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CALIBRATION OF A FISSION GAS MONITOR*

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Abstract—A method for calibrating beta-sensitive gas-monitoring equipment is presented which avoids precise mixing of concentrated active gases with diluent air. This method depends upon neutron activation of the argon in P-10 proportional counting gas and passage of the active gas successively through the monitor chamber and a proportional counter chamber. The latter serves for the determination of specific activity of the monitor gas. The method is satisfactory for specific activities corresponding approximately to maximum permissible concentration (MPC) levels.

INTRODUCTION

In monitoring for gross airborne activities, the separate determination of particulate and gaseous activities is frequently desirable. The particulates can be determined by conventional filter techniques, which in conjunction with suitable detectors serve for the separate determination of gross alpha, beta and gamma activities. In most situations, true gaseous alpha contaminants are not of interest. Because of these considerations, the calibration method discussed herein is limited to gaseous beta-gamma activities. The monitor chambers discussed employ a chamber volume of roughly 1 ft^3 and thin window GM or scintillation detectors mounted internally. The calibration method is not limited to such arrangements however. These detectors were selected because their response reflects disintegrations rather than emitted energy.

Most gaseous activities, such as mixed fission gases and their daughters, emit both beta and gamma radiation, but not in any fixed ratio for all isotopes. These contaminants are so complex and time dependent that it is seldom feasible to identify the separate activities and their relative contribution to the total air contamination. An estimate of their total contribution must suffice in most instances. The concomitant gamma radiation is customarily ignored because of the generally lower detection efficiencies involved plus the fact that beta emission usually accompanies disintegration.

In principle, the response of monitoring systems can be estimated analytically on the basis of geometrical and electrical considerations, detection efficiencies, average beta absorption, etc.§[1,2] In most practical situations, however, the required corrections and assumptions are such that some sort of calibration is necessary in order to obtain a reasonably good estimate of monitor response. This calibration has to be a compromise in view of the range of beta energies exhibited by the fission gases. The use of argon-41 for this purpose seems to be as satisfactory a choice as any other.

The low-energy betas originating in the more remote parts of the monitor chamber are less likely to impinge on the detector surface and produce a count than are those of greater energy. This difficulty can be minimized by reducing the physical dimensions of the monitor chamber but this also reduces the amount of activity which can actuate the detector. As a consequence the chamber has to be of reasonable size in order to

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§ While intended for gamma radiation some geometric relations shown could be used for beta radiation, with proper assumptions.

get statistically significant results at the low MPC levels of interest. Beta particles having energies in excess of 0.2 MeV can penetrate a total air distance of about 1 ft. A portion of those betas having lower energies are also detected depending upon their point of origin in the chamber. On the basis of these qualitative considerations, the calibration factor should not show marked variation with energy for those beta activities having maximum energies of 0.6 MeV or more, inasmuch as such betas have average energies exceeding 0.2 MeV. Computed calibration factors for right cylinders[2] indicate that this conclusion is approximately valid. An inspection of a typical tabulation of the beta energies for the fission gases and their daughters reveals that many, but by no means all, show maximum energies in excess of 0.6 MeV.[3]

CALIBRATION METHOD

The usual method of calibrating air monitors depends upon the admixture of known amounts of some gaseous activity to a known volume of air with a later determination of the monitor response with this contaminated air. This approach permits the use of gaseous activities having known beta energies. Thus, any effects of beta energy upon the calibration can be readily studied within the limits imposed by the availability of suitable reference activities. This method is particularly desirable where a single contaminant is of interest in the monitoring system.

There are some disadvantages in using this method, however. These include:

(1) The specific activity of the added contaminant must be determined separately or else depend upon the supplier's evaluation. The time, cost and equipment required for this independent determination may be objectionable.

(2) The specific activity of the added contaminant may be well above MPC levels and thereby constitute some hazard if the concentrated contaminant is accidentally released.

(3) Some chemical processing to derive the active gas may be required except where active inert gases are employed.

(4) Precise volume measurements are required in the actual dilution. With high initial specific activities this may present a problem.

(5) Intimate mixing with the air volume is required.

(6) The contaminants generally have to be obtained offsite.

(7) Appreciable absorption of active contaminants on the container walls may occur for other than the inert gases.

The method herein suggested depends upon the thermal neutron activation of the argon in P-10 proportional counter gas (90 per cent argon, 10 per cent methane) such that the resulting specific activity lies in the range of interest. Some details for two activation schemes are included in the next section. This P-10 mixture is readily obtainable commercially and is usually available wherever proportional counters are in use. After activation, this gas is passed successively through an ordinary proportional counter chamber of known volume and the monitor chamber being calibrated. The monitor chamber can employ any of the detectors previously mentioned or it can be operated as an ionization chamber. The specific activity of the P-10 gas is determined simply and directly by the ratio of the net count rate of the proportional counter and the product of its volume and the dpm-μc constant. Thus the specific activity of the gas in the proportional counter (PC) is given by

$$\frac{\text{PC net counts/min}}{\text{PC volume in cm}^3 \times 2.2 \times 10^6\ \text{dpm}/\mu\text{c}}$$

The only assumption involved is that all beta disintegrations in the counter volume are recorded as counts irrespective of where these disintegrations occur therein. Obviously, this condition will not be altogether valid for those gas systems where any separation of daughter particulate activities or any adsorption of gaseous activities on the counter walls can occur. Any such separation effects will influence the gas monitor also.

Once this measure of specific activity and the corresponding monitor response has been obtained, the calibration factor for a particular monitor chamber is readily computed. This should be valid for all other specific activities, provided that the monitor counting system is not overloaded and the beta particles have the requisite energy. Overloading is unlikely at the activity levels of practical concern.

About 99 per cent of the argon-41 undergoes coincident beta and gamma decay, the maximum beta energy being 1.2 MeV and the photon energy being 1.3 MeV.[3] The remaining disintegrations yield a beta particle having a maximum energy of 2.48 MeV. Most of the betas, therefore, have sufficient energy to effect a monitor count for the suggested chamber sizes, irrespective of the point of origin in the monitor chamber. The gamma contribution to the monitor response should be small for a thin scintillator and is likely to be trivial in the case of a thin-window Geiger detector. As far as the proportional counter is concerned, the argon-41 acts essentially as a pure beta emitter. Its half-life (110 min) is short enough to permit calibrations over a fair range of activities if this is desired.

Obviously the specific activity in both the monitor and the proportional counter chambers must be the same in order to obtain a proper calibration constant. Ordinarily, there is considerable disparity in size between the proportional and the gas monitor chambers, these being about 60 and 14,000 ml in the present instance. The greater this disparity, the greater the uniformity in specific activity must be in order to have a representative sample in the proportional chamber. Reversing the flow direction through the proportional and monitor chambers provides a good check on the uniformity of the specific activity in each. Prior flushing with inert P-10 gas is advisable for complete air removal.

Another satisfactory procedure which requires considerably less total activity is to first fill both chambers with inert P-10 gas. The active P-10 gas is then added to the monitor chamber such that its gas pressure is somewhat above atmospheric. After thorough mixing (e.g. with an internal fan) some of this active mixture is bled through the proportional counter chamber for flushing purposes. The calibration is then made as before. A suitable correction for the higher pressure in the monitor chamber can be made if desired.

While the chief virtue of this method lies in its simplicity, there are some disadvantages:

(1) The calibration is limited to argon-41 activity. Hence, the magnitude of beta energy effects must be separately evaluated analytically or by adding other gaseous activities.

(2) The P-10 gas density is greater than that of air in the ratio of about 38 : 29. The linear beta absorption coefficient in P-10 gas is thus greater than for air.

These are not believed to be serious objections with respect to monitoring the inert fission gases and their daughters as air contaminants. As already indicated, the exact nature and composition of these activities will be unknown. The complexities in mode of decay and rate of decay of such activity mixtures precludes an exact specific activity evaluation by any type of gas monitor. As a consequence, corrections for monitor response at various beta energies are unwarranted. Furthermore the proper MPC values are just as uncertain, thus necessitating the use of mixture contamination limits.

By the same token, correcting the calibration constant for the difference in density between P-10 gas and air is also of questionable value. This is especially true in view of the fact that this constant, as obtained by the calibration method suggested above, necessarily gives conservative values for air activity. Owing to the smaller beta absorption coefficient for air, a given activity level in air must give a larger monitor response than will the same level in P-10 gas.

APPARATUS AND PROCEDURE

Diagrams of the apparatus used in two typical calibrations are presented in Fig. 1(a) and (b). The chamber and detector details are indicated for each case. Both chambers have an internal volume of roughly 0.5 ft^3. The steel walls of the cylindrical chamber are 3 in. thick. The resulting background with the 4 Geiger detectors was about 70 counts/min. The chamber is provided with a small internal fan to achieve adequate mixing. The spherical chamber has a thin sheet-aluminium wall. Its background, for the particular detector adjustments used, was about 145 counts/min. Since no provision for internal mixing was made, it was necessary to depend upon adequate flushing with active P-10 gas. Both chambers were well flushed with inert P-10 gas before calibration.

For the cylindrical monitor a PoBe source having an emission rate of about 4×10^6 neutrons/sec was used to activate the P-10 gas. Polyethylene tubing 0.25 in. o.d. wound on a thimble to give a coil length of about 8 in. and a

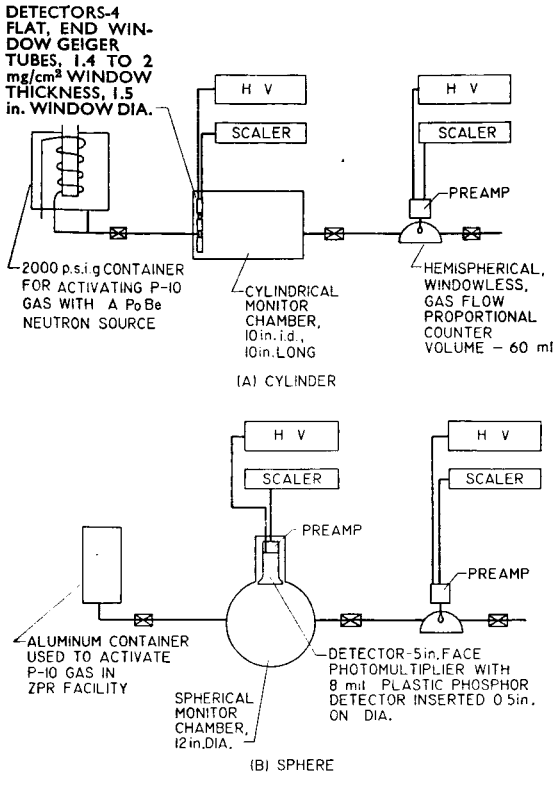

FIG. 1. Schematic diagram showing calibration apparatus.

thickness of about 3 in. was used as the moderator. This coil was placed inside a container designed for 2000 psig. P-10 gas to be activated was admitted at 500 psig, both to the interior and exterior sides of this polythylene coil. The PoBe source was placed in the thimble and left there for several half-lives. After activation, the gas in the tubing and some of that outside, was passed into the monitor chamber to give a final pressure somewhat above atmospheric. After mixing, this active P-10 mixture was bled through the proportional counter chamber and its specific activity determined.

In the case of the spherical chamber, the P-10 gas was activated in an aluminium tank 4 in. in diameter and 16.5 in. long at about 50 psig, using the thermal flux (about 10^7 neutrons/(cm²)(sec)) available on the exterior of the NASA Zero Power Homogeneous Reactor Facility. Higher specific activities were attainable by this method.

RESULTS AND DISCUSSION

Calibration data for the cylindrical and spherical monitor chambers are presented in Figs 2 and 3, respectively.

In order for the monitor response to be exactly proportional to the specific activity at all activity levels in the monitor chamber, all calibration points when plotted on rectilinear paper

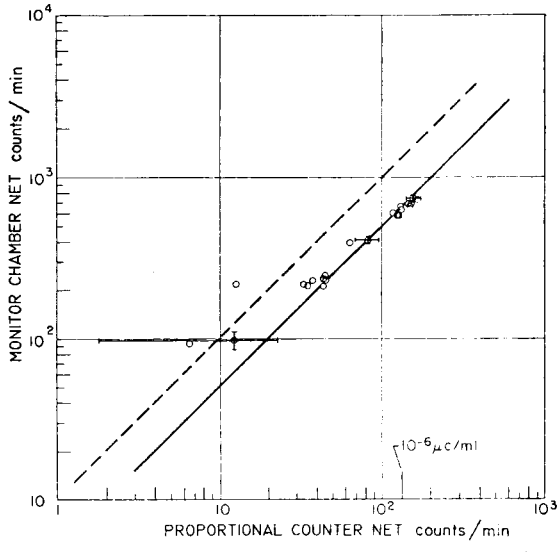

FIG. 2. Comparison of proportional counter and cylindrical gas monitor count rates

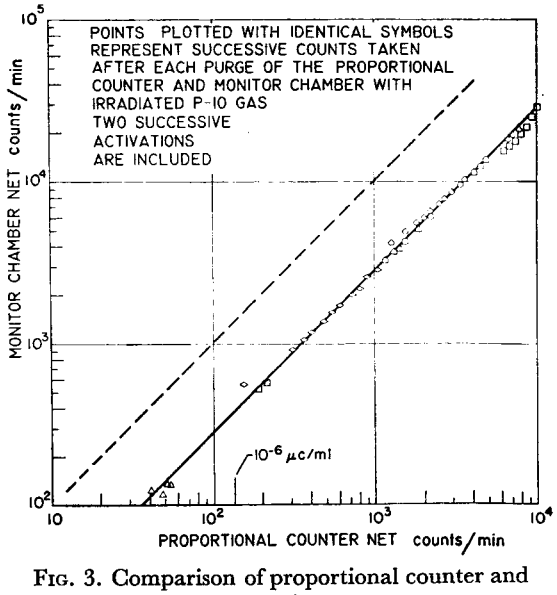

FIG. 3. Comparison of proportional counter and spherical gas monitor count rates.

must lie on a straight line which passes through the origin. In other words,

Monitor response in net counts/min

$$= k \times \text{specific activity}$$

$$= k \times \frac{\text{PC net counts/min}}{\text{PC volume} \times 2.2 \times 10^6 \text{ dpm}/\mu\text{c}}$$

where k is the desired calibration constant.

In Figs 2 and 3, the logarithm of the monitor net count rate is plotted against the logarithm of the proportional counter net count rate using log-log paper. The equation for this line is accordingly

$$\log(\text{monitor counts/min}) = \log(\text{PC counts/min}) + \log k - \log(\text{PC volume}) - \log(2.2 \times 10^6)$$

The last three terms on the right side of this equation are constants for a given calibration system. Only one of these, namely $\log k$, is determined by the monitor chamber and its detector. As k varies from one monitor system to another, the lines representing the calibration data should properly lie parallel to one another and to the dotted line through the even decade points. This property of log-log plots is of some service in drawing the best calibration line.

The straight lines through the calibration data points in Figs 2 and 3 are drawn in accordance with this principle of parallelism. Except for three points plotted for low activities in Fig. 2, most of the experimental data in both figures agree quite well with the calibration lines shown.

The actual error magnitudes at the 95 per cent level are shown at three activity levels in Fig. 2. Except at the very low activity levels, most of the points lie within the expected deviations from the calibration line selected for representing the experimental data.

The proportional counter net count rate corresponding to an activity level of 10^{-6} μc/ml is indicated in Figs 2 and 3 for orientation purposes. The minimum practical activity level for which either monitor is usable may be assumed to be that at which their net count rate equals their respective background count rate. In the case of the cylindrical monitor (Fig. 2) this limit is about 10^{-7} μc/ml. For the spherical monitor this limit is about 4×10^{-7} μc/ml. These do not necessarily represent the ultimate limits but rather those for which these particular systems are considered satisfactory.

The data plotted in Fig. 3 represent several additions of activated P-10 gas to the monitor chamber, some of which involved activation of separate batches of P-10 gas. The fact that all points lie so close to the calibration line must indicate that the added activity was well distributed throughout the sphere. If this were not so it is difficult to see how the activity in the proportional counter chamber could be so representative of that in the sphere in all cases. No special provision for internal mixing in the spherical chamber was employed in this case other than that provided by the incoming activated gas stream.

The validity of the assumption stated earlier that all disintegrations in the proportional counter chamber were counted has not been exhaustively investigated. The available literature has not been of much help in this respect. HOGREBE[4] does claim that the counting losses near the wall of a cylindrical chamber of about the same diameter are of the order of a few per cent. Some qualitative tests with a C-14 point source in a proportional chamber of about four times the diameter show that losses in excess of 10 per cent are unlikely. It is believed that the losses in the proportional counter chamber used in these calibrations were substantially less. Errors of this magnitude are certainly acceptable in terms of the intended use of these monitor chambers.

SUMMARY

Monitoring equipment for the measurement of gaseous activities in air ordinarily requires some sort of calibration. A simple method using conventional equipment is described, and some typical calibration data presented. The method depends upon the neutron activation of the argon in P-10 proportional counter gas and the determination of its specific activity in such a counter. The same gas is also passed through the monitor chamber and its response noted.

REFERENCES

1. V. V. BOCHKAREV and V. A. BAZHENOV, Some Methods of Measuring β-Active Gases. *Metrology of Radionucleides*, p. 327. International Atomic Energy Agency, Austria (1960).
2. T. ROCKWELL, III, *Reactor Shielding Design Manual*, Chap. 9. TID 7004 (1956).
3. D. STROMINGER, J. M. HOLLANDER and G. T. SEABORG, *Rev. Mod. Phys.* **30**, 2 (1958).
4. K. HOGREBE, The Absolute Measurement of the Radioactivity of Gases. *Metrology of Radionucleides*, p 301. International Atomic Energy Agency, Austria (1960).