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METHODS FOR MEASUREMENT OF ^{222}Rn EMISSIONS
FROM UNDERGROUND URANIUM MINES

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U.S. Environmental Protection Agency
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METHODS FOR MEASUREMENT OF ^{222}Rn EMISSIONS
FROM UNDERGROUND URANIUM MINES

Forward:

The following three methods are intended for use in monitoring the emission rate (activity per unit time) of ^{222}Rn in ventilation air exhausted from active underground uranium mines. The methods include procedures for making 1) concentration measurements, 2) flow-rate measurements, and 3) emission rate calculations. The general format follows the usage found in the Code of Federal Regulations -Title 40, Part 60, Appendix A.

METHOD 1--DETERMINATION OF RADON-222 EMISSION RATES FROM UNDERGROUND
 URANIUM MINES; GRAB SAMPLING METHOD FOR MEASURING RADON-222 CONCENTRATIONS
 IN MINE EXHAUST AIR

Introduction

A method is described that uses scintillation flasks for grab sampling and
measuring the concentration of ^{222}Rn in air. There is a variety of commercial
and shop made flasks and counting equipment that can be used with them, and
any system which uses a scintillation flask which can satisfy the criteria
below may be used as a measurement system. Since this technique is to be used
to evaluate the average ^{222}Rn emission rate from air streams that often are
characterized by significant temporal variations of radon concentrations, it
will be necessary to average samples collected at several times to achieve the
necessary precision and accuracy, as outlined below. It will also be necessary
to measure the flow-rates of exhaust air at approximately the same times that
samples are collected.

1. *Principle and Applicability*

1.1 Principle. Air containing gaseous radon is withdrawn from each mine
vent using either an in-line probe or a vent-outlet probe which uses the velo-
city of the air stream to force a sample through the collection apparatus. A
fraction of the air is filtered and drawn into a scintillation flask. After
allowing the ingrowth of ^{218}Po, ^{214}Bi, ^{214}Pb and ^{214}Po, which are the short
lived daughters of radon, the radon is determined by counting the scintilla-
tions produced in the zinc sulfide coating on the flask walls by alpha-particles
within the flask.

1.2 Applicability. This method applies to radon-222 (radon) emissions. It
does not measure radon-220 (thoron) emissions. Both long-term and short-term
variations in emissions occur at uranium mines. This procedure will yield
estimates of the average emission rate for a calendar quarter when several
samples are collected over an interval of several days within one quarter. To
estimate an annual average, the measurements must be repeated quarterly and
the results for each quarter must be averaged. Since existing uranium mine
vents have several configurations, two sampling techniques and equipment are
specified - one for in-stack sampling where accessible sampling ports are pre-
sent and the other for sampling at the outlet of the vent.

2. *Sensitivity and Range*

2.1 Sensitivity. The sensitivity of this method depends on the size of the
scintillation flask and its background scintillation rate, which normally in-
creases with use because of the accumulation of ^{210}Pb and its daughter ^{210}Po
from the decay of radon in air samples stored in the flasks. When new, back-
ground counting rates from about 0.1 counts per minute (c/m) up to 1 c/m have
been reported for commercially available flasks, depending on make and model.
Typically, the backgrounds of 0.1 c/m are associated with small flasks which
have a capacity of about 100 ml and response factors of about 0.5 c/m per pCi/l.
Backgrounds of 1 c/m are associated with larger flasks with capacities up to
1.4 liters and response factors of about 6 c/m per pCi/l.

The limiting sensitivity is defined here as that concentration which is needed
to achieve the overall precision standard of 22% total error at the 95% confi-
dence level. As will be discussed under Paragraph 4, sensitivity is limited
by the statistical variations inherent in counting radioactive decay events as
well as three other sources of variation. Using reported estimates for the
magnitude of these terms, this method has a minimum sensitivity of about 10
pCi/l for a 30-minute counting time for 100 ml scintillation flasks with back-
grounds of 1 c/m and efficiencies of 0.5 c/m per pCi/l.

2.2 Range. The maximum concentration that can be determined is limited by the maximum counting rate that can be measured without unacceptable losses caused by the pulse pair resolution of the instrument. The pulse pair resolution time is the time required for the instrument to process and record a count before a second count can be detected and processed. As counting rates get progressively higher, the time between individual pulses gets shorter and shorter. Eventually a significant fraction of the pulses are lost because the instrument can't detect them until it has completed the processing of the preceding pulses. Corrections can be made for such losses, but it is best to limit the counting rate so that the maximum correction will be less than 10% because such corrections are normally only approximate.

Typical laboratory scintillation counters have pulse pair resolution times ranging from about 2 microseconds up to 20 microseconds. However, some commercial counting systems may have longer resolving times because they are designed for portability or for environmental samples with low activity. For resolving times less than 20 microseconds, count-rates up to 3×10^5 counts per minute can be tolerated. This is equivalent to a radon concentration of about 1.5×10^5 pCi/l for a 100 ml flask.

3. *Interferences*

3.1 Moisture Entrapment. Air from some uranium mine vents contains significant quantities of entrained water droplets that will be collected by the sample delivery system. Normally, if a small quantity of entrained water is present, it will be stopped by the inlet filter. However, when a visible spray of water is present, the droplets can plug the inlet filter or pass through it into the scintillation flask where the water will affect the detection efficiency of the scintillator for alpha particles. Therefore, filters should be checked to be sure that they have not become plugged after the sample has been collected, and the interior of the sampling flask should also be checked for liquid water. Visible droplets on the window of the flask indicate the presence of excess moisture. In such cases, a water spray removal system must be employed in the sampling apparatus.

3.2 Particulate Matter. Occasionally the quantity of particulate matter present in mine ventilation air may be sufficient to block the inlet filter. For this reason, high efficiency fiberglass filters have been specified for the sample collection system because they plug more slowly than other high efficiency filters. If air is no longer passing through the filter after the scintillation flask has been filled and removed from the sampling line, the filter is plugged. Another sample should be collected when dust levels are lower, or a larger area in-line filter should be used.

3.3 Static Charge. Scintillation flasks often acquire static charges on their surfaces during handling. Such charges can drastically change the counting efficiency of the flask by attracting the charged ions of radon progeny born in the flask to areas of the flask where the detection efficiency for the alpha-particles emitted in their decay is significantly different from normal. This is especially true for scintillation flasks constructed of plastic materials which are inherently non-conductors of electrical charge. Static charge can also occur on the glass viewing window of metallic flasks and on inlet tube connections. For this reason, flasks which have non-conducting parts must be treated to make them at least temporarily conductive. Some manufacturers apply coatings to the interior of such surfaces to make them conductive. All nonconducting exterior surfaces must be sprayed with an antistatic spray before using the flasks. This must be done at least three hours before counting to permit the decay of any daughter ions attracted to charged flask areas prior to spraying. The flasks should not be rubbed or wiped between spraying and counting.

3.4 Daughters of ^{220}Rn. In some cases, there is a possibility that significant quantities of ^{220}Rn will be present in uranium mine air from deposits of thorium at the mine site. Although ^{220}Rn will decay to insignificance before counting because it has a half life of only 56 seconds, its longer lived daughters, ^{212}Pb and ^{208}Tl, will form within the scintillation flask between sample collection and counting. This activity will be an insignificant fraction of the total activity unless the thorium content of the ore exceeds 5% of its uranium content by weight. For mines that have thorium assays which average below 5 weight % of the uranium content, no corrections for ^{220}Rn daughters are necessary. For mines that do not have such assay information, or that have assay information indicating that the thorium content of the ore exceeds 5% of its uranium content, a test must be performed to determine if a correction for the ^{220}Rn daughters will be necessary. The correction must be determined by re-counting one of the scintillation flasks, collected at each vent of the mine, three times at four-hour intervals after the initial count (one of the intervals may be as long as 20 hours if required by working shifts). The resulting count-rates should be plotted on semi-logarithmic graph paper. The final count should be decay corrected to the time of the initial count using the half-life of ^{222}Rn. If ^{220}Rn daughters are present, the count-rate of the initial count will be higher than predicted by the extrapolation of the later counts. The difference between the measured activities can give an indication of the amount of ^{220}Rn daughters present. Because this technique is sensitive to changes in count-rate, it is essential that the operator verify that the flasks being used do not leak significantly during the period of the evaluation and that procedures to minimize static charge have been used.

If the decay study of one set of samples indicates that the excess activity from the first count was less than 10% of the decay corrected activity from the later count, then the normal counting and delay intervals can be used for that vent. If the excess exceeds 10%, the recommended procedure must be modified on all subsequent samplings to permit a 2-3 day holding period before counting. After that time the daughters of ^{220}Rn will have decayed to less than 4% of their initial activity and can be assumed to be an insignificant fraction of the ^{222}Rn activity, except for the highly unlikely event that the thorium content of the ore exceeds its uranium content, (in which case, the delay interval must be extended until counts taken at successive 24 hour intervals decrease with the characteristic half-life of ^{222}Rn).

4. *Precision and Accuracy*

4.1 Accuracy. At the present time (Aug., 1984) there are no recognized primary national or international standard radon sources. Laboratories performing radon calibrations normally use the equipment and method described in the ASTM standards (12.1). In that method, radon ingrown in a sealed flask containing a standard solution of ^{226}Ra, traceable to the National Bureau of Standards (NBS), is purged from the solution and transferred to the scintillation flask using a prescribed transfer apparatus. The radon ingrown in the sealed flask between purges can be computed from the ^{226}Ra concentration and the time interval.

Several governmental laboratories that routinely perform radon measurements using the flask method have been participating in an ongoing intercomparison study to evaluate the accuracy and precision of their measurements (12.2, 12.3). These laboratories send four scintillation flasks to the Environmental Measurements Laboratory (EML) of the U. S. Department of Energy. There, the flasks are tested for leaks and filled from a large chamber containing air mixed with radon. The scintillation flasks are returned to the laboratories where the radon concentrations are measured. Fisenne (12.2, 12.3), reported the results

of the first four intercomparisons. Those results indicated that there were difficulties with leaks and/or the calibrations of the radon flasks. The range of average results for eight laboratories was 22% in the first intercomparison, and 26% in the second. This scatter in calibrations is greater than would be predicted on the basis of the precision of replicate measurements and is also much greater than the errors normally estimated for NBS radium standards. For this reason, the calibration of scintillation flasks by the ASTM method is not recommended at this time.

Currently, two laboratories have tentatively been designated to provide calibrated mixtures of radon in air based on their in-house calibration techniques. EML can provide radon concentrations appropriate for environmental measurements. The Bureau of Mines maintains a laboratory in Denver, Colorado which can provide radon at concentrations typical of those encountered in uranium mine air. For calibrations, scintillation flasks must be shipped to that laboratory where they will be filled with the standardized mixture.

4.2 Precision. Analysis of the errors contributed by various sources inherent in this method indicates that it is practical to estimate the quarterly average radon concentration in air from a uranium mine vent with a total error of 22% at the 95% confidence level. Therefore, an arbitrary limit of 22% has been set for this total error. The total % error, $(1.96\ s_T)$, can be derived from the following expression:

$$s_T = \left(s_c^2 + s_m^2 + s_a^2 + s_s^2\right)^{1/2}$$

where,
    s_T = the total % standard deviation of the method
    s_c = the % standard deviation from counting statistics
    s_m = the inherent % standard deviation from replicate measurements
    s_a = the stated % standard deviation of the calibration standard
and,
    s_s = the standard deviation of the measured radon concentrations resulting from the temporal variations of concentrations present in air from a mine vent.

This formula must be adjusted for the numbers of repeated sampling and replicate flasks that must be collected from a mine vent to achieve the required precision, as will be described later in this section.

The % standard deviation from counting statistics, s_c.
The formula for s_c is as follows:

$$s_c = \frac{100\ (EC + 2B)^{1/2}}{ECt^{1/2}}$$

where,
    E = the counting efficiency, counts/minute per pCi/l,
    C = the measured radon concentration, pCi/l,
    B = the background count-rate, counts per minutes,
and t = the length of sample and background counts, minutes

Example:
For a scintillation flask with a background of 1 c/m, an efficiency of 0.5 c/m per pCi/liter and counting intervals of 30 minutes for both sample and background measurements,

$$s_c = \frac{100\,(0.5C + 2)^{1/2}}{0.5C(30)^{1/2}}$$

$$s_c = \frac{(667C + 2670)^{1/2}}{C}$$

For $C = 10$ pCi/l, $s_c = 9.7\%$.

The inherent error of the flask method, s_m.

This error must be determined by filling several scintillation flasks with a
constant known source of radon and calculating the standard deviation of the
measured radon concentrations using the ordinary statistical formula, and not
counting statistics (9.7). Results reported by Jackson (12.4) indicate that
the maximum inherent error should be about 7% for this method. The measured
value of s_m must not exceed 7%.

Precision of calibrations, s_a.

This term should be obtained from precision
estimates supplied by the calibration laboratory of the Bureau of Mines. If %
standard deviations are not supplied, the reported standard deviations and
average concentrations should be used to compute s_a (using the same formula
used for calculating s_m from the measured standard deviation (9.7).

Variability of source term concentrations. s_s.

The variability of concen-
trations at individual mine vents cannot be determined in advance of measure-
ments. For the purpose of this method the average value of s_s reported by
Jackson (12.4) for short term temporal variations of radon concentration
measured at 150 vents of 27 operating mines will be assumed to apply.
$s_s = 24\%$.

Since the error associated with temporal variations of the source term alone
exceeds the prescribed limit for total error, this method requires that samples
be collected from each vent on more than one occasion. The collection times
should be spaced at intervals. It is also necessary to collect a duplicate
set of flasks at each vent at each sampling time to assure that the scintilla-
tion flasks have been properly filled and have not leaked. Pairs of measure-
ments differing by more than the maximum allowable error (11.1) should be re-
peated at about the same collection time on another day. The replicate measure-
ments will affect s_c, s_m and s_s, but s_a will not be affected. A total of six
acceptable pairs of samples must be collected.

To account for the total number of samples which will be collected, the formula
for the total error must be modified. The standard deviation of the average
of several measurements is equal to the standard deviation of the individual
measurements divided by the square-root of the number of measurements. Since
s_c and s_m are affected by the total number of samples measured, but s_s is only
affected by the number of repeated samplings, the total % error becomes:

$$\text{Total \% Error} = 1.96\,s_T = 1.96\left(\frac{s_c^2}{12} + \frac{s_m^2}{12} + s_a^2 + \frac{s_s^2}{6}\right)^{1/2}$$

where
12 = the total number of samples (6 times 2) and
6 = the number of repeated samplings.

Using the previous estimates of s_c, s_m, s_a, and s_s, the maximum total error is as follows:

$$\text{Total Error} = 1.96 \left(\frac{(9.7)^2}{12} + \frac{(7)^2}{12} + (4)^2 + \frac{(24)^2}{6} \right)^{1/2}$$

$$= 22\%$$

5. *Apparatus*

5.1 Sampling Apparatus (See Figures 1 through 4)

5.1.1 In-Stack Grab Sampling Probe. This unit must comply with specifications given in 10CFR60 Appendix A, Method 3, (2.1.1 and 2.1.2). (See Figure 1).

5.1.2 Vent-Outlet Sampling Probe. The probe must be made of metal or plastic components joined with flexible tubing. Joints must be clamped. It must incorporate a funnel at the inlet to collect sufficient air to maintain a positive air flow. The funnel must have a diameter of at least 10 cm at the larger end and no less than 1 cm at the smaller end. The funnel should be connected to a stainless steel tube of 1.3 cm (1/2 inch) diameter and should be formed with a curve at one end with a radius of approximately 23 cm (9 inches) so that the funnel can be inserted into the outlet of a vent. The overall length should be about 1.8 meters (6 feet) and the short end should be about 38 cm (15 inches) from the top (see Figure 2).

5.1.3 Water Separator. When water is entrained in the ventilation air, a separator for removing the water must be attached to the sampling probe at the bottom of the long end. Flexible tubing should be used for attachment. Joints should be leak tight. Water separators may be constructed of metal, glass, or plastic. However, glass impingement separators are not recommended for field work unless properly housed, because of the possibility of breakage. Water separators can be obtained from commercial sources or can be shop-made.

5.1.4 In-line Filter. The in-line filter apparatus must use high efficiency fiberglass filters with an active filter area of at least 7.5 cm^2. Filters must be at least 99.9% efficient for standard 0.3 μm DOP particles in air. Filter papers must pass at least 20 liters/minute at 2.75 psi head pressure. Filter housings must be leak tight and suitable for fiberglass filters. To prevent tearing when using units with replaceable filter elements, the compressive connections with the edge of the filter paper must not rotate as the assembly is tightened. Filter housings which prevent the filter paper from tearing with flow in either direction are recommended but not essential. Plastic or metal tubing connectors and adaptors with leak tight ribbed or serrated tapers may be used. At least two filter units will be needed. Ready-to-use one-piece filtration devices with suitable characteristics may be used if integrity of filter and seals are verified.

5.1.5 Flowmeter. The flowmeter must be a rotameter that is direct reading for air and has a minimum scale reading 1 liter/minute or less and a maximum reading between 9 and 15 liters/minute. The scale accuracy must be certified by the manufacturer or a test laboratory to ± 10% or less of full scale.

5.2 Scintillation Flasks. At least four scintillation flasks of the same type must be available for measurements. Scintillation flasks may be constructed of metal, glass, or plastic. Flasks must be equipped with a flat transparent viewing window. Viewing windows must not be larger in diameter than the diameter of the face of the photomultiplier tube used in the counting system. Flasks must be coated with silver-activated zinc sulfide phosphor on all interior surfaces except the viewing window. They must be equipped with

PROBE

FLEXIBLE TUBING
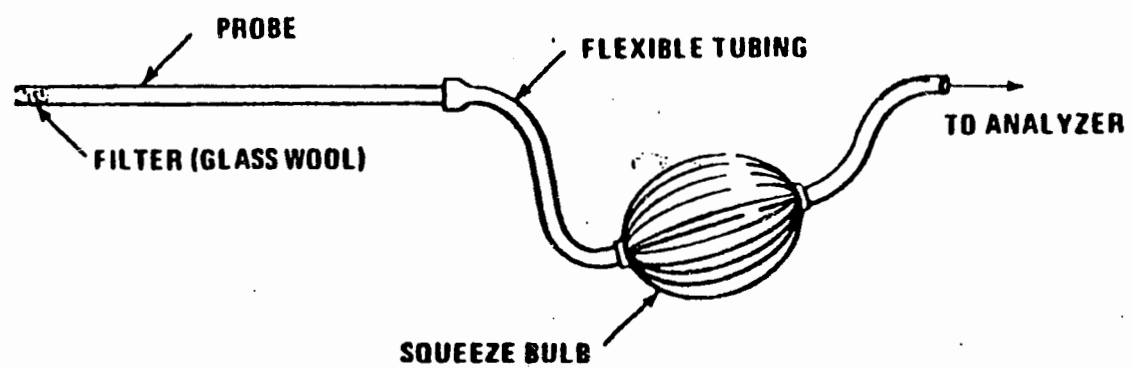
FILTER (GLASS WOOL)

TO ANALYZER

SQUEEZE BULB

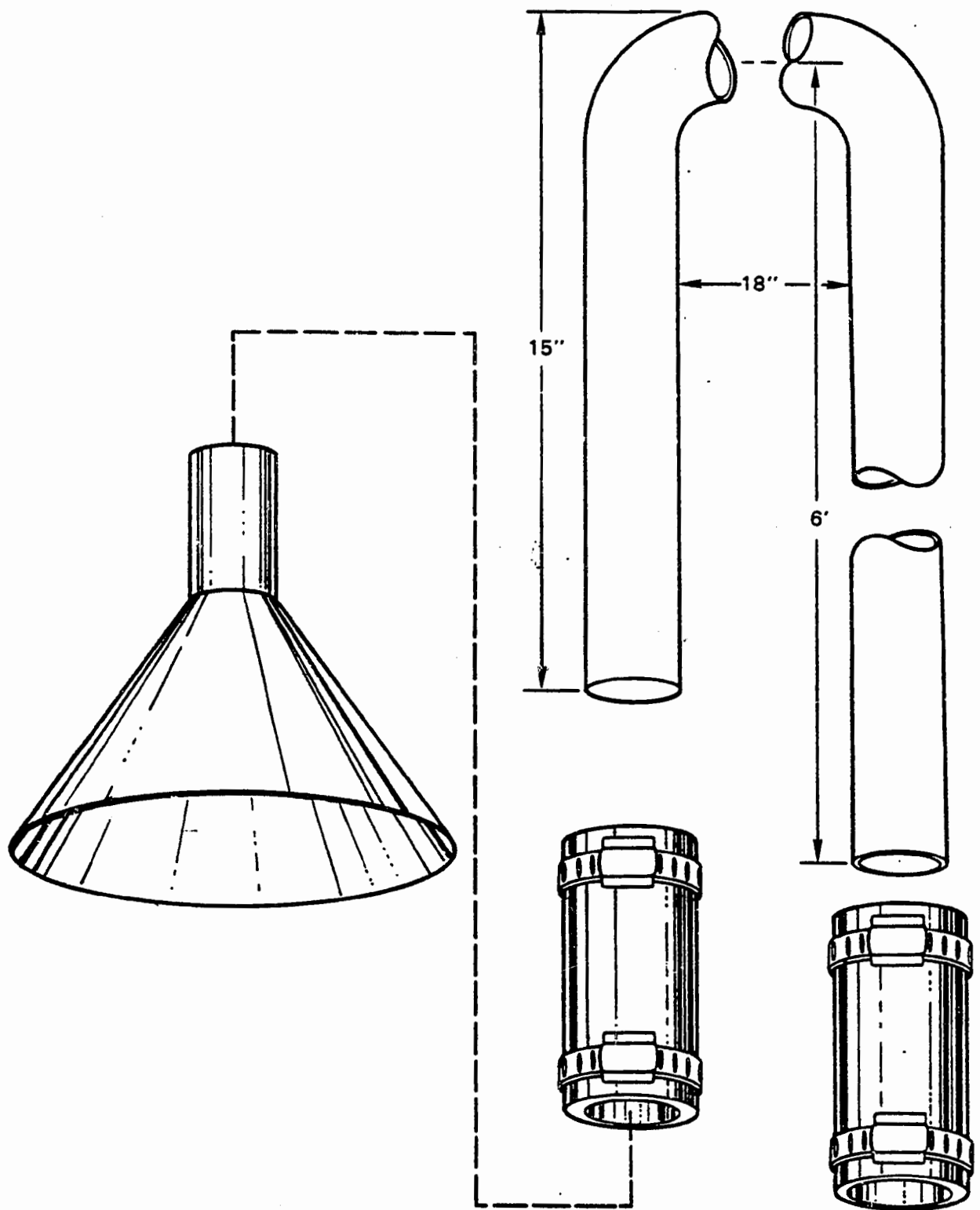FIGURE 1. In-stack grab sampling probe

FIGURE 2. Vent-outlet sampling probe
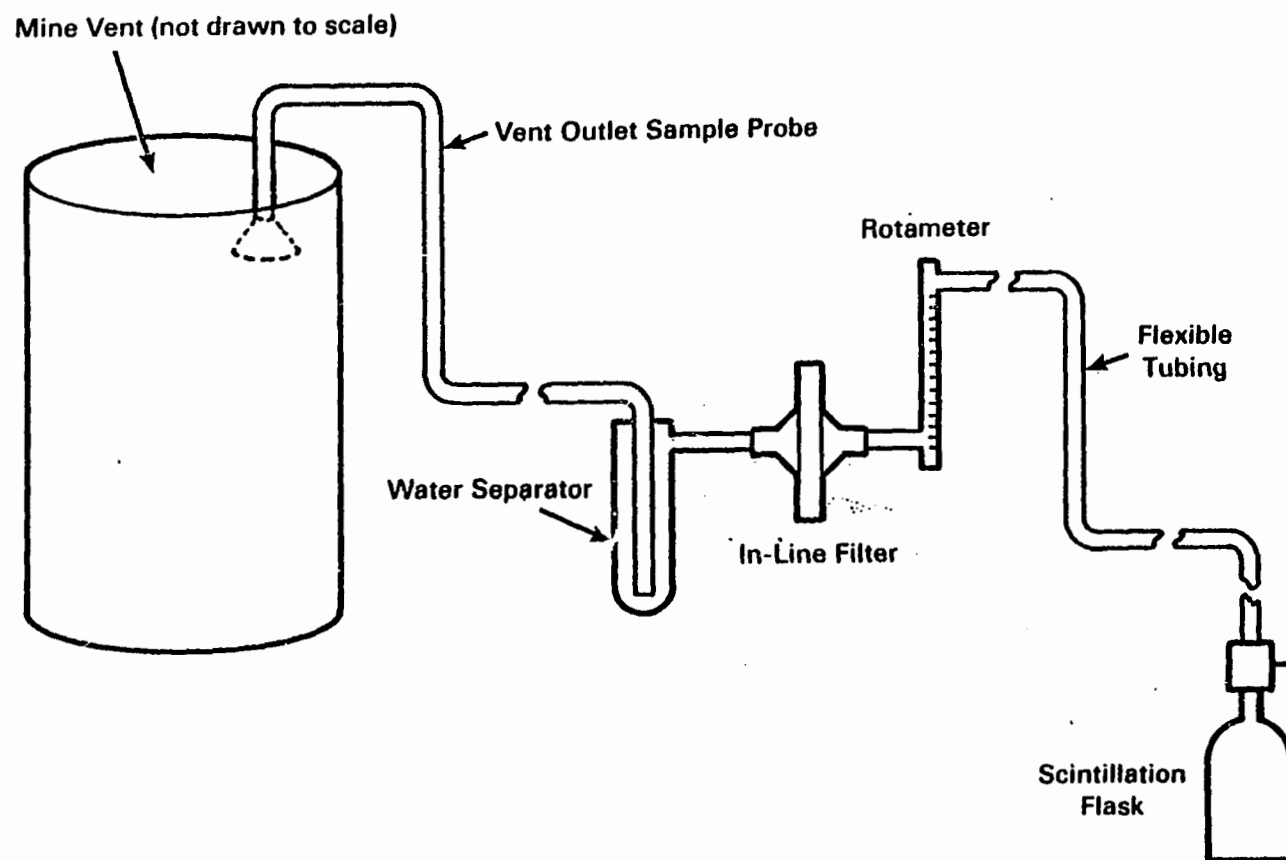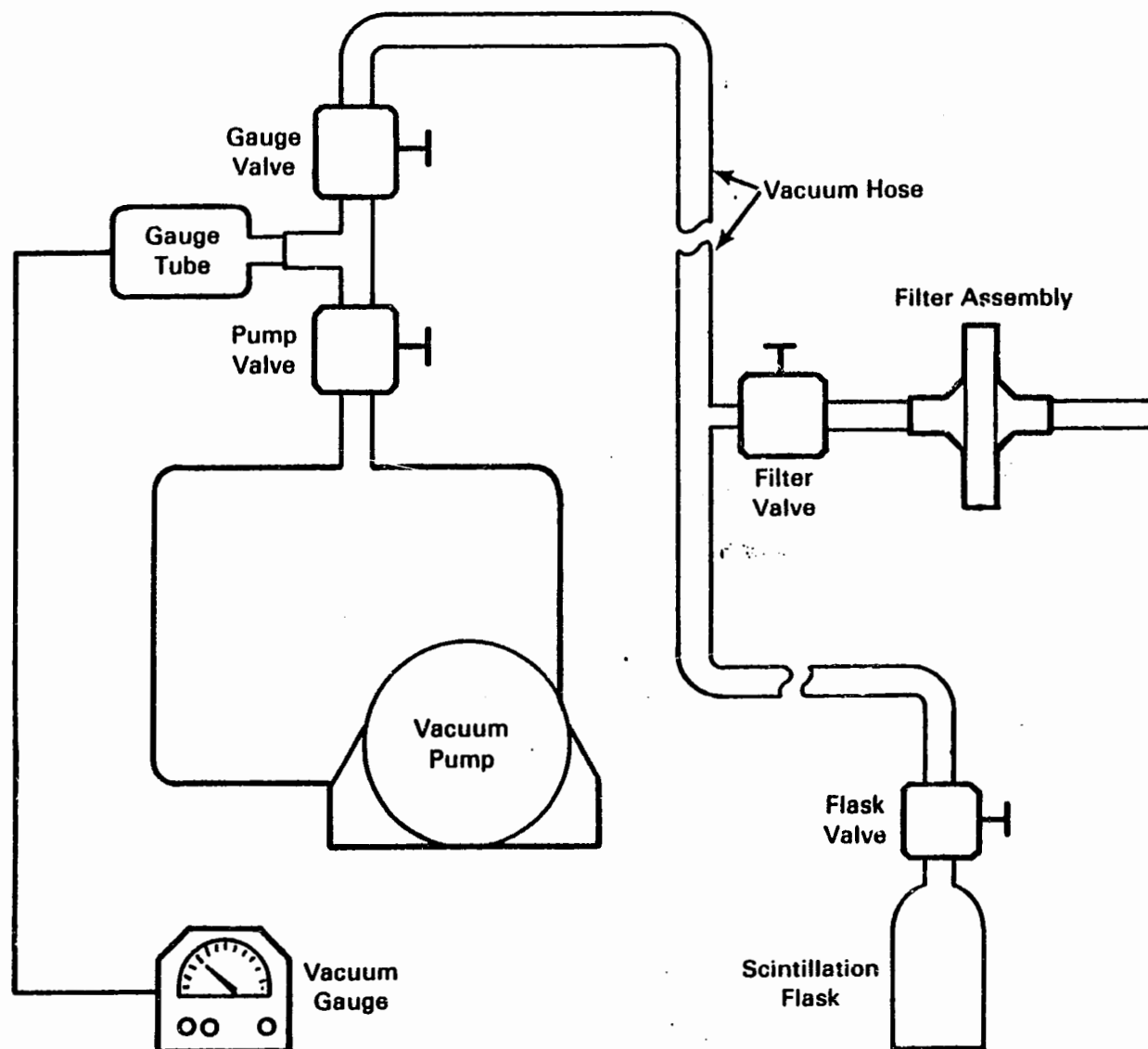
FIGURE 3. Vent-outlet radon sampling apparatus

FIGURE 4. Flask evacuation assembly

Stopcock

Zinc Sulfide
Scintillator
Coated

Metal Walls

Clear Silica or Pyrex
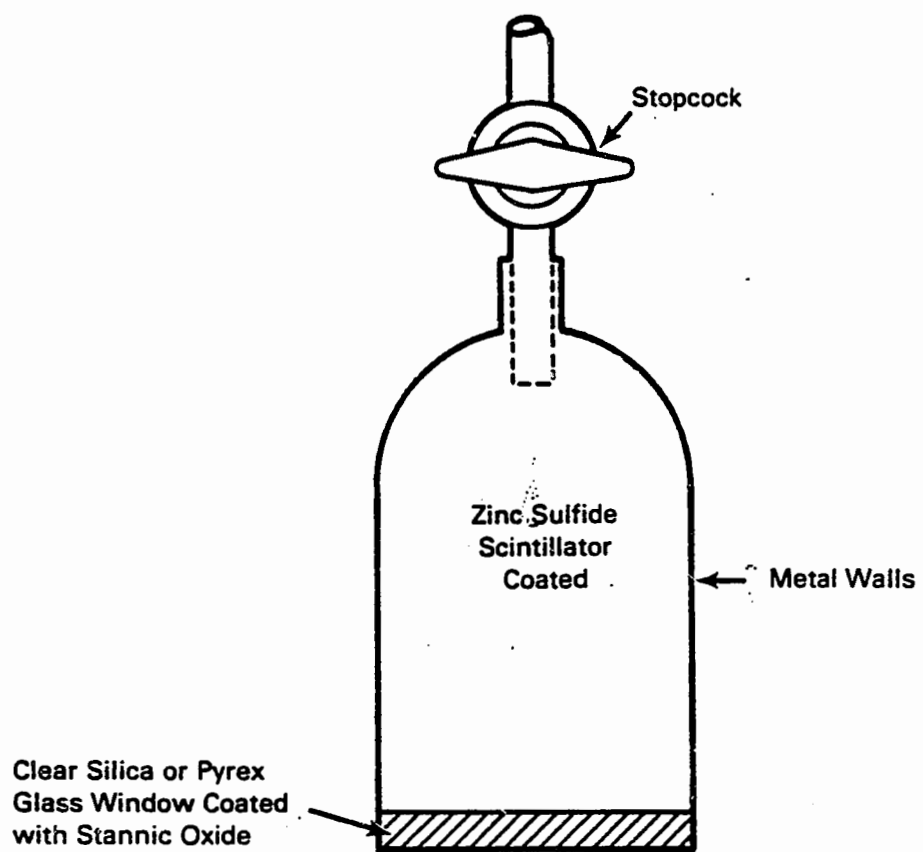Glass Window Coated
with Stannic Oxide

FIGURE 5. Scintillation flask

one or two valves or stopcocks suitable for high vacuum and positive pressures up to 14.7 psig. Flasks may have any combination of efficiency, volume, and background counting rate that produces a relative counting error s_c (4.2) no greater than 10% with air containing 10 pCi ^{222}Rn/liter using a 30 minute counting interval on the counting system in use. A typical small scintillation flask is shown in Figure 5.

5.3 Counting System. The counting system must have at least the following components.

5.3.1 Light Tight Counting Chamber to house photomultiplier tube and scintillation flask. The counting chamber must be equipped with a photomultiplier tube base and a resister string capable of supplying appropriate bias voltages to the photomultiplier (P.M.) tube dynode stages and anode. Radio frequency and magnetic shielding for the P.M. tube must be provided. Suitable connectors for applying high voltage to the P.M. tube must be provided. The chamber must be equipped to supply a pulse signal from the P.M. tube anode that is decoupled from high voltage. A preamplifier may be included in the electrical circuit.

5.3.2 Photomultiplier (P.M.) Tube. The P.M. tube must be capable of operation at overall bias voltages of at least 1400 volts, and must have a peak spectral response for emission wavelengths of 440-450 μm. P.M. tubes must have flat end viewing windows.

5.3.3 High Voltage Power Supply. The power supply should be capable of delivering the D.C. voltage requirements of the photomultiplier tube and the current requirements of the resister string of the tube base. High voltage must be filtered, to give <10 mV ripple peak to peak 5 hz to 50 mHz and regulated to provide ≲ 0.0025% variation in the output voltage for any combination of line and load variation within the operating voltage and current requirements of the photomultiplier tube and tube base in normal counting operations. Voltage settings must be indicated and adjustable in steps of ≲25 volts; any setting must be repeatable to within ± 0.5%. Temperature stability must be ≲ ± 50 ppm/°C from 0 to 50°C. Voltage and current requirements of the power supply must be compatible with either the power supplied by the low voltage power supply to the counting system, or by 110V AC lines.

5.3.4 Amplifier. The counting system amplifier must be compatible with the pulses from the output of the photomultiplier tube base (voltage, time constant and electrical impedence). Amplifier overall gain must be sufficient to produce signal pulses larger than the noise discrimination level used in either the amplifier output or the scaler input when the photomultiplier tube is operating in its normal voltage range. Gain must be externally adjustable or preset for optimum system performance. If preamplification is not provided by the tube base (5.3.1), a suitable preamplification stage must be provided in the amplifier with an externally accessable signal test point at its output. Gain drift must be ≲ 0.005%/°C. The amplifier must provide pulse shaping networks, and baseline restoration suitable for zinc sulfide scintillator count-rates expected from radon concentrations up to 1.5 X 10^5 pCi/liter. It must have an operating temperature range from 0 to 50°C.

5.3.5 Scaler/Discriminator/Timer. Scaler input requirements must be compatible with amplifier output pulses and impedence. The scaler must have a preadjusted input discriminator, or if a discriminating stage with an indicating external adjustment to eliminate spurious counting from noise or other erroneous pulses. If the amplifier contains an indicating output discriminator, or if a separate discriminator stage is provided, the scaler discriminator need not be indicating. Scaler circuits must have at least 20 MHz count-rate capability. The count readout must be at least six decades with either LED or LCD display.

An overflow indicator must be provided if less than seven decades are displayed. Leading zeros must be suppressed. If a separate timing unit is used, the scaler must be provided with a gate to allow the timer to turn it on and off. A test feature which illuminates all segments must be provided for readouts. The timer must be crystal timebase controlled, with an inherent accuracy of at least ± 0.0025%, and setable in minutes, with increments no larger than 1 minute and a maximum interval that is not less than 60 minutes. The timer must be accurate to within ± 0.01% over an operating temperature range of 0 to 50°C. The timer must provide gating signals compatible with the scaler.

5.3.6 Cabinet/Power Supply. The amplifier, scaler, (discriminator), and timer must be housed in a suitable cabinet that has provisions for housing the power supply where necessary. The cabinet must provide all power needs of the equipment it houses with suitable regulation of the needed power. Ventilation must be adequate to prevent internal temperatures greater than 50°C with all housed equipment operating, in an environment which is less than 35°C.

5.4 Vacuum Pump. A small one or two stage rotary vane vacuum pump must be used. The ultimate blank-off vacuum must be 20 microns or less and free air pumping rates must be at least 20 l/min. The exhaust outlet must be connectable to tubing or hose no larger than 3/4" I.D. to permit exhausting to outdoors.

5.5 Vacuum Gauge. The vacuum-gauge must be a thermocouple type with a range of 0-1000 microns of mercury (0-5000 microns optional) and a direct meter readout. It must be equipped with a metal gauge tube with a male npt thread connector, threaded to metal or plastic tee adapter. The metal gauge tube and tee adapter are to be coupled to vacuum pump shut-off valve (5.8).

5.6 Pressure Gauge, Air. The range of the pressure gauge must be 0-15 psig. It must be accurate to 3% of its scale range.

5.7 Compressed Air Regulator. The regulator must have single or double stage control and a flow indicator. The regulator must conform to the following specifications: maximum inlet pressure: 3000 psi; outlet pressure range: 4-80 psi; flow range: 0.2-2 slpm. The inlet connector must be CGA 346 or other as used for air cylinders specified in Paragraph 6.2. Regulator must be equipped with shut-off and flow control valves.

5.8 Vacuum/Pressure Valves (three required). The valve shut-off, and connections must be leak tight. The internal flow restriction must provide a maximum flow of at least 20 SCFM air at a pressure drop of 10 psi. Valves isolate the vacuum pump, (5.4) vacuum-gauge, (5.5) and the outlet of in-line filter (5.1.4).

5.9 Explosion Shield. The explosion shield must be suitable for scintillation flask pressurization or evacuation operations.

5.10 Barometer (optional). The barometer must be calibrated for the altitudes at the sampling site against a mercury barometer.

5.11 Oscilloscope (optional). The oscilloscope must provide a bandwidth 100 M Hz or more; a minimum sensitivity of 5 mV/div; fastest sweep of no more than 20 ns/div; a triggered sweep. It must provide sweep speeds from 20 ns/div to at least 50 ms/div and calibrated verticle scale from 5 mV/div to at least 5 V/div.

6. *Reagents*

6.1 Anti-Static Spray. Any commercial anti-static spray in an aerosol can may be used. Test a small area to be certain that spray constituents do not attack plastic components of scintillation flasks.

6.2 Air, Dry Compressed . Compressed air must comply with the following specifications: maximum dew point -75°F, or maximum moisture 3 ppm; maximum pressure 2200 psi; outlet connector compatible with regulator (5.8). This air must be aged at least 30 days before use, to allow ^{222}Rn to decay.

7. *Standards*

7.1 Calibration Sources. <u>Caution</u>. Radioisotope standards are available from commercial and governmental suppliers. However, the possession, transportation, and use of radioisotopes are regulated by federal, state and local statutes. For possession of many radioisotopes, licenses are required. Persons not specifically trained in the safe handling of radioactive materials should not perform analyses involving their use. Compliance with institutional, local, state, and federal regulations concerning the procurement and use of radioactive materials is the responsibility of the laboratory using this method.

7.1.1 Radon-222. Institutionally calibrated ^{222}Rn in air is available from the U. S. Bureau of Mines, Denver Research Center. Flasks to be calibrated must be appropriately packaged to prevent damage and shipped to the center where they will be filled with air containing radon. A range of concentrations is available. For the purpose of this method, a concentration of 1000 ± 200 pCi/liter should be requested. The shipping address is:

> U. S. Bureau of Mines
> Denver Research Center
> Bldg. 20, Denver Federal Center
> Denver, CO 30255
> Attention: Radiation Hazards Group

Prior to sending flasks, a written request for calibration should be sent to the Research Center. The type of flask to be filled, its approximate volume, the desired concentration, and the approximate date when the calibration will be needed, should be specified. Details can then be worked out between the laboratories. The radon concentration, internal pressure, relative humidity of fill air, calibration date and time, and the estimated accuracy of the calibration at one standard deviation will be needed from the research center.

A convenient regenerative source of radon in air using ^{226}Ra in solution can be constructed (12.1) or a radon generator using ^{226}Ra in a solid substrate can be purchased commercially. These sources can be used to make periodic checks of counter characteristics but must not be used for flask calibrations.

7.1.2 Control Source Scintillation Standard. Point source reference standards of alpha-particle emitting radionuclides are available from the National Bureau of Standards or commercial sources. These standards are prepared from radioisotopes which require a federal or state license for repossession. In Table 7.1 are listed two appropriate alpha-particle emitting point source standards and potential suppliers (<u>Other commercial suppliers may be available. The listing of a commercial supplier does not constitute endorsement by the U.S. Environmental Protection Agency or its contractors</u>).

TABLE 7.1

Point Source Alpha-Particle Emitting Standard Sources

Radionuclide	Nominal Activity Range	Supplier
Plutonium-238	11 - 540 nCi (specify)	U.S. Bureau of Standards
Americium-241	50nCi	New England Nuclear Corp.

Standards purchased for this method must have a nominal activity in the range from 10-50 nCi.

After receipt, the alpha-particle emitting control source should be mounted on a disc of transparent plastic coated with silver activated zinc sulfide scintillator. Material of this type is available from William B. Johnson and Associates, Research Park, Montville, N.J. 07045. Other suppliers may be available. See endorsement disclaimer above.

These scintillation control sources should be prepared by mounting the alpha-particle emitting source(s) with emitting surface facing the scintillator surface. (Note: if the two-source technique will be used to determine the instrument dead time (9.3), the alpha particle emitting sources should not be permanently attached to scintillator discs until after measuring the dead time. With that technique, both sources are mounted precisely on a scintillator disc and the counting rate measured. Then each must be mounted separately in the exact original positions and each counted.) After dead time measurements are taken, each source must be permanently attached to a scintillator disc. A diagram of one possible method for construction of this control source is shown in Figure 6. Specific dimensions of the scintillator disk are not critical as long as it is larger in diameter than the mount of the alpha-particle emitting point source, and equal to or less than the diameter of the face of the photomultiplier (P.M.) tube used for counting scintillation flasks. If the outside diameter of the scintillator disk is the same as the face of the P.M. tube, it will be easier to reproducably mount the source at one location on the P.M. tube. An index line should be marked on the edge of the source so that its orientation can be reproduced in counting.

8. *Safety Precautions.* The laboratory is responsible for compliance with all applicable federal, state, local, and institutional health and safety regulations in performing this method. Appropriate eye, ear and hand protection and protective clothing should be worn. Some of the safety considerations when sampling uranium mine vents are as follows:

8.1. Operations involving handling pressurized or evacuated scintillation flasks involve risk of explosion or implosion. Care should be used to prevent dropping or damage. Damaged scintillation flasks should not be used until repaired. Appropriate explosion shields should be used when pressurizing or evacuating.

8.2. The sound pressure level near some operating mine vents can be sufficient to cause hearing imparement, so effective ear protection should be worn in these cases.

8.3. Concentrations of ^{222}Rn in air emitted from some mine vents may be orders of magnitude greater than the concentrations specified for occupational exposures. Although potential exposure intervals are very brief, for those occasions where there is risk of excessive inhalation exposure, appropriate respiratory protection should be worn.

8.4. When sampling or measuring flow rates from operating vents, it is necessary to be aware of the location of air moving components, so that accidental contact with moving parts will be avoided.

8.5. Since the air velocity from mine vents can be of the order of several hundred meters per second, entrained sand, dust and gravel can be thrown from the vent outlet with considerable force. Appropriate eye protection should be worn.

8.6. The operating voltage for some fan motors is as high as 440V. Care should be taken to avoid sources of electric shock.

9. *Pretest Procedures*

9.1 Leak-testing Scintillation Flasks

Scintillation flasks must be tested for leaks before and after calibrating and at weekly intervals when samples are being collected at daily intervals

Point Source Attached
with Pressure Sensitive
Plastic Tape

Alpha Particle Emitting
Point Source (Activity
Facing Scintillator)

Top Surface Coated
with Silver Activated
Zinc Sulfide Phosphor

Transparent
Plastic Film

Reference Line for Positioning Source
Drawn on Edge

Pressure Sensitive
Plastic Tape

Point Source

Alpha-Particles

Zinc Sulfide
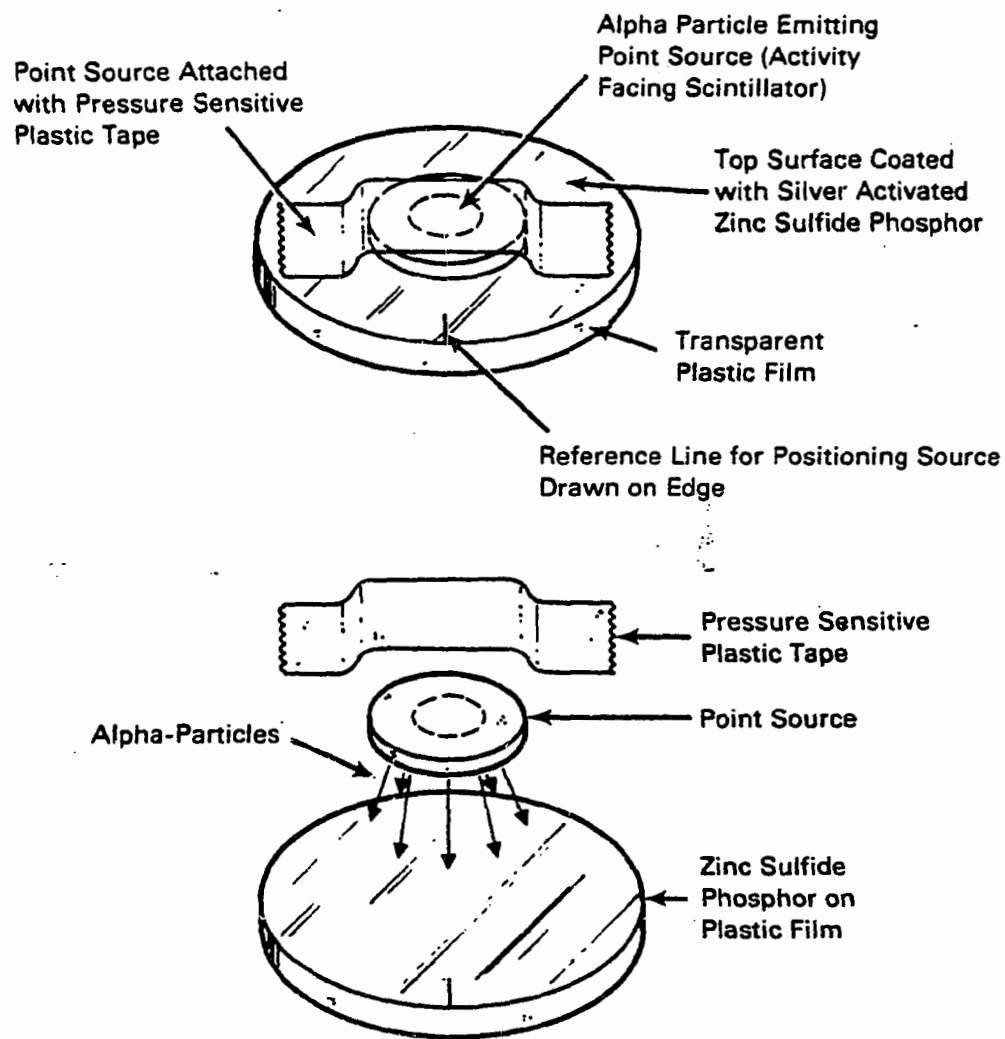Phosphor on
Plastic Film

FIGURE 6.   Suggested scintillating control source assembly details

or oftener. Both tests (9.1.1) and (9.1.2) should be performed initially, then (9.1.1) can be used for routine testing. Before vacuum leak testing, the vacuum system between the pump shut-off valve and the gauge, and filter assembly must be sufficiently leak tight that pressures do not drift more than 5% in a one-hour period at total pressures below 800 microns of Hg. Before pressure testing, verify that scintillation flasks are designed to withstand high vacuum and internal pressures exceeding 14.7 psig because of the explosion hazard. During vacuum and pressure testing, scintillation flasks should be behind a suitable explosion shield.

9.1.1 Vacuum Testing. Assemble the vacuum pump, gauge, and one of the on-line filters. Attach a vacuum valve (5.8) to the pump (5.4) inlet, with the flow direction into the pump. Attach the vacuum-gauge (5.5) to the valve inlet using a tee fitting. Attach a second valve to the third leg of the tee fitting. Connect a flexible tube to the inlet of the second valve. Attach a tee fitting to the flexible tubing . To one leg of the tee, attach the third vacuum valve (5.8). To the vacuum valve attach the outlet of an in-line filter (5.1.4). To the other leg of the tee, attach a length of flexible tubing which can be attached to the scintillation flask inlets (see Figure 5.) These connections may be made with flexible tubing and ribbed connectors or with threaded connectors. All connections must be vacuum tight with the scintillation flask inlet plugged (see above). Attach a scintillation flask to the vacuum pump, gauge and filter assembly. Evacuate the flask to give a measureable vacuum less than 800 microns. Record the pressure, close the scintillation flask valve and store for at least 16 hours. Reattach the flask to the pump assembly. Evacuate all lines to the initial internal pressure of the flask. Close valve between the vacuum pump and gauge and open the scintillation flask valve. Residual pressure in the scintillation flask must not have increased to more than 1000 microns. If a greater pressure is measured, determine source of the leak and repair it.

9.1.2 Pressure Testing. Attach the output of the compressed air regulator (5.7) to the inlet of filter assembly. Close the gauge valve. Insert a tee fitting between a scintillation flask and the pump assembly inlet hose and install the pressure gauge (5.6). With regulator shut-off valve closed, set the pressure of the regulator output to 4 psi. Open the regulator shut-off valve and flask valve. Slowly open the flow control valve and pressurize the flask to 4 psi on the pressure gauge attached to the tee. Close the flask valve and regulator shut-off valve. Remove the flask and store it for at least 16 hours. Re-attach the flask to the tee. Open the regulator shut-off valve and again adjust line pressure to 4 psi on gauge attached to tee. Close the regulator shut-off valve and open the flask valve. The pressure should be at least 3.9 psi. If a pressure leak is observed, the leak point may be determined using a leak-testing bubble solution or by emersion of the flask in water. Repair pressure leaks before using the flask.

9.2 Optimizing Electronic Set-points. Unless the optimum settings for counting scintillation flasks have been previously determined, the operating high voltage (H.V.) and set-points for the counting system must be established. The H.V. applied to the P.M. tube must be within the operating range for the tube specified by the manufacturer. In no case should the H.V. setting be above the maximum specified for the tube.

Place a scintillation flask filled with air containing at least 800 pCi ^{222}Rn/liter on the P.M. tube in the light tight chamber. Close the chamber and wait 5 minutes. Attach a suitable oscilloscope to the test point between the preamplifier output and amplifier input. Turn on the H.V. Increase the H.V. while observing pulses from preamplifier. Pulses will increase in size

as voltage increases. Increase the H.V. until the largest pulses from pre-amplifier begin to clip (flatten at their top). Then decrease the voltage slightly until no clipping is observed. Record this initial H.V. setting.

If the amplifier has adjustable gain, set the control(s) to the approximate center of the gain span. Adjust the discriminator between amplifier and scaler to minimum discrimination. Take a count and record the count-rate. Increase the discriminator setting by 10% of full scale. Take a count and record the count-rate. Repeat over the full range of the discriminator. At minimum discrimination the highest count-rates should be observed. The count-rate should decrease with increasing settings.

In the center of the span there should be a range of settings where count-rates change by about 5% between settings. (This is the discriminator plateau.) The set-point for the discriminator is the setting at about the center of the plateau. If count-rate does not drop by at least a factor of two between the discriminator plateau counts and those at the maximum setting, the amplifier gain setting should be reduced by about 10%. Re-set the discriminator to maximum and measure the count-rate. Continue decreasing amplifier gain by 10% steps and determine the point where the count-rate is about a factor of two lower than it is on the discriminator plateau region. This is the ampli-fier set-point. Then take a new series of measurements with 10% increments of the discriminator. Set the discriminator at the set-point where the count-rate is in the center of the discriminator plateau region as defined above. If there was no discriminator plateau on the original set of measurements, and count-rates fell by a factor of two or more at lower discriminator settings, the amplifier gain should be increased in 10% steps until the count-rate at a discriminator setting of 30% of full scale is less than 10% lower than the count-rate at a setting of 20% of full scale. This is the approximate ampli-fier set-point. Then the count rate should be remeasured at each discriminator setting to establish the discriminator plateau and set-point. The amplifier set-point can be readjusted slightly if necessary to optimize the upper end of the plateau curve.

After the set-points for the discriminator and amplifier gain have been established, the H.V. plateau should be determined. Set the amplifier and discriminator to their set-points. Lower the H.V. by 300 volts from the initial set-point, and measure the count-rate. Increase H.V. in 100 volt steps, measuring the count-rate at each step. The count-rate will increase rapidly at first, then increase more slowly. When the count-rate again in-creases rapidly, discontinue increasing the H.V. Reduce the H.V. by 200 V.

Make a linear plot with the count-rate as the ordinate and the H.V. as the abscissa. The region of the curve through the data which has the minimum slope is the H.V. plateau. The slope of the H.V. plateau should be less than 5% increase of count-rate per 100 V increase of H.V. The length of the H.V. plateau should be at least 100 V. If necessary, intermediate points on the plateau can be measured at 50 V intervals. When measurements are com-pleted, turn off the H.V. and remove the flask. The useful settings for H.V. are approximately in the center of the H.V. plateau but should not ex-ceed the initial H.V. setting determined by oscilloscope testing, by more than 50V. A voltage plateau for the instrument background should also be measured using a new flask filled with aged air (9.5). A series of overnight background counts should be taken, one at each of the previously determined H.V. plateau set points. The optimum H.V. setting will have the minimum background count for a voltage setting that is on the original plateau. If necessary, the H.V. set-point can be at any point that is more than 50V from the extremities of the H.V. plateau. The optimum settings and all sample,

background, and calibration counts made at those settings must be recorded. For nominally identical scintillation flasks, these settings may be assumed to be the same from flask to flask. Otherwise, optimum settings for H.V. should be determined for each kind of flask, but amplifier and discriminator settings should not be readjusted.

9.3 Instrument Dead Time. Unless the instrument dead time for the scintillation flask counting system has been provided by the manufacturer, it must be determined by the operator. The dead time should be determined using one of the methods given by the National Council on Radiation Protection and Measurements (12.9). Zinc-sulfide scintillation counters may be assumed to have non-extendable dead time effects, over the range of count-rates expected for mine-vent monitoring. One or two alpha-particle emitting source(s) mounted on zinc sulfide phosphor disc(s) (7.1.2) will be needed for this calibration.

The source-pulser method is recommended for its relative accuracy and simplicity; otherwise, the two-source method is suggested.

Once the dead time has been established, the expression relating the true count-rate to the observed count-rate is:

$N = n/(1-n\tau)$

where,

   N = the corrected count-rate,

   n = the observed count-rate,

and, τ = the dead time in the same time units as count-rates.

When τ has been measured, this equation can be used to determine the corrected count-rate for any given measured rate.
Using the expression:

$$n = \frac{0.01}{1.01\ \tau} ,$$

the count-rate corresponding to a 1% correction ($N/n = 1.01$) can be determined. For count-rates below that quantity, the dead time correction may be neglected.

9.4 Control Source Checks. The control source (7.1.2) is used to determine that the counting efficiency of the counting system remains constant after calibration. The count-rate for the control source is established during calibrations. Daily counts of the control source are made when samples are being analyzed to establish that the efficiency has not changed significantly.

After the H.V. operating point has been established for scintillation flasks, the control source must be counted at that H.V. Before flask calibration counts are taken, position the control source on the P.M. tube, close the light tight counting chamber, wait five minutes, adjust H.V. to the set-point and take a count of sufficient duration to accumulate at least 20,000 counts. Record the count. Turn off the H.V. Remove the source and then replace it and repeat the counting process. Repeat removing, replacing and counting the control source ten times. Remove the source, and measure the background counting rate for the empty counting chamber using the same counting interval. Compute the count-rate for all counts. Subtract the empty chamber background count-rate from all control source count-rates. Then compute the average count-rate for the control source and compute the relative standard deviation of the net count-rate. If the relative standard deviation exceeds 1% of the average, the variability of control source counts is excessive. In that case, it is necessary to determine whether the source positioning is inconsistent or the counting system is malfunctioning. Once the source of excess variation has been corrected and a set of 10 control

source measurements with acceptable variability has been accumulated, prepare a control chart with the average count-rate for that counting system and limits of ±2% of the average count-rate shown on the verticle scale and the dates of measurements shown on the horizontal scale.

Each day before samples are counted, make one measurement of the control source and the empty-chamber background using the same time interval as previously. Plot the net count-rate on the control chart. If the net count-rate is within ±2% of the previous average, the system is in control and may be used for samples. If a control source count-rate falls outside the limits, repeat the count one time. If it falls within limits, the instrument may be used. If it does not, a counter malfunction is likely, and the cause of the changed counting efficiency must be determined and corrected. If the first count falls outside of limits on two consecutive days the cause must also be determined and corrected.

9.5 Backgrounds. Prior to calibration or sampling, the background count-rate for each flask must be determined using scintillation flasks filled with compressed air that has been aged for at least 30 days to permit the decay of any radon initially present.

9.5.1 Evacuation and Filling. For flasks with two inlet valves, evacuation is unnecessary, and flushing may be accomplished with the vacuum-pump and gauge isolated from the system, using aged compressed air, through the filter inlet. For flasks with a single inlet valve, the following procedure must be followed. Attach the vacuum pump, gauge and filter assembly (Figure 4) to the flask using flexible tubing. With the flask valve closed, evacuate the system. Turn on the thermocouple gauge. While pumping, open the valve on the scintillation flask and evacuate to less than 1000 microns pressure. Close the valve on scintillation flask. Close the vacuum-guage valve and open the filter-assembly valve. Open the valve on scintillation flask and allow it to fill with filtered ambient air. Close the filter assembly valve; then open the vacuum-gauge valve and evacuate the flask to less than 1000 microns pressure. Repeat filling and flushing two more times. Evacuate the flask to less than 1000 microns pressure and close the flask and vacuum-guage valves. Attach the outlet of the compressed air regulator to the filter inlet. Adjust the regulator pressure to 4 psi with regulator shut-off valve closed. Open the filter assembly and vacuum-gauge valves and pump the filter assembly to less than 1000 microns. Shut off the gauge valve. Open the regulator shut-off valve and flask-valve and slowly open the flow control valve of the regulator. Allow the flask to fill until flow rate drops. Close the flask valve and regulator shut-off valve. Remove the flask from the assembly and open the flask valve briefly to allow pressure to equilibrate with ambient atmospheric pressure.

9.5.2 Background Count Measurement. Spray the exterior of the evacuated flask with anti-static spray and store overnight. Place flask in the counting chamber taking care to center the window of the flask on the photomultiplier tube. Select one orientation for all flasks (e.g. valve handle facing forward) so that the geometry is consistent. Close the chamber. Allow five minutes for the phosphorescence of the scintillator to decay to insignificance and adjust H.V. to the set-point and initiate a 30-minute background count. Record the counts registered, the count interval, flask number, and counting date and time. Compute background counts per minute and record. The background count-rate for each flask may be written on the flask or a label attached to the top of the flask. However, static charges may be induced when removing gummed labels, so they should not be removed until after the sample has been collected and counted.

9.6 Calibration

Prior to any sampling run, calibrate the system using the following procedures. A complete calibration should be performed before making field measurements and at six month intervals. Send all scintillation flasks used for field measurements (at least four flasks) to the Bureau of Mines to be filled with a known quantity of ^{222}Rn in air (1000 ± 200 pCi/l), (7.1.1.) When flasks are received, examine them for damage in shipping, spray all exterior surfaces with anti-static spray. Delay counting for at least three hours after spraying. Place a scintillation flask on the P.M. tube in the counting chamber and close the lid. After five minutes, adjust the H.V. to the set-point. Preset the time to at least 30 minutes, and initiate the count. Record the time of the count, flask identification code and length of the count. After completion of the count, record the count registered on the scaler. Turn off the high voltage, remove the flask and replace it with the next flask to be calibrated, and repeat the process. After all flasks have been counted one time, repeat the process a second time for all flasks. After 1-2 days, recount all flasks.

If the elevation at the measurement site is more than 1000 feet higher or lower than the elevation in Denver, Colorado, the pressure within the scintillation flask will be significantly different from the pressure normally present when flasks are filled during sampling. This can make a small difference in the counting efficiency of the flasks. If the elevation is lower, filtered ambient air may be allowed to bleed into each flask through a length of tubing. (The radon concentration in the ambient air should be negligible compared to that inside the flask.) Close flasks as soon as possible after filling to minimize outward diffusion of the flask contents.

If the altitude at the location of sampling is more than 1000 feet higher than at Denver, CO., it will be necessary to bleed a portion of the air from each flask to achieve pressure equilibrium. Bleed air from the flask to the outdoors or a suitable fume hood through a 4-5 foot length of tubing. Close the flask as soon as possible. The dilution of the flask contents will be negligible and the net effect will be the removal of a fraction of the radon-air mixture. The fraction removed can be determined from the barometric pressures and temperatures at the time of filling and at the sampling site. If a local weather station cannot provide the barometric pressure, it must be measured with a barometer (5.11).

After bleeding or filling flasks, repeat the original counting procedure again, taking three counts spaced by at least 1 to 2 days. After counts have been completed, efficiency factors should be computed before evacuating flasks.

9.6.1 Correction for loss of Flask Contents after Bleeding. The correction factor for computing the fraction of the initial radon-air mixture remaining in the flask is as follows:

$$F = \frac{P_2 T_1}{P_1 T_2}$$

where:

F = the fraction of initial air containing ^{222}Rn remaining in the flask

P_1 = the barometric pressure of the fill air at the time of filling, mm Hg,

P_2 = the barometric pressure at the sampling site, mm Hg,

T_1 = the absolute temperature of the fill air at the time of filling, $^{\circ}$K, and,

T_2 = the absolute temperature at the location when bleeding air, $^{\circ}$K.

9.6.2 Efficiency Calculations. The following formula must be used to compute flask efficiencies:

$$E = \frac{[C_S - C_B(t_S/t_B)](\lambda) \exp(\lambda_T)}{[1-\exp(-\lambda t_S)](F)(P)\ [1-C_S(\tau/t_S)]}$$

where,
E = the flask efficiency, counts/min per pCi ^{222}Rn/liter,
C_S = the observed standard count during the counting interval
C_B = the observed background count prior to filling,
   $\exp(X)$ is the Xth power of the base of natural logorithm
λ = the radioactive decay constant for ^{222}Rn $(1.259 \times 10^{-4}$ min$^{-1})$,
T = the interval between the start of the count and the date/time of the fill at calibration, minutes,
t_S = the length of the standard count interval, in minutes,
t_B = the length of the background count, in minutes,
F = the fractional loss of air for flasks bled at the sampling site, (F=1.0 unless flask pressure was reduced by bleeding before the count)
P = the calibration concentration of the ^{222}Rn in the flask at the time of fill calibration, pCi/liter,
and,
τ = the dead time determined for flasks, minutes.

Tabulate counting efficiencies and respective counting dates and times. For each flask compute the average and relative standard error of the average for the original set of three counts and also the second set if flask air pressure was increased or decreased. Use ordinary statistical formulas and not errors based on counting statistics. The statistical formula to use is as follows:

$$E_{\bar{x}} = \pm(100) \left[\frac{n \sum_{i=1}^{n} X_i^2 - (\sum_{i=1}^{n} X_i)^2}{(n-1)} \right]^{1/2} \frac{1}{\sum_{i=1}^{n} X_i}$$

where,
$E_{\bar{x}}$ = the relative standard error of the average efficiency, %,
n = the number of replicate counting measurements used to determine the average efficiency, (3 in this case),
and,
X_i = the efficiency determined from the ith count, c/m per pCi/l.
The relative standard errors of each average must be less than ±3%. If errors exceed 3%, determine if there is a decrease in efficiency with time for each set of counts. Such decrease indicates that a flask is probably leaking. Scattered efficiencies indicates that there has probably been a problem with static charge buildup on flasks. In the latter case, an additional set of counts may be taken after again spraying with anti-static spray and waiting at least three hours. If flask internal pressures have been adjusted, this additional set should correspond to the set taken after adjustment. A new overall average and relative standard error of the average may be taken after discarding the efficiency of any single count which represents an extreme value. This relative standard error must not exceed ±3.

If the relative error of the efficiency for any flask exceeds 3%, the flask should be evacuated, checked for leaks, and leaks repaired, and returned to the Bureau of Mines for recalibration. The efficiency of a flask should be constant under constant conditions of internal pressure. A variation of measured efficiencies can be caused by changes in internal pressure, the distribution of radon progeny within the flask or the presence of a barrier such as liquid water on the scintillation surface. The average efficiency for the set taken after any required pressure adjustment will normally differ from the original average by less than 15%. If pressure was increased, the efficiency should decrease, and if pressure was decreased, the efficiency should increase. The ratio of the efficiency after pressure adjustment to the original efficiency for flasks of the same nominal construction must be consistent. Any flask with a ratio differing from the average ratio for other flasks of the same construction by more than 5% must also be re-calibrated.

Average efficiency factors with less than 3% relative standard deviation may be used in sample computations for the corresponding flask. If adjustment of the internal pressure of flasks was necessary, the average efficiency for each flask after the pressure adjustment must be used in sample computations. After calibration counts are complete, evacuate and flush the flasks with ambient air, exhausting their contents outdoors or to a fume hood.

Test all flasks for leaks (9.1).

9.7 Replication Errors (s_m)

Replication error is the inherent error of a set replicate measurements when a constant concentration of radon is measured with several flasks under conditions such that the inherent counting errors are negligible. Unless a source of radon, known to have a fixed concentration, is available at the laboratory, flasks should be returned to the calibration facility of the Bureau of Mines after the original calibration. At least four flasks must be used in this test. When flasks have been filled and returned, the radon concentrations should be measured and each corrected to a single reference time using the original calibrations. Counting intervals must be long enough to accumulate at least 20,000 counts for each flask so that the error from counting statistics is negligible. Then the standard deviation of the replicate concentration measurements should be calculated using the usual statistical formula (9.6.2). s_m must be equal to or less than 7%, which is the assumed maximum inherent error for this method.

10. *Sampling and Analysis Procedure.*

10.1 General Considerations. This section describes the use of apparatus and protocol for sampling and measuring ^{222}Rn concentrations. To establish the average characteristic emission rate during a particular quarterly interval, it is necessary to measure both the ^{222}Rn concentrations and vent flows from all vents of a mine. Because of the short-term variability of the emission rate, grab-sampling measurements must be repeated on six occasions and averaged. Duplicate grab samples must be taken on each occasion. The sampling schedule must be adjusted to include emissions under typical operating conditions and to exclude occasions when non-typical short-term variations are probable. All six measurements must be completed in a calendar quarter of a year. Samples must not be collected at the following times:

1. Within one hour of shift change or blasting.

2. During a period when mine ventilation is other than that typical during active mining. (e.g. a shut-down or flow-reversal).

3. Within two hours after shut-down or reversal of vent fans.

4. During or within 24 hours after passage of a local weather disturbance (storm) that produces an atypical barometric pressure.

Sampling times must be spaced so as to obtain a representative sampling of the air concentrations present during normal operating conditions. One of the six samples must be collected between 1:00 and 3:00 P.M. to include the normal daily concentration maximum reported by Jackson (12.4). The remaining samples should be collected at uniform intervals during working shifts. At mines which have more than one exhaust vent, samples should be collected from all exhaust vents before resampling any vent.

Since some existing mine vents are not equipped with in-line sampling ports, two alternate sampling methods are given. For sampling from vents with existing sampling ports, the in-line sampling technique should be used.

10.2 Sample Collection. Use scintillation flasks that have been sprayed with anti-static spray. Measure flask background count-rates before using. For flasks with a single valve, evacuate before sampling. Flasks with two valves need not be evacuated.

10.2.1 Sampling vent stacks with access sampling ports. A modification of the in-stack grab sampling procedure specified in Method 3 of Appendix A, 40CFR Part 60 (12.8) must be used. When entrained water is present, fit the water separator (5.1.3) to the outlet of the squeeze bulb (5.1.1). Then attach the inlet of the in-line filter assembly (5.1.4) either directly to the squeeze bulb outlet or to the outlet of the water separator. Insert the probe into the access port and position as instructed in Method 3 of (12.8). Squeeze the bulb several times to completely flush the sampling system. (At least enough times to pump 10 times the normal volume of the system). Attach a scintillation flask (5.7) to the outlet of the in-line filter using flexible tubing. Open the inlet valve of an evacuated flask or both valves of dual inlet flasks. Squeeze the bulb a sufficient number of times to pressurize or flush the scintillation flask. Flushing should be continued until at least 10 flask volumes have been pumped through. Close the flask valve(s). Remove the flask and attach a second flask. Repeat filling or flushing the flask. After both samples have been collected, partially open one valve of each for 1-2 seconds, then close, to allow the internal pressure to equilibrate with ambient atmospheric pressure. Record the flask identification codes, sampling site, and the date and time of sampling in a field notebook. The vent flow must be measured immediately preceding or following sample collection. Return flasks to the laboratory for counting. Avoid rubbing flask exterior surfaces to minimize the possibility of inducing static charges.

10.2.2 Sampling Vent Stacks without Access Ports. Assemble vent outlet sampling apparatus (5.2). When entrained water is present, fit the water separator (5.1.4) to the outlet of the probe (5.1.2) with flexible tubing. Then attach the inlet of the in-line filter assembly (5.1.4) either directly to the probe outlet of the outlet of the water separator. Connect the flow-meter (5.1.5) to the outlet of the in-line filter. Flexible tubing should be attached to the outlet of the flow-meter to permit attaching the scintillation flasks. See Figure 3.

Before inserting the funnel end of the probe into the outlet of a vent, observe the interior of the vent to determine the locations of moving parts and obstacles. A suitable ladder may be needed. Cautiously insert the funnel end of the probe into the air stream and into the vent. With the outlet of the probe parallel to the air stream, the plane of the face of the probe funnel inlet will be facing to the stream. The bend of the metal probe tube will limit the depth that the probe can be inserted into the vent. It is necessary to insert the probe as deeply as possible without touching moving parts. Do not insert deeper than the limit created by the bend in the metal tubing. Air will be forced through the probe and attached equipment. Adjust

the position of the probe to obtain the maximum possible flow, indicated by the flow-meter. Any sampling flow-rate greater than 2 l/min will supply a sample in a reasonably short time. Attach a scintillation flask inlet valve to the tubing on the outlet of the flow-meter. Slowly open the inlet valve of the scintillation flask. For evacuated flasks, allow to fill slowly so that the fill rate does not exceed the rate of supply. For flasks with dual inlet valves, open the second valve and allow the flask to flush for the interval necessary to pass at least 10 flask volumes. When evacuated flasks are filled, the flow indication will drop to zero. Close flask valve(s) and remove from the hose. Attach a second scintillation flask and repeat the sampling. After both samples have been collected, partially open the inlet valve for 1-2 seconds, then close, to equilibrate the interior pressure with the ambient atmospheric pressure. Record the flask identification codes, sampling site and the date and time of sampling in a field notebook. The vent flow must be measured immediately preceding or following sample collection. Return the flasks to the laboratory for counting. Avoid rubbing the flask exterior surfaces to minimize the possibility of inducing static charges.

10.3 Counting Scintillation Flask. Wait at least three hours after filling scintillation flasks to permit the ingrowth of the short-lived daughters of ^{222}Rn. The counting system must be in control (9.3) before using. Place a scintillation flask in the light tight chamber in the same orientation to the P.M. tube that was used during calibration. Close the light tight chamber, turn on H.V. and adjust to the set-point. Wait 5 minutes. Adjust the timer for a 30-minute count interval. Initiate the count. Record the time that the count started, the flask identification code, sampling site, and the date and time of sampling. When the count is completed, record the number of counts. Turn-off the H.V. and change samples. Repeat the counting procedure for all samples. Calculate the radon concentrations before pumping samples from scintillation flasks. If a pair of duplicate measurements differs by more than the maximum allowable error (e_{max}) given below, recount both flasks and recalculate radon concentrations and allowable errors. Record all results. Any pair of the measurements from two flasks not differing by more than e_{max} can be averaged and the average used as the acceptable measurement for a given sampling. If no pair of measurements are acceptable, the sampling must be repeated. If there is a possibility that ^{220}Rn is present in mine air recount flasks as indicated in Paragraph 3.4 and use the results to determine an appropriate delay time between sampling and counting.

11. *Calculations*. Carry out all calculations retaining one extra decimal place beyond that present in the acquired data. Round off the numbers following calculations, retaining three significant figures.

11.1 Allowable Error. The formula for the allowable error (e) between replicate samples is as follows:

$$e = 200 \left[\frac{(C_S/t_S^2 + C_B/t_B^2)}{(C_S/t_S - C_B/t_B)^2} + \left(\frac{s_m}{100} \right)^2 \right]^{1/2}$$

where,
  e = the allowable error between duplicates (%)
  C_S = the gross sample count including background,
  t_S = the sample count interval, minutes,

C_B = the background count interval, minutes
t_B = the background count interval, minutes
and
s_m = the measured inherent limiting precision (9.7) of scintillation flask measurements, %.

The allowable error or each of the pair of flasks must be calculated. The maximum allowable error (e_{max}) is the larger of the two values of e.

11.2 Radon Concentration. The formula for calculating the concentrations of radon is as follows:

$$ P = \frac{[C_S - C_B t_S / t_B] \; \lambda \; [\exp(\lambda T)]}{[1 - \exp(-\lambda t_S)] \; E \; [1 - C_S(\tau / t_S)]} $$

where,

P = the ^{222}Rn concentration in pCi/liter,

λ = the decay constant for ^{222}Rn ($\equiv 1.259 \times 10^{-4}$ min^{-1}),

$\exp(X)$ = the X power of the base of the natural logarithm

T = the interval between sample collection and the start of the sample count, minutes,

E = the efficiency factor for the scintillation flask, c/m per pCi/l,

and

τ = the dead time for the counting system in minutes. (τ may be assumed to be zero for sample count-rates which give a dead time correction less than 1% (9.3).

After calculating the average concentrations for pairs of sampling flasks with acceptable allowable error, for each of six samplings, record all results on a sample record sheet. (See Figure 7 for an example.)

FIGURE 7.   Example of Radon Count/Calculation Record Table.

Record of Radon Concentration Measurements

Company_____

Mine_____

Vent Identification_____

Sample 1.

   Sampling Date/Time_____

Background Counts

Flask	Date/Time	Length of Count	Total Counts

Flask	Instrument	Date/Time	Length of Count	Total Count	Radon Conc.	Allowable Error

Test A. Minimum % Range Between Duplicate Flasks_____%. Test
B. Maximum % Allowable Error_____%. If "B" > "A". Average
^{222}Rn concentration from Test "A" Flask Measurements_____
pCi/liter. IF "A" > "BB". Resample.

12. *References*

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METHOD 2 - VENT FLOW MEASUREMENTS

1. Principle and Applicability.
1.1 Principle. To determine the rate of ^{222}Rn emissions from underground uranium mines, both the average radon concentration in the air and the volumetric flow rate must be measured. The volumetric flow rate measurements must be taken at the above ground portion of the mine exhaust vent using the reference technique found in Method 1 of Appendix A, 40CFR Part 60, (See Method 1, 12.8 for reference). A traverse of air velocity measurements must be taken in the accessible portion of the vent which best conforms to the requirements of that method.
1.2 Applicability. Method 1 (12.8) specifies that it can not be used when (1) flow is cyclonic or swirling, (2) a stack is smaller than about 0.3 m (12 in.) in diameter or 0.071 m^2 (113 in.^{2}) in cross-sectional area or (3) the measurement site is less than two stack or duct diameter downstream or less than a half diameter upstream of a flow disturbance. However, measurement points at existing mine vents rarely conform to item (3) and occasionally fail to conform to items (1) and (2). At some vents no in-line access ports have been provided for sampling and velocity measuring traverses. At some mines, air is exhausted from large entry portals where no stack is present. Procedures for a detailed assessment of the applicability of this method are given in the EPA Quality Assurance Handbook for Air Pollution Measurement Systems-Volume 3. Stationary Source Specific Methods, EPA-600/4-77-027b August, 1977. This volume also gives more detail about measurement and sampling considerations than is present in (12.8). If it is determined that the reference method is not applicable to a measurement site, suitable modifications to the sampling site must be made or the administrator must be consulted about alternative measurement methods, such as the anemometer technique given in Method 14 of (12.8).
2. Computations. For flow measurements from mine vents, the computation of average stack gas velocity conforms to Paragraph 5.2 of Method 1 (See Method 1, 12.8 for reference). However, ^{222}Rn concentration measurements are made after equilibrating the internal pressure of the sampled air in scintillation flasks with the local barometric pressure. The normal computation for stack gas volumetric flow given in Paragraph 5.3 of the reference method converts the velocity to the equivalent volume of dry air at standard barometric pressure and temperature. For the purpose of this method, it will not be necessary to make this conversion. The applicable formula is as follows:

$$Q_e = \frac{3.6 \times 10^6 \, V_s A P_s}{P_{bar}}$$

where,
   Q_e = volumetric ambient stack gas flow rate corrected to local barometric pressure, liter/hour),
   3.6×10^6 = conversion factor, (sec/hr)(liters/m^3),
   v_s = Average stack gas velocity, m/sec,
   A = Cross sectional area of stack, m^2,
   P_s = Absolute stack gas pressure, mm Hg,
        $(P_s = P_{bar} + P_g)$

P_{bar} = barometric pressure at measurement site, mm Hg,
and
P_g = stack static pressure, mm Hg.

2.1 Summary Table. A summary table must be prepared listing the mine, vent number, and flow rates calculated for each of the six sampling intervals.

METHOD 3--COMPUTATION OF RADON EMISSION RATES

1. Principles and Limitations
1.1 Principals. Radon emission rates must be computed for each mine vent
from the average radon concentration measured in Method 1 and the flow rate
measured in Method 2. An emission rate must be computed for each of six
sampling occasions. Then the six emission rates must be average to give the
average rate for that vent during one quarter of a year. The annual average
emission rate is the average of four quarterly averages.
1.2 Limitations. Limitations of the measurement processes are discussed
for methods 1 and 2. The computational process produces average emission
rates relative to the barometric pressure at the sampling site. Thus, the
emission rates are useful for predicting local concentrations of radon as a
result of atmospheric dilution. (If exposure rate estimates at distant sites
are required, radon concentrations and stack flow rate determinations would
have to be corrected to the equivalent dry air at standard temperatures and
pressures.)
2. Calculations. Carry out calculations retaining one extra decimal figure
beyond that present in the required data or previously calculated values.
Round-off results to three significant figures.
2.1 Summary Table
Prepare a summary table listing the radon concentration and flow rate for
each of the six sampling occasions at each vent of the mine. An example of
such a table is shown in Figure 8. Reserve the final column for the results
of emission rate calculations.

Figure 8. Sample Summary Table

Summary Table
Operator:_____ Mine Identification:_____
Quarterly Interval:___4/84 - 7/84___

Vent	Sampling Date/Time	Vent/Flow (1/hr)	^{222}Concentration (pCi/1)	^{222}Rn Emission rate (1/hr)
1	6-3-84/08:40	8.49×10^7	528	
1	6-4-84/11:25	8.62×10^7	541	
1	6-3-84/14:25	8.52×10^7	823	
1	6-4-84/16:15	8.47×10^7	613	
1	6-3-84/18:30	8.53×10^7	548	
1	6-3-84/21:10	8.41×10^7	534	
2	6-4-84/09:20	5.13×10^7	1560	
2	6-3-84/11:35	5.21×10^7	1610	

2.2 Emission Rate Calculation. The emission rate for any sampling of a
mine vent should be calculated using the following equation:

$$E_{vt} = 10^{-12} (C_{vt} F_{vt})$$

where:
   E_{vt} = the hourly emission rate of ^{222}Rn from vent v at sampling time t,
       Ci/hr
   C_{vt} = the measured average ^{222}Rn concentration in air from vent v at
       sampling time t, pCi/liter

F_{vt} = the measured average volumetric stack gas/low rate for vent v at sampling time t, liter/hr.

After calculation, emission rates should be placed in the summary table.

2.3 Calculation of quarterly average vent emission rate. The quarterly average emission rate for each mine vent is the arithmetic mean of the six measurements taken at that vent (all taken within a 30 day interval).

$$\bar{E}_v = 1/6 \sum_{t=1}^{6} E_{vt}$$

where:
E_v = the quarterly average emission rate of vent V, li/hr,
E_{vt} = the emission rate of vent V at the tth sampling, Ci/hr.

2.4 Calculation of quarterly average mine emission rate. The total emission rate for a mine is the sum of the average emission rates determined for all exhaust vents for that quarter. The emission rate is computed as follows:

$$\bar{E}_{mq} = \sum_{v=1}^{6} \bar{E}_v$$

where:
$\bar{E}_{mq}$ = the quarterly average ^{222}Rn emission rate for mine m, with n vents, Ci/hr.

2.4 Calculation of annual average mine emission rate. The annual average emission rate for a mine is the arithmetic mean of four quarterly average emission rate measurements taken at intervals of about three calendar months. The formula for calculating the annual average is as follows:

$$\bar{E}_{ma} = 1/4 \sum_{q=1}^{4} \bar{E}_{mq}$$

where:
$\bar{E}_{ma}$ = the annual average ^{222}Rn emission rate for mine m, Ci/hr.