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An alpha-gamma coincidence spectrometer based on the Photon-Electron Rejecting Alpha Liquid Scintillation (PERALS®) system

J.R. Cadieux, G.A. Fugate*, and G.S. King III
Savannah River National Laboratory, Aiken, South Carolina 29808, USA

Abstract

An alpha-gamma coincidence spectrometer has been developed for the measurement of selected actinide isotopes in the presence of high beta/gamma fields. The system is based on a PERALS® liquid scintillation counter for beta/alpha discrimination and was successfully tested with both high purity germanium and bismuth germanate, gamma-ray detectors using conventional analog electronics.

Keywords: coincidence, liquid scintillation, PERALS

1. Introduction

The detection and measurement of many long-lived, actinide isotopes in environmental samples continues to be one of the more difficult problems in radioanalytical chemistry. The metrology of alpha-emitting radionuclides from neptunium through curium is of particular interest for monitoring nonproliferation, detecting releases to the environment, and controlling special nuclear materials. The conventional analysis of many of these isotopes requires extensive sample purification and preparation prior to assay of short half-life nuclides by alpha spectrometry and longer half-life nuclides by mass spectrometry. A number of the actinides do have gamma or X-ray emissions associated with their alpha decays but they are generally at energies below 100 keV. These transitions are often heavily converted, resulting in low intensities (numbers of photons per decay), limiting their analytical utility. The analytical problem is amplified when high levels of beta-emitting, fission and activation products are also present in the samples. The bremsstrahlung and gamma-ray emission from these interferences can overwhelm the low energy region of the gamma-ray spectrum. The ^{241,243}Am and ^{244}Cm radionuclides have useful gamma transitions but their chemistries are very similar to the lanthanides isotopes and ^{90}Y which are produced in high yields in the fission process. Their chemical separation from the lanthanides and yttrium, typically by column chromatography, is long and meticulous/exacting. The analysis of ^{237}Np can be even more difficult challenge that is not solvable by chemical separation if a significant amount of ^{239}Np activity is present. The ^{239}Np is a beta/gamma emitter with a 2.4 day half-life that is produced in high yield by the neutron capture of ^{238}U and can dominate the gamma-ray spectrum of the actinide fraction of a freshly irradiated uranium sample for a month or longer.

There has been a concerted effort in recent years to improve the sensitivity of gamma-ray measurements in laboratory and field instruments by using coincidence counting techniques to actively suppress the gamma-ray background [1-3]. The International Atomic Energy Agency has explored the use of an alpha-gamma coincidence spectrometer (AGCS) to suppress the gamma-ray background and

* Corresponding Author. E-mail address: Glenn.Fugate@SRNL.DOE.GOV (G.A. Fugate)

provide high selectivity for americium and plutonium isotopes.[4] The system combines an alpha spectrometer with a low energy gamma-ray detector and demands simultaneous detection of alpha and gamma pulses within the resolving time of the system (typically a few microseconds). Their study used a standard silicon detector for alpha spectrometry to detect the alpha events and generate a gating pulse for the gamma spectrometer. High resolution alpha detectors have good discrimination (factors of greater than 10^4) from beta particles and the use of the alpha signal as a gating requirement eliminated much of the external gamma-ray background activity and internal gamma-rays from beta decays in the sample. However this AGCS has a number of limitations since it still requires adequate chemical separation to produce a chemically pure, thin source, suitable for alpha spectrometry. It also restricts the amount of sample to 50 - 100 micrograms or less on the alpha plate to limit straggling of the alpha particles in the deposit which drastically affects the alpha energy resolution. In addition the alpha source to silicon detector path must be in vacuum and tightly coupled to the gamma-ray detector to preserve the overall system's efficiency which is mechanically challenging. Even when all of these criteria are satisfied the typical detection efficiency for alpha particles is about 30% or less.

This work has taken a different route to providing an alpha gating signal for a low-energy, gamma-ray spectrometer for measuring actinides and other alpha emitters. An AGCS has been developed based upon a liquid scintillation counter (LSC), alpha spectrometer, the PERALS® system from ORDELA (Oak Ridge Detector Laboratory, Inc.) [5]. The PERALS® unit is a self-contained LSC specifically designed for alpha spectrometry [6], housed in a three, nuclear instrument module wide unit. This system was originally developed over 30 years ago for measurement of ^{210}Po and ^{235}U in bioassay samples [7, 8]). It uses tailored organic extracting scintillators to chemically bind the element of interest and transfer it from the aqueous phase into a non-water soluble mixture or cocktail. To enhance peak resolution, oxygen is purged from the solution and the sample is sealed in a small Pyrex™ tube that is inserted into a reflective chamber in front of a high quality phototube. In the energy deposition process, the alpha particles produce triplet states in the molecules of the cocktail whose light pulses then take longer to decay in the scintillating cocktail than those in singlet states, produced by beta or gamma interactions. The resultant alpha pulses have longer decay times that can be electronically selected by pulse shape analyses with beta/gamma and alpha pulses separated by about 30 nanoseconds in the time domain. PERALS® samples purged with argon and closed with a vinyl cap are typically stable for counting for periods of days to weeks; flame sealed standards have been stable for over a decade.

Time to pulse height circuitry in the PERALS® converts the decay times of the beta and alpha signals into two distinct voltage pulses, the larger of the two amplitudes corresponding to the alpha particle interactions. A pulse shape discriminator control allows easy selection of the alpha events. The system achieves a full width at half maximum resolution of about 300 keV at an alpha energy of 6 MeV. The alpha detection efficiency is almost 100% and alpha/beta discrimination is better than 1000 to 1 due to the pulse shape analysis. The LSC's capacity of 50 to 100 milligrams per sample is almost a factor of 1000 greater than alpha deposits for conventional, high resolution alpha spectrometry, providing higher sensitivity for long half-life nuclides with low specific activities such as ^{232}Th, ^{238}U and ^{237}Np.

Over the years numerous methods have been developed for the analysis of a number of alpha emitting nuclides using the PERALS® systems [9]. Extensive work has been devoted to measuring natural occurring radioactive isotopes of uranium and thorium along with their decay chain progeny: radium, radon and polonium [10-14]. This spectrometer has found useful applications in environmental research and monitoring [10, 11, 13, 15-17], and it has been accepted as a standard method for the analysis of trace uranium in drinking water and environmental samples [18, 19] by the ASTM and the US EPA (Standard Method 3125). Specific methods for many of the actinides have been developed and evaluated [20-25]. The PERALS® has been compared with other measurement methods, many of which employ systems and/or devices that were orders of magnitude more complicated and expensive, with the advantages and disadvantages of these approaches having been well characterized [26, 27]. One of the drawbacks to quantifying alpha emitting nuclides with the PERALS® has been the limited energy resolution of LSC as compared to measurements with silicon detectors which has restricted some of the practical analytical applications for actinides. The alpha energy resolution for the PERALS® is about a factor of 10 lower than the standard silicon alpha detector. The beta discrimination is also 10 to 100 times lower than that of conventional alpha spectrometry. Considerable effort has been dedicated to compensating for these deficiencies [26, 28-30], but the limited alpha energy resolution has remained a disadvantage for the PERALS® [28, 29]. The analog electronic system, though still effective, is a design that is over 30 years old. Recent progress in digital electronic systems hardware and digital pulse processing would suggest that significant progress could be made by modernizing the pulse analysis section of the system. Over the past decade, staff at the Savannah River National laboratory have updated several specialized low-level counting systems with digital pulse processing, improving the analytical results provided by these systems. Of particular use is the ability to capture and store every digitized event. Post processing and analysis of the entire data set has substantially increased the information from a single measurement and, in turn, it's precision. For the AGCS coincidence data, digital processing has the potential to provide multi-parameter spectra for each analysis. Rather than just using the alpha information to generate a gate signal, individual alpha events could be evaluated by energy and matched with the coincident gamma-ray spectrum. Additionally, the data set could also be analyzed for simultaneous beta/gamma coincidences that are normally excluded.

Another advantage of the coincidence counting method is the ability to directly determine the absolute disintegration rate of the source from the counting data without prior calibration to measure efficiencies of the detectors. This technique has been known [30] and used in a variety of applications including safeguards measurements [31, 32], production of primary standards [33, 34], or improving nuclear decay data schemes[35-37]. In practical applications, employing either beta-gamma or gamma-gamma coincidences, there are numerous corrections that are required depending upon the desired accuracy.[38] However in alpha-gamma coincidence there is very little cross-talk between the detectors and most of the corrections are not necessary; therefore the accuracy is typically limited by the uncertainty in the non-coincident gamma-ray counting rate.

2. Equipment

The PERALS® system uses a carefully designed sample holder to couple the sample to the phototube [39] providing essentially quantitative transfer of the scintillation light. An ORDELA Model 8100AB-HV

PERALS® Spectrometer was modified with a thin-walled sample holder (Figure 1). The aluminum block on the front which housed the sample holder was replaced with a chamber fabricated from a low density plastic (Bakelite). The black area is the sample chamber which was machined out in a circular shape centered on the sample to position the low energy photon (LEP) detector in intimate contact against the PERALS® sample with a high geometric coverage. An Ortec™ GLP Series Planar Germanium detector 36 mm in diameter by 13 mm thick was used as the gamma-ray detector. It was mounted in a horizontal cryostat and had a 0.25 mm beryllium window and a dead layer of only 0.3 µm with effective sensitivity down to less than 10 keV. The LEP detector was aligned directly against the PERALS® chamber (Figure 2). The AGCS was designed for maximum sensitivity in the 20 to 200 keV energy range.

The pulse shape voltage output from the alpha pulses was used as the input signal for an Ortec model 550A Single Channel Analyzer (SCA) with the SCA window set to the same voltages as the pulse shape discriminator (PSD) on the PERALS®. The output of the SCA was stretched and delayed by an Ortec model 416A Gate and Delay Generator whose output triggered the coincidence logic on an Amptek MCA-8000 multichannel analyzer (MCA). A block diagram of the electronic setup is shown in Figure 3. Alternatively, a specific alpha energy from the PERALS® can be selected for gating by setting the SCA to a window that corresponds to the pulse height output of the alpha energy spectrum.

A bismuth germanate (BGO) scintillation detector was also tested with the coincidence system in the place of the high purity germanium spectrometer for applications where a cryogenically cooled detector isn't practical. The BGO crystals are rugged, have a high atomic number and density for gamma-ray absorption and are routinely used in the field. A BGO of an acceptable size (5 cm diameter by 5 cm thick) crystal was available for this work. The output from the integral phototube on this detector was input to an Ortec model 113 preamplifier followed by a model 571 linear amplifier.

3. Experimental

The system was setup and tested using a 75 Bq ^{226}Ra standard (extracted and sealed in a counting tube). The short lived progeny in equilibrium in its decay chain through ^{214}Po produces four separate alpha energy peaks from 4.8 to 7.6 MeV. The associated beta/gamma activity in the source is from the decays of ^{214}Pb and ^{214}Bi. Figure 4 shows an oscilloscope trace of the pulse shape outputs for the ^{226}Ra and Figure 5 is the stretched and delayed timing pulses relative to the gamma-ray spectroscopy amplifier input to the MCA. Of the four alpha emitting nuclides in the chain, only ^{226}Ra has a coincident gamma-ray transition (186 keV, 3.6% intensity). The gated and ungated spectra from the sample for equal counting times are shown in Figure 6. In the ungated sample, the low energy portion of the spectrum is dominated by the continuum of Compton scattered photons from beta/gamma emitters in the sample and the external gamma-ray background in the room. The bottom spectrum has the alpha gated spectrum with an increase in the signal to noise ratio on the order of 1000.

A 70 Bq ^{241}Am standard was analyzed in the AGCS, employing the low energy gamma-ray (60 keV, 36% intensity) in coincidence with its alpha decay. A 1.6 MBq ^{137}Cs gamma-ray source was placed on the PERALS® chamber about 5 cm from the LEP detector face to simulate either co-extraction of short lived fission products or data collection in an environment with a high gamma-ray background. Having an

external source was convenient to allow the correct co-incidence timing to be determine for ^{241}Am while the gamma-ray and Compton electrons make a reasonable approximation for beta emitting fission product behavior in the sample. Spectra were again collected in both gated and ungated modes (Figure 7) with the external source producing 2-3% dead time in the MCA running in the ungated mode. The ratio of the 662 keV ^{137}Cs peaks in the ungated versus the gated spectra was about 3500 to 1. The region of the alpha gated spectrum below 40 keV shows coincidental detection of small amounts of the 32-36 keV barium K X-rays from the ^{137}Cs, but the neptunium L X-rays at 14 and 18 keV and the low intensity (2.4%), 26 keV gamma transition are clearly resolved.

Data from the ^{241}Am source was used to test the direct calculation of the disintegration rate (D) from the three measured count rates: the gamma count (Cγ); the alpha count (Cα); and the alpha-gamma coincidence count (Cαγ). These count rate values are related to the corresponding D by the detector efficiencies, εα and εγ, and the decay fractions, I_α and I_γ, by the following three equations.

$C\gamma = \varepsilon\gamma \times I_\gamma \times D$

$C\alpha = \varepsilon\alpha \times I_\alpha \times D$

$C\alpha\gamma = \varepsilon\gamma \times \varepsilon\alpha \times I_\gamma \times I_\alpha \times D$

In its simplest form the D can then be calculated by:

$D = (C\alpha \times C\gamma) / C\alpha\gamma$. Equation (1)

The three measured rates were Cγ = 1.49 ± 0.02 sec^{-1}, Cα = 72.99 ± 0.06 sec^{-1} and Cαγ = 1.56 ± 0.01 sec^{-1}. From equation (1), this gives a measured disintegration rate of 69.9 ± 0.8 Bq which agrees very well with the decay corrected, reference value for this source of 70.1 ± 0.2 Bq.

To evaluate the AGCS's detection sensitivity for low-level measurements, a 2 Bq sample of ^{237}Np was processed for measurement in the PERALS® AGCS. The neptunium was oxidized to the +6 state with potassium bromate in 1 molar nitric acid solution. The aqueous phase was pre-extracted with xylene to remove any bromine generated in the oxidation step and neptunium was extracted into the organic phase (high purity p-xylene) with di-2ethylhexyl phosphoric acid. The air was purged from the sample with argon. The sample was measured several days to weeks after extraction, allowing the ^{234}Pa to grow back in while other granddaughters were not present in significant quantities to further complicate developing appropriate coincidence timing for the gate. The ^{237}Np was measured for periods of up to a day with the external ^{137}Cs source at 5 cm in both gated and ungated modes. The energy region below 200 keV is shown for both cases in Figure 8. The low energy transitions at 29 keV (15%) and 86 keV (12%) are both present in the gated spectrum along with the ^{233}Pa K and L X-rays. A recent study thoroughly evaluated the quantitative analysis of $^{237\text{-}239}$Np by gamma-ray spectrometry [40] but employed a cadmium absorber to reduce the intense activity below ~80 keV. In the AGCS spectrum, the 29 keV line of ^{237}Np is quite distinct even in the presence of the externally induced, gamma-ray background. The 312 keV gamma-ray (39%) from the ^{233}Pa daughter which had grown into the sample was not visible in the spectrum since it is in coincidence with a beta decay. A reagent blank was

generated by extracting a sample of high purity acid, purging and sealing it. The blank was measured for alpha-gamma coincidences several times for 1 to 2 day periods in the ambient background. The low energy region between 20 and 200 keV had negligible activity (1 to 2 counts per day in a typical peak integration region) with the exception of the Pb K X-ray energy at ~75 keV which had a small but discernible peak. This gives detection limits for actinides such as ^{237}Np and ^{241}Am of 1 to 2 mBq.

As several recent studies have examined the use of solid state scintillators or detector for detection of xenon nuclides[34, 41-45], the behavior of the PERALS® coincidence system was also investigated in beta/gamma operation with a ^{133}Xe sample. The SCA window was adjusted to cover the region of the beta/gamma pulses in the pulse shape output, and the ancillary electronics were set to generate the gating signal from the beta pulses. The five day xenon isotope was absorbed on a few activated charcoal beads after purification by gas phase chromatography. The beads were suspended in 1 cm^3 of a commercial liquid scintillation cocktail (Ultima Gold™) and allowed to settle with the xenon transferred into the organic liquid. Xe-133 has an 81 keV gamma-ray (36.9% intensity) and two ^{133}Cs K X-rays around 30 keV. Spectra of the sample with and without beta coincidence gating are shown in the Figure 9. Coincidence counting reduced the Compton scattering background in the low energy region by a factor of ~ 200. The beta-gamma mode of operation would have applications in measuring traces of ^{238}Np [46] in the presence of high concentrations of ^{239}Np as these isotopes have similar half-lives.

Finally, additional studies were performed to test the system with lower resolution, room temperature detector to demonstrate that specific isotopic measurements can still be made with the loss of resolution by performing the measurements in alpha(or beta)/gamma coincidence. The system employed a BGO gamma detector with the ^{226} Ra source in both the gated and ungated mode (Figure 10). Due to the faster timing of the scintillation gamma-ray components, the gate width and delay setting were reduced and dead time of the system was acceptable even though the crystal was larger than desirable for this application. The resolution was only about 10% FWHM but the 186 keV peak is clearly quantifiable in the alpha gated spectrum.

4. Conclusions

The PERALS® has distinct advantages in this application. The analog pulse shape discrimination in the LSC is effective method for eliminating much of the gamma-ray background both internal and external to the sample. This system significantly reduces the need for external passive shielding of the gamma-ray detector, freeing the system to be located in any laboratory rather than being confined to a heavy shielded or specialized counting facility. The sample preparation is simple and straight forward. The LSC based AGCS can process and analyze a sample in under an hour a marked improvement in time and effort compared with conventional alpha spectrometry. Merging the alpha selectivity of the PERALS® and the simplicity of alpha LSC with the high resolution of low energy photon spectrometry of the actinides appears to be a very viable solution for the rapid measurement of actinides and other alpha emitting nuclides.

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Figure 1. Modification of PERALS® sample chamber from aluminum (shown on left) to Bakelite (close up shown on right) to reduce absorption of low energy gamma-rays.

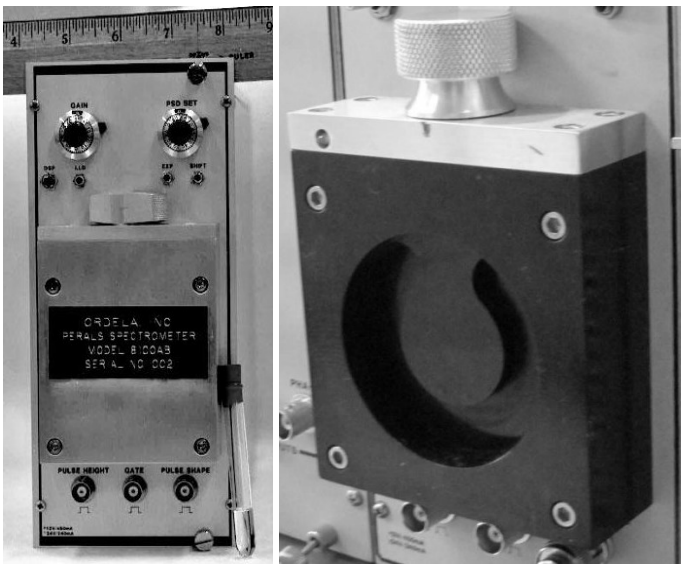


Figure 2. Picture of alpha-gamma coincidence spectrometer. The PERALS® unit if on the left with the sample chamber being the black box located on the front of the module. A side looking high purity germanium detector is located in front of the sample chamber.

Figure 3. Block diagram of alpha-gamma coincidence spectrometer components. The PERALS® output is fed to a single channel analyzer (SCA) to trigger a gate pulse. The high purity germanium (HpGe) spectrometer signal is amplified and input to a multichannel analyzer (MCA) whose acquisition is controlled by the gate pulse.

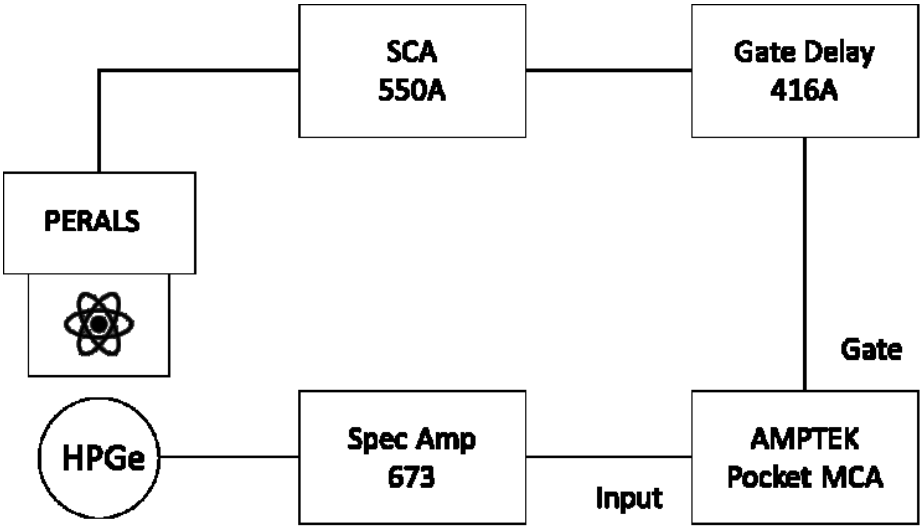


Figure 4. Oscilloscope traces of pulse height and shape from PERALS®. The lower trace shows the instrument response to the energy of the event. The upper trace shows the pulse decay timing for both beta and alpha events with alpha events having the longer lifetime.

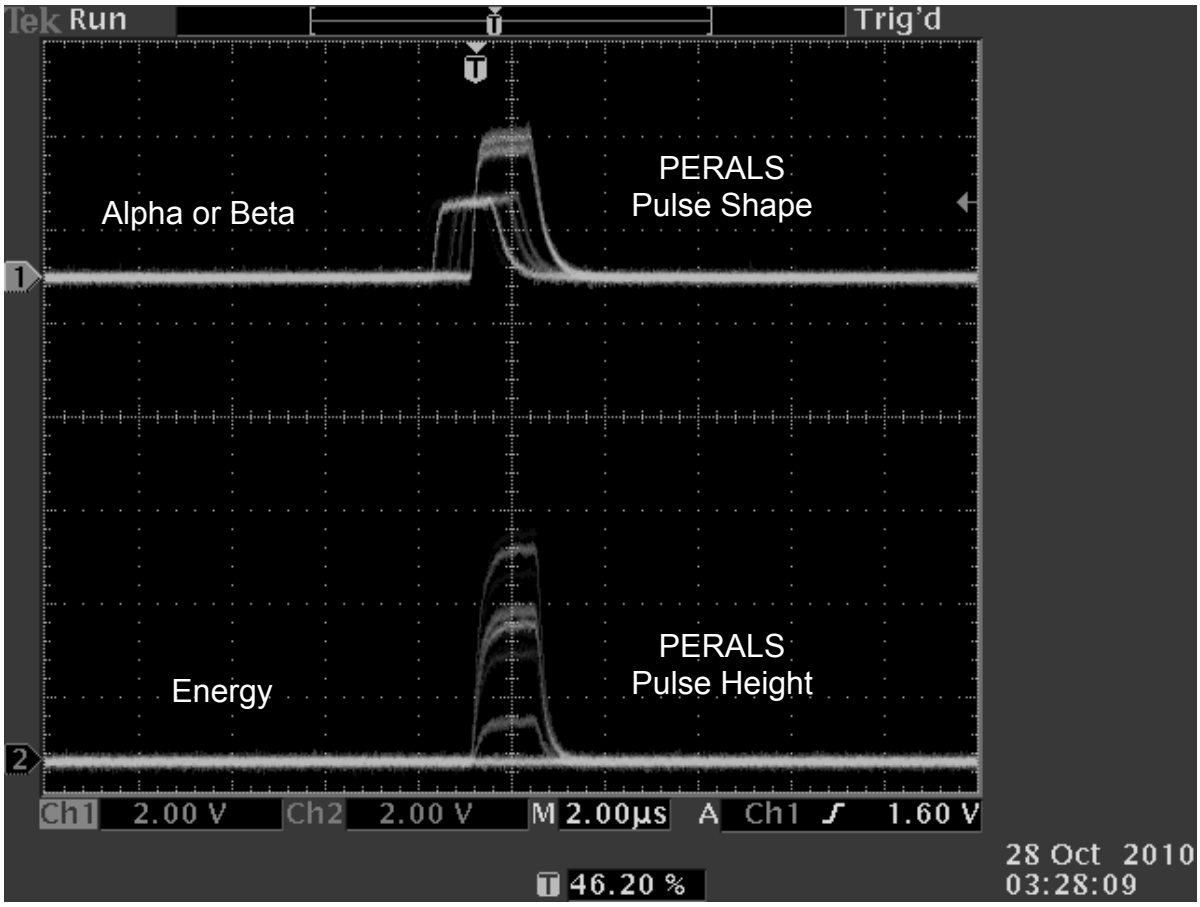


Figure 5. Oscilloscope traces of alpha-gamma coincidence measurements showing steps: (from top to bottom) PERALS® detection pulse shape which discriminates between alpha and beta events, SCA pulse triggered by an alpha event, delay gate pulse, and gamma response.

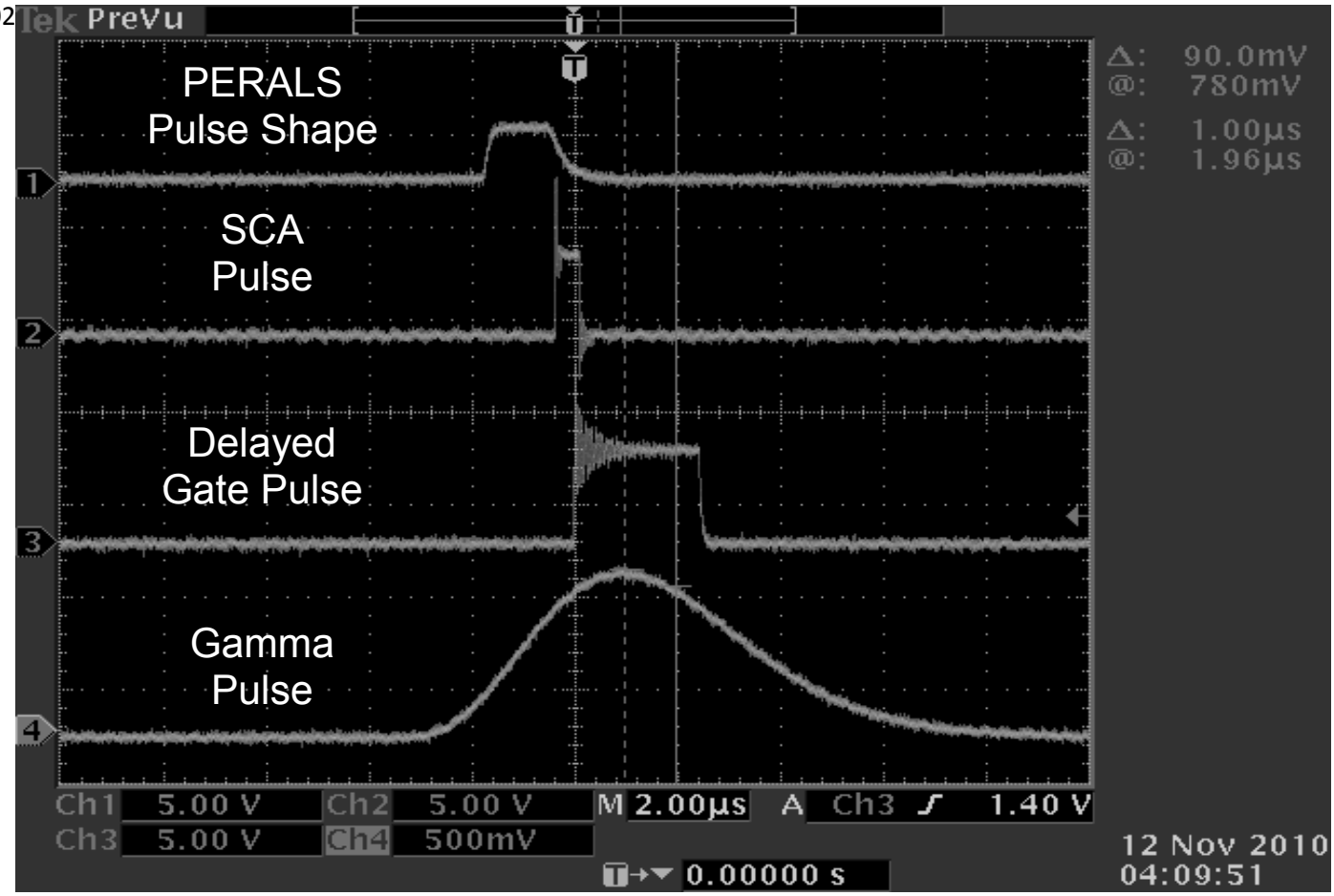


Figure 6. Gamma-ray spectra collected for similar count times by the alpha-gamma spectrometer with (bottom) and without (top) alpha coincidence detection of 75 Bq of ^{226}Ra in the presence of 1.6 MBq ^{137}Cs. Prominent lines in the top spectra from ^{137}Cs at 662 keV, ^{214}Pb at 352 keV and the majority of the Compton continuum are absent when coincidence is employed, revealing the ^{226}Ra peak at 186 keV.

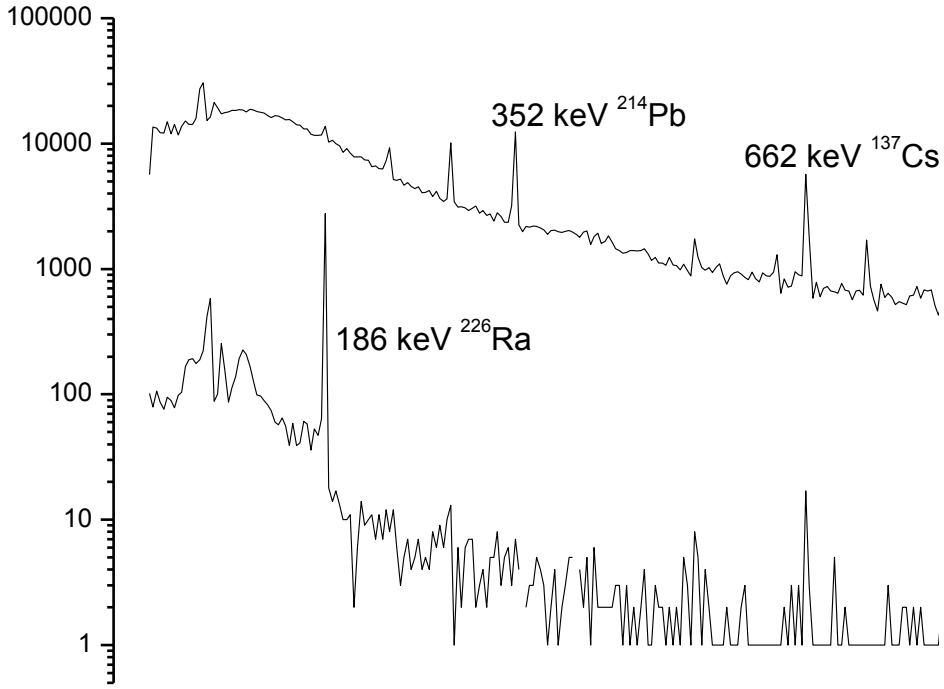


Figure 7. Gamma-ray spectra collected for similar count times by the alpha-gamma spectrometer with (bottom) and without (top) alpha coincidence detection of 70 Bq of ^{241}Am in the presence of 1.6 MBq ^{137}Cs. Prominent lines in the top spectra from ^{137}Cs at 662 keV and the majority of the Compton continuum are absent when coincidence is employed, revealing the ^{241}Am peak at 60 keV.

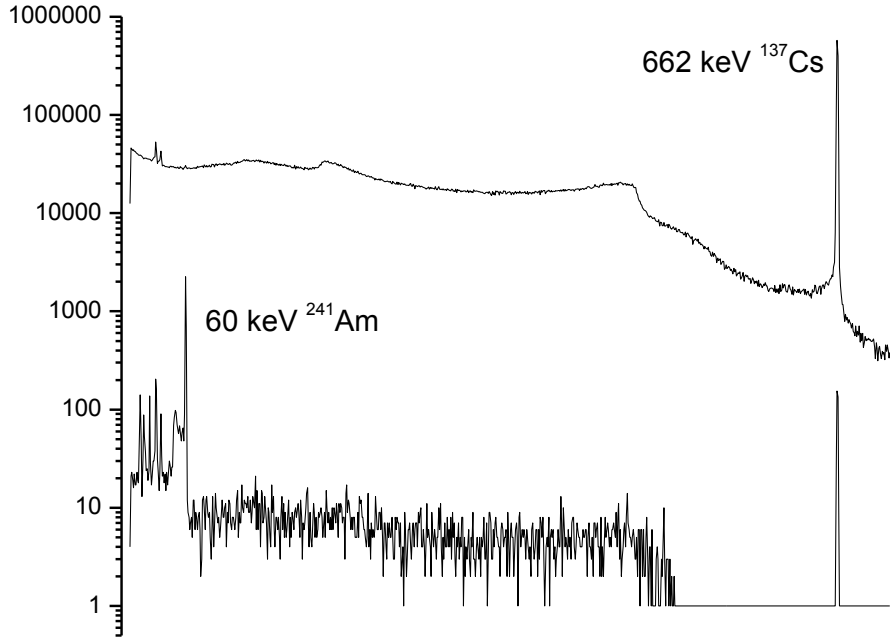


Figure 8. Gamma-ray spectra collected for similar count times by the alpha-gamma spectrometer with (bottom) and without (top) alpha coincidence detection of 2 Bq of ^{237}Np in the presence of 1.6 MBq ^{137}Cs. The majority of the Compton continuum are absent when coincidence is employed, revealing the ^{237}Np gamma-ray peaks at 29 and 86 keV as well as several of the L and K (13.3, 92.2 and 95.8 keV) X-rays of ^{233}Pa.

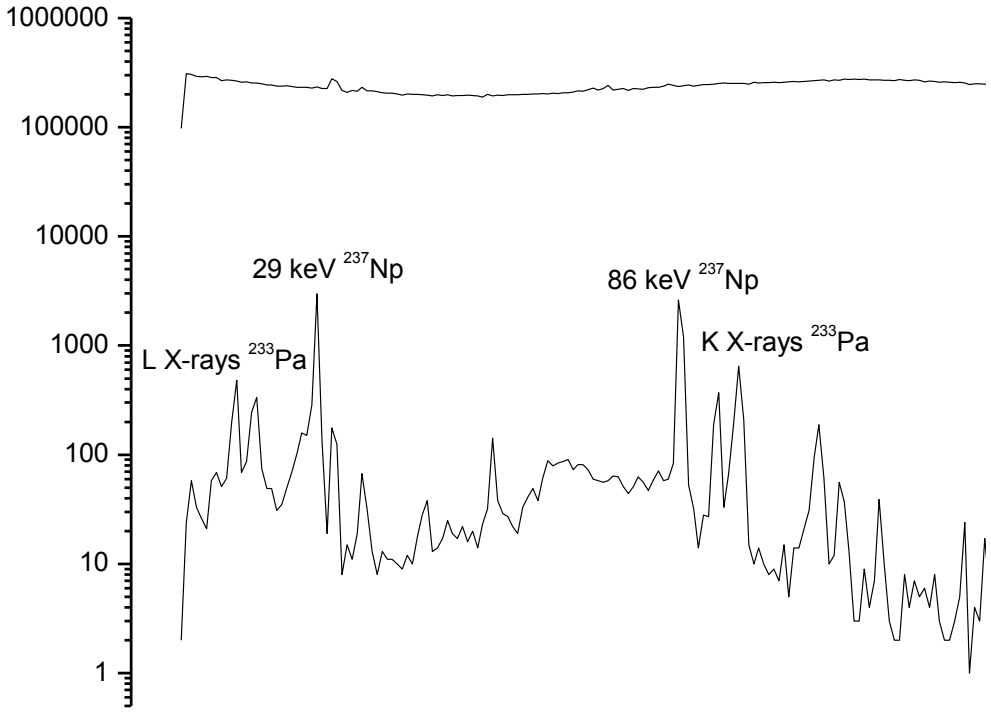
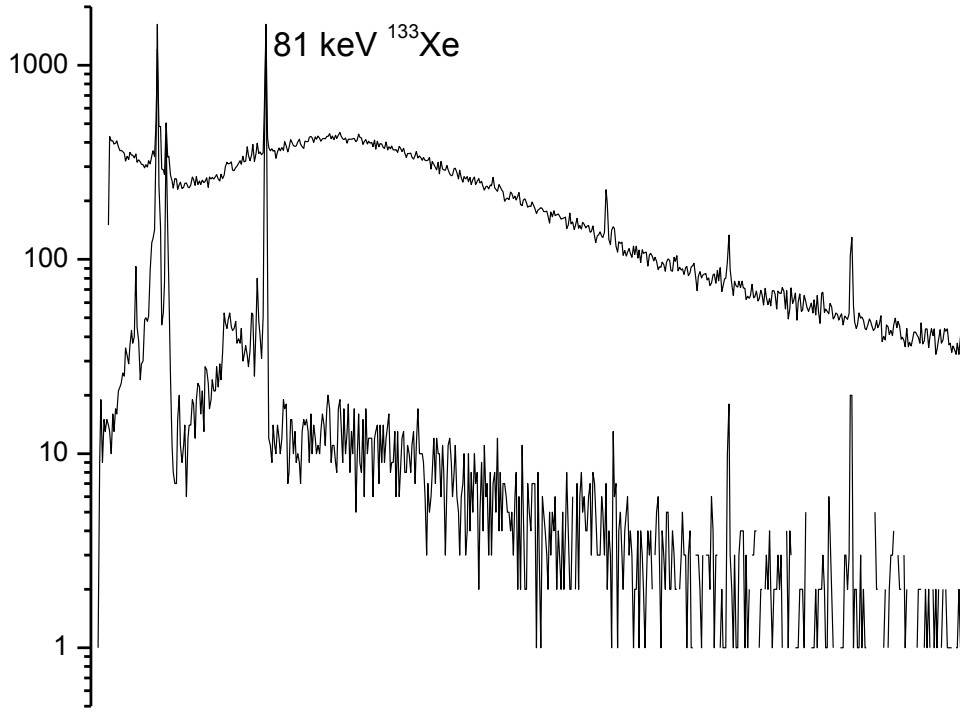
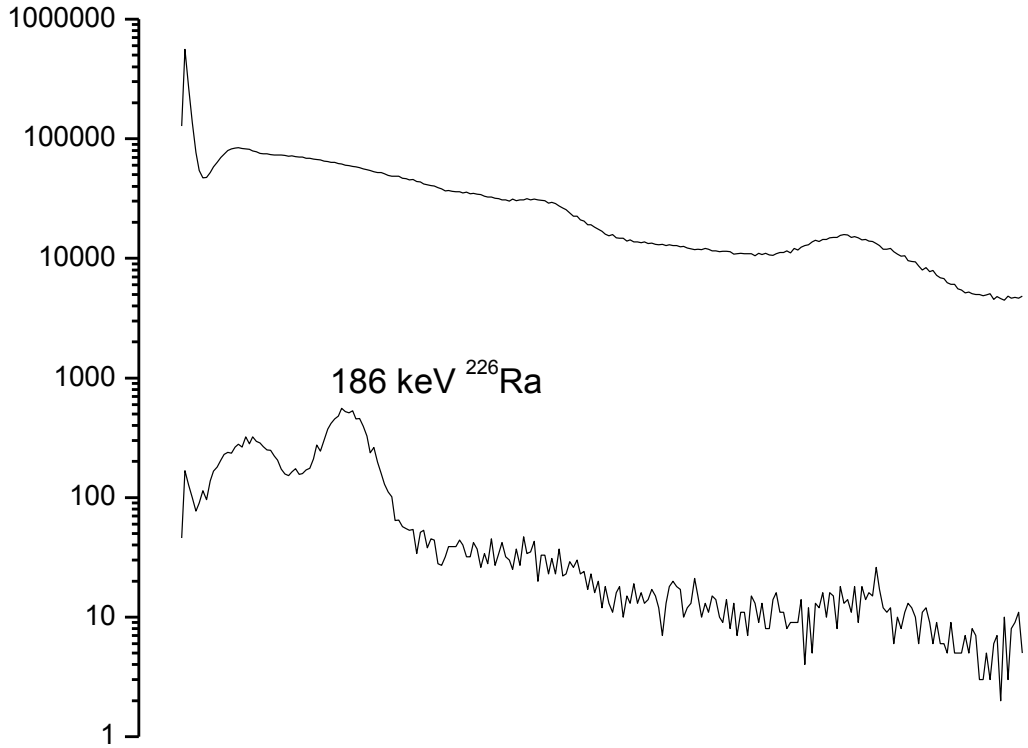


Figure 9. Gamma-ray spectra collected for similar count times by the spectrometer operating in beta-gamma mode with (bottom) and without (top) beta coincidence detection of ^{133}Xe in the presence of 1.6 MBq ^{137}Cs. The majority of the Compton continuum are absent when coincidence is employed, revealing the ^{133}Xe gamma-ray peaks at 81 keV.



433 Figure 10. Gamma-ray spectra collected with (bottom) and without (top) alpha coincidence detection of
434 ^{226}Ra in the presence of 1.6 MBq ^{137}Cs employing a BGO detector.



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