RP710[a]

Laboratory Method for Gross Alpha and Beta Activity Determination

1.0 Scope and Application

This method is used to rapidly screen a variety of matrices for both high and low activities of alpha and beta emitting radionuclides in waters, air filters, soils, sludges, waste waters, and solvents. The purpose of this method is three-fold: 1) to provide adequate information concerning the activity within samples, and thus determine if further, more detailed analyses are required, 2) to support accountability of radioactive material and ensure that a receiving laboratory's radioactive materials licenses or limits are not exceeded, and 3) to ensure that Department of Transportation regulations concerning the transport of radioactive materials have not been exceeded.

Gross screening analyses are not expected to be as accurate nor as precise as more detailed radiochemical separations. Rather, they are intended to provide rapid information associated with a particular action level with minimal chemical preparation. Additionally, these types of analyses are not intended to give "absolute" activity measurements, but rather "order-of-magnitude" estimates.

This method is intended to be complementary to other gross alpha/beta "rapid" screening methods, such as field methods or certain liquid scintillation methods. It is intended to provide more detailed information when samples are thought (or known) to be close to a given action level. In addition, this method does not produce any by-products (other than the sample itself), an advantage as opposed to liquid scintillation methods (i.e., scintillation cocktail).

2.0 Summary of Method

Samples are leached with acids, and organic matter is oxidized, if necessary. An aliquot of the sample or leachate is then evaporated to dryness on a stainless steel planchet and counted for alpha and/or beta radioactivity on a low-background gas-flow proportional counter. Calculated activities depend on sample mass and are based on calibration of specific isotopic standards.

(a) This method was consolidated by W. P. Brug (Los Alamos National Laboratory, Los Alamos, New Mexico). Appendixes 1 and 2 were prepared in collaboration with K. D. McCroan (National Air Radiation Environmental Laboratory, Montgomery, Alabama).

3.0 Interferences and Limitations

The problems discussed below should be recognized as inherent interferences and limitations of the method.

- High hygroscopic salt content in evaporated samples can cause the sample mass to fluctuate due to moisture absorption. To minimize this interference, the salts are converted to oxides by heating the sample intensely until it glows with a characteristic dull-red color.

- Self-absorption and scattering will occur when large mass, high density solid samples are analyzed for gross alpha activity.

- Volatile radioisotopes of C, ^{3}H, Tc, Po, Cs, and adsorbed Rn may be lost when samples are heated, especially to a dull-red heat.

- The crosstalk correction method for differentiating beta from alpha radiation may not adequately protect from misidentification for samples containing very low-energy alpha or high-energy beta energies.

- Moderate to high levels of gamma radiation can be misinterpreted as alpha or beta activity by gas-flow proportional counters; this interference can be minimized by using a guard detector and employing anti-coincident counting techniques.

- Proportional counters operated in the "windowed" mode are limited to detecting (beta) particles with an energy of 0.1 MeV or greater. To detect "soft" betas, the counter must be operated in the "windowless" mode.

4.0 Safety

Samples may contain a variety of hazards in addition to radioactivity and should be handled accordingly. Reagents and apparatus used in this method are standard for chemistry labs, and counting rooms and standard safety practices apply. For more information, one may refer to Chapter 4, Safety.

5.0 Apparatus and Materials

5.1 Sample Preparation

- Infrared heating lamp

- Hotplate

- Transfer pipettes

- Glass (Pyrex or equivalent) beakers

- Watch glasses to fit beakers

- Mylar film: 0.0064-mm (0.00025-in.) maximum thickness; or acrylic solution: 50-mg Lucite dissolved in 100-mL acetone, or other appropriate means to fix activity to planchet

- Stainless steel planchets

- Centrifuge and centrifuge tubes or filtering apparatus

5.2   Sample Counting

- Gas flow proportional counting system or scintillation detection system

- Guard detector operated in the anti-coincidence mode (usually a part of a typical proportional counter)

- Ionizing gas (counter specific, P-10 for example)

6.0   Reagents

- Nitric acid (conc, 70%)

- Nitric acid (12 M) • hydrochloric acid (1 M), store in a polyethylene bottle

- Nitric acid (8 M)

- Hydrochloric acid (conc)

- Hydrogen peroxide (30%)

7.0   Sample Collection, Preservation, and Storage

Aqueous samples should be acidified with HNO_3 so that the pH is less than 2. Adding the acid precludes the formation of precipitates and thus analyte loss. In addition, lowering the pH will minimize the adsorption of radionuclides on container walls by creating relatively non-polar surfaces.

No preservation is necessary for soil, sludge, air filter, or organic samples before analysis.

8.0   Procedure

Aliquot size. A general guideline for the aliquot size is that it should be chosen such that the coverage density (g/cm^2) on the planchets does not exceed 5 mg/cm^2 for gross alpha determinations and 10 mg/cm^2 for gross beta determinations.

Sample drying. The sample should be heated carefully so spattering does not occur. To avoid spattering, samples are first heated with a low temperature, and then the temperature is slowly ramped upwards. If the mass of the sample fluctuates or appreciable salt content is expected, the mass should be stabilized by heating the sample until it glows with a characteristic dull red color, which converts the hygroscopic salts into their respective oxide forms.

Fixing activity to planchet. Some sample residues contain particles that can become airborne and thus need to be fixed to the planchet to prevent sample cross-contamination and/or detector contamination (especially when counting in the "windowless" mode). Suggestions include covering the sample with a thin (0.0064-mm thickness) mylar film, or treating it with an acrylic solution (e.g., a few drops of 50-mg Lucite dissolved in 100 mL acetone). Whatever technique is employed (if any), the efficiency curves and crosstalk curves generated should be based on the sample configuration.

8.1   Waters

The waters analysis method presented here is intended to be equivalent to other standard methods {SW-846 Method 9310 (EPA 1986); EPA Method 900.0 (Krieger and Whittaker 1980); *Standard Methods for the Examination of Waters and Wastewaters* Method 7110 (Clesceri et al. 1992)}.

8.1.1   If the sample is known or suspected to contain significant chloride (acid or salts), the chloride should be converted to nitrate before transferring to a stainless steel planchet. *(Chlorides will attack the stainless steel, depositing heavy metal ions on top of the sample, thereby increasing inelastic scattering. Subsequently, no correction can be made for this effect.)* Chloride salts can be converted to nitrates by adding 5 mL 16 M HNO$_3$ and evaporating to near dryness (chloride ions will be oxidized to gaseous chlorine). One repeat of this step is usually sufficient.

8.1.2   A sample should be evaporated directly on a tared counting planchet, or for larger samples, it should be evaporated in a beaker and transferred quantitatively to a counting planchet, using 8 M HNO$_3$ to wash down beaker walls.

8.1.3 The sample is dried by heating on a hot plate or under an infrared lamp. If hygroscopic salts are present, the sample should be heated until it glows with a characteristic dull red color to stabilize the mass.

8.1.4 The dried planchet is weighed, and the net residue mass is determined.

8.1.5 The sample is fixed to the planchet, if necessary, and counted for an appropriate time.

8.2 **Sludge**

8.2.1 An aliquot of sludge is placed into a tared beaker, and the wet weight is determined if required. The sample is dried (under an infrared lamp or drying oven for example) and weighed to determine the dry weight.

8.2.2 Approximately 40 mL of 12 M HNO_3 • 1 M HCl are added to 1 g of dried sample (i.e., 0.5 g of dried sample requires approximately 20 mL of acid solution). The sample is covered with a watchglass and digested on a hotplate for about 20 min.

8.2.3 The sample is removed from the hotplate, and the leachate is separated by centrifuging, filtering, or decanting.

8.2.4 Steps 8.2.2 and 8.2.3 are repeated. The resultant leachates are combined.

8.2.5 Approximately 20 mL of 8 M HNO_3 per 1 g of dried sample are added to the remaining residue in the beaker. The sample is covered with a watchglass and digested on a hotplate for an appropriate time (i.e., roughly 10 min).

8.2.6 After cooling, the leachate is separated by centrifuging, filtering, or decanting and is combined with the previously collected leachates and brought to a known volume with 8 M HNO_3.

8.2.7 An aliquot of the leachate is dried onto a tared stainless steel planchet. The sample is heated until it glows with a characteristic dull red color to stabilize the mass, if necessary, and the sample is cooled. The residual weight is recorded.

8.2.8 The sample is fixed to the planchet, if necessary, and counted for an appropriate time.

8.3 **Organics**

This section is applicable to all organic samples that can be evaporated to dryness by simple convective heating; other samples, such as high boiling point organics and oils, should be determined by other methods.

8.3.1 An aliquot of the sample (e.g., 5 mL) is pipetted onto a tared stainless steel planchet.

8.3.2 The sample is dried by heating on a hot plate or under an infrared lamp. If hygroscopic salts are present, the sample is heated until it glows with a characteristic dull red color to stabilize the mass.

8.3.3 The dried planchet is weighed, and the net residue mass is determined.

8.3.4 The sample is fixed to the planchet, if necessary, and counted for the appropriate time.

8.4 **Air Filters**

8.4.1 The filters are mounted directly on a planchet (no chemical processing) with the loaded face of the filter exposed.

8.4.2 The sample is counted for an appropriate time.

8.5 **Soils**

8.5.1 An aliquot (e.g., 2 g) of homogenized soil is placed into an appropriately sized tared beaker, and the weight is determined. The soil may be dried (under a heat lamp for example) and the dry weight determined.

8.5.2 The sample is covered with conc HNO_3, and 3 to 5 drops (per 1 g of dried sample) of conc HCl are added to the beaker. The sample is swirled carefully and allowed an appropriate amount of time (generally overnight) to leach at room temperature. Alternatively, the sample can be heated with stirring to reduce leaching time to about 30 min.

8.5.3 The sample is heated until the yellow-orange NO_2 fumes no longer appear, indicating HCl is no longer present.

8.5.4   The soil residue is separated from the leachate (e.g., by centrifuging, filtration, etc.). The soil residue is rinsed with (~ 5mL) 8 M HNO_3, adding the rinse to the leachate.

8.5.5   The leachate is either quantitatively transferred to a tared planchet, or it is brought to a known volume, and an aliquot is transferred to a tared planchet.

8.5.6   The solution is evaporated on the planchet to dryness under a heat lamp. The heating rate is controlled so that the sample does not spatter.

8.5.7   The planchet is allowed to cool; then it is reweighed, and the weight is recorded.

8.5.8   The activity is fixed to the planchet, if necessary, and counted for an appropriate time.

8.6   **Sewage, Waste Water, and Aqueous Slurries**

8.6.1   An appropriate aliquot of the sample is poured into a tared beaker, reweighed, and the wet weight is determined.

8.6.2   The sample is evaporated to dryness and re-weighed. The dry weight is then determined.

8.6.3   The sample is heated on a hotplate and carefully treated with conc HNO_3 and H_2O_2 until all organic material has been oxidized.

The sample should not have a black tarry consistency (due to remaining organics), but may still have a grayish or reddish appearance. If the sample is black or tarry, the wet ashing (as described in the previous step) should continue to complete the oxidation of organics.

8.6.4   Approximately 20 mL of conc HNO_3 and approximately 10 mL of conc HCl are added, and it is boiled until all HCl has been driven off (the yellow-orange NO_2 fumes will no longer appear). Continued addition of conc HNO_3 and evaporation may be necessary to eliminate Cl^- from the matrix.

8.6.5   Evaporation is continued until approximately 3 mL of solution remain.

8.6.6   Approximately 10 to 15 mL of 8 M HNO_3 are added to the sample.

8.6.7   Any residue is separated from the leachate (e.g., by centrifuging, filtration, etc.). The residue is rinsed with 8 M HNO_3, adding the rinse to the leachate.

8.6.8   The leachate is either quantitatively transferred to a tared planchet, or it is brought to a known volume, and an aliquot is transferred to a tared planchet.

8.6.9   The solution is evaporated on the planchet to dryness under a heat lamp. The heating rate is controlled so that the sample does not spatter.

8.6.10   The planchet is allowed to cool, reweighed, and the weight is recorded.

8.6.11   The activity is fixed to the planchet, if necessary, and counted for an appropriate time.

8.7   **Calibration and Standards** *(A more in-depth discussion of efficiency and crosstalk determination is presented in Appendix 1.)*

To ensure accurate measurements, it is necessary to determine both detector efficiency and crosstalk as a function of sample mass. (Efficiency determination as a function of sample mass is equivalent to determining a zero-mass efficiency with an associated self-absorption factor.) While most modern detector systems allow the crosstalk to be minimized via manual discriminator (or window) adjustment, it is only at the expense of reduced detection efficiency. If instrumental parameters are adjusted to minimize or eliminate crosstalk, crosstalk curves still need to be generated to ensure that crosstalk factors are negligible throughout the entire analytical sample mass range. Finally, when the sample mass exceeds coverage density guidelines, or crosstalk is deemed significant, the curves should be determined to generate the requisite correction factors. The same data used to generate the efficiency curves can also be used to generate the crosstalk curves.

8.7.1   Calibration standards should be prepared from matrices similar to those to be measured, and the mode of fixing the activity to the planchet should also be the same as that used for the sample. Two tracer solutions are used: one from a measured amount of any National Institute of Standards and Technology (NIST) traceable alpha emitting secondary standard (for example ^{238}Pu/^{239}Pu) and the other from a measured amount of NIST traceable beta emitting standard (for example ^{90}Sr/^{90}Y). *Some laboratories have suggested using a pure alpha emitting standard, such as ^{210}Po, to accurately determine alpha to beta crosstalk. Other alpha standards also have gamma and x-ray emissions associated with them, which will be detected as beta counts, and thus overestimate the crosstalk factor. Other laboratories argue that using standards that contain x-rays etc. more accurately represent real world samples and thus better estimate detector performance. If a measured sample contains no interferences (x-rays etc.), then the resultant activity will be underestimated. The decision of which type of standard to be used is left up to the individual laboratory, but one must be aware of the limitations associated with the choice selected.*

The resultant solutions should have sufficient activity so that acceptable counting statistics from a standard mount can be achieved with a reasonable count time. (For example 10,000 total counts have an associated 1% counting error, and 100 counts have a 10% counting error.)

8.7.2   To generate the calibration curves (alpha efficiency, beta efficiency, alpha-to-beta crosstalk, and beta-to-alpha crosstalk) as a function of sample mass, two groups of matrix (e.g., soil) standards (one for alpha and one for beta) are prepared with the same amount of radioactive tracer while the amount of residue is varied so that the masses encompass the target sample masses.

8.7.3   The individual measured alpha and beta efficiencies are calculated as follows:

$$\varepsilon_x(m) = \frac{N_x}{D_x}$$

where

$\varepsilon_x(m)$ = measured efficiency for mass, m
x = α or β (dependent on the emission source)
N_x = measured net count rate in channel x
D_x = known disintegration rate for either an alpha or beta standard.

Additionally, the alpha to beta crosstalk factors are determined using the alpha standards, and the beta to alpha crosstalk factors are determined using the beta standards. These factors are calculated as follows:

$$X_{\alpha\beta}(m) = \frac{N_\beta}{N_\alpha}$$

$$X_{\beta\alpha}(m) = \frac{N_\alpha}{N_\beta}$$

where

$X_{\alpha\beta}(m)$ = alpha to beta crosstalk for mass m
$X_{\beta\alpha}(m)$ = beta to alpha crosstalk for mass m
N_α = measured net alpha counts
N_β = measured net beta counts.

8.7.4   Equations or graphical plots should be derived to describe all four calibration curves (note: these curves are generally not linear). This is usually done with the help of a computer or some plotting device. Therefore, the desired data for any arbitrary sample mass can be interpolated from any one of these curves.

9.0 **Calculations** *(A more complete and rigorous discussion of the calculations is provided in Appendix 2.)*

If crosstalk has been determined to be negligible, then all the references to $X_{\alpha\beta}$ and $X_{\beta\alpha}$ are equal to zero and thus can be disregarded. However, the crosstalk forms of the equations are the most general.

9.1 The crosstalk corrected net count rates are calculated by the following equations:

$$N_{X\alpha} = \left(\frac{C_\alpha}{t_{C_\alpha}} - \frac{B_\alpha}{t_{B_\alpha}}\right) - X_{\beta\alpha}(m)\left(\frac{C_\beta}{t_{C_\beta}} - \frac{B_\beta}{t_{B_\beta}}\right)$$

$$N_{X\beta} = \left(\frac{C_\beta}{t_{C_\beta}} - \frac{B_\beta}{t_{B_\beta}}\right) - X_{\alpha\beta}(m)\left(\frac{C_\alpha}{t_{C_\alpha}} - \frac{B_\alpha}{t_{B_\alpha}}\right)$$

where

$N_{X\alpha}$ and $N_{X\beta}$	=	crosstalk corrected net alpha and beta count rates, respectively
C_α and C_β	=	measured alpha and beta sample counts
$t_{C\alpha}$ and $t_{C\beta}$	=	alpha and beta sample count time
B_α and B_β	=	measured alpha and beta background counts
$t_{B\alpha}$ and $t_{B\beta}$	=	alpha and beta background count time
$X_{\alpha\beta}$ and $X_{\alpha\beta}$	=	alpha to beta crosstalk and beta to alpha crosstalk for a given mass, m.

9.2 The equations for the alpha activity, A_α, and the beta activity, A_β are

$$A_\alpha = \frac{N_{x\alpha}}{\varepsilon_{x\alpha}(m) \bullet Q} \bullet k$$

$$A_\beta = \frac{N_{x\beta}}{\varepsilon_{x\alpha}(m) \bullet Q} \bullet k$$

where

A_α	=	alpha activity
A_β	=	beta activity
$N_{x\alpha}$	=	crosstalk corrected net alpha count rate
$N_{x\beta}$	=	crosstalk corrected net beta count rate
k	=	factor to convert to the desired reporting units

Q $\quad=\quad$ sample size (volume or mass)

$\varepsilon_{X\alpha}(m) \quad=\quad$ crosstalk corrected alpha detector efficiency at mass m

$\varepsilon_{X\beta}(m) = \quad$ crosstalk corrected beta detector efficiency at mass m.

Note: the crosstalk corrected efficiencies are given by

$$\varepsilon_{X\alpha} = \varepsilon_\alpha(m) \cdot \left[1 - X_{\alpha\beta}(m)\, X_{\beta\alpha}(m)\right]$$

$$\varepsilon_{X\beta} = \varepsilon_\beta(m) \cdot \left[1 - X_{\alpha\beta}(m)\, X_{\beta\alpha}(m)\right]$$

9.3 The alpha and beta counting errors, $\sigma_{N_{X\alpha}}$ and $\sigma_{N_{X\beta}}$, are calculated as

$$\sigma_{N_{X\alpha}} = \sqrt{\left(\frac{C_\alpha}{t_{C_\alpha}^2} + \frac{B_\alpha}{t_{B_\alpha}^2}\right) + X_{\beta\alpha}^2(m)\left(\frac{C_\beta}{t_{C_\beta}^2} + \frac{B_\beta}{t_{B_\beta}^2}\right) + \sigma_{X_{\beta\alpha(m)}}^2 \left(\frac{B_\beta}{t_{B_\beta}} - \frac{C_\beta}{t_{C_\beta}}\right)^2}$$

$$\sigma_{N_{X\beta}} = \sqrt{\left(\frac{C_\beta}{t_{C_\beta}^2} + \frac{B_\beta}{t_{B_\beta}^2}\right) + X_{\alpha\beta}^2(m)\left(\frac{C_\alpha}{t_{C_\alpha}^2} + \frac{B_\alpha}{t_{B_\alpha}^2}\right) + \sigma_{X_{\alpha\beta(m)}}^2 \left(\frac{B_\alpha}{t_{B_\alpha}} - \frac{C_\alpha}{t_{C_\alpha}}\right)^2}$$

9.4 The total error or the uncertainty in the alpha and beta activity calculations, σ_{A_α} and σ_{A_β}, can be expressed by the following equations:

$$\sigma_{A_\alpha} = \sqrt{\left(\frac{\sigma_{N_{x\alpha}}}{k^{-1} \cdot \varepsilon_{x\alpha}(m) \cdot Q}\right)^2 + \left(\frac{A_\alpha \cdot \sigma_{\varepsilon_{x\alpha(m)}}}{\varepsilon_{x\alpha}(m)}\right)^2 + \left(\frac{A_\alpha \cdot \sigma_Q}{Q}\right)^2}$$

$$\sigma_{A_\beta} = \sqrt{\left(\frac{\sigma_{N_{x\beta}}}{k^{-1} \cdot \varepsilon_{x\beta}(m) \cdot Q}\right)^2 + \left(\frac{A_\beta \cdot \sigma_{\varepsilon_{x\beta(m)}}}{\varepsilon_{x\beta}(m)}\right)^2 + \left(\frac{A_\beta \cdot \sigma_Q}{Q}\right)^2}$$

where

$\sigma_{A_\alpha} \quad = \quad$ error in the alpha activity

$\sigma_{A_\beta} \quad = \quad$ error in the beta activity

$A_\alpha \quad = \quad$ measured alpha activity

$A_\beta \quad = \quad$ measured beta activity

$$\sigma_{\varepsilon X\alpha(m)} \quad = \quad \text{uncertainty in the crosstalk corrected alpha detector efficiency/self-absorption factor}$$

$$\sigma_{\varepsilon X\beta(m)} \quad = \quad \text{uncertainty in the crosstalk corrected beta detector efficiency/self-absorption factor}$$

$$\sigma_{Q} \quad = \quad \text{uncertainty in the sample quantity.}$$

10.0   Quality Control

Quality control (QC) and instrument verification vary greatly between laboratories, and this section is only meant to provide guidance. However, minimal QC guidelines (from EPA SW-846 Method 9310) are 1) all QC data should be maintained and made available for easy reference; 2) one method blank per sample batch should be employed; and 3) one sample duplicate should be run for every 10 samples. Control samples such as matrix spikes, blank spikes and/or performance evaluation samples should also be run periodically.

11.0   Method Performance

The following single laboratory based performance data measurements were made on aqueous, soil, and filter samples. The known analyte concentration levels are listed, along with the obtained precision and bias and associated number of replicate samples. No performance data are available for organic, sewage, or sludge matrices.

Gross Alpha Single Laboratory Performance Data

Matrix	Known Activity Range	Bias (%) Minimum	Maximum	Average	Median	Precision (%) (Std Dev of Bias)	Total Analyses
Water	0-0.37 Bq/L	-13.04	69.81	42.01	50.95	±28.16	10
	0.37-01.1 Bq/L	-46.67	26.05	-18.20	-26.67	±28.21	9
	1.9 kBq/L	-5.88	19.61	7.13	5.88	±5.91	24
Soil	0-0.19 Bq/g	-77.55	150.00	17.87	6.25	±78.82	8
	0.19-0.56 Bq/g	-34.21%	60.71	15.00	17.56	±31.93	10
Air Filter	0.78 Bq/filter	-19.05	0.00	-10.19	-9.52	±4.71%	50

Gross Beta Single Laboratory Performance Data

Matrix	Known Activity Range	Bias (%) Minimum	Maximum	Average	Median	Precision (%) (Std Dev of Bias)	Total Analyses
Water	0.19-1.9 Bq/L	-27.66	15.63	-6.33	-4.41	±13.68	14
	0.005-2.3 kBq/L	-10.32	8.06	1.85	1.617	±3.61	24
Soil	0-0.19 Bq/g	-17.65	93.02	29.58	18.37	±40.99	11
	0.19-0.56 Bq/g	-46.43	23.46	1.60	1.14	23.12	7
Air Filter	2.3 Bq/filter	-22.58	-16.31	-19.52	-19.35	±1.54	50

12.0   References

Clesceri, L. D., A. E. Greenberg, and R. R. Trussell, eds. 1992. "Method 7110, Gross Alpha and Gross Beta Radioactivity (Total, Suspended, and Dissolved)." *Standard Methods for the Examination of Water and Wastewaters*. American Public Health Association, 18th edition.

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13.0   Reviewer Modifications/Suggestions

A reviewer suggested that a modification of the sludge, air filters, and soils preparation be as follows:

> A single or double mixed acid digestion may be performed with 25 mL of 16 M HNO_3, 5 mL of 12 M HCl, and 2 to 3 mL of HF to moist salts. The solution is heated in 15 to 20 mL of 8 M HNO_3 to dissolve the contents, transferred to a centrifuge tube, centrifuged and decanted into a 100 mL volumetric flask, and diluted to 100 mL with deionized water. The sample is evaporated onto a tared stainless steel planchet.

Appendix 1

Self-Absorption and Crosstalk Curve Determinations

Alpha and Beta Self-Absorption/Efficiency Determinations

The presence of appreciable sample mass results in attenuation of alpha and beta particles and loss of counting efficiency. This phenomenon is more significant for alpha particles than for beta particles because of the more highly ionizing nature and consequent shorter path of alpha particles in matter. For example, a 3 MeV alpha particle only has a range of approximately 1.6 cm in air, while a 1 MeV electron will travel approximately 3 m.

The presence of additional sample mass can result in complete attenuation of the alpha particle (self-absorption with loss of counting efficiency) or alternatively, degradation of the alpha particle energy to the point that it deposits energy equivalent to a beta particle (cross-talk, again with loss of alpha particle counting efficiency). To compensate for these effects, the instrument efficiency and crosstalk for both alpha and beta particles must be determined as a function of sample mass.

Determination of alpha/beta self-absorption curves. The parameters that are required for the determination of alpha and beta activities are derived from curves that plot both the absolute detector efficiency and alpha/beta self-absorption as a function of the sample mass residue. Note: this method combines the absolute zero-mass detector efficiency, $\varepsilon(0)$, with a self-absorption factor, $A(m)$, to yield the detector efficiency/self-absorption factor, $\varepsilon(m)$.

$$\varepsilon(m) = \varepsilon(0) \cdot A(m)$$

This is equivalent to methods that determine these two factors separately since $\varepsilon(0)$ is a constant. From the preceding equation, a boundary condition must exist so that $A(0) = 1$, which gives the zero-mass or absolute efficiency of the instrument.

At 2.5 mg/cm^2 surface coverage (50 mg for a 2 in. planchet), a distinct inflection point occurs in the self-absorption curve, which makes it more viable to model this phenomena with two curves (one for 0 to 50 mg and the other for 50 mg and higher). The theoretical origins for this inflection point can be traced to the fact the 2.5 mg/cm^2 is roughly the surface coverage necessary to completely attenuate an alpha particle originating from the bottom of the planchet with a trajectory normal to the planchet surface.

Fitting Functions. While many fitting functions can be used, some of the more common equations are a) linear, b) exponential, and c) 1/x.

$$\text{a)} \qquad \varepsilon(m) = a \cdot m + b$$

$$\text{b)} \qquad \varepsilon(m) = a \cdot e^{-b \cdot m}$$

$$\text{c)} \qquad \varepsilon(m) = \frac{a}{2 \cdot m}$$

where

a, b	=	adjustable parameters
m	=	sample mass
ε(m)	=	detector efficiency/self-absorption factor

The parameters for the fitting function are generally found by least-squares regression. Note: each portion of the curve can be fitted with a unique function. The following example shows the low- mass portion of the curve being fitted by a linear function and the high-mass portion being fitted as 1/x.

$$\varepsilon(m) = \left. \begin{array}{ll} b - \dfrac{m}{2 \cdot a} & m \le 50 \text{ mg} \\[2em] \dfrac{a}{2 \cdot m} & m > 50 \text{ mg} \end{array} \right\}$$

Crosstalk

In simultaneous gross alpha/beta determinations, two types of counting efficiencies can be defined mathematically. First of all, the traditional counting efficiency, $\varepsilon_x(m)$, where x = α or β, which is simply the ratio of the measured counts in a particular channel to the known disintegration rate, is

$$\varepsilon_x(m) = \frac{N_x}{D_x}$$

where

$\varepsilon_x(m)$	=	measured "traditional" efficiency for mass, *m*
x	=	α or β (dependent on the emission source)
N_x	=	measured net count rate in channel x
D_x	=	known disintegration rate for either an alpha or beta standard.

The second type of detection efficiency is the detection of an emission event of one type in the other channel (i.e., an alpha particle is mistakenly counted in the beta channel or window). This "cross-efficiency" term is unique to simultaneous gross alpha/beta measurements. The

"efficiency" of mistakenly measuring an alpha particle in the beta channel, $\varepsilon_{\alpha\beta}(m)$, is very similar to the previous equation, except it is the ratio of the measured beta count rate to the known alpha disintegration source. Likewise, the beta-alpha efficiency, $\varepsilon_{\beta\alpha}(m)$, is the ratio of the measured alpha count rate to the known beta disintegration source. Mathematically, these two "cross-efficiencies" can be summarized as

$$\varepsilon_{xy}(m) = \frac{N_y}{D_x}$$

where

$$\begin{aligned}
\varepsilon_{xy}(m) &= \text{measured "cross" efficiency at mass, } m \\
x,y &= \alpha \text{ or } \beta \ (x \neq y) \\
N_y &= \text{measured net count rate in channel y} \\
D_x &= \text{known disintegration rate for either an alpha or beta standard.}
\end{aligned}$$

To simplify the calculation and correctly deduce the actual count rates (crosstalk corrected count rates) from the measured count rates, two crosstalk terms, $X_{\alpha\beta}(m)$ and $X_{\beta\alpha}(m)$, are defined as the ratio of the respective "cross-efficiency" to the "traditional efficiency."

$$X_{\alpha\beta}(m) = \frac{\varepsilon_{\alpha\beta}}{\varepsilon_\alpha} = \frac{N_\beta}{N_\alpha}$$

$$X_{\beta\alpha}(m) = \frac{\varepsilon_{\beta\alpha}}{\varepsilon_\beta} = \frac{N_\alpha}{N_\beta}$$

where

$$\begin{aligned}
X_{\alpha\beta}(m) &= \text{alpha to beta crosstalk for mass } m \\
X_{\beta\alpha}(m) &= \text{beta to alpha crosstalk for mass } m \\
N_\alpha &= \text{measured net alpha counts} \\
N_\beta &= \text{measured net beta counts.}
\end{aligned}$$

As can be seen from the previous set of equations, the crosstalk terms simplify to a ratio of net counts of the alpha and beta channels for a single measurement of a given disintegration source. Therefore, to experimentally measure the alpha to beta crosstalk for a given mass, $X_{\alpha\beta}(m)$, an alpha source with a standardized disintegration rate is prepared at the desired mass and counted for an appropriate amount of time. The crosstalk term is calculated simply by dividing the net measured beta counts by the net measured alpha counts. The beta to alpha crosstalk term, $X_{\beta\alpha}(m)$, can be determined in a similar fashion by using a beta standard. Note: since particle scattering should increase in a non-linear relationship with sample mass, crosstalk curves must be derived as a function of mass in a similar fashion to the self-absorption curves.

Typically, the counting system should be set up so that less than 1% of the beta events are counted in the alpha channel. This is accomplished by varying the discriminator window values and the operating voltage values. Note: not all systems provide this type of variability in instrument setup. In such cases, the system has been optimized by the manufacturer to minimize the alpha to beta crosstalk factor or to provide a correction factor within the data reduction algorithm.

Appendix 2

Derivation of the Measured Alpha and Beta Disintegration Rate, Activity, and Error Calculations

Observed Net Count Rate

At the end of each counting cycle, the net count rate is calculated for the alpha and beta channels. The net count is the counts/min with background subtracted. In general, the count rate is calculated as

$$N_x = \frac{C_x}{t_{C_x}} - \frac{B_x}{t_{B_x}}$$

where

x	=	α or β channel
C_x	=	measured gross counts in channel x
t_{Cx}	=	sample counting time in channel x
B_x	=	measured background counts in channel x
t_{B_x}	=	background counting time in channel x
N_x	=	measured net x count rate.

Measured Alpha and Beta Disintegration Rate

Due to the crosstalk phenomena, the measured disintegration rate is represented by two linear equations.

$$N_\alpha = \varepsilon_\alpha(m)D_\alpha + \varepsilon_{\beta\alpha}(m)D_\beta$$

$$N_\beta = \varepsilon_\beta(m)D_\beta + \varepsilon_{\alpha\beta}(m)D_\alpha$$

where

$\varepsilon_\alpha(m)$	=	measured alpha efficiency for mass, m
$\varepsilon_\beta(m)$	=	measured beta efficiency for mass, m
$\varepsilon_{\alpha\beta}(m)$	=	alpha-beta cross-efficiency for mass, m
$\varepsilon_{\beta\alpha}(m)$	=	beta-alpha cross-efficiency for mass, m
D_α	=	actual alpha disintegration rate
D_β	=	actual beta disintegration rate
N_α	=	measured net alpha count rate
N_β	=	measured net beta count rate.

These equations have two similar terms. The first term is the "traditional" counting situation where the measured count rate is a result of the actual disintegrations multiplied by the detection efficiency. The second term incorporates the cross-efficiency term. This term gives

the measured counts from the other disintegration source that ideally should not have been measured in the given channel. The sum of these two terms is simply the observed count rate for each channel. The two quantities that need to be calculated are D_α and D_β – the actual sample alpha and beta disintegration rates, respectively. In this simple linear system, two equations and two unknowns exist. The desired quantities can be calculated by simple substitution and some algebra as

$$D_\alpha = \frac{N_\alpha - \left[\dfrac{\varepsilon_{\beta\alpha}(m)}{\varepsilon_\beta(m)}\right] \cdot N_\beta}{\varepsilon_\alpha(m) \cdot \left[1 - \dfrac{\varepsilon_{\alpha\beta}(m)\,\varepsilon_{\beta\alpha}(m)}{\varepsilon_\alpha(m)\,\varepsilon_\beta(m)}\right]}$$

$$D_\beta = \frac{N_\beta - \left[\dfrac{\varepsilon_{\alpha\beta}(m)}{\varepsilon_\alpha(m)}\right] \cdot N_\alpha}{\varepsilon_\beta(m) \cdot \left[1 - \dfrac{\varepsilon_{\alpha\beta}(m)\,\varepsilon_{\beta\alpha}(m)}{\varepsilon_\alpha(m)\,\varepsilon_\beta(m)}\right]}$$

Substituting the crosstalk terms for the efficiency ratios in the previous equations yields the desired result:

$$D_\alpha = \frac{N_\alpha - X_{\beta\alpha}(m)\,N_\beta}{\varepsilon_\alpha(m) \cdot \left[1 - X_{\alpha\beta}(m)\,X_{\beta\alpha}(m)\right]}$$

$$D_\beta = \frac{N_\beta - X_{\alpha\beta}(m)\,N_\alpha}{\varepsilon_\beta(m) \cdot \left[1 - X_{\alpha\beta}(m)\,X_{\beta\alpha}(m)\right]}$$

Note as the crosstalk values approach zero, these equations simplify into the more typical case where the measured dpm is equal to the measured net count rate divided by the detection efficiency.

Crosstalk Corrected Count Rate and Efficiency

By closely examining the preceding dpm equations, D_α and D_β, the numerators are the crosstalk corrected net count rates, $N_{X\alpha}$ and $N_{X\beta}$, and the denominator is the crosstalk corrected efficiency/self-absorption factors, $\varepsilon_{X\alpha}(m)$ and $\varepsilon_{X\beta}(m)$.

$$\varepsilon_{X\alpha} = \varepsilon_\alpha(m) \cdot \left[1 - X_{\alpha\beta}(m)\,X_{\beta\alpha}(m)\right]$$

$$\varepsilon_{X\beta} = \varepsilon_\beta(m) \cdot \left[1 - X_{\alpha\beta}(m)\,X_{\beta\alpha}(m)\right]$$

By substituting for the observed count rates into the crosstalk corrected count rates, the final desired equations are obtained:

$$N_{X\alpha} = \left(\frac{C_\alpha}{t_{C_\alpha}} - \frac{B_\alpha}{t_{B_\alpha}} \right) - X_{\beta\alpha}(m)\left(\frac{C_\beta}{t_{C_\beta}} - \frac{B_\beta}{t_{B_\beta}} \right)$$

$$N_{X\beta} = \left(\frac{C_\beta}{t_{C_\beta}} - \frac{B_\beta}{t_{B_\beta}} \right) - X_{\alpha\beta}(m)\left(\frac{C_\alpha}{t_{C_\alpha}} - \frac{B_\alpha}{t_{B_\alpha}} \right)$$

Uncertainty in the Crosstalk Corrected Counting Rates

Using the fact that the counting error is simply the square root of the number of counts, the uncertainties for the crosstalk corrected counting rates, $\sigma_{N_{X\alpha}}$ and $\sigma_{N_{X\beta}}$, are calculated as

$$\sigma_{N_{X\alpha}} = \sqrt{ \left(\frac{C_\alpha}{t_{C_\alpha}^2} + \frac{B_\alpha}{t_{B_\alpha}^2} \right) + X_{\beta\alpha}^2(m)\left(\frac{C_\beta}{t_{C_\beta}^2} + \frac{B_\beta}{t_{B_\beta}^2} \right) + \sigma_{X_{\beta\alpha(m)}}^2 \left(\frac{B_\beta}{t_{B_\beta}} - \frac{C_\beta}{t_{C_\beta}} \right)^2 }$$

$$\sigma_{N_{X\beta}} = \sqrt{ \left(\frac{C_\beta}{t_{C_\beta}^2} + \frac{B_\beta}{t_{B_\beta}^2} \right) + X_{\alpha\beta}^2(m)\left(\frac{C_\alpha}{t_{C_\alpha}^2} + \frac{B_\alpha}{t_{B_\alpha}^2} \right) + \sigma_{X_{\alpha\beta(m)}}^2 \left(\frac{B_\alpha}{t_{B_\alpha}} - \frac{C_\alpha}{t_{C_\alpha}} \right)^2 }$$

Activity Calculations

The activity and error calculations are performed at the end of each counting cycle. The activity of the sample is in pCi per unit volume (mass). The equations for the alpha activity, A_α, and the beta activity, A_β, are

$$A_\alpha = \frac{N_{x\alpha}}{\varepsilon_{x\alpha}(m) \bullet Q} \bullet k$$

$$A_\beta = \frac{N_{x\beta}}{\varepsilon_{x\alpha}(m) \bullet Q} \bullet k$$

where

A_α	=	alpha activity
A_β	=	beta activity

$N_{X\alpha}$	=	crosstalk corrected net alpha count rate
$N_{X\beta}$	=	crosstalk corrected net beta count rate
k	=	factor to convert to the desired reporting units
Q	=	sample size (volume or mass)
$\varepsilon_{X\alpha}(m)$	=	crosstalk corrected alpha detector efficiency/self-absorption factor at mass, *m*
$\varepsilon_{X\beta}(m)$	=	crosstalk corrected beta detector efficiency/self-absorption factor at mass, *m*

Activity Error Calculations

The uncertainty in the alpha and beta activity calculations, σ_{A_α} and σ_{A_β}, can be expressed by the following equations:

$$\sigma_{A_\alpha} = \sqrt{\left(\frac{\sigma_{N_{x\alpha}}}{k^{-1} \bullet \varepsilon_{x\alpha}(m) \bullet Q}\right)^2 + \left(\frac{A_\alpha \bullet \sigma_{\varepsilon_{x\alpha(m)}}}{\varepsilon_{x\alpha}(m)}\right)^2 + \left(\frac{A_\alpha \bullet \sigma_Q}{Q}\right)^2}$$

$$\sigma_{A_\beta} = \sqrt{\left(\frac{\sigma_{N_{x\beta}}}{k^{-1} \bullet \varepsilon_{x\beta}(m) \bullet Q}\right)^2 + \left(\frac{A_\beta \bullet \sigma_{\varepsilon_{x\beta(m)}}}{\varepsilon_{x\beta}(m)}\right)^2 + \left(\frac{A_\beta \bullet \sigma_Q}{Q}\right)^2}$$

where

$\sigma_{A\alpha}$	=	error in the alpha activity
σ_{A_β}	=	error in the beta activity
A_α	=	measured alpha activity
A_β	=	measured beta activity
$\sigma_{\varepsilon_{X\alpha}(m)}$	=	uncertainty in the crosstalk corrected alpha detector efficiency/self-absorption factor
$\sigma_{\varepsilon_{x\beta}(m)}$	=	uncertainty in the crosstalk corrected beta detector efficiency/self-absorption factor
σ_Q	=	uncertainty in the sample quantity.