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Self-attenuation as a function of gamma ray energy in naturally occurring radioactive material in the oil and gas industry

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HIGHLIGHTS

- Self attenuation of gamma-rays.
- Naturally occurring radioactive material in oil and gas industry.
- Ra-226, Ra-228 and Pb-210.

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ABSTRACT

Self-attenuation correction factors were experimentally determined using radioactive point sources in combination with a subject material of naturally occurring radioactive material (NORM) obtained from oil exploration waste products. The self-attenuation correction factors were taken across a range of gamma ray energies from 41.73 to 1408.0 keV. It is noted that the greatest amount of self-attenuation occurs in the energy regime below 200 keV and rises to near zero attenuation at higher energies for these types of samples. For the 46.5 keV gamma ray of ^{210}Pb there can be an underestimation of 62%.
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1. Introduction

Gamma ray spectroscopy is an invaluable tool when used to assay materials containing or contaminated by radionuclides. By analyzing a gamma ray spectrum of a particular material, it then becomes possible to determine the concentration of radioisotopes within that material. This is, of course, direly important in multiple fields including radioactive waste management, health physics, and dosimetry among others. Thus, it becomes critically necessary to correctly and accurately measure the gamma ray spectrum for a given material. This is problematic for those materials that are particularly dense or else contain large quantities of higher Z elements such as iron, strontium or barium as found in some instances of NORM found in the oil and gas industry (Landsberger et al., 2013). In the case of these materials, gamma rays emitted by the radioisotopes present in the material are attenuated by the material itself before being measured with a detector apparatus (Boshkova and Minev, 2000).

Any gamma ray that is emitted by the decay of a radioisotope in a material has a distinct probability of undergoing various interactions with the material and being attenuated prior to being counted on a detection system. These interactions with the material result in scattering of the gamma ray, absorption, or loss of energy to the material (e.g. Compton scattering). The attenuation of these gamma rays by the emitting material results in an underestimation of the intensity of photopeaks in the spectrum for these materials, particularly at low energies of less than 100 keV (Robu and Giovani, 2009). The extent of this gamma ray self-attenuation is dependent on both the geometry of the measured sample as well as the linear attenuation coefficient, μ, for the material in the sample. The linear attenuation coefficient is itself reliant on the properties of the material, such as density and composition, and the energy of the gamma rays in question. The intensity of a gamma ray source passing through an attenuating material is given as the following:

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$$I(E) = I_0(E)e^{-\mu(E)x}, \quad (1)$$

where $I(E)$ is the attenuated intensity of the gamma rays emitted at energy E, $I_0(E)$ is the intensity of the gamma rays at energy E prior to attenuation, $\mu(E)$ is the linear attenuation coefficient of the attenuating material at energy E, and x is the linear thickness of the material.

While it is possible to determine the value of μ for a particular material through calculation and computational simulation, it is not practical to do this in many cases during the course of field study of large quantities of NORM samples due to the need to ascertain the chemical composition of each sample. Instead, it is usually more useful to determine the quantity μx through experimental methods and to utilize this as a means of calculating the self-attenuation correction factor for a material across a broad energy range (Haddad and Albyiat, 2009). By using the μx quantity, the self-attenuation correction factor, F_{att}, is then given as

$$F_{att} = \frac{\mu x}{1 - e^{-\mu x}}. \quad (2)$$

This can also be rewritten in terms of the measured gamma ray peak intensities as

$$F_{att} = \frac{-\ln(I/I_0)}{1 - (I/I_0)}. \quad (3)$$

(Robu and Giovani, 2009; Al-Masri et al., 2013) where F_{att} is the self-attenuation correction factor, I and I_0 are the count rate of external point source through the sample and standard, respectively. In our method, standard point sources of ^{152}Eu, ^{214}Am and ^{109}Cd were positioned above a sample which is located on the detector, and the number of counts in the full energy peak is measured. The purpose of this work is to employ a simple methodology in order to determine the self-attenuation correction factor for a range of gamma ray energies such that a relation can be formed for one sample of NORM in the oil and gas industry and that might be easily adapted to other similar material samples.

2. Experimental methodology

The intensity of gamma rays at a specific energy is easily taken from the peak counting rate for that energy in a gamma ray spectrum. Therefore, it is possible to develop the previously mentioned self-attenuation correction factors for a given gamma ray energy by measuring the peak counting rate for photons emitted by a radioactive point source as it has been attenuated through the sample of interest; in this case a sample of NORM. In order to determine the initial non-attenuated intensity of the point source, it is necessary to obtain a gamma ray spectrum of the source in which it is centered over an empty Petrie© dish (6 cm diameter and 1.5 cm height) sample container exactly matching the container of the NORM 20-gram sample. This is a method similar to one previously employed using gamma ray calibration sources (Parker, 1984). This is done to reduce geometric dependence of the resulting self-attenuation relation. Previous studies have focused on determining the effect of geometric orientation of the source–sample-detector arrangement using both computational modeling and numerical analysis methods (Abbas, 2001; Conway, 2011; Lepy, 2010).

The attenuated intensity of the source is then the difference between the peak counting rate at a given energy for the source affixed to the top of the NORM sample and the NORM sample in isolation from the source. The majority of the desired gamma and x-ray energies range from 40.1 to 1408.0 keV can be obtained by using a ^{152}Eu point source. In addition the ^{241}Am (59.4 keV) and the ^{109}Cd (880 keV) sources were also used. It should be noted that it is helpful to not choose gamma ray peaks for measurement that are present in both the point sources and the isolated sample of interest.

3. Results and discussion

The NORM sample measured was taken from waste produced from oil drilling processes in the western Texas region. The sample and source configuration previously described were counted in a standard Petrie© dish (6 cm diameter and 1.5 cm height) for periods of 4 h or 14,400 s each, culminating in the generation of four gamma ray spectra including background, sample-detector, sample-point source-detector, and point source-detector measurements. A standard 30% efficient hyperpure germanium detector with a thin beryllium window in a well shielded “clam shell” configuration was employed. These data were collated into the self-attenuation results given in Table 1 and plotted in Fig. 1 below.

From Fig. 1 is evident that the level to which gamma rays are attenuated in the NORM sample is highest at lower energies. This is to say that gamma rays much above 400 keV in these NORM samples tend to transmit more than 90%. However, other types of environmental or geological samples that have higher Z elements in high abundance may attenuate photons more severely at all energies. It is important to note that in many radioactive samples, NORM included, there are several isotopes of interest in the region below 200 keV that would be undercounted without regarding this self-attenuation factor, particularly isotopes such as ^{210}Pb (46.5 keV) and ^{2236}Ra (186.3 keV).

The equation of the overall relation of self-attenuation to energy is then given as the function:

$$\frac{1}{F_{att}} = 0.0548 \ln E^3 - 1.0008 \ln E^2 + 6.0882 \ln E - 11.366 \quad (4)$$

In fitting the best curve to the data, it is important to note not only the shape of the lower energy section of the curve, but also the point at which the gamma rays are no longer attenuated in the material and the value $1/F_{att}$ goes to 1. It is expected that assays of other similar materials would require only analysis of the energy spectrum prior to this point of pure transmittance, and as such could potentially shorten analysis conducted on those materials.

Table 1
Self-attenuation factors as a function of energy.

E (keV)	$1/F_{att}$ [c]
39.8[a]	0.2561
45.4[b]	0.3863
59.25	0.5638
88.0	0.7598
121.8	0.8824
344.3	0.9256
411.1	0.9334
443.9	0.9967
688.8	1.0150
778.9	0.9578
867.4	1.0058
963.75	1.0184
1212.9	0.9948
1408.0	1.0439

[a] K_α x-ray.
[b] K_β x-ray.
[c] Eq. (3).

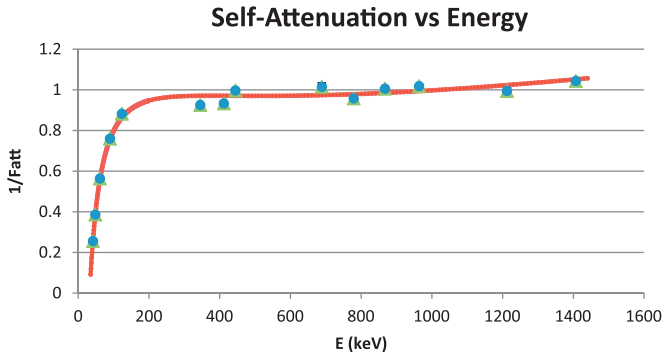

Fig. 1. Self attenuation vs energy from the data in Table 1. It is apparent that the amount of attenuation tapers off quickly with increasing gamma ray energies. Typical uncertainties based in the transmitted and attenuated counting statistics is estimated to be ~2–3%.

4. Conclusions

The ability to accurately measure concentrations of radioisotopes in a sample is dependent on using the self-attenuation correction for that material. This self-attenuation correction factor is itself highly energy dependent, where gamma rays with lower energy will be attenuated much more effectively in a sample. This work has presented a simple means by which to determine the self-attenuation-energy relation for a sample of material. In particular, it was determined that the bulk of the self-attenuation of gamma rays occurs below 120 keV, and steadily rises to near perfect transmittance of gamma rays at higher energies. For the 46.5 keV gamma ray of ^{210}Pb there can be an underestimation of 60%. This method should be quickly adaptable for use with other materials in order to determine the self-attenuation relations for those materials.

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