CONF-970424--2
SAND97-0798C
SAND--97-0798C

DIRECT ANALYSIS OF AIR FILTER SAMPLES FOR ALPHA EMITTING ISOTOPES

A. H. Mohagheghi *, F. Ghanbari *, S. B. Ebara *,
M. E. Enghauser *, and S. N. Bakhtiar **

* Sandia National Laboratories, P.O.Box 5800, Albuquerque, NM 87185-0651, USA
** Westinghouse WIPP, 1509 Lincoln Drive, Carlsbad, NM 88220, USA

The traditional method for determination of alpha emitting isotopes on air filters has been to process the samples by radiochemical methods. However, this method is too slow for cases of incidents involving radioactive materials where the determination of personnel received dose is urgent. A method is developed to directly analyze the air filters taken from personal and area air monitors. The site knowledge is used in combination with alpha spectral information to identify isotopes. A mathematical function is developed to estimate the activity for each isotope. The strengths and weaknesses of the method are discussed.

Introduction

This report describes an alpha spectroscopy method for the rapid assessment of alpha emitting contaminants on unprocessed air filters. This technique is potentially useful for situations where the identities and ratios of different contaminants are not known and cannot be determined using gamma spectroscopy due to the weakness or lack of photon emissions from the sample.

There are a number of cases that a rapid analysis of air filters for alpha emitting isotopes are needed when there is a significant amount of alpha activity of an air filter. The air filter results are one of the indicators used to determine the uptake of radiation by radiological workers. In order to determine a possible received dose, the isotope and its activity are needed. However, for some projects (e.g. nuclear cleanup projects), the isotopic composition and ratios are not well known. This usually requires radiochemistry to separate various isotopes and determine their activities. The traditional radiochemical process involves physical preparation, elemental separation for each suspect element, sample mounting, counting, and analysis. The radiochemical method takes, at best, days to complete and is costly. The analysis is further complicated by the presence of large amounts of fission products on most air filters that require extra steps in sample handling and chemistry. The technique described in this method is used to resolve these problems.

When collecting air samples, particles are accumulated in multiple layers. Therefore, when the sample is counted on an alpha spectroscopy system, the particles located in the deeper regions exhibit greater attenuation before reaching the detector. This results in a sample spectrum with a broad energy distribution. When the sample is counted under a complete vacuum, the particles emitted from the uppermost surface are not attenuated and the emission energy can be determined by the edge of the peak.

Once the alpha emission energy is matched to the

DISCLAIMER

appropriate nuclide(s), it is necessary to determine the nuclide's peak area and perform an efficiency correction to estimate the activity of the nuclide. For samples containing multiple nuclides, overlapping spectrum peaks make it difficult to determine the peak area associated with each nuclide by simply setting regions of interest. An asymmetrical function is used fit to the peaks and to de-convolute the area for each isotope. An equation and a complimentary computer model are used to determine the counting efficiency. These steps are used to identify isotopes and to estimate their activities within a few hours.

Theory

The basic premise of the described method is that particles are accumulated in multiple layers during the collection process. Therefore, the alpha particles from the upper layers do not experience any significant energy loss and arrive at the detector at their correct energy. The alpha particles emitted in deeper layers lose some of their energy and have a larger energy spread (see figure 1).

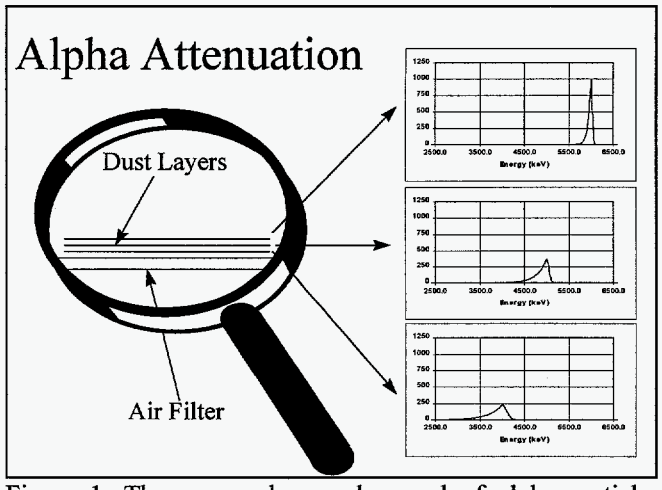

Figure 1. The energy loss and spread of alpha particles traveling through matter.

The primary source of energy loss for alpha particles passing through matter is electronic excitation and ionization. The relativistic quantum mechanical treatment by Bethe gives [1]:

$$\frac{dE}{dx} = D\left(\frac{z}{\beta}\right)^2 n\left[\ln\frac{2mc^2\beta^2\gamma^2}{I} - \beta^2\right], \qquad (1)$$

where

D = 5.0989×10^{-25} MeV-cm^2,
z = Projectile atomic number,
b = Projectile speed (v) / Light speed (c),
n = Electrons per cm^3 in the target,
m = Electron rest mass, and
I = Mean ionization potential.

The energy loss depends only on the charge and velocity of the projectile, but not its mass. As the particle energy increase from near zero, the energy loss increases due to the $1/\beta^2$ factor. As the projectile energy continues to increase, the ln term in equation (1) begins to dominate and the energy loss starts to decrease. This is referred to as the region of relativistic rise. The combination of the two terms causes a peak (called the Bragg peak - see figure 2) to occur where the rate of energy loss is maximum.

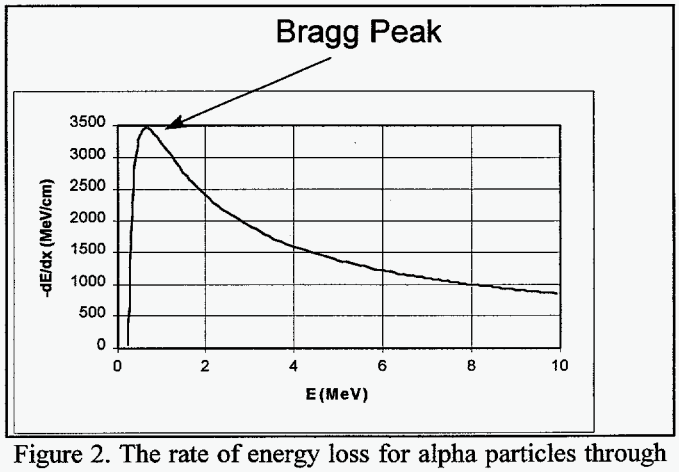

Figure 2. The rate of energy loss for alpha particles through soil (modeled as SiO_2).

The transport of a 5 MeV alpha particle was modeled using equation 1. The alpha particles'

rate of energy loss is shown in figures 3 where -dE/dx changes smoothly with thickness up to about 4000 ug/cm2. The energy loss rate then reaches a maximum at a thickness of about 4400 ug/cm2 and begins to fall rapidly.

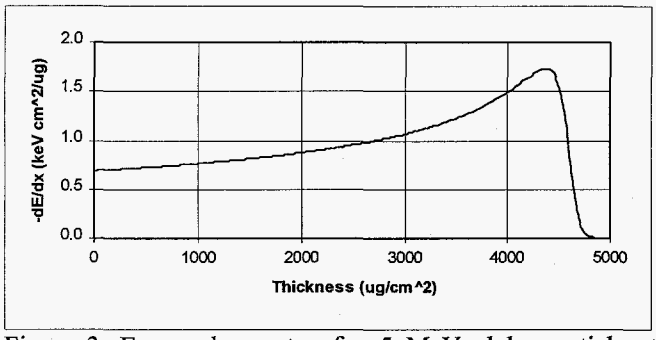

Figure 3. Energy loss rate of a 5 MeV alpha particle at various dust layers.

The particle's energy as a function of thickness is shown is figure 4 where the energy falls smoothly until it reaches a thickness of about 4500 ug/cm2. For extremely low energies, the alpha particle velocity becomes comparable to the bound electrons in the target material, violating one the simplifying assumptions use to drive equation 1. At this stage, atomic shell corrections are needed. By requiring that the energy loss rate be kept in the linear region, keeping equation 1 valid, and keeping the minimum alpha energy greater than 2 MeV (to stay above the β spectrum), the maximum surface density is around 3500 ug/cm2. This gives a maximum of about 69 mg of dust on a typical two inch air filter for the direct counting method to work.

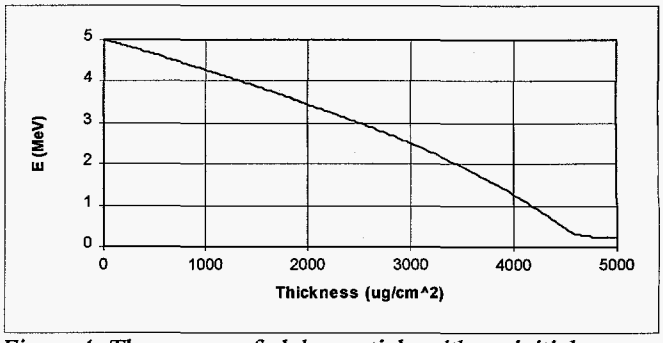

Figure 4. The energy of alpha particle with an initial energy of 5 MeV through various dust layers.

Once an alpha spectrum has been acquired, an asymmetrical function is used to fit the spectrum. The function is given by:

$$y_i = A_i e^{-0.5(x-c_i)^2/\sigma_h^2} \qquad x \geq c_i$$

$$y_i = A_i e^{-0.5(c_i-x)/\sigma_l} \qquad x < c_i \tag{2}$$

where

A_i = Amplitude for peak i,
x = Alpha Energy,
c_i = Centroid for peak i,
σ_h = Peak width on high energy side, and
σ_l = Peak width on low energy side.

The asymmetrical function provides a curve that is a Gaussian function on the high energy side of the peak centroid modeling the un-attenuated alphas originating in the top layers. Equation 2 is an exponential function on the low energy side of the peak centroid simulating the exponential attenuation of alpha energies emitted from deeper layers. A graph of equation 2 for an amplitude of 1000 counts, centroid of 5000 keV, σ_h of 30 keV, and σ_l of 300 keV is shown in figure 5.

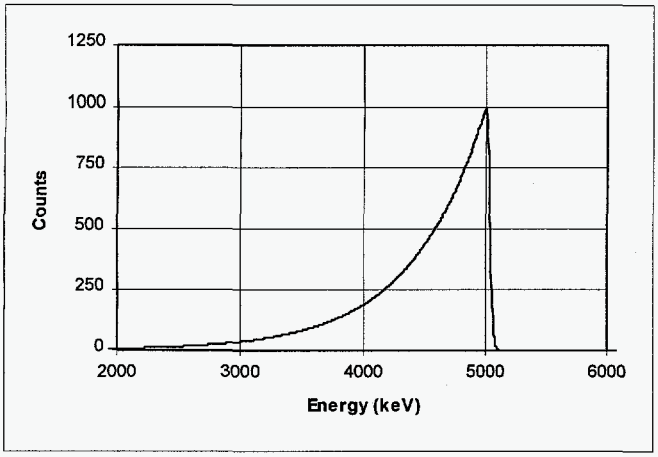

Figure 5. A graphical representation of equation 2.

For multiple isotopes, equation 2 is simply summed over the number of peaks (n) in the spectrum.

$$Y = \sum_{i=1}^{n} y_i \,, \tag{3}$$

Once the peak locations and areas have been determined the counting efficiency needs to be determined. For simple geometries where the particles are uniformly distributed on an air filter and the sample radius is less than the detector radius, the solid angle seen by the source is given by Sergè [4]:

$$\Omega = 2\pi\left(1-\cos\alpha\left\{1+\frac{3}{2}\frac{r^2}{d^2}\left(\frac{\sin 2\alpha}{4}\right)^2\right\}\right), \tag{5}$$

where

$\alpha = \tan^{-1}(a/d)$,
a = detector radius,
r = source radius, and
d = source to detector distance.

The counting efficiency is then given by:

$$\text{Efficiency} = \Omega/4\pi, \tag{6}$$

For cases where the particle distribution is not uniform, or the source radius is larger than the detector, a computer model is developed to estimate the counting efficiency. Briefly, the source is divided to a number of pixels, the efficiency for each pixel is computed by assuming it is a point source, and finally the efficiencies for all the pixels are added and normalized to estimate the source efficiency.

Finally, the peak areas may determined by integrating equation 2 for each peak and dividing by the counting time and the computed counting efficiency.

Verification

A test was conducted to test the adequacy of the method. An mixed alpha containing Am-241, Pu-239, Cm-244, and U-233 was chosen for this test. The source was made by electroplating the isotopes on a stainless steel disk with a radius of 1.0 cm. The counting system for this test consisted of an ion implanted surface barrier detector (500 mm^2 area), a vacuum chamber, and the associated electronics and software. The source was counted at a distance of 0.5 cm from the detector for 100 minutes at a pressure of 430 torr (328 ug/cm2). The detector had a radius of 1.26 cm. This essentially simulates a case where all the alpha particles are emitted from the deeper layers and are all therefore attenuated. The resulting spectrum as expected shows four widened peaks (see figure 6).

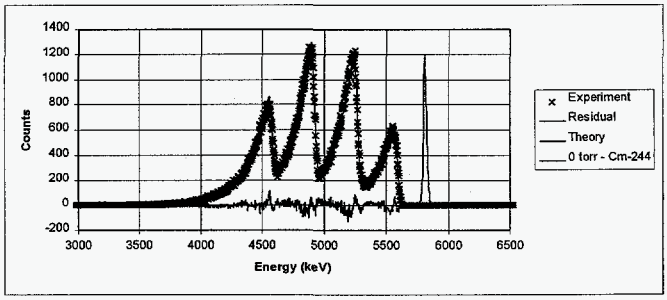

Figure 6. The electroplated standard counted under a pressure of 430 torr. The narrow peak on the right is the location of the Cm-244 peak at 0 torr.

The experimental data is fit to equation 3 with four terms. The efficiency is calculated using equation 5 and the results are shown in table 1.

Isotope	U-233	Pu-239	Am-241	Cm-244
Centroid (keV)	4553	4898	5246	5563
Amplitude	683	1126	1142	624
s_h	23			
s_l	87			
Reduced c^2	0.33			
Measured dpm	2068	3410	3458	1890
Known dpm	2088	3384	3516	1812
Bias	-1%	1%	2%	4%

Table 1. Results of fitting the test spectrum.

The reduced χ^2 of 0.33 verifies that the proposed formula represents the data well. The above test demonstrates that the modified asymmetrical function works well in estimating the relative fractions for samples containing multiple nuclides where the sample distribution is uniform and the maximum thickness is below 3500 $\mu g/cm^2$.

Results

The first sample is a two inch air pre-filter that is used during a nuclear cleanup project involving high burn up mixed oxide spent fuels. A non-destructive cleaning unit is employed that uses high velocity carbon dioxide pellets to remove surface contamination. The contamination becomes airborne and is collected by a high efficiency particulate air filter in the unit by high velocity air flow.

The sample was first counted at a distance of 1 cm from the detector for 1000 minutes. The resulting spectrum is shown in figure 7. The sample has a large amount of beta activity with the Cs-137 electron conversion peak clearly showing. There are also three major alpha peaks and two small ones visible in the spectrum.

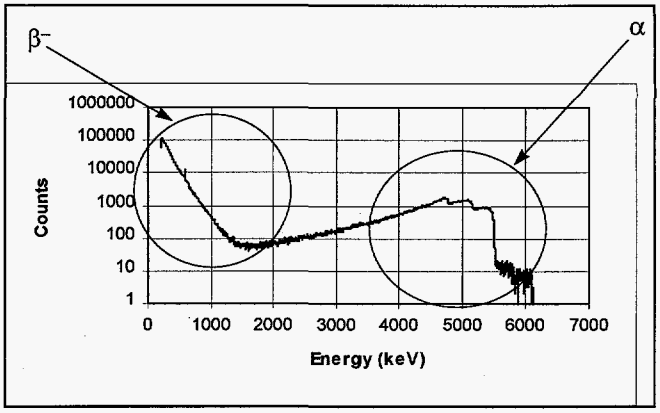

Figure 7. Sample 1 spectrum.

A close examination of the filter revealed that the dust loading is not uniform and is concentrated around the edges. In order to obtain a better counting geometry and to obtain better statistics on the smaller peaks, the sample was lowered and counted at a distance of 4.4 cm from the detector for 6000 minutes. The resulting alpha spectrum (see figure 8) is fit to equation 3 with five terms and the results are shown in table 2.

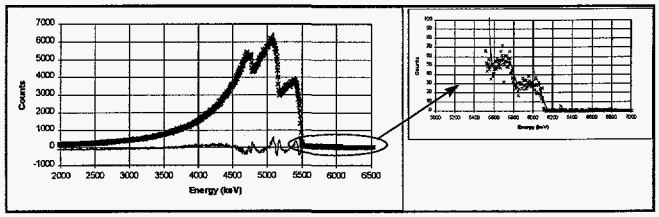

Figure 8. The alpha spectrum for sample 1. The dots are the observed points, the solid line is the fitted equation, and the fit residuals are shown at the bottom.

Peak	1	2	3	4	5
Centroid (MeV)	4.72	5.08	5.41	5.75	6.04
Amplitude	1475	4101	4024	35	35
σ_h	49				
σ_l	314				
Reduced χ^2	6.6				
Activity (dpm)	2245	6242	6125	53	53

Table 2. Fit results for sample 1.

The next step in the analysis is to match the observed peaks to potential isotopes. The major sources of radioactivity are experimental reactor fuels (highly enriched Uranium mixed with Plutonium) with a high burn up value. A list of major alpha emitting isotopes that can be produced in this process are shown in table 3. For highly enriched Uranium, the major source of activity is U-234 due its short half-life compared to U-238 and U-235. The resolving power of the given spectrum is estimated to be around 100 keV, implying isotopes with energies that are closer than 100 keV, show up as a single peak.

Isotope	τ (years)	Energy (MeV)	
U-234	2.5E5	4.78	→ Peak 1
Pu-239	2.4E4	5.16	} Peak 2
Pu-240	6.6E3	5.17	
Am-241	432	5.49	} Peak 3
Pu-238	88	5.50	
Cm-244	18	5.81	→ Peak 4
Cm-242	0.5	6.11	→ Peak 5

Table 3. List of major alpha emitting isotopes in a high burn up MOX fuel.

The second sample was also taken during a similar nuclear cleanup project, but, the contamination source was different. The sample has a much greater dust loading on it with most of the activity concentrated on one side. The sample is counted at a distance of 4.4 cm from the detector for 1000 minutes and the resulting spectrum is shown in figure 9.

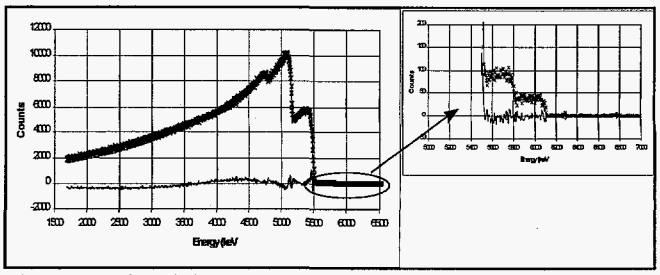

Figure 9. The alpha spectrum for sample 2. The dots are the observed points, the solid line is the fitted equation, and the fit residuals are shown at the bottom.

The spectrum is fit to equation 3 with five terms and the results are shown is table 4. The main difference from sample 1 is the large peak width on the lower energy side due to higher amount of particulates on the filter.

Peak	1	2	3	4	5
Centroid (MeV)	4.74	5.12	5.45	5.77	6.07
Amplitude	600	5000	5900	57	40
σ_h	24				
σ_l	900				
Reduced χ^2	4.2				
Activity (dpm)	2591	21592	25479	248	175

Table 4. Fit results for sample 2.

Summary

In summary, the proposed method provides for rapid identification and quantification of alpha emitting isotopes on an air filter. The advantages of the method are a rapid determination of a possible dose received by radiological workers that minimizes down time, minimizes hazardous radioactive wastes associated with radiochemistry, allows to focus resources where it's needed. The method is sensitive to particulate distribution on a filter and some care must be take it into account for activity determination. Finally, the same method may be applied for direct counting of soil samples and other matrices [5].

The authors thank Robert Reese and Fernando Dominguez for performing most of the

measurements, Susan Longley and Charles Potter for technical discussions. This work was supported by the United States Department of Energy under contract DE-AC04-94AL85000. Sandia is a multi-program laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the US DOE.

References

1. R. C. Fernow, *Introduction to Experimental Physics*, Cambridge University Press, 1986.
2. J. E. Turner, *Atoms, Radiation, and Radiation Protection*, MCGraw-Hill, 1992.
3. M. J. Koskelo, W.C. Burnett, P.H. Cable, *Radioactivity & Radiochemistry Journal*, Vol. 7, No. 1, p. 18 , 1996.
4. E. Sergè, *Experimental Nuclear Physics, Volume III*, John Wiley & Sons, 1959.
5. K. E. Meyer, A. C. Lucas, *DOE Methods for Evaluating Environmental and Waste Management Samples*, DOE/EM-0089T, 1995.
6. Department of Energy Health Physics *Manual of Good Practices for Uranium Facilities*, Idaho National Engineering Laboratory, DE88-013620, EGG-2530, UC-41, 1988.
7. Environmental Protection Agency, *Environmental Radiation Dose Commitment: An Application to the Nuclear Power Industry*, EPA-520/4-73-002, Washington, D.C., 1974.