Alpha Coincidence Spectroscopy studied with GEANT4

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***Abstract*—The high-energy side of peaks in alpha spectra, e.g. ^{241}Am, as measured with a silicon detector has structure caused mainly by alpha-conversion electron and to some extent alpha-gamma coincidences. We compare GEANT4 simulation results to ^{241}Am alpha spectroscopy measurements with a passivated implanted planar silicon detector. A discrepancy between the measurements and simulations suggest that the GEANT4 photon evaporation database for ^{237}Np (daughter of ^{241}Am decay) does not accurately describe the conversion electron spectrum and therefore was found to have discrepancies with experimental measurements. We describe how to improve the agreement between GEANT4 and alpha spectroscopy for actinides of interest by including experimental measurements of conversion electron spectroscopy into the photon evaporation database.**

***Index Terms*—alpha spectrometry, GEANT4, conversion electrons.**

I. INTRODUCTION

THE deconvolution of ^{239}Pu from ^{240}Pu and ^{238}Pu from ^{241}Am has always been a challenging task for traditional alpha spectroscopy in environmental monitoring. This challenge is due to the differences of the dominant alpha energies being less than the intrinsic energy resolution of the alpha spectroscopy system. Current deconvolution software for alpha spectrometry can manage with alpha energy overlap but still the ^{239}Pu/^{240}Pu isotopic ratio is reported [1], [2].

We are investigating a coincidence technique (alpha-conversion electron (CE) and alpha-gamma) that could potentially provide a means to simultaneously assay these isotopes individually using traditional detection techniques, e.g. silicon implanted planar technology (PIPS), a multi-wire proportional counter or time projection chamber (TPC). This technique is based on developing an efficient detector apparatus which will allow the coincident detection of the alpha particle from a parent isotope and the conversion electron from the nuclear de-excitation from the daughter isotope. The alpha and CE spectrum is unique to the individual isotopes of interest and provides a means to disentangle the isotopic activity on a faster time scale than the methods currently used (e.g. chemical separation and alpha spectrometry and thermal ionization mass spectrometry).

As a first step to understanding the potential of using this coincidence technique the measured energy spectrum from a large area PIPS detector from ^{241}Am has been analyzed. The experimental conditions have been modeled in GEANT4 (geant4.9.5.p02) [3]. The source term utilizes the G4RadioactiveDecay module and the General Particle Source (GPS). The total energy deposited in the silicon returned by GEANT4 was convoluted with a Gaussian and one exponential [4] after convolution there are discrepancies between the measured and simulated energy spectrum. This discrepancy seems to be caused by inaccurate CE coefficients in GEANT4 PhotonEvaporation database. This database is accessed by G4RadioactiveDecay to calculate the emission probability of gamma and CEs during the relaxation of the daughter isotope from the original nuclear decay.

II. EXPERIMENTAL MEASUREMENTS

A clean, small area (7 mm diameter), thin sample of ^{241}Am ($\sim$0.1 Bq) was prepared and counted with a Canberra commercial grade alpha spectroscopy system [5] which contained a 450 mm^2 area (i.e. A450-18AM) Canberra passivated implanted planar silicon (PIPS) detector. The alpha spectroscopy electronics used here was only sensitive from 3 - 6 MeV which does not allow a direct measurement of the conversion electron spectra. However, it is widely accepted that the structure on the high-energy side distribution of ^{241}Am alpha spectroscopy is mainly due to alpha-CE coincidences [1], [6], [7] and not alpha-photon coincidences. This is due to the high detection efficiency of a silicon detector of an electron as compared to a photon. Therefore, one can indirectly study the CE electron spectrum by measuring the alpha spectrum.

The ^{241}Am sample was counted for $\sim$48 hours at a distance of 7 mm from the PIPS detector. The energy spectra acquired with the ^{241}Am source and the apparatus is shown in Fig. 1. Results of a Gaussian fit to the main alpha peak of ^{241}Am energy spectrum on Fig. 1 yield a FWHM of 19.9 keV and a mean energy of 5502 keV. The discrepancy of the mean alpha particle energy, which should be 5486 keV, is due to an incorrect energy calibration of the electronics. This does not pose a problem for the analysis performed in this work (see Section III).

III. SIMULATIONS AND DISCUSSION

GEANT4 was used to model the experimental conditions and the energy spectrum of silicon. The geometry of the source term was modeled with the General Particle Source

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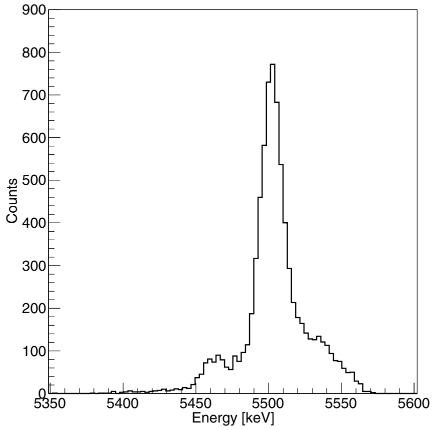

Fig. 1. The experimental energy spectrum from the large area PIPS detector after 48 hours of counting the 0.1 Bq ^{241}Am sample. The FWHM of the 5486 keV alpha from ^{241}Am is 19.9 keV. The structure on the high-energy side of the energy spectrum due to alpha-CE coincidences in the silicon detector.

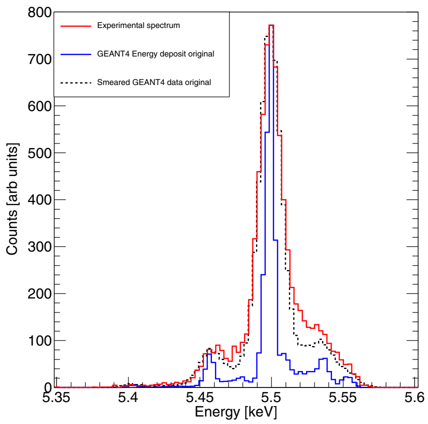

Fig. 2. The results of the GEANT4 simulation total energy deposit with the original Photon Evaporation database. Measured spectrum total energy deposited in the silicon detector, blue line: simulation spectra without resolution convolution, black dotted: convolution of simulation with detector resolution and electronic noise. Both simulated distributions have been peak normalized to the experimental measurement and have the same bin size.

TABLE I

THE MODIFICATIONS TO THE GEANT4 PHOTON EVAPORATION DATABASE. THE FIRST ROW IS THE EXISTING DATA OF THE 59.5 KEV LEVEL TO GROUND TRANSITION FOR ^{237}NP FROM THE PHOTONEVAPORATION2.2 DATABASE. THE 'MODIFIED' ROW CONTAINS THE VALUES WHICH HAVE BEEN INSERTED INTO THE DATABASE AND ACCOUNT FOR THE RESULTS DISCUSSED IN THE TEXT.

Data	**Level (keV)**	**L_1**	**L_2**	**L_3**	**N+**
a93.237	59.54	0.2468	0.2265	0.2284	0.06096
Modified	59.54	0.2496	0.2292	0.0845	0.06331

(GPS) package which allows the creation of complex source geometries with macro-driven commands. The particles of the source term were generated by the G4RadioactiveDecay module. G4RadioactiveDecay is based on data from the Evaluated Nuclear Structure Data File (ENSDF). ^{241}Am alpha decays to different energy levels of the daughter isotope ^{237}Np. The nuclear de-excitation of ^{237}Np, if left in an excited state from the alpha decay, produces either gamma or CE emission. G4RadioactiveDecay computes the emission of these particles by accessing the Photon Evaporation database. This database contains nuclear energy levels, transition probability, total internal conversion probability and subshell CE probabilities amongst other data for a given isotope. It should be noted, that most if not all, of the data contained in the Photon Evaporation database pertaining to CE probabilities is based on theoretical calculations using the measured gamma emission of the isotope. It should also be noted that in order for G4RadioactiveDecay to accurately calculate the gamma emission intensity, "internal conversion" has to be activated.

The result of using the PhotonEvaporation2.2 database with the GEANT4 simulation described above is shown in Fig. 2 for 1×10^6 ^{241}Am nuclear decays. The smearing function has been optimized to reduce the residual between measurement and simulation on the main alpha peak. The smeared simulation does correlate well with the experimental measurement for the alpha peak events at 5.486 MeV. However, there are discrepancies on the high-energy of the main alpha peak. Several adjustments to the simulations were made in an attempt to account for this discrepancy, e.g. the dead layer on the silicon detector, geometrical source parameters and the physics list were all adjusted. However, none of these modifications was found to improve the agreement. Qualitative improvement between simulation and measurements was only found by modifying the photon evaporation database itself. The changes to the database are shown in TABLE I.

Since, 84.6% of the alpha decay of ^{241}Am populates the 59.5 keV level of ^{237}Np, only this row in the photon evaporation database was modified. The modified data for the L_1, L_2, L_3 and N+ shells were calculated based on data from DeVol et al. [8] and BrICC [9]; the M shell was not modified. The work by DeVol et al. reported measurements of CE emission probabilities for ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Am. However, DeVol did not report an emission probability for the L_1 shell of the 59.5 keV level. This is probably due to the fact that the difference in electron energy between the L_1 and L_2 for this level is only $\sim$0.8 keV which would have not been resolvable with the experimental equipment. Therefore, the sum of the emission probability of the L_1 and L_2 shell has been assumed as the reported value of the L_2 in that work. The emission probability for the L_1 shell was calculated by using this sum of probabilities along with the ratio of the L_1 to L_2 shell as calculated by BrICC.

After the database was modified, the simulation was re-run, the energy deposit was smeared with the same function as in Fig. 2 and is shown in Fig. 3. On a qualitative level, the modifications of one energy level of ^{237}Np in the PhotonEvaporation database in GEANT4 have reduced the discrepancies between measurements and simulations of the energy spectrum of a silicon detector. Also, the statistical uncertainty in the simulation is small compared to the discrepancy which existed between the simulated and measured spectra before the modifications to the database were made.

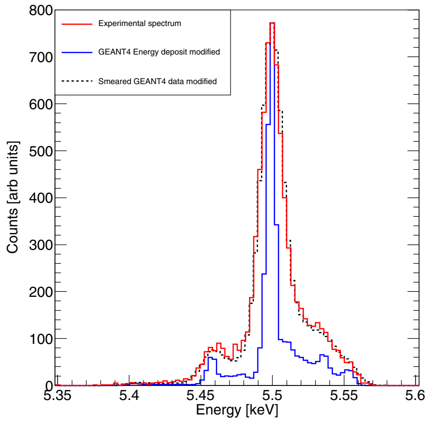

Fig. 3. Same as Fig. 2, but with the Modified CE emission probabilities shown in TABLE I.

IV. CONCLUSIONS

Measurements of the ^{241}Am alpha-particle energy spectrum taken with a silicon PIPS detector revealed structure on the high-energy side of the main alpha peak that are due to alpha-CE coincidences which occur in the detector. Preliminary GEANT4 simulations of the ^{241}Am measurement revealed discrepancies between the simulation and measurement in the high-energy side of the dominant alpha peak, a region in which coincident CE play a significant role. It has been shown that excellent qualitative agreement between simulation and measurement can be achieved by modifying the GEANT4 PhotonEvaporation database. We will proceed with these modifications as measurement data becomes available to support the alpha-CE coincident technique being explored at Pacific Northwest National Laboratory.

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