the measuring period

I-131 concentration in air	Set measuring period		I-131 activity on measuring filter	R_d
(nCi/m^3)	(sec)	(min)	(nCi)	(cps)
2	3×10^2		0.833	1.04
2	6×10^2	10	1.67	2.08
2	10^3		2.78	3.47
2	1.98×10^3	33	5.50	6.87
2	3×10^3		8.33	10.4
2	6×10^3	100	16.7	20.8

Tab. VI Statistical error of R_d due to preloading of the measuring filter for various measuring periods

R_o		ΔR_d (2σ) as a function of the measuring period ++						Total collection time t_g for the buildup of R_o at different concentrations +		
		measuring period (sec)						I-131-concentration in air (nCi)		
		3×10^2	6×10^2	10^3	1.98×10^3	3×10^3	6×10^3			
cps	nCi +	cps	cps	cps	cps	cps	cps	2	0.2	0.04
10	8	0.52	0.37	0.28	0.20	0.16	0.12	0.8 h	8.11 h	1.79 d
30	24	0.89	0.63	0.49	0.35	0.28	0.20	2.41 h	1.04 d	6.35 d
100	80	1.63	1.15	0.89	0.64	0.52	0.37	8.11 h	3.92 d	∞
300	240	2.82	1.94	1.55	1.10	0.89	0.63	1.04 d	22.8 d	"
10^3	800	5.16	3.65	2.83	2.01	1.63	1.15	3.92 d	∞	"
3×10^3	2400	8.94	6.32	4.89	3.48	2.82	2.00	22.8 d	"	"
10^4	8000	16.3	11.5	8.94	6.35	5.16	3.65	∞	"	"

+ for a negligible background count rate

++ calculated for the difference measurement of R_d from two values

DISCUSSION

DEMPSEY: Would tritium give you any background concern in reactors when it does build up in time in the air?

WILHELM: That wouldn't be counted at all.

CONTINUOUS MONITORING OF NOBLE GASES IN REACTOR STACK EXHAUST

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Abstract

A system at the Savannah River Plant continuously monitors radioactive noble gases (^{41}Ar, ^{133}Xe, ^{135}Xe, ^{85m}Kr, ^{87}Kr, ^{88}Kr) discharged to exhaust stacks from each of three heavy-water moderated reactors. A description of the system, calibration technique, and early operating experience are discussed.

A flowing sample of stack exhaust air, passing through a 2.8-liter stainless steel sample cell, is monitored for noble gases by a lithium-drifted germanium, Ge(Li), detector coupled to a multichannel analyzer. Isotopes of noble gases are counted and release data are updated each ten minutes by teletype printout. The system is calibrated by determining the counting efficiency vs gamma energy relationship for both the detector and the sample cell. These relationships are determined with International Atomic Energy Agency standard point sources, National Bureau of Standards ^{133}Xe and ^{85}Kr standard gas sources, and sources containing mixed radioactive noble gases. Accuracy of calibration for the absolute efficiency of the system is ±15% at two standard deviations. Operating experience shows that the system functions reliably and provides useful data on release of radionuclides to the atmosphere.

Introduction

Of the 1.27 mrems calculated annual radiation exposure contributed by Savannah River Plant atmospheric releases to an individual residing at the plant boundary, approximately 30%(1) is attributed to noble gases released from the reactors. Sources of noble gases released are: venting gas to control reactor blanket gas pressure, leakage of blanket gas, and activation of reactor ambient air. Dilution, dispersion, and decay reduce levels of radioactivity from noble gases to well below the present guides for release to the environs.

Because release rates of noble gases vary as a function of reactor power, fuel cladding integrity, and reactor charge, total gas releases can be determined most accurately by continuous monitoring. As one part of an overall plant effort to improve monitoring of radioactivity released to the environs, a study was made to determine the feasibility of using a lithium drifted germanium, Ge(Li), gamma detector and pulse height analyzer system for continuously monitoring noble gas releases. Feasibility of the system was established and a decision was made to install such a monitor in each of the three reactors.

Discussion

Measurement Technique

Noble gas radioactivity is measured gamma ray spectrometry to obtain both qualitative and quantitative data. Qualitative analysis is possible since each radioactive noble gas emits gamma photons of characteristic energy. These photons are detected by a solid-state gamma detector and differentiated according to energy by a multichannel pulse height analyzer. Quantitative measurements of noble gas radioactivity are possible since the decay rate of each isotope is constant. Activity in curies per liter of air flowing out ot the stack is computed from analyzer count rate data using calibration constants measured for the system.

Gas Sample

Radioactive noble gases from each plant reactor are released to the atmosphere through HEPA and charcoal filters to a 61-meter high stack. A continuous air sample for the noble gas monitors is extracted from the stack at the 45-meter level. The sample is passed into a sample cell, analyzed and returned to the stack (figure 1). The 2.8 liter sample cell (figure 2) is a right circular cylinder made of stainless steel. To obtain good sample exchange, air flows into the bottom of the cell and out the top at a rate of 6 liters per minute, thus exchanging the 2.8 liter sample volume twice a minute. A 0.25-mm thick stainless steel window recessed in the bottom of the cell provides transmission of gamma radiation to the detector with minimal absorption. The sample cell is mounted with the window directly above the Ge(Li) gamma detector. Both the sample cell and detector are shielded by 10 cm of lead to reduce background radiation.

Detector Components

Figure 3 is a block diagram of all the monitor's electronic components, most of which are housed in an air-conditioned cabinet (figure 4) to maintain stable temperature and low relative humidity.

The solid-state detector (figure 5) is a commercially manufactured Ge(Li) detector cooled to liquid nitrogen temperature. The detector is of true coaxial geometry with an absolute efficiency of 1×10^{-4} count per second per gamma per second for 1.333 MeV cobalt-60 gamma photons. The resolution at 1.333 MeV is 2.72 KeV full width at half-maximum, and the peak-to-compton ratio is 28 to 1. The detector is protected from loss of coolant by a "low liquid nitrogen level" alarm which shuts down the high voltage supply to the detector and sounds an audible alarm if nitrogen level becomes low.

A preamplifier and amplifier-baseline restorer amplify and shape electronic pulses from the detector so the pulses will be in a useable form when they reach the multichannel analyzer. These instruments provide high electronic gain yet add a minimum amount of electronic noise to the detector signal.

The multichannel analyzer is a 4096-channel unit which sorts incoming voltage pulses according to voltage amplitudes. Since the voltage of each pulse is proportional to the energy of a gamma photon, a spectrum of the incident photon energies is produced by the analyzer. The spectrum covers energies from 13 to 1856 KeV with a conversion gain of 0.45 KeV per channel. Logic circuitry within the analyzer can be easily programmed to add counts accumulated in several channels for regions of interest in the spectrum. Such regions are selected for each isotope peak and for a corresponding background spectrum level.

Data Accumulation

Data are accumulated for twenty-four hour intervals with intermediate teletype printout of data at the end of each ten minutes of counting. The final printout from a twenty-four hour period is used to determine net counts for each isotope released through the stack. The release rate in curies per day is determined by multiplying the net counts per second by calibration constants and stack flow rate. At the end of each twenty-four hour cycle the analyzer automatically resets itself and begins another accumulation. The intermediate data printouts at ten minute intervals allow correlation of short-term release data with reactor operation. All data are recorded at a central monitoring panel.

Method of Calibration

Though a Ge(Li) detector produces voltage pulses that have accurate proportionality to incident gamma photon energy, the efficiency for photon capture by the detector varies considerable with gamma energy. Since standardized sources of short half-life noble gases were not available for calibrating the monitor, it was calibrated by indirect techniques.

First an accurate measurement of efficiency was made for various radioactive point sources located 25.4 cm from the detector. The point sources were obtained from the International Atomic Energy Agency, National Bureau of Standards, and Savannah River Plant, and had gamma energies from 60 KeV to 1333 KeV(2) (table 1). Figure 6 plots detection efficiency in counts per gamma versus gamma energy in KeV, and clearly illustrates the variation in detection efficiency at various energies.

After establishing detection efficiences for point sources, NBS standard gas sources of relatively long half-life ^{133}Xe and ^{85}Kr (contained in small glass sample vials) were counted at 25.4 cm. Correcting only for gamma attenuation of the glass vials, the detection efficiencies for ^{133}Xe and ^{85}Kr showed good agreement with the efficiencies extrapolated from the point source efficiency curve (figure 6). The glass sample vials were thus shown to be satisfactory calibration sources. Since the reactor blanket gas system contains high concentrations of radioisotopes representative of those being released from the reactor stack (see table 2), samples of this gas were transferred into the glass vials and calibrated to obtain standard sources. Gamma activity for each isotope present in these sources was calculated using efficiency values extrapolated from the point source efficiency curve. The calibrated sources were then released into the stainless steel sample cell and counted. Efficiencies in counts per gamma were established for each noble gas isotope by dividing counts per second measured in the cell by gammas per second calculated for the standards. These efficiencies were then the total efficiencies for the sample cell and detector. This process was repeated a number of times to verify reproducibility. Figure 7 shows total system efficiencies in counts per gamma vs gamma energy in KeV.

Calibration Accuracy

The accuracy of calibration is ±15% at the 95% confidence level and applies to samples within the minimum and maximum sensitivities shown in table 3. The table also shows total errors for stack release rates; these errors include stack flow rate errors and statistical deviation of count rates. The minimum sensitivities in table 3 were established by doubling the background count rate for each isotope, taking the square root, and doubling that number. The maximum sensitivity was fixed by an accumulation limit of 10^6 counts per integral region of interest in the analyzer.

Early Operating Experience

The noble gas monitors have been running online and providing accurate, continuous release data for five months. Other than minor component failures, no abnormal system breakdown has resulted from field operation of a detection system which would normally be operated under laboratory conditions. Field operating personnel, rather than specially trained technicians, maintain and operate the equipment and make release calculations according to written procedures.

The monitors have also served several other useful purposes. For example, monitoring has allowed computer comparison of integrated release data and daily grab samples of stack gas (table 4). Nearly all of the results were within statistical and systematic errors for each of the two sampling systems. The comparison demonstrated that previous release rates based on grab samples represented practicable release data during normal operating conditions; however, grab samples do not provide auditable records nor accurately represent release rates during all operating conditions.

The continuous monitor also allows close analysis of release rate during varied operating conditions. For example, figure 8 shows typical ^{133}Xe release measured over an eight-hour period. The most significant variation in release rate under routine operating conditions occurred during blanket gas venting where release rate increased by a factor of five. However, the short duration of this high release made its overall contribution to total daily release small, 3%.

Continuing Programs

In addition to providing basic stack release data in an auditable form, the noble gas monitors will continue to be used to study the effects of reactor operation on release rates. The monitors will also aid in current stack dispersion studies by providing real time release rate data.

References

1. "Environmental Monitoring in the Vicinity of Savannah River Plant, Annual Report for 1973," DPSPU-74-30-1; prepared for the U. S. AEC by E. I. du Pont de Nemours & Co.

2. Nuclear Data Tables 8, 445-666 (1971) "Catalogue of Gamma Rays Emitted by Radionuclides," M. A. Wakat.

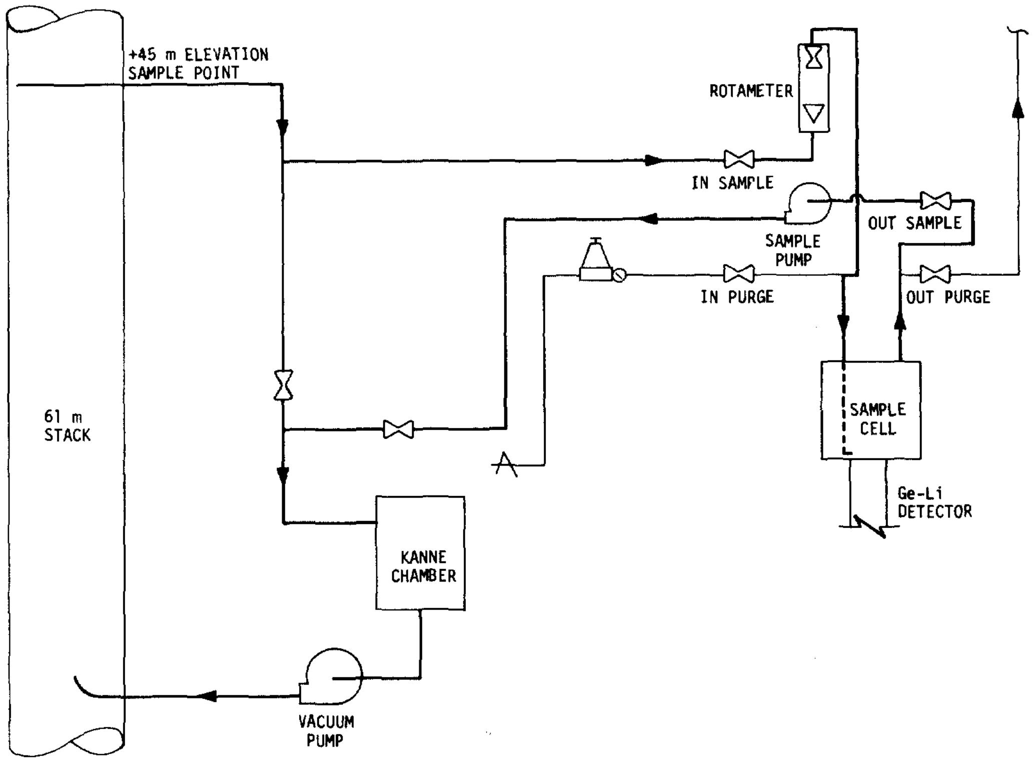

FIGURE 1. NOBLE GAS MONITOR SAMPLING SYSTEM

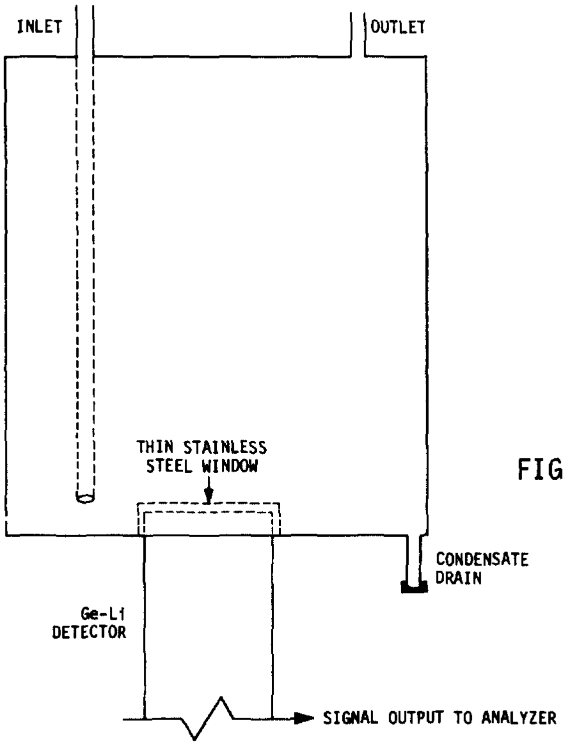

FIGURE 2. SAMPLE CELL, 2.8 LITERS

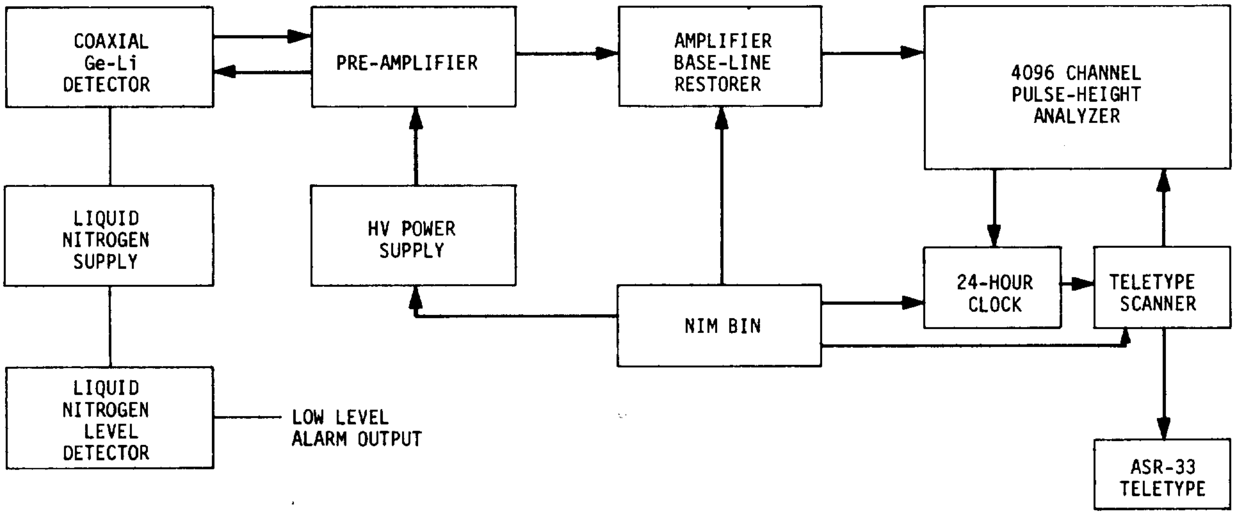

FIGURE 3. GAMMA RAY SPECTROMETER

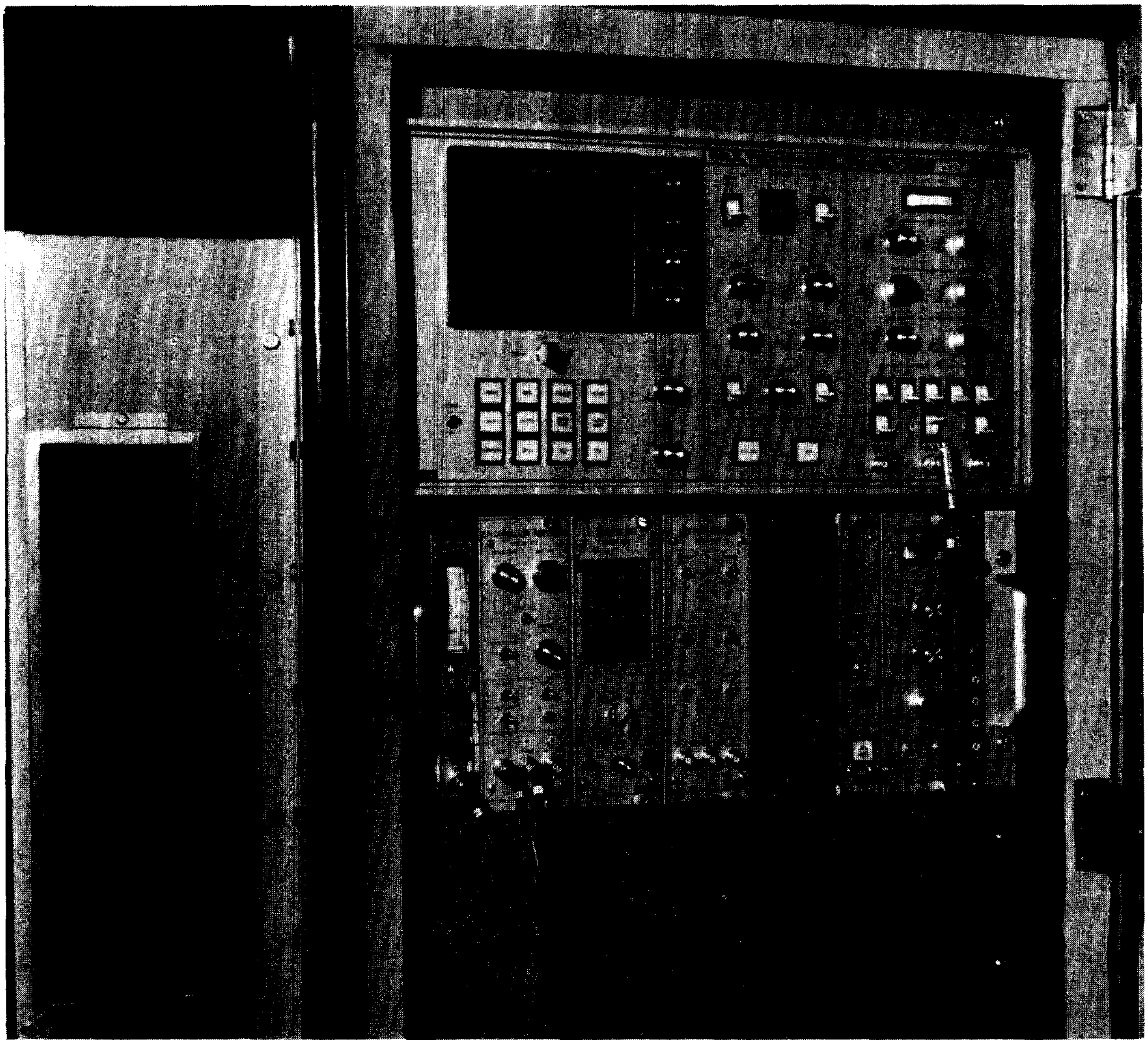

FIGURE 4. NOBLE GAS MONITOR INSTRUMENTATION

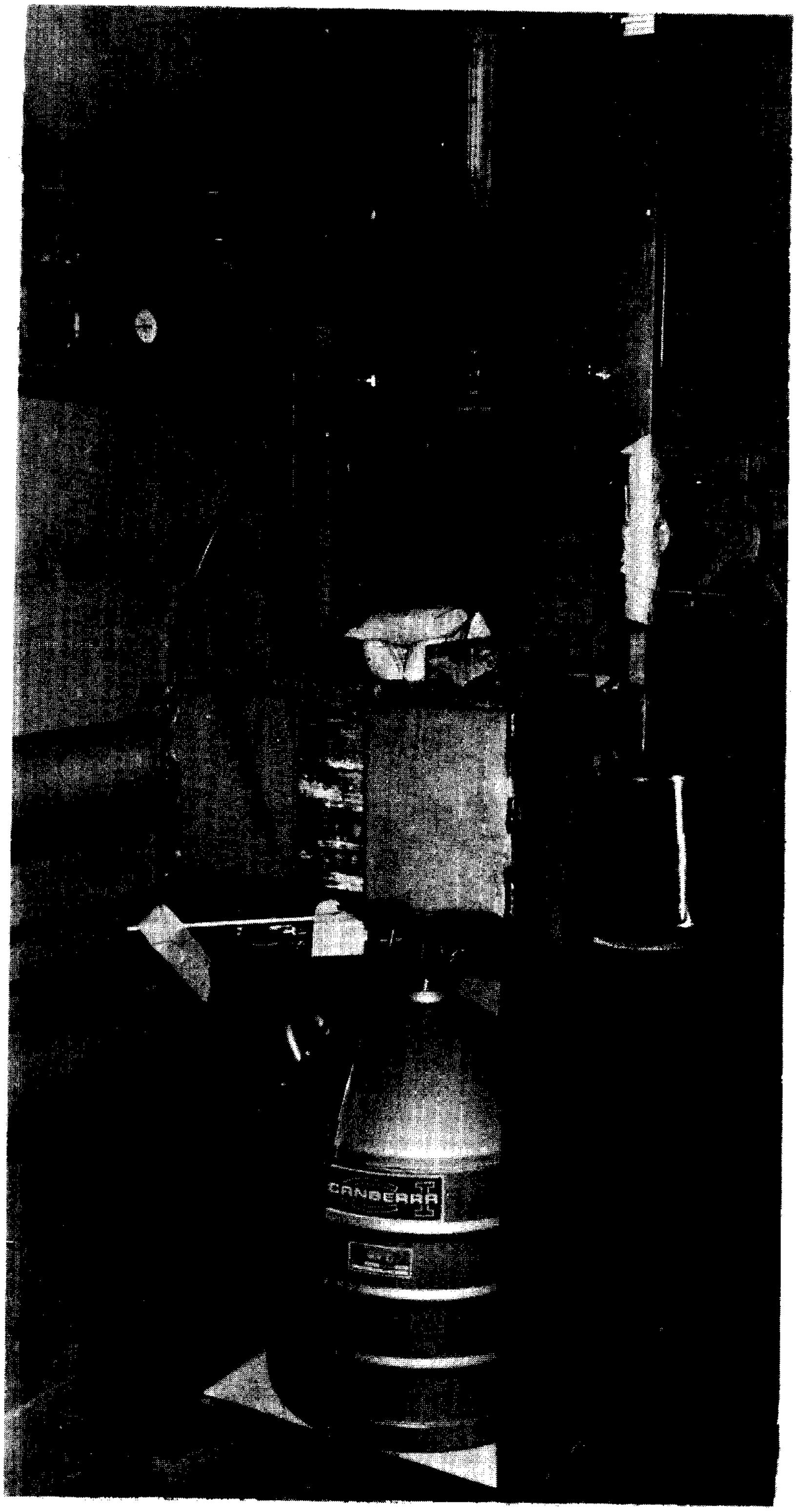

FIGURE 5. Ge-Li DETECTOR ENCLOSURE AND VALVE PANEL

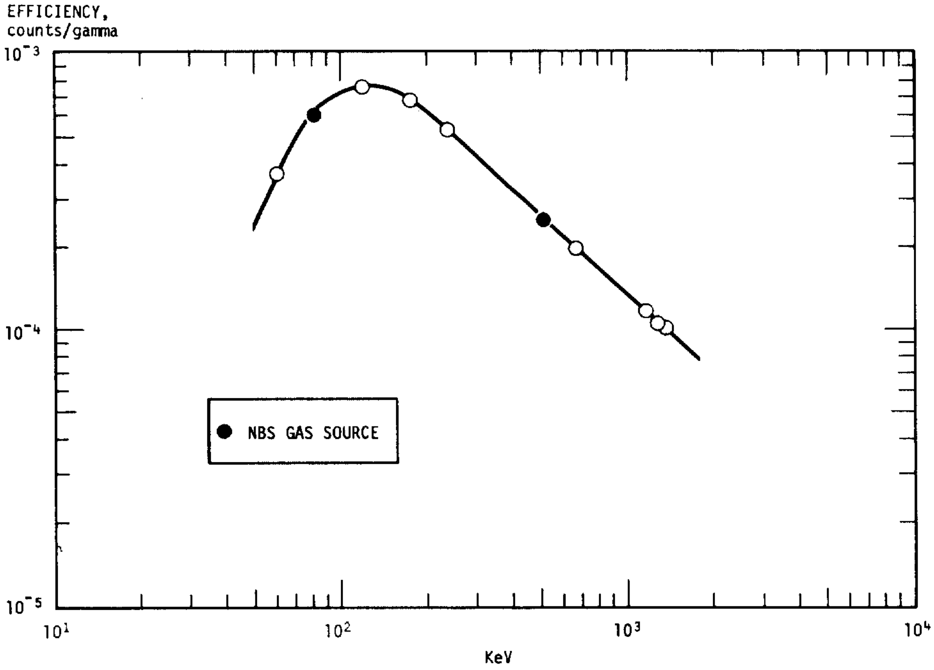

FIGURE 6. DETECTOR EFFICIENCY FOR POINT SOURCES AT 25 Cm

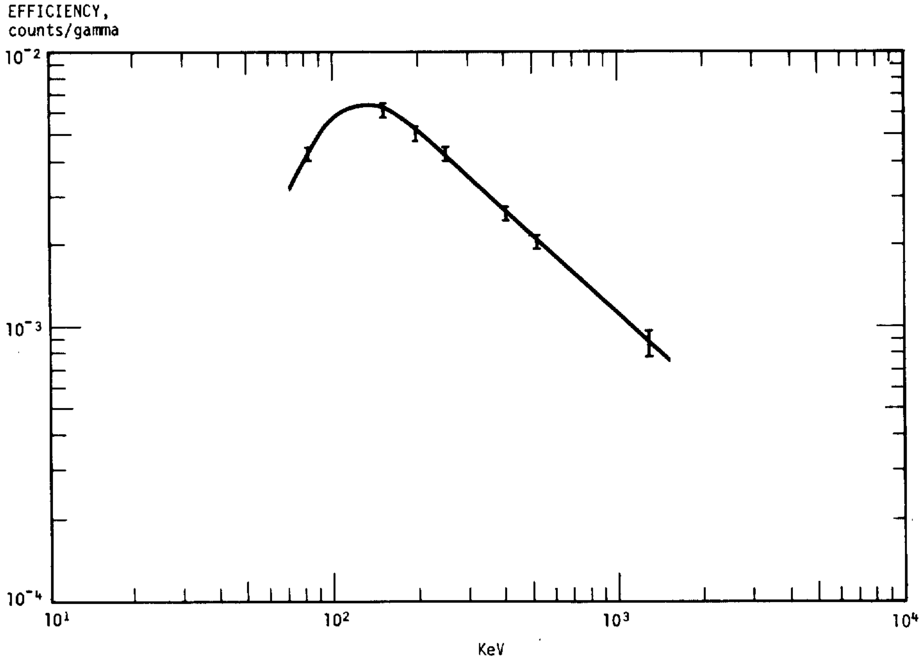

FIGURE 7. TOTAL SYSTEM EFFICIENCIES

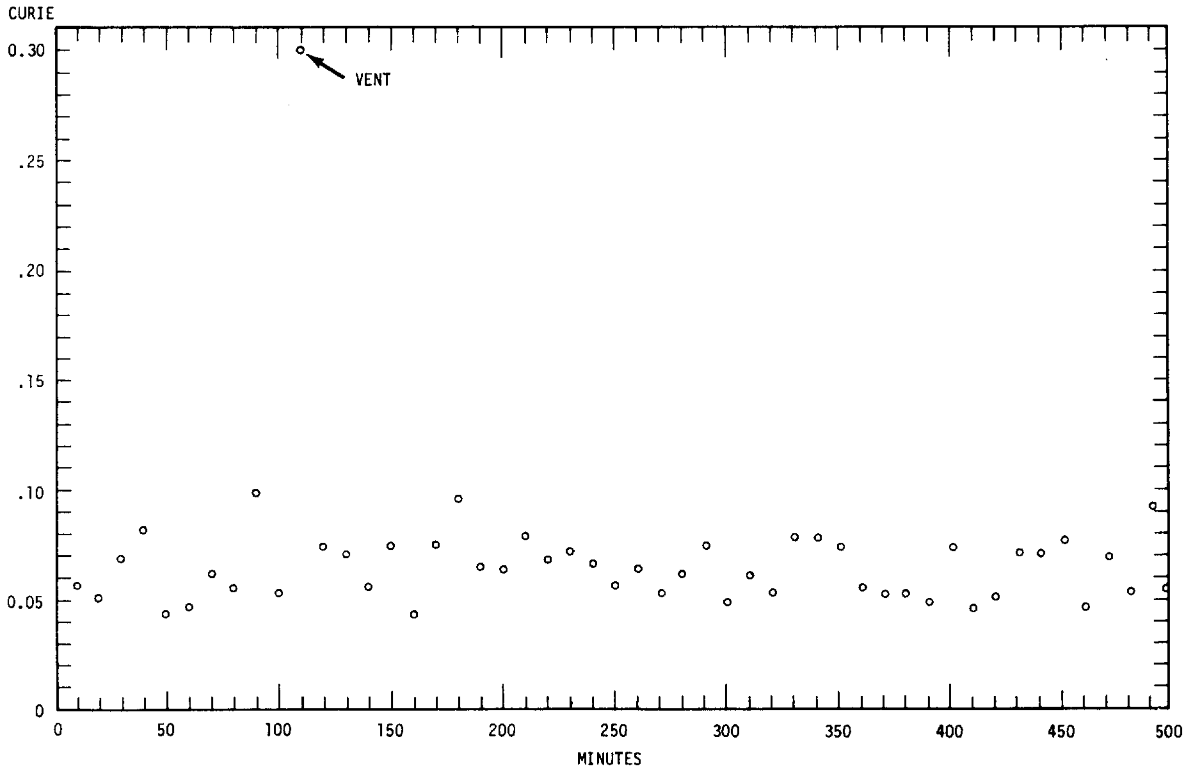

FIGURE 8. TYPICAL RELEASE PERIOD FOR ^{133}Xe

SOURCES USED FOR DETERMINATION OF DETECTOR EFFICIENCY AT 25 CM

ISOTOPE	ENERGY, KeV
^{241}Am	59.6
^{133}Xe	81.0
^{57}Co	121.0
^{125}Sb	176.4
^{228}Th	238.4
^{85}Kr	514.0
^{137}Cs	661.6
^{60}Co	1173
^{22}Na	1275
^{60}Co	1333

TABLE 1

ISOTOPES PRESENT IN SAMPLES TAKEN FROM BLANKET GAS SYSTEM

ISOTOPES	ENERGY, KeV
^{133}Xe	81.0
^{85m}Kr	149.5
^{88}Kr	196.1
^{135}Xe	248.8
^{87}Kr	403.0
^{41}Ar	1293.6

TABLE 2

CURIES DETECTABLE IN 24 HOURS OF COUNTING TIME

ISOTOPE	MINIMUM SENSITIVITY	MAXIMUM SENSITIVITY
^{133}Xe	0.331 ± 56%	44,100 ± 22%
^{85m}Kr	.128 ± 56%	15,900 ± 22%
^{88}Kr	.278 ± 56%	41,900 ± 22%
^{135}Xe	.103 ± 56%	17,900 ± 22%
^{87}Kr	.193 ± 56%	59,000 ± 22%
^{41}Ar	0.121 ± 56%	82,000 ± 22%

TYPICAL RELEASES FOR 24-HOUR PERIOD

ISOTOPE —	^{133}Xe	^{85m}Kr	^{88}Kr	^{135}Xe	^{87}Kr	^{41}Ar
CURIES —	9.14	2.75	4.57	4.57	2.63	110

TABLE 3

CONTINUOUS MONITOR RELEASE DATA VS. GRAB SAMPLE ANALYSES, AVG CURIES/DAY FOR 5-DAY PERIOD

	ANALYSIS	
ISOTOPE	CONTINUOUS MONITOR	GRAB SAMPLING
^{133}Xe	9.69	11.50
^{85m}Kr	2.57	2.89
^{88}Kr	4.33	3.66
^{135}Xe	4.77	4.20
^{87}Kr	2.43	5.15
^{41}Ar	108.0	102.0

TABLE 4

DISCUSSION

DYER: Do you do anything with a spectrometer at all?

M.B. WOOD: No. We are going to go to a computerized analysis of the entire spectrum.

DYER: What type of counted background subtraction do you use?

M.B. WOOD: We set up a spectrum on either side of the peak, and just subtract that.

CANTELOW: How do you avoid the problem of drift in the amplifier and detector?

M.B. WOOD: The amplifier is kept at a constant temperature by an air conditioning system. We haven't seen any appreciable amount of drift, but we check the analyzer every few days to make sure specific peak channels are in operating condition. This alerts us if the values start looking bad.

KOVACH: Have you ever found any krypton-85?

M.B. WOOD: Never.

EMISSION MONITORING SYSTEMS FOR PLUTONIUM FACILITIES

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I. Abstract

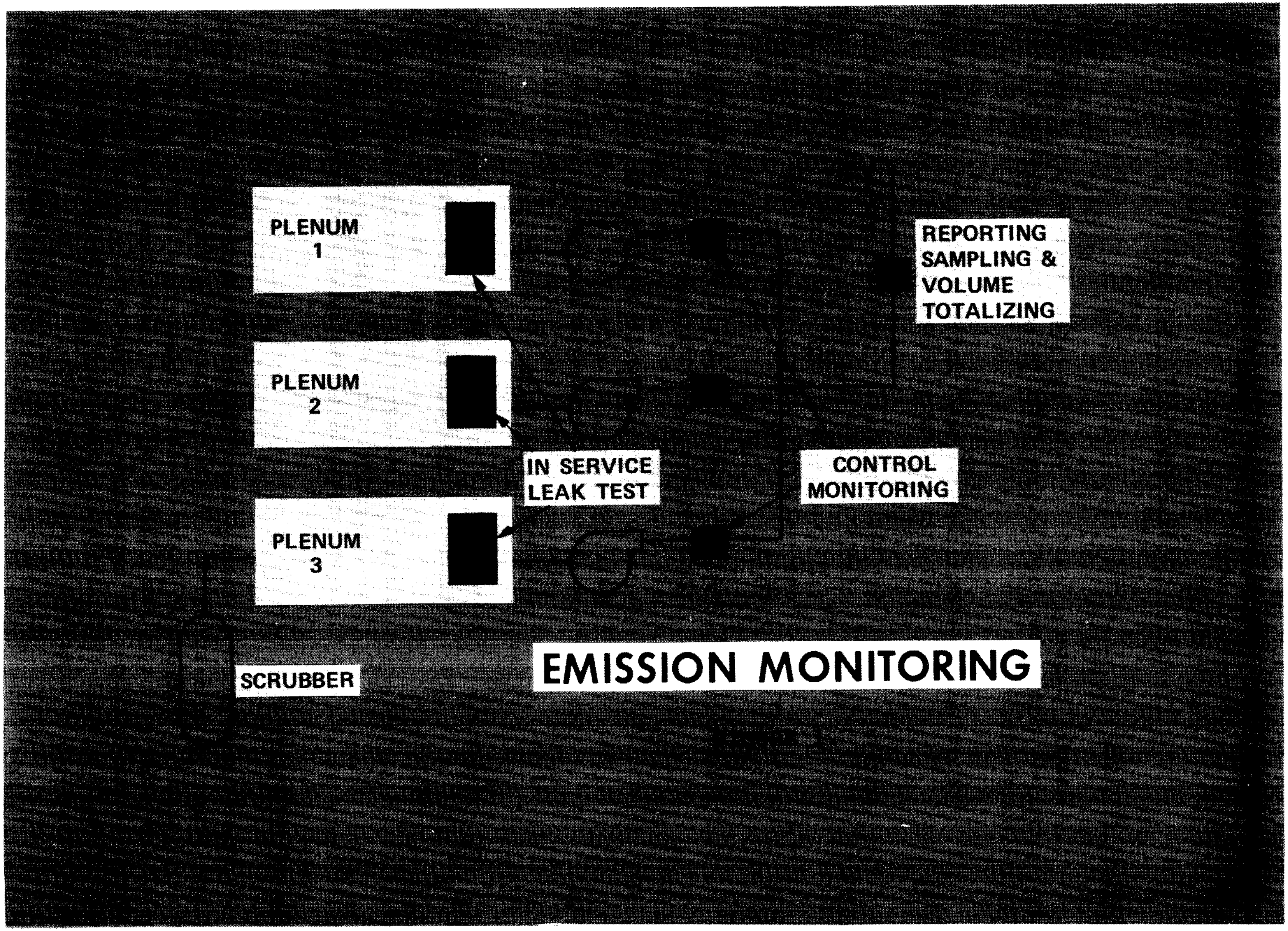
The primary purposes of monitoring gaseous effluent are to insure that environmental discharges conform to specified guidelines and to evaluate the performance of the air cleaning system. This paper describes a system, at a plutonium facility, whose important characteristics are:

1. continuous monitoring with central readout of the amounts of radioactive and chemical pollutants,
2. extensive fixed sample collection, and
3. measurement of the total effluent volume.

Also described is a technique for in-service contamination surveys of filter plenums. The system meets the requirements set forth in the "Minimum Design Requirements for New Plutonium Facilities."

II. Introduction

Effective and accurate measurement of emissions becomes more difficult as air cleaning systems increase in size. Those at Rocky Flats are quite large, a typical system consisting, for example, of three multistage filter plenums and a scrubber -- one plenum for process area room air, one for fume hoods, and one scrubber and plenum for gloveboxes. These plenums, with capacities of several thousand cubic feet per minute (cfm) each, service various atmospheres from room air to inert gases. The exhaust from these plenums is manifolded into a common exhaust point. Radiometric surveys of the plenums are taken routinely to assure filter integrity. Air monitoring is performed after final filtration from each individual plenum. Finally, samples for formal reporting purposes are collected from each exhaust point. (Fig. 1)



PLENUM
1

PLENUM
2

PLENUM
3

IN SERVICE
LEAK TEST

REPORTING
SAMPLING &
VOLUME
TOTALIZING

CONTROL
MONITORING

SCRUBBER

EMISSION MONITORING

This paper deals primarily with monitoring for particulate radioactive emissions as the standards for chemical pollutants are not completely defined at this time. Equipment for radiation monitoring is described below.

III. Fixed Sample Collection

Sample Probes.

The sample probes, used to withdraw representative portions of the airstream from the duct, were fabricated according to the following guidelines:

1. Material - Stainless steel tubing was utilized because of strength and corrosion resistance.
2. Sizing - A 3/8" minimum outside diameter (OD) was specified for rigidity. The inside diameter (ID) was sized isokinetically, by varying the wall thickness, to a 2 cfm ± 20% sample rate.
3. Fabrication - The tip was tapered (1/2") from the OD to the ID to minimize airstream disturbance. A minimum radius of six inches was required on the bend prior to the filter holder. Varying lengths were used inside the duct in an attempt to sample a representative cross section. The length of the probe outside the duct was held to a minimum.
4. Mounting - The probe is attached to a five-inch square flange. The flange is gasketed and mounted to the duct with studs and wing nuts. This facilitates removal of the assembly for probe inspection and cleaning. An offset stud prevents misalignment of the probe.

Filter Holders.

The filter holder in use is a Rocky Flats design. The cone shape prior to and after the filter allows for uniform particle distribution. The inlet side of the holder is attached directly to the sample probe. The outlet side is connected by flexible tubing to an orifice. A manometer is connected across the orifice providing a visual indication of the sample flow rate which may be adjusted by a needle valve. (Fig. 2).

Central Vacuum.

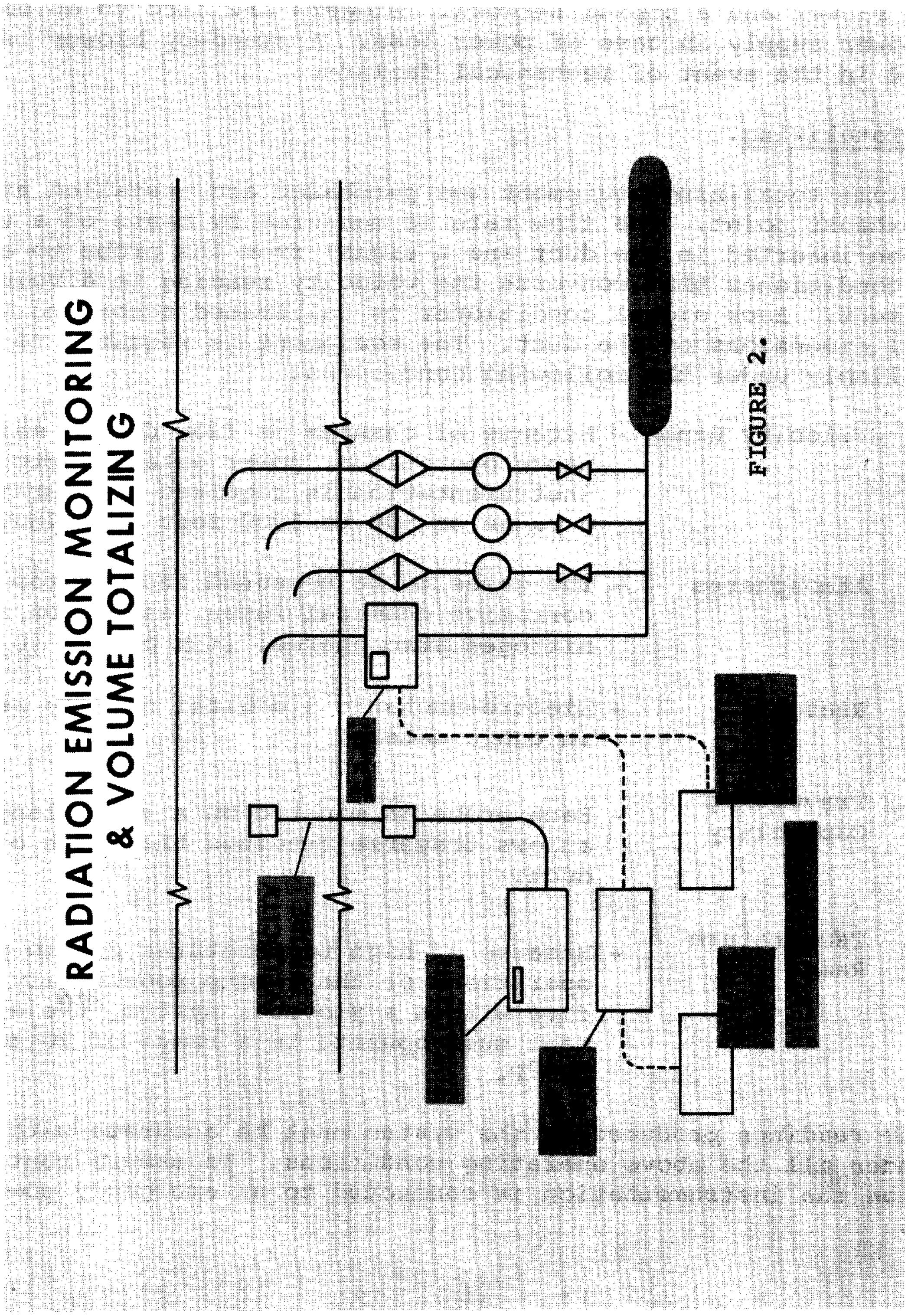
Vacuum for all air sampling in a building is supplied by a central blower and a piping network. Blowers are tied to an emergency power supply in case of power loss. A stand-by blower is utilized in the event of mechanical failure.

Volume Totalizing.

Volume totalizing equipment was purchased and installed at every major exhaust point. The flow rate is measured by means of a velocity probe inserted in the duct and a signal from the probe to a signal conditioner that converts the velocity reading to a volume measurement. Each signal conditioner is calibrated according to the physical dimensions of the duct. The equipment is required to function reliably under the following conditions.

1. Velocity Range - Because of changes in flow due to maintenance operations, power outages, etc., the instrumentation is required to operate in a range of 500 to 5000 feet per minute.
2. Atmospheres - The gases to be measured include room air, corrosive chemical fumes, and argon and nitrogen atmospheres. ($<$5% oxygen [O_2]).
3. Readout - Electro-mechanical digital display reading in cubic meters.
4. Traversing Capability - Each probe is mounted on a slip flange that allows traverse readings the width of the duct.
5. Temperature Range - Because of high temperatures due to process operations or the remote possibility of a fire within a glovebox system, the equipment must operate in a range of 70 to 200°F.

The readings produced by the system must be accurate within ± 5% under all the above operating conditions. To assure continuous operation the instrumentation is connected to an emergency power supply.



RADIATION EMISSION MONITORING & VOLUME TOTALIZING

FIGURE 2.

IV. Sample Processing

Sample Counting.

The samples are collected periodically and transported to a counting laboratory. They are first analyzed for total long-lived alpha by direct counting of the filter media. Subsequent specific plutonium (Pu) analysis is made by chemical isolation of the plutonium, electroplating the solution on a planchet, and alpha pulse height analysis.

Data Evaluation.

The result of the sample counting gives the average concentration of radioactive discharges for that particular sampling period. These data times the total effluent volume, as accumulated by the volume totalizing equipment, give total emissions for the sampling period.

V. Continuous Air Monitoring

To provide continuous monitoring of the exhaust gases, the following equipment was purchased with the specifications as listed below:

1. Type - Single channel pulse height analyzer continuous air monitors (SAM's) were selected.
2. Scanning Provision - The SAM's are capable of a digital output to a data acquisition unit.
3. Emergency Power - A rechargeable battery pack within the unit provides power when normal power is lost. These batteries will provide continued operation for approximately 20 hours under normal conditions and approximately 4 hours during alarm situations.

Data Acquisition Units.

A multiplexing scanner and data logger is utilized to present a central readout of the various SAM units in a building. The primary specifications for the acquisition units are:

1. Scan Rate - All SAM's are scanned approximately once every second. A visual display indicates SAM location and number of counts for each unit at a rate of approximately one per second. Any unit may be observed on demand.

2. Printout - A standard line printer is activated whenever a unit registers an alarm condition. The data printed are time, station activity, and alarming station(s) printed in red. Automatic printout occurs every eight hours to provide record data. Printout of data is also available on demand.

The data acquisition unit is tied to emergency power to assure operation during abnormal conditions.

Function of SAM System.

The selective continuous air monitor provides for detection of alpha particles of a specific energy and the rejection of other energies. The sensitivity of this instrument to plutonium is increased because of the reduction of count rate from naturally occurring radon-thoron daughters. The air sample is collected on a 47 millimeter (mm) glass fiber filter media. Readout is provided by a rate meter. An adjustable meter relay actuates a local alarm when a specific amount of activity is collected on the filter media. The alarm is also annunciated to the Utilities control center, guard station outside the building, and the Radiation Monitoring operations office. The scanner in the Radiation Monitoring operations office also provides a permanent record of the date, time, sample number and count rate. These air monitors operate at a 10% geometry (4π) at a normal operating background of one to five counts per minute (cpm). The air monitors sample from the effluent exhaust stream after final filtration.

The alarm point is normally set at 15 cpm. The time necessary to reach this count rate will depend on the concentration of the contaminant and the background count rate. For example, assuming ^{239}Pu to be the isotope of interest and the background is zero, for a concentration of 2×10^{-12} microcuries per cubic centimeter (μCi/cc) and a sample rate of two cfm, the time necessary to accumulate an equivalent of 15 cpm would be 10 hours, at which time the alarm would activate. If the background were five cpm, then an accumulation of an additional 10 cpm would activate the alarm in approximately six and one-half hours. It should be noted that the SAM's primary purpose and function is for detecting gross amounts of radioactive material during accident-type conditions, thereby providing earlier detection and allowing for immediate corrective action.

VI. Radiometric Sampling Techniques

To maintain integrity of filtering systems and to provide for early detection of small leaks, contamination surveys are taken on a periodic schedule determined by the Radiation Monitoring department. Dry swipes are taken in the air locks and plenums on the floor, walls, ceilings, doors, and framework of High Efficiency Particulate Air (HEPA) filters. The dry swipe method consists of smearing over a surface of approximately one square foot with a 55 mm diameter filter paper. The dry swipes are then counted for alpha particle emissions in a scintillation counter. In addition, direct readings are taken on the downstream side of the filter media surface with an alpha survey meter. Any readings significantly above the expected background level are then investigated and followed up by dioctylphthalate aerosol (DOP) testing when necessary. Periodically, air samples are also taken between the filter stages. These samples will indicate gross changes in radioactivity of the between-stage atmospheres. This provides an indication of any change in efficiency of the individual stage.

VII. Emergency Air Sampling

Fixed sampling equipment is stored in a cargo carrier for emergency sample collection. In the event access to a building and/or sample collection point is lost, sampling can be performed outside the structure. Equipment stored in the cargo carrier includes:

1. portable electrical generators,
2. portable air sample pumps,
3. sample probes and filter holders, and
4. associated sampling equipment.

Ports are installed in exhaust stacks outside the structure to accept sample probes should the need arise.

VIII. Chemical Pollution System

The total sampling system for chemical pollutants includes: sampling system conditioner, analyzers, and data acquisition for the emitted pollutants that are pertinent to the problems present in the Denver Air Control area. Data obtained are transmitted to a central data logging system for permanent records and for supplying computer information to print out processed data. The sample conditioner system consists of a Beckman reflux permeation distillation dryer or equivalent. For pollutants, as indicated, the following analyzers are used: (Fig. 3)

Total Hydrocarbons - Beckman Instrument -- Model 400 or equivalent

Flame Ionization

CHEMICAL POLLUTION MONITORING EQUIPMENT

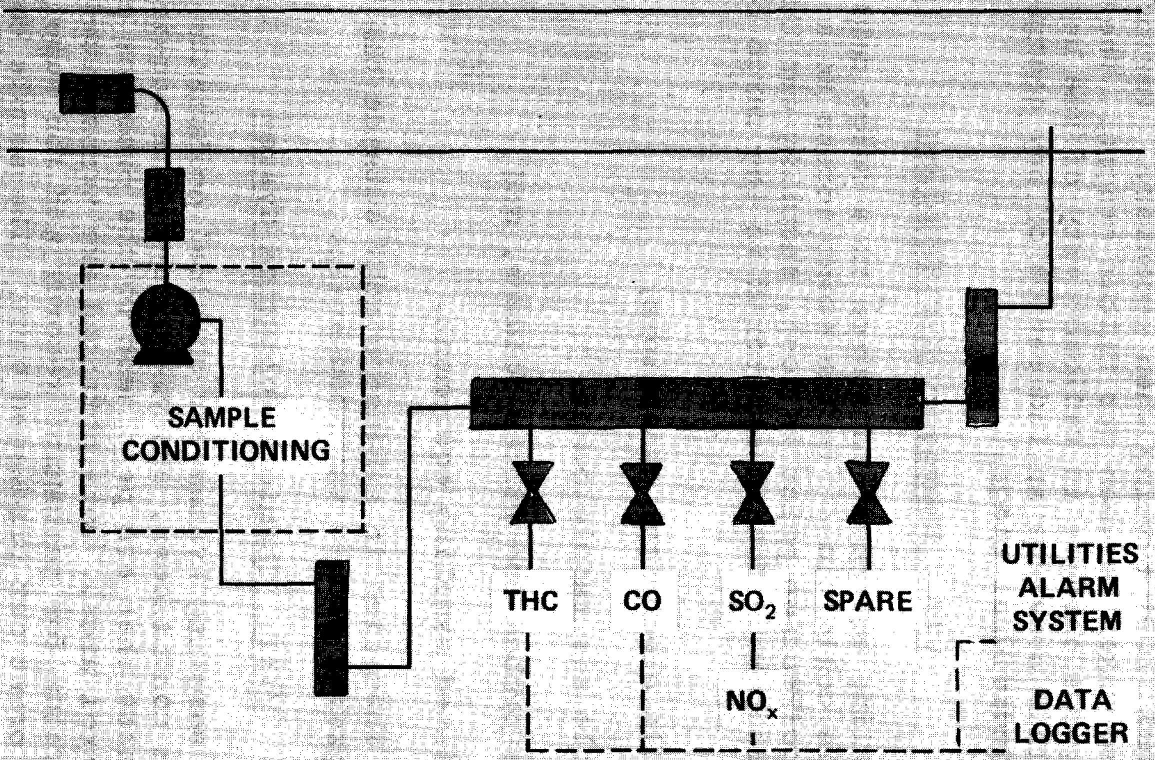


FIGURE 3.

NO_x and SO_2 - Environmetrics Inc. -- Model N5 300 or equivalent
Electro Chemical Sensor type

CO - Beckman - Model 865
Infrared Nondispersive

The data acquisition system takes analog readings from all of the analyzers of this system and logs them on standard Phillips tape cassettes. Equipment consists of:

Martex Instruments Inc. -- Model DAL Data Logger or equivalent
Model DAR Reader or equivalent

VIII. Advantages of the System Described

The radiometric surveys of the filter plenums provide in-service leak testing. Detailed surveys can indicate small filter leaks before detection by air sampling. They can be a valuable test of plenum integrity when used in conjunction with DOP testing.

Data collected at the individual plenum control points provide further evaluation of air cleaning efficiency. The fixed air sample collection gives information regarding low-level concentrations of radiation, while the SAM's provide constant surveillance for excessive releases.

Samples collected at the reporting point supply information on concentrations of radioactive and chemical pollutants. Those concentrations, in conjunction with total effluent volume, provide total release data. The chemical pollution equipment and the SAM's provide continuous monitoring for excessive releases.

IX. Conclusion

Use of the methods and equipment discussed is the most practical approach to monitoring a large ventilation system. Some future changes might be:

1. Larger detectors for the SAM's resulting in more rapid response to lower concentrations of plutonium.
2. Advances in chemical pollution monitoring equipment.
3. Scanning of volume totalizing equipment for central readout of total effluent volumes.

DISCUSSION

SHIPP: You said that you limited the problem of thoron and radon. I was wondering how you got rid of partial absorptions which tend to throw your sub-count down at the level of plutonium?

FREIBERG: That's true. The sub-count in a normal air sample that you take during the day in a working area might have as many as 3 to 5 hundred counts per minute. We have found on the pulse height analyzer that 5 to 15 is a very low level. Thoron and radon are in the 6mev region, and the tail, by the time it gets down there, is very small. What we're doing is setting the pulse height analyzer.

KOVACH: Do you have air straighteners in the sampling stream ahead of your detector to be sure you are relating to single point samples from the duct?

FREIBERG: That's correct, we have straighteners where we don't have quite the duct length we would like. Also, we don't have just one sample location.

DEMPSEY: I keep hearing that we can't measure the efficiency of more than one HEPA filter in series. It seems to me that what you're doing here is very close to getting a real number for that situation by measuring the plutonium. If you quantitate what you are doing, do you think that it would be possible to get a real reading on these efficiencies?

FREIBERG: We have done some of this periodically on routine samples between each filter station, mainly to see if there is any change in the degree of penetration in first bank of filters. If we notice a large difference, we know that we have a leak. We find from experience, that if you continuously monitor the surface of these filters on the cold side, very small quantities of plutonium will show up. You will detect leaks more readily than with a DOP test. There will be about a 6 million count per minute of plutonium before you notice a leak with a DOP test. We can pick it up in very short order. We find we catch most of our leaks in this way. When we do find a leak, we go into DOP testing.

THE EVALUATION OF FIELD TEST EQUIPMENT FOR HALIDE AND DOP TESTING

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Abstract

The Nucon Testing Services Department, field testing at power reactor sites, has performed tests using R-11, R-12, and R-112 in conjunction with gas chromatographs and direct reading halide detectors. The field operational experience with these detector systems, thus sensitivity, precision, and manner of field calibration,are presented.

Laboratory experiments regarding ^{3}H tagged methyl iodide for in place leak testing of adsorber systems indicate a low hazard,high reliability process for leak testing in facilities where atmospheric cross contamination occurs.

I Introduction

The Nucon Field Testing Services Department, field testing at power reactor sites, has performed tests using various types of detectors and test agents. Among the different types of test agents are halides R-11, R-12, R-112, and DOP. The different detector systems used include chromatographs, ultra-violet and open-diode halide detectors and forward light scattering chambers for field testing.

II State of Art Review

Common refrigerants have been used as test agents for many years. Much of the knowledge of these agents came from work performed at the Savannah River Laboratory. R-12 was the first to be tried. Test results showed that R-12 worked well on new carbon. Unfortunately, the tests also showed that for a moisture content of greater than five percent, the test agent would begin to "bleed through" the filter under test during the time it was being checked for mechanical leaks. Thus an accurate measure of the mechanical leak was difficult to ascertain. These tests were run using a gas chromatograph which has a sensitivity to R-12 of 0.03 vppm. With this sensitivity, it was necessary to use an upstream concentration of approximately 500 vppm. This figure was based on maintaining an upstream/downstream ratio of at least 10,000 to 1 in order to test to 99.99% efficiency. Generation of the R-12 vapor caused no problem. It's boiling point of -29.8°C meant that it was already a gas at room temperature and could simply be metered out in the proper quantity with no external heat source.

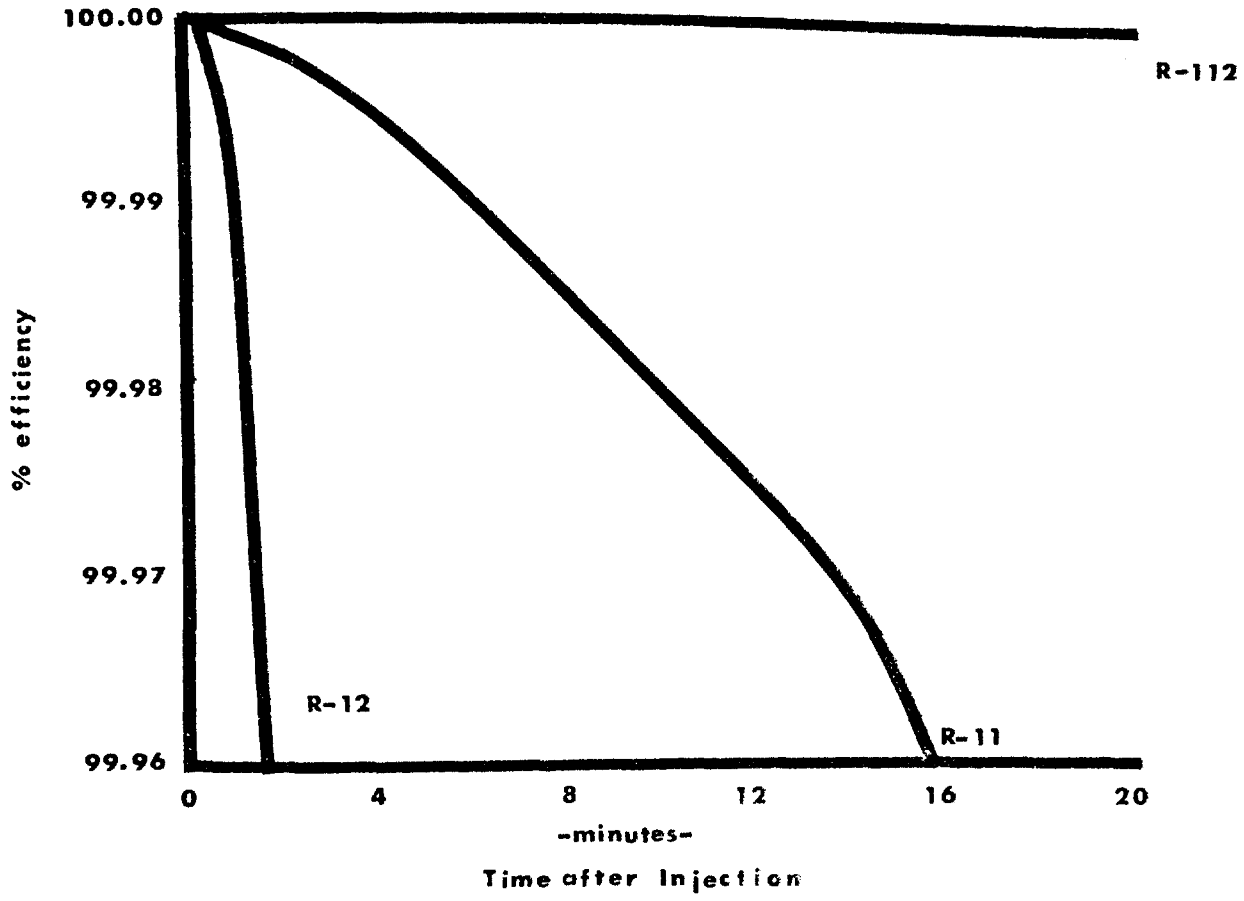
Because the gas chromatograph had a low sensitivity to R-12, it was decided to try R-11. It was found that the chromatograph had a sensitivity of 0.0003 vppm to R-11. This increase in sensitivity of two orders of magnitude meant that the upstream concentration could be decreased by two orders of magnitude and still maintain the necessary 10,000 to 1 ratio. Again a series of tests were performed on scale adsorber cells. The results of these test showed that R-11 was useful for leak-checking new 1.0 inch deep carbon cells with up to 12.5%

adsorbed H_2O. Higher moisture contents caused a more rapid bleed-through of the R-11 due to the high moisture content. Thus, depending on the time needed to adequately test the adsorber, the results may have been inaccurate and not a true measure of mechanical leak. Generation of the R-11 vapor was a little more difficult than the R-12 vapor. Its boiling point of 23.8°C (M.P. -111°C) meant that it was a liquid at room temperatures. Thus, in order to generate the R-11 vapor it would have to be heated above its boiling point. This required a more complex means of control on the generator since more variables had to be considered, i.e. temperature, pressure, and flow.

A third refrigerant, R-112, was then tried. The chromatograph sensitivity was found to be 0.001 vppm. This meant minimum upstream concentration of 10 vppm had to be maintained. The conclusions drawn from the subsequent small scale tests indicated a high degree of efficiency on new carbon. The bleed-through phenomenon did not become significant until the moisture content reached a level of approximately 30% adsorbed H_2O. This made R-112 appear to be ideal for leak testing in the field because the typical average carbons contained about 27% wt. moisture. Generation of the R-112 vapor was a major drawback. Its high melting point of 26°C meant that it was a solid at room temperature. Thus it would have to be raised above not only its melting point, but also its boiling point of 92.8°C. This meant that much more heat would have to be added in order to generate the R-112 vapor.(1) (2)

Another major drawback to R-112 is the fact that it is very difficult to remove from the new carbon. Under normal operating conditions it could take several weeks to significantly reduce the amount of R-112 retained on the carbon. Therefore it is restricted to 1.0 inch deep carbon beds although early tests on 2.0 inch deep beds were also performed using R-112. It should be noted that R-112 is no longer commercially produced in the United States. A comparison of breakthrough times is shown in Figure 1.

The early standard for field testing was the use of the gas chromatograph with an electron capture detector. This device uses a chromatographic column and an electron capture diode to separate and detect the various gases in the sample. This device acquires a sample once every minute from the sample source. The gas is then separated, ionized, and detected with the output of the detector recorded on a strip chart. This sampling time delay is a major drawback to the machine in that a phenomenon could occur during the analysis time and for mechanical leak establishment even 30 sec integration of results can indicate misleading results. Also in order to get an accurate plot back to zero time a minimum of four readings would have to be taken. This could cause a serious problem when using R-11 with a "wet" adsorber. The test agent could start to bleed-through before the second reading could be taken. The use of multiple dilution flowmeters to decrease upstream concentration to measurable levels can also add more error to the results. Thus a greater uncertainty in filter leak rates can exist. To overcome the problem of switching flowmeters for upstream and downstream, two chromatographs could be used. This presents another problem. It has been found that it is very difficult to co-calibrate two gas chromatographs. Besides being sensitive to halides the chromatograph is highly sensitive to O_2 and oxygen containing compounds such as CO_2. When using a chromatograph, one must be able to distinguish between O_2 containing compounds and the test agent used. Thus the accuracy of the chromatograph is entirely dependant on the operators skill at interpreting the peaks on the chromatograph's output.



EFFECT OF REFRIGERANT TYPE ON BREAKTHROUGH FOR 2.0 INCH BC 727 AT 40 FPM CONTAINING 25% WT MOISTURE AT 25°C

FIGURE 1

The standard in field testing of HEPA filters is the forward light scattering chamber. This device works on the principal of the amount of light scattered through a blackend chamber. This light falls on a photomultiplier and the output of this MPT is proportional to the amount of light falling on it. This provides a convenient way of measuring the concentration of the DOP used to leak check the HEPA filters. The most common problem in this device is the regulation of the high voltage needed to drive the MPT. Usually a clipping type regulator is used. It was found though, that because of the harmonics generated by this method of regulation,that several of the electronic components tended to overheat. To cure this problem several capacitors were added to the regulator to clean up the output waveform of current commercial units.

III New Equipment Development

Because of the drawbacks of the equipment previously available, Nucon undertook development of its own field test unit. It is called the F-1000 system, shown in Figure 2. It is composed of an electronics mainframe, remote sensor heads for halide and DOP, and two stand-alone generators, one for halide and one for DOP. The F-1000 Halide system uses an ultra-violet type detector for its upstream detector. Its range is from 20 vppm up to 1000 vppm. The downstream detector uses an open diode as the detecting element. Its range is from 0.02 vppb up to 20 vppm with the use of a 10 to 1 dilution flowmeter for concentrations from 1.0 to 20 vppm. These detectors are mounted in remote sensor heads and connected to their respective mainframe modules by a multiconductor cable. This cable allows the sensors to be located as near to the sampling port as is practical thus minimizing any delay time due to long sample lines. An internal pump continuously draws the sample through the sensor cell thereby giving a continuous readout of the duct concentration. The main advantage to this system is the direct readout in actual R-11 (or other calibrated) concentration. It contains two digital readouts which give the measured concentration direct in vppm. There is no need to consult a calibration curve to determine duct concentration.

The DOP section of the F-1000 uses the same basic forward light scattering chamber with a few refinements of commercially available units. The major refinement over other units is the addition of a custom designed regulator for the MPT power supply. This power supply regulates the high voltage within 0.1% at 900 volts DC over line voltages from 100 to 125 volts AC. This regulation adds greatly to the overall stability of the DOP sensor.

Another advantage is the fact that the DOP system electronics can be housed with the Halide electronics making it a complete detector system. This has the advantage of using the digital readout used on the Halide with the DOP. Thus the readout is directly in percent penetration.

The two generators are able to operate independently of the detector system. An optional interconnecting cable allows the generators to be keyed from the mainframe console. These generators are rated for use on systems up to 40,000 CFM capacity. The other commercially available generators have not been found to generate sufficient DOP at the 100 μg/liter rate above 20,000 CFM flows.

IV Calibration

Field calibration of the halide detectors can be accomplished in three ways. First, the units may be calibrated before they are shipped to the test site.

NUCON F-1000 HALIDE AND DOP FIELD TEST SYSTEM

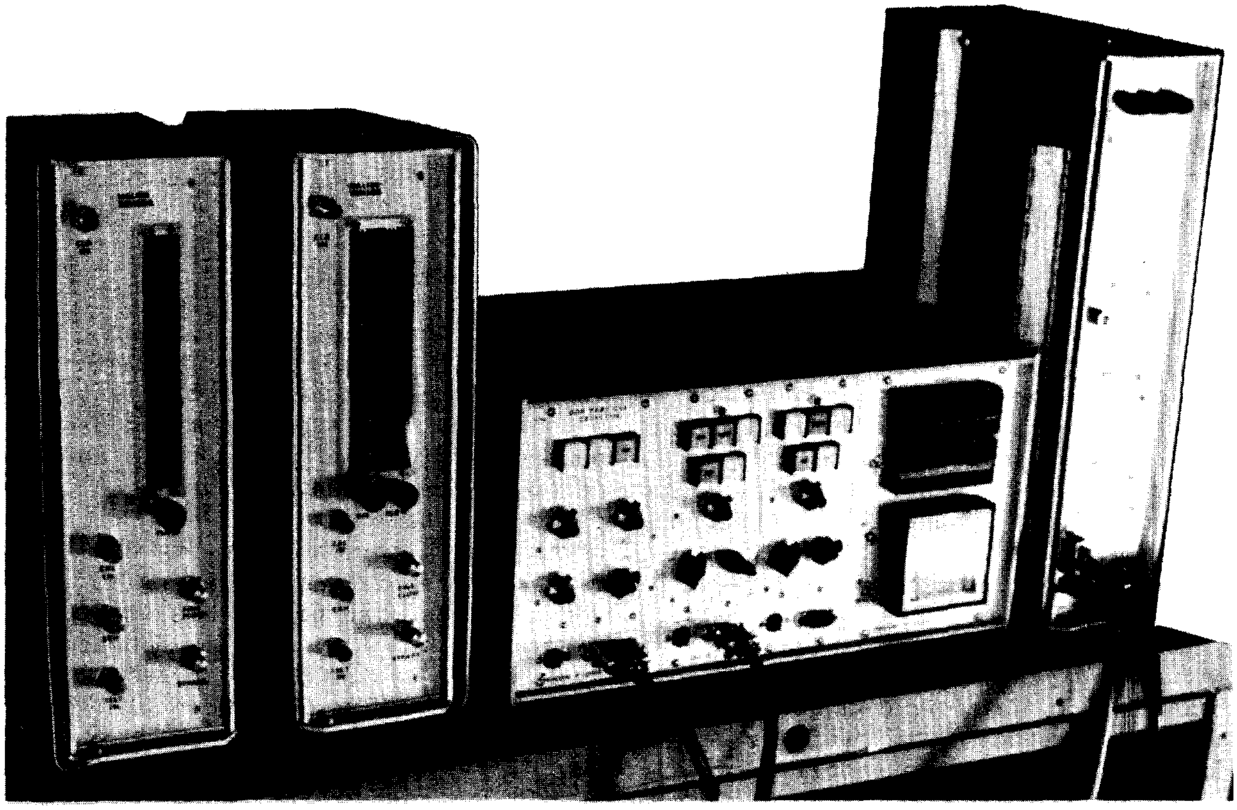


FIGURE 2

Here there is the possibility that the units will be knocked off calibration by transportation. The second method is to send a tank of standard calibration gas to the testing site. In this case there could be a problem if the tank is shipped by air. Airlines are restricting what type of items are allowed to be handled as baggage. The equipment can be sent as baggage but the tank of calibration gas might have to be sent by some other means . This would increase the chance of the calibration gas getting lost or delayed in shipment in which case time would be lost waiting for it. The third, and most practical, is the use of permeation tubes for field calibration. These devices use a saturated bed which releases its trapped compound at a controlled rate, dependant of course on temperature and air flow through the tube. These devices are small and can be packed along with the equipment for field calibration.(3)

The DOP detector requires no frequent calibration to tracable standards as the Halide detectors do. Since it is a relative reading rather than a quantitative reading instrument. For relative calibration, the instrument is set to read 100% using the upstream air. Then it is moved to the downstream sample port and the relative amount of penetration is measured. However, periodic calibration using a small wind tunnel with a 100 μg/liter DOP concentration is desirable.

V Use of Radioactive Compounds for Leak Testing

Recent experiments performed at Nucon explored the possibility of using some compound other than R-11 or R-12 for retesting filter systems. The main problem with retesting filters is the fact that they cannot be effectively tested due to high halide background concentration until the test agent from the previous test is blown and/or significantly reduced. This time period could be anywhere from 24 hours to a full week depending on conditions and test agent. Laboratory tests show that it would be possible to retest a filter using tritium tagged methyl iodide. In these tests an ion chamber with a sensitivity of 1 $\mu Ci/m^3$ was used as the downstream detector. Based on this sensitivity an upstream concentration of 10,000 $\mu Ci/m^3$ had to be used. The test setup appears in Figure 3. It was found that with new carbon no detectable amount of tritium appeared downstream of the test cartridge. To prove that the system could indeed detect a leak, a test was prepared with an intentional leak path as in Figure 4. The leak path was made by inserting a very small plastic tube down the center of an environmental iodine sampling cartridge as shown in Figure 4. Again an upstream concentration of 10,000 $\mu Ci/m^3$ was injected ahead of the filter;this time a peak of 14 $\mu Ci/m^3$ was detected after the filter. This peak represents a 0.14% leak through the filter which agreed with theoretical calculations.

Another series of test were run using full size filters. First the filters were checked using the CH_2TI. Then the same filters were checked using R-11. All filters tested were found to be 99.99% efficient using both test agents. Thus it seems possible to use the tritium tagged methyl iodide as a test agent for field testing. Also since the ion chamber is insensitive to halide gases, it would be possible to use this method for retesting adsorbers without any interference from the previous testing agent.

VI Typical Field Test Problems

One of the most unusual and difficult filter systems which Nucon has tested is the Spent Fuel Pit exhaust system at one of the nuclear power plants. This

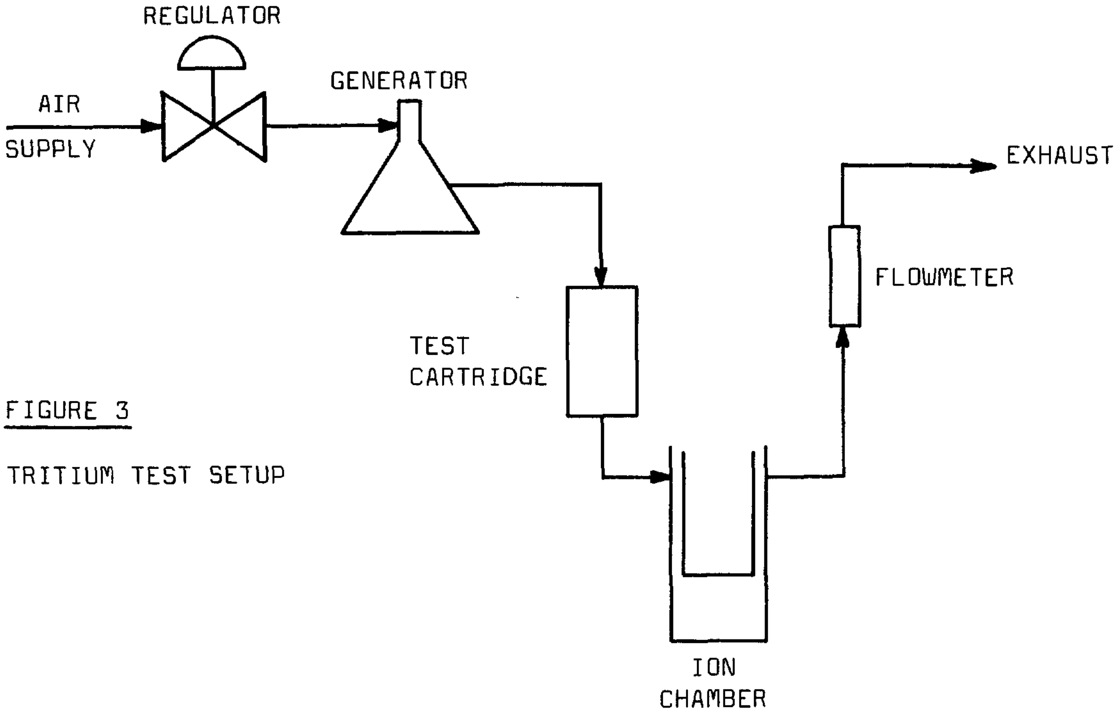


FIGURE 3

TRITIUM TEST SETUP

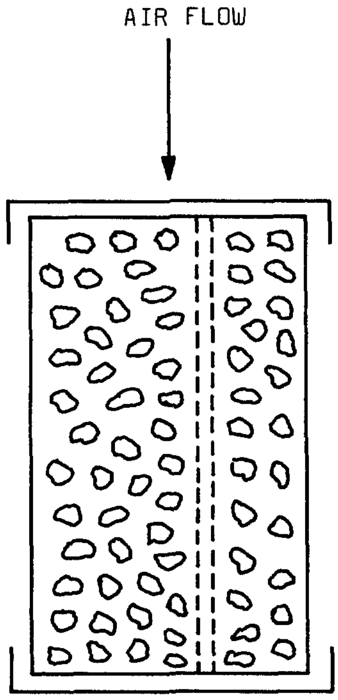


FIGURE 4

TEST CARTRIDGE WITH LEAK PATH

filter system was not installed when the plant was originally constructed. It was installed after start-up in order to conform to AEC regulations. The system consists of thirty-three oversized flatbed filters mounted in groups of three and forming one wall overlooking the SFP. The outlet of these filters formed a common plenum. The ideal way to test these filters would be to blank off the banks that were not being tested and inject into the one that was being tested. Because of the airflow capacity, this could not be done. Instead a special shroud had to be fabricated to cover one bank. This shroud assured that all the test gas was going into the filters being tested. By moving the shroud to each bank individually, the entire filter system was successfully tested.

In calculating the final downstream concentration, a dilution factor of 11 to 1 had to be used. The factor is derived from the fact while one bank is being tested, the other ten were pulling room air into the common plenum. One problem encountered in obtaining the downstream sample was the inaccessibility of the sample port. Because of the design of the system it was necessary to crawl along the top of the filter plenum for about thirty feet and then stand up among the outlet ducts to reach the common duct. The lack of lighting in the area added to the problem, although it was only necessary to reach that point to install the sample hose which was left installed for future testing.

References

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2. Muhlbaier, D.R. "Nondestructive Test of Carbon Beds for Reactor Containment Application Progress Report: July 1964 - September 1964." E.I. du Pont de Nemours & Co., Savannah River Laboratory, Aiken, S.C. USAEC Report DP-950 (1965)

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CHAIRMAN'S SUMMATION:

I would like once again to thank our four speakers, Mr. Wilhelm, Mr. Wood, Mr. Freiberg, and Mr. Schreiber for their presentations. I think they were all well prepared and excellently delivered.

When I opened this session, I indicated a desirability to have the monitoring system work efficiently and reliably under normal conditions; operation under normal conditions of monitoring is essential. As Mr. Freiberg pointed out, operation under off-normal conditions is absolutely necessary. Our monitoring systems must be developed to operate under all conceivable off-normal conditions. I would conclude this session by saying, if we are going to err, let's be certain we learn from our errors. We have the information to correct our mistakes.