Calibrating Germanium Detectors for Assaying Radioiodine in Charcoal Cartridges

Daniel M. Montgomery
Analytics, Inc.

Monitoring radioiodine in ambient air or gaseous effluent streams requires concentration of radioiodine from large volumes of air to achieve the sensitivity required by various regulatory agencies. This is accomplished by passing an air sample through a particulate filter followed by a charcoal cartridge. Since radioiodine may be present in various chemical forms including elemental iodine, organic iodide species, and other inorganic species, the charcoal cartridges are impregnated with KI or TEDA (tetraethylenediamine) to convert organic iodide species to forms that are collected on charcoal. In the presence of high noble gas activity (i.e., accident monitoring), a silver zeolite cartridge is recommended over a charcoal type cartridge since noble gases are not retained on silver zeolite but are highly retained on charcoal and would significantly lower the sensitivity for radioiodine.

The preferred method to analyze the filter and cartridge is to count them separately with a germanium gamma-ray spectrometer (counting the filter and cartridge together is discouraged). Then, the activities on the filter and cartridge are combined to give the total radioiodine activity. The contribution of particulate radioiodine is generally small; however, studies[1] have shown that particulate radioiodine may constitute as much as 40% of the total. The fraction of particulate radioiodine is variable and dependent on the source of radioiodine and other conditions such as pre-release treatment of the effluent stream (e.g., HEPA filter, charcoal train). Because of the low-activity levels, the filter and charcoal cartridge are normally counted in close proximity to the gamma-ray detector (1 cm or less). The cartridge is counted with the inlet side facing the detector—"face" counting. This practice minimizes the counting time necessary to achieve a given sensitivity (LLD); however, it may introduce significant errors in the measurement as will later be discussed.

Efficiency calibrations for filters are rather straight forward. Filter standards with an eight radionuclide mixture (^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113}Sn, ^{137}Cs, ^{88}Y, and ^{60}Co) covering the energy range from 88 keV to 1836 keV are commercially available or can be prepared from a standard solution of this mixture. It is important that the activity be uniformly distributed across the same active area as the samples to be counted. Significant coincidence summing may occur for ^{88}Y and ^{60}Co, and corrections may be necessary in the energy range from 898 keV to 1836 keV, depending on the detector efficiency and the proximity of the sample to the detector. The radionuclides ^{131}I, ^{133}I and ^{135}I, with the longest half-lives (8.0 d, 20.9 h and 6.7 h, respectively), emit strong gamma rays (at 364, 529 and 1260 keV, respectively) that are essentially free of coincidence summing even when the sample is counted near the detector housing. If the 1260-keV gamma ray from ^{135}I is used for quantification, coincident summing corrections for the standard may be necessary. However, an alternative would be to use the 420-keV gamma ray from ^{135}I which is outside the energy range where coincidence summing corrections of the standard are necessary.

Efficiency calibrations for charcoal cartridges are more complex since the distribution of iodine in the cartridge is dependent on many factors including: the iodine species, humidity, temperature, sample flow rate, and length of exposure. These factors can all affect the distribution and the penetration depth of the radioiodine in the cartridge. Charcoal cartridges can be counted directly on the detector, or the charcoal can be removed from the container, homogenized and counted.

The practice of removing the charcoal before counting has, in principle, the advantage of yielding a homogeneous sample. A calibration standard can be prepared by adding a known quantity of mixed gamma-ray standard to the appropriate mass of charcoal and thoroughly mixing to ensure homogeneity. Disadvantages of this technique include: a lower counting efficiency than when "face" counted; a longer analysis time due to disassembly of the cartridge and mixing of the charcoal in another container, and possible sample loss during transfer. Due to these problems, most nuclear power plant counting rooms do not use this technique.

Based on my experience with the Nuclear Regulatory Commission (NRC) and as a consultant to the nuclear power industry, the most common practice for counting charcoal cartridges is to "face" count the sample within a few centimeters of the detector. The preparation of an appropriate standard requires some knowledge of the radioiodine distribution in the charcoal cartridge samples. Based on the discussion above, the distribution may be quite dependent on the particular monitoring situation (i.e., environmental monitoring, long term stack monitoring, grab samples for respiratory protection purposes), and environmental conditions.

This paper focuses on ^{131}I; however, other short-lived radioiodine species may be present in effluent samples. These include: ^{132}I, $t_{1/2}$ = 2.3 hours; ^{133}I, $t_{1/2}$ = 20.9 hours; ^{134}I, $t_{1/2}$ = 50.2 minutes, and ^{135}I, $t_{1/2}$ = 6.7 hours. In most nuclear power plants the concentrations of short-lived radioiodines in gaseous effluents are low due to the holdup time associated with the effluent treatment systems. However, if short-lived radioiodine radionuclides are detected, NRC requires quantification and reporting of these radionuclides. Therefore, it is wise to calibrate the detectors over a wide enough energy range to permit analysis of all the iodine radionuclides (i.e., up to 1300 keV). Additionally, the software utilized by most commercially available gamma-ray spectroscopy systems require 8-10 efficiency/energy pairs at appropriate energies to accurately define an efficiency equation over the energy range from 88 to 1836 keV.

One method of preparing charcoal and silver zeolite cartridge standards involves disassembling the cartridge and gravimetrically adding a known quantity of the eight radionuclide mixture (as described above) to a portion of the charcoal. The amount of spiked charcoal is based on the depth of spiked charcoal desired when reloaded into the cartridge. The spiked charcoal is homogenized and placed back in the cartridge with a barrier between the active and inactive layers to prevent mixing. The active layer can be varied from approximately 5 mm to the total active depth of the cartridge (homogeneous).

Most laboratories use a "face" loaded cartridge—which means that the activity is loaded preferentially on the inlet or front "face" of the cartridge. Experience has shown that a 5-mm "face" loaded standard, counted with the inlet or "face" side toward the detector cryostat for the entire counting period, will generally provide an adequate efficiency calibration for charcoal cartridges. However, a better alternative is to count the standards (and samples) with the inlet side down ("face" side) for half of the counting time and then with the inlet side up ("flip" side) for the remainder of the counting time. The spectrum is collected over the entire counting interval. This will give results, for both standards and samples, that are less sensitive to the distribution of iodine.

The latter part of this article provides experimental data on the effect of radioiodine distribution on detector efficiencies for counting with inlet side down ("face" counting) and counting by the "flip" technique. Detector efficiencies may vary considerably for different models and types of cartridges. Calibrations are generally valid only for the specific type or brand of cartridge used for the calibration.

	3 mm from detector			3 cm from detector			6 cm from detector		
		Efficiency, %			Efficiency, %			Efficiency, %	
^{131}I Load	FB Ratio	Face	Flip	FB Ratio	Face	Flip	FB Ratio	Face	Flip
0-mm	2.82	4.08	2.76	2.08	1.34	1.00	1.75	0.61	0.48
2-mm	2.55	3.90	2.72	1.92	1.28	0.97	1.68	0.58	0.46
5-mm	2.25	3.59	2.59	1.73	1.23	0.97	1.56	0.56	0.46
10-mm	1.66	3.00	2.42	1.44	1.10	0.93	1.32	0.51	0.45
15-mm	1.33	2.73	2.38	1.22	1.01	0.92	1.20	0.49	0.45
25-mm	1.04	2.45	2.41	1.02	0.95	0.92	1.00	0.46	0.46

Flip efficiency refers to counting efficiency when the source is counted with the inlet side facing the detector for half of the counting period and the inlet facing away for half of the counting period.

Face efficiency refers to counting efficiency when the cartridge is counted with the inlet side facing the detector.

FB is the ratio of activity observed counting with the inlet side facing the detector to the activity observed when counting the inlet side facing away from the detector.

Table 1 Measured front-to-back ratios and counting efficiencies.

There are several unpublished studies[2,3] investigating the distribution of radioiodine in charcoal cartridges under actual monitoring conditions. Investigations by Dale Nix[2] at TVA's Browns Ferry Nuclear Plant showed that radioiodine was found within the front 5 mm and the average depth was about 2 mm. These studies included various monitoring locations in the plant and included cartridges that were in place for up to one week. Investigations by McFarland[3] at a radioiodine processing facility showed that greater than 95% of the radioiodine in charcoal cartridge samples was within the front 5 mm. These samples were collected in ambient air over a period of 2-3 days. Spiking the first 5 mm on the inlet side ("face") of a charcoal cartridge with the radionuclide standard, therefore, provides a reasonable approximation of the expected distribution (and activity gradient continuously decreasing from the front "face" to the back).

Loading effects on the measured counting efficiency for radioiodine were investigated by spiking charcoal cartridges with ^{131}I at various depths and counting at three different distances from a germanium detector. Commercially available TEDA impregnated charcoal cartridges, with approximate dimensions of 5.7-cm wide by 2.54-cm thick, were used in this work. The thickness of the radioiodine distribution corresponded to 0-mm, 3-mm, 5-mm, 10-mm, 15-mm, and 25-mm (homogeneous). All samples were prepared gravimetrically from ^{131}I solutions calibrated to within ± 3%. The 0-mm depth was prepared by distributing the activity on a filter using a pattern of drops over the active area to approximate a uniform distribution. The filter was placed in the cartridge, in front of the charcoal bed on the inlet side ("face"). Tests of the 0-mm cartridge showed that ^{131}I did not volatilize or redistribute during the test period of approximately 2 days. Other thicknesses of ^{131}I spiked charcoal were prepared by adding a known amount of ^{131}I activity to a predetermined amount of charcoal, homogeneously mixing it, and replacing it in the inlet side ("face") of the cartridge to provide the desired depth. A barrier was inserted to separate the ^{131}I active from the inactive charcoal. The remainder of the inactive charcoal was replaced and the cartridge resealed.

The counting system consisted of a 30-cm^3 germanium detector interfaced to a computerized multichannel analyzer system. After each count, the net peak area and efficiency for the ^{131}I, 364.8-keV gamma ray were determined. Each cartridge standard was counted with the "face" down for 1000 seconds and processed to determine the net counts and efficiency. The cartridges were then turned over with the "face" side up ("flipped") and counted for an additional 1000 seconds by resetting the ADC live time to 2000 seconds, without erasing the spectrum. The cumulative count was then processed to give the net

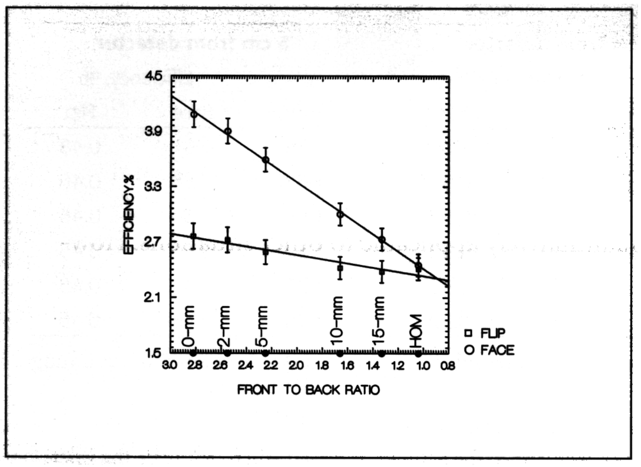

Figure 1 Efficiencies vs front-to-back ratios for "flip" and "face" counting 3 mm from detector.

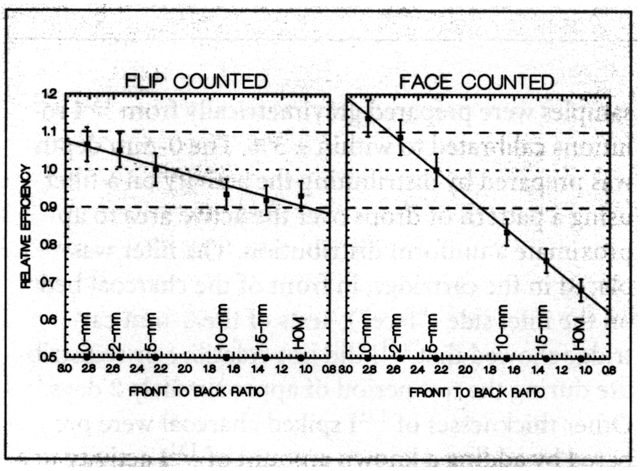

Figure 2 Relative efficiencies vs front-to-back ratios for "flip" and "face" counting 3 mm from detector.

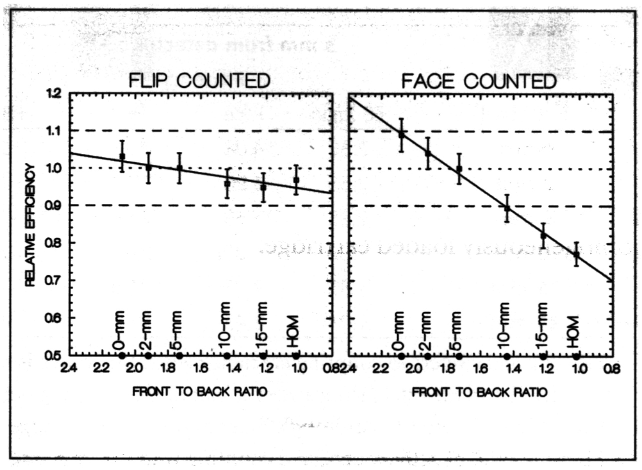

Figure 3 Relative efficiencies vs front-to-back ratios for "flip" and "face" counting 3 cm from detector.

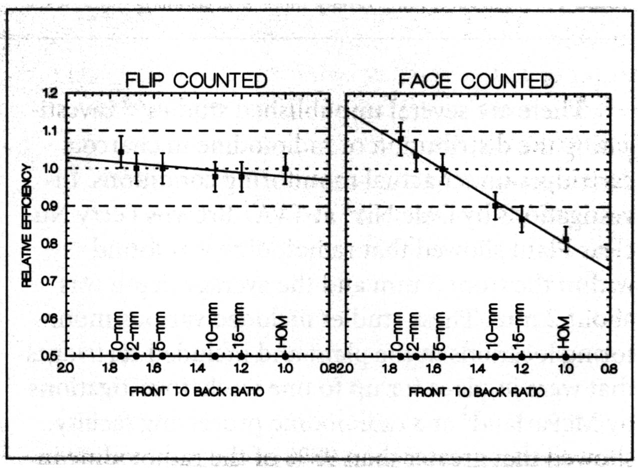

Figure 4 Relative efficiencies vs front-to-back ratios for "flip" and "face" counting 6 cm from detector.

counting rate and efficiency for the 364.8-keV gamma ray. Front-to-back ratios were determined from the following formula:

$$FB = \frac{FC}{TC - FC}$$

where FB = front-to-back ratio, FC = counts of the 364.8-keV gamma ray from counting with "face" down, and TC is the total counts of the 364.8-keV gamma ray from the "face" and "flip" counts.

The front-to-back ratio represents the ratio of the counting rate for a cartridge counted with the "face" side down, to the counting rate for a cartridge counted with the "face" side up ("flipped"). This ratio correlates with the distribution of radioiodine in the cartridge with the highest value of FB for "face" loaded cartridges. The value of FB decreases as the depth of loading increases and approaches unity for homogeneously loaded cartridges (the ratio may not be exactly one, since many cartridges are not manufactured so that the front and back are precisely the same).

The measured front-to-back ratios and counting efficiencies are presented in Table 1. Counting efficiencies for cartridges loaded at various depths were plotted against the front-to-back ratios. These results are presented in Figure 1, for a source to detector distance of 3.0 mm. As expected, counting cartridges "face" down in close proximity to the detector (Figure 1) is very sensitive to loading. The 0-mm loaded cartridge has an efficiency that is 1.70 times that of a homogeneously loaded cartridge.

The effects of the loading at source-to-detector distances of 3 mm, 3 cm and 6 cm, for both "flip" and "face" counting, are shown in Figures 2-4, where relative efficiencies are plotted against the front-to-back ratios. The counting efficiencies were normalized to 1.00 for cartridge efficiencies with a 5-mm thick loading. This assumes that the cartridge efficiency, with the 5-mm thick loading, is most representative of the expected loading for actual charcoal cartridge samples. Upper and lower limits were drawn at relative efficiencies 1.10 and 0.90 to represent an allowable deviation limit of plus and minus 10% from the 5-mm value. The value of 10% is somewhat arbitrary; however, a 10% systematic error would still allow one to meet the acceptance criteria used by the U.S. NRC's Confirmatory Measurements Program. The most restrictive NRC criteria would allow a measurement to deviate up to 15% from the NRC measured value.

The problems associated with counting samples very close to the detector are illustrated in Figure 2. Although all points are within the limits when "flip" counted, only the 2-mm cartridge would be within the limits when "face" counted. Careful comparison of Figures 2, 3 and 4 show that the deviation from the 5-mm cartridge efficiency decreases as the source-to-detector distance increases. At a source-to-detector distance of 3 cm, all points are within 5% for "flip" counting and 3 values are outside of the 10% limits for "face" counting. At a source-to-detector distance of 6 cm, all points are within 5% of the 5-mm value for "flip" counting, and all points except for the 15-mm and homogeneously loaded cartridges are within the 10% limits for "face" counting.

As demonstrated by the above measurements, potential errors associated with a variable distribution of radioiodine in cartridge samples, can be minimized by using the "flip" method of counting even at a sample-to-detector distance of 3 mm.

Because the data presented in this paper are dependent on many factors such as the detector dimensions, the type of charcoal cartridge used, and sampling conditions, these results may not be quantitatively applicable to other situations. However, these studies demonstrate the value of the "flip" counting method for a variety of conditions. Site specific studies may be conducted to determine the distribution of radioiodine in samples collected under actual operating conditions using the techniques presented in this article. The most appropriate loading for a calibration standard can then be selected.

If a high degree of accuracy is desired or is necessary, a series of efficiency curves or tables can be determined for different distributions (i.e., 5-mm-, 10-mm-, 15-mm-loaded cartridges) using mixed gamma-ray standards. The ratio of front-to-back activity for radioiodine would be calculated for each sample and then the appropriate efficiency curve or table would be utilized for the analysis.

When "face" counting of cartridges is practiced, it may be a good idea to periodically measure the ratio of front-to-back activity in charcoal cartridge samples to ensure that calibrations and counting conditions are appropriate to meet the desired measurement accuracy.

Acknowledgements

The author thanks Estie W. Belvin for her assistance in preparation of the charcoal cartridge standards and John McCorvey for counting the standards.

References

1. P.G. Voillequé, et al.,"Evaluation of the Air-Vegetation-Milk Pathway for ^{131}I at the Quad Cities Nuclear Power Station", *U.S. Nuclear Regulatory Commission Report* NUREG/CR-1600, November 1981.
2. Personal Communication (1990), D.N. Nix, Tennessee Valley Authority, Sequoyah Nuclear Power Plant, P.O. Box 2000, Soddy-Daisy, TN 37379.
3. Personal Communication (1988), R.C. McFarland, Analytics, Inc., 1380 Seaboard Industrial Blvd., Atlanta, GA 30318.

R&R