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Operational Experience with Two Tritium Effluent Monitoring Systems

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OPERATIONAL EXPERIENCE WITH TWO TRITIUM EFFLUENT MONITORING SYSTEMS

by

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ABSTRACT

The Los Alamos National Laboratory has designed, built, and operated two new tritium stack monitoring systems: the wide-range tritium effluent monitor, with a useful range of a few microcuries per cubic meter to $10^8 \ \mu \text{Ci/m}^3$, and an improved Kanne chamber and new electrometer, called a Model 39 Electrometer-Chargemeter. Both tritium chambers reduce sensitivity to tritium contamination, assure a fast response, and convert easily to microcuries by an integrating chargemeter with digital readout. We discuss the calibration of these monitors and point out the advantages of these chambers over conventional systems.

I. INTRODUCTION

Los Alamos National Laboratory handles large amounts of tritium gas annually. The limitations of conventional tritium effluent monitoring systems prompted the development of improved designs, and in this report we discuss two of these improved systems.

II. WIDE-RANGE TRITIUM EFFLUENT MONITOR

The wide-range tritium monitor has been in routine use at the Laboratory's tritium processing facility since January 1980, where large, unexpected gaseous releases of tritium are possible; this instrument is designed for such an application.

A. Wide-Range Detector

A schematic of the ionization chamber is shown in Fig. 1. A series of parallel grids form two active volumes,

labeled low- and high-range chambers. The low-range section has a geometric volume of $\sim 1.0 \ l$; the high-range-section geometric volume is $\sim 0.1 \ l$. The high-range section has reduced grid spacing compared with that of the low-range section, which increases the electric field strength and reduces recombination effects at high concentrations. A deionizer is built into the intake of the chamber to remove ions from the air before they enter the active volume. Also, a particulate filter at the intake removes dust and heavy-ion pairs.

B. Reduced Chamber Effect

In all tritium effluent monitoring systems, internal chamber contamination from tritiated oil or from condensation of tritium water vapor is a problem. The resulting increase in background current significantly increases the lower limit of detectability and makes effluent release calculations difficult and erroneous at low concentrations. The wide-range detector is designed to minimize the contamination effect through internal electrode spacing. At Los Alamos, because of the



Fig. 1. Schematic of a wide-range tritium monitor.

reduced barometric pressure (\sim 75% of sea level), the maximum range of the 18-keV betas is about 10 mm in air. Based on this, the inner wall of the chamber was designed to be 10 mm from the active volume. The chamber walls are at the same voltage as the collection grids; therefore, tritium-contaminated walls will not contribute to the signal.¹ By making the electrodes out of fine grids, the surface area was effectively reduced, thus further reducing the contamination effect.

C. Los Alamos Model 600 Electrometer-Chargemeter

The Model 600 Electrometer-Chargemeter measures current and integrates charge. There are two electrometers: one accepts a current signal from the lowrange chamber and the other accepts a signal from the high-range chamber. Each electrometer feeds its own four-decade logarithmic amplifier. Mixing the outputs of the two logarithmic amplifiers achieved the final eightdecade analog current output. The low-range electrometer spans 1 fA to 10 pA, and through a discriminator circuit, the high-range electrometer takes over at 10 pA to 100 nA. A normalized chamber current is displayed in analog form on the face of Model 600. Figure 2 shows the chassis, the low-range electrometer amplifier, and the wide-range chamber.

Charge is measured by taking the linear outputs from the electrometer amplifiers, feeding these signals into a voltage-to-frequency converter, and counting the pulse train. This procedure makes digital signal integration possible, which is a measure of charge.

D. Operational Experience

Since its installation the wide-range tritium effluent monitor has measured monthly releases ranging from 1.5 $\times 10^7$ to 5.3 $\times 10^9$ µCi. The largest accidental acute release experienced so far has been a few hundred curies, resulting in a maximum concentration of ~350 mCi/m³. The electrometer automatically ranged into the second decade of the high-range electronics. For a backup, this system has a conventional Kanne chamber and sixdecade picoammeter monitoring the effluent, providing an opportunity to compare the results and evaluate the



Fig. 2. Model 600 chassis, electrometer oven, and chamber.

response time of the two systems. In general, the widerange system responded faster, thus tracking the total release more accurately. For the 24-h period that included this release, the total integrated effluent was 12.6% higher than that registered by the conventional monitoring system. Advantages of using the wide-range system when an acute release occurs are (1) results are available almost immediately because of the integrating capability, (2) there is more confidence in the data because response time is fast, and (3) the upper limit of response (100 Ci/m³) assures adequate documentation of unexpectedly large releases.

The length of sampling line was minimized to take advantage of the 1-s current/meter time constant that tracks sudden, high-tritium concentrations. It is advantageous to keep that distance as short as possible.

The chamber was microphonic, but shock mounting the low-range electrometer and the chamber eliminated mechanical noise. The tritium effluent monitoring system has an inherent current that is composed of current produced from gamma interactions with the ion chamber, current resulting from variations in the environmental radon levels, and current produced from chamber contamination (if any). Also, an offset current from the electrometer is necessary; that is, a slight positive bias current of several femtoamperes is needed



Fig. 3. Wide-range chamber response curve.

from the electrometer to keep the analog meter from being driven negative. Typical inherent background currents over a 6-month period in 1982 ranged from 8 to 30 fA, which correspond to a tritium equivalent concentration of 9 to 32 μ Ci/m³ with a mean minimum detectable concentration of 12 μ Ci/m³. Because of the background fluctuation, we took daily fresh air background samples to establish the currents resulting from radon-level fluctuations or contamination.

E. Calibration

The wide-range tritium monitor was first calibrated at the Los Alamos Gamma Calibration Range. We used radiation fields of 1.9×10^{-3} to 1.2×10^3 R/h and recorded the output current, which was automatically normalized to the 1-& chamber. The results of the gamma calibration are shown in Fig. 3. At the range change, marked on the figure, a slight deviation from a linear line occurred. It is believed that a submillivolt offset in the zero of the high-range electronics caused this deviation.^t

The wide-range monitor was then calibrated to tritium gas. See Fig. 4 diagram. Because detectability ranged from a few microcuries per cubic meter to $10^8 \ \mu \text{Ci/m}^3$, we had to use two tritium sources and a calibrated instrument for the tritium calibration/linearity check. For the low end of the range (up to 400 $\mu \text{Ci/m}^3$), a Johnston* calibrator was used as the source. For the higher

*Johnston Laboratories, Inc., 3 Industry Lane, Cockeysville, MD 21030.



Fig. 4. Typical calibration diagram. Johnston Laboratories, Inc., Cockeysville, Maryland, manufactures the Triton 111.

concentrations (2.5 mCi/m³ and higher), a high-concentration (>500· μ Ci/m ℓ) gas sample was introduced into the closed loop system using a hypodermic needle, and the current reading was plotted as a function of the calibrated Johnston 111 meter reading. The calibration/linearity check of the wide-range monitor is performed to 10⁶ μ Ci/m³, and the complete response curve mentioned above is shown in Fig. 3.

III. IMPROVED KANNE TRITIUM EFFLUENT MONITOR

The way in which Kanne chamber currents are measured has changed very little over the past two decades. The current is fed to an electrometer, and its logarithmic output, usually 10^{-13} to 10^{-7} A, drives a six-decade strip-chart recorder. The quantity of tritium passing through the Kanne chamber is calculated by hand-integrating the area under the logarithmic trace, which is time consuming and can introduce inaccuracies from individual interpretations.

Now, by the use of the improved Kanne Chamber and the Los Alamos-built Model 39 Electrometer-

Chargemeter, all the guesswork and hand integrations are a procedure of the past. The improved chamber and associated electronics are now the Laboratory's principal tritium effluent monitoring system.

A. Improved Kanne Chamber

In the late 1970s, Los Alamos redesigned the Kanne chamber to reduce the problem of internal contamination from tritiated oil and tritiated water vapor and to make decontamination easier.² The conventional Kanne chamber, described by Hoy,³ consists of three concentric cylinders with the inner and outer cylinders at or near ground potential, whereas the intermediate cylinder is operated at ~200 V. The region between the outer and intermediate cylinder acts like an ion trap. The inner region is the ion chamber and the inner cylinder is the collecting electrode. The chamber was redesigned to reduce the effective surface area of those cylinders on which contamination would affect the signal from the chamber. Figure 5 is a detailed drawing of the improved



Fig. 5. Detailed drawing of the improved Kanne chamber.

Kanne chamber; a wire cylinder replaces the highvoltage cylinder of the conventional Kanne chamber. A 6.4-mm-diam aluminum rod replaces the conventional 76-mm-diam cylinder electrode. Because of theoretical recombination effects at high concentrations, we are considering a larger (>6.4-mm-diam) collecting electrode with minimal surface area.

B. Reduced Contamination Effect

The redesign of the conventional Kanne chamber, as well as the wide-range chamber redesign, reduced contamination by taking into account the range of tritium betas. To reduce contamination in the Kanne chamber, we kept all surfaces outside the high-voltage cylinder (connecting rods and chamber wall) >1 cm from the wires. Because the outer support rings are grounded, the ionization resulting from their contamination terminates in the ring and does not contribute to the signal. Thsupported end of the collector electrode is shielded (Fig. 5) to prevent increased signal from contamination on the support rings or chamber end. The sensitive area of a conventional Kanne chamber is ~11 000 cm²; however, the sensitive area of the improved Kanne chamber is <266 cm², a 40-fold reduction in sensitive area.¹

The improved design of the Kanne chamber eases the decontamination process; passing a heating current through the high-voltage wires decontaminates them. If necessary, the central collecting electrode can easily be removed for decontamination or replacement.

The improved Kanne chamber does not use an internal deionizer and one must be provided. An external high-efficiency particulate air (HEPA) filter removes dust and oil.

C. Los Alamos Model 39 Electrometer-Chargemeter

The Los Alamos Model 39 Electrometer-Chargemeter and temperature-controlled oven containing the preamplifier are shown in Fig. 6. We may choose any one of three full-scale current ranges with a selector switch; the selected range then spans four decades of electrometer output. The electrometer measures currents as low as 1 fA (10^{-15} A) and integrates these currents. However, the electrometer sensitivity normally is switched one decade lower than 1 fA because of background currents, and for normal operation the most sensitive range becomes 100 pA, full scale.



Fig. 6. Model 39 chassis and preamplifier oven.

D. Operational Experience

Improved Kanne chamber background currents at Los Alamos vary considerably with location and environmental factors. Over an 8-month period in 1982, one system displayed a mean fresh-air background current of 100 fA (1.0×10^{-13} A) with a range of 40 to 180 fA and a mean tritium-equivalent background concentration of 1.8 μ Ci/m³. This variation results from a mild radon contribution, verified by corresponding barometric pressure readings. At the other extreme, a Kanne chamber system in an underground storage vault showed background chamber currents ranging from 150 fA (1.5 \times 10^{-13} A) to 900 fA (9.0 × 10^{-13} A). This finding corresponds to a tritium-equivalent background concentration ranging from 2.7 to 16.4 µCi/m³, respectively, and resulting from large and variable radon contributions. The mean fresh-air background current was 210 fA (2.1 \times 10⁻¹³ A). We found the environmental factors influencing background current inversely proportional to the barometric pressure and attributed this relationship to radon.

E. Calibration

The initial calibration for the improved Kanne chamber system defined the response as $1.8 \times 10^7 \mu \text{Ci/cm}^3$ per ampere compared with $2 \times 10^7 \mu \text{Ci/cm}^3$ per ampere for the conventional chamber.^{1,2} The implied effective volume for the improved chamber is 56.6 ℓ .

IV. FEATURES COMMON TO BOTH SYSTEMS

Both the Model 600 and Model 39 chargemeter systems have a readout that covers a 10-decade digital display, from 10^{-12} to 10^{-2} C/digit full scale. Chassis readout has three decades of digital indicators and exponent multipliers. Charge readout selection is manual or automatic: the three most significant digits with nonzero information are displayed along with the correct exponent multiplier.¹

Both electrometers can suppress unwanted signal currents (for example, from chamber contamination), which reduces the accumulation of charge over long periods of time.

The two systems allow short distances between the ionization chambers and their associated electrometer amplifiers for optimal low-current measurements. The main chassis for both systems may be remote from their points of measurement.

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