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Modeling β-γ coincidence spectra of ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe

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In support of the Comprehensive Nuclear-Test-Ban Treaty (CTBT), improvements have been made to the model of the Automated Radioxenon Sampler/Analyzer (ARSA) β-γ coincidence detector for radioxenon monitoring. MCNPX is used to simulate the detector response for all the electrons and photons emitted from ^{131m}Xe, ^{133}Xe, ^{133m}Xe, ^{135}Xe, and ^{137}Cs signals. A MatLab code was written to incorporate the MCNPX results in the calculation of β-γ coincidence spectra. These will aid in the development of the Spectral Deconvolution Analysis Tool (SDAT)[1] and to calibrate β-γ coincidence systems. The models developed for this work include improvements over previous models in their ability to address Compton scattering in the β-cell, and the β-distribution offset in the 31 keV γ-ray region for ^{133}Xe.

Introduction

The Comprehensive Nuclear Test Ban Treaty (CTBT) requires international monitoring for evidence of nuclear weapons testing. Of special interest in the monitoring of atmospheric radionuclides are the noble gas fission products because of their natural physical property of resisting filtering, even when released underground. The radioxenon isotopes ^{131m}Xe, ^{133}Xe, ^{133m}Xe, and ^{135}Xe are radioactive noble gases released in high concentrations in fission and have half-lives on the order of days (Table 1), making them prime candidates for detection.[2] Determination of the ratios of the radioxenon isotopes helps to distinguish between the thermal fission that occurs in a nuclear power reactor and the fast fission that drives a nuclear weapon.[3]

The two measurement methods for atmospheric monitoring that are allowed by the CTBT are high resolution γ-spectroscopy and β-γ coincidence spectrometry. The Automated Radioxenon Sampler/Analyzer (ARSA) developed at Pacific Northwest National Lab (PNNL) is a β-γ coincidence detector designed to measure low-level signatures of atmospheric radioxenon released in nuclear weapons tests.[5] The ARSA detector uses four BC-404 plastic scintillators for β-detection and two NaI(Tl) γ-detectors for γ- and X-ray detection. The gas sample cell is placed within the scintillator which is in turn encased in the NaI(Tl) detectors. The electronics of the ARSA are designed to count in a matrix any coincident pulse in the plastic scintillator and the NaI(Tl) detectors. The tally location in the matrix is given by the β-energy and the γ-energy (Fig. 1). The NaI(Tl) detector is calibrated with a ^{152}Eu test source, and the scintillator efficiency is calibrated through the analysis of the Compton scattering from a ^{137}Cs source.

The goal of this research is to produce accurate models of the individual radioxenon isotopes that will be used in the Spectral Deconvolution Analysis Tool[1] to determine relative isotopic concentrations in real samples. One specific area that needed improvement is the modeling of a 45 keV shift in the β-spectrum of ^{133}Xe caused by a coincident conversion electron with the decay β. A model was also produced for the response expected from Compton scattering of a ^{137}Cs γ-ray in the plastic scintillator and detection of the scattered photon in the NaI(Tl) detector.

Experimental

The experiment consists of first modeling the geometry of the ARSA detector in MCNPX. The ARSA's β-γ coincidence detection ability is modeled by measuring the pulse-heights of γ- and β-interactions in the NaI(Tl) and β-cells individually. The F8, or pulse-height light tally, is used in MCNPX so that both β- and γ-interactions in each detector are counted. Each isotope is modeled individually to get a set of calibration data for deconvolution of the real data that contains all of the isotopes.

Modeling each radioxenon of interest

By producing an accurate set of β-γ coincidence data for the different radioxenons of interest, we can use a deconvolution algorithm like SDAT for determining the amounts of each in a real sample of unknown concentrations. These data sets are created by individually modeling each major γ-ray and each major β-particle that are in coincidence.

For example, the 81 keV γ-ray from the decay of ^{133}Xe is modeled in MCNPX and the resulting m x 1 array representing the spectrum in the NaI(Tl) detector is multiplied by the 1 x n array representing the spectrum produced in the model of the β-particle with an endpoint energy of 346 keV that occurs in coincidence with the γ-ray. The values of m and n are defined by the number of energy bins used for each detector, for this experiment,

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both were set to 255. One histogram is produced modeling the coincidence of the 81 keV γ-ray and the 346 keV β-particle. Another histogram is produced modeling the coincidence of the 31 keV X-ray and a 391 keV max β-particle that is the result of summing in the β-cell of the 346 keV endpoint β-particle and the 45 keV conversion electron associated with the 31 keV X-ray. These two histograms are weighted for appropriate decay probabilities and summed to give the model for β-γ coincidence spectra of ^{133}Xe (Fig. 2). This process is repeated for ^{131m}Xe, ^{133m}Xe, and ^{135}Xe, and the results are shown in Figs 3 and 4, respectively.

Comparison of a real sample containing mostly ^{133}Xe with the model of ^{133}Xe shows the model to be a good approximation. The β-spectra in coincidence with the 81 keV γ-ray in the real sample and the model are compared in Fig. 5. Similarly, the γ-spectrum taken from the β-channel corresponding to an 81 keV β-event in both the real sample and the model are shown in Fig. 6.

Modeling the calibration of the system using ^{137}Cs

The calibration of the β-cell is done using the Compton scattering of the 661.7 keV γ-ray from ^{137}Cs. The Compton scatter of a γ in the scintillator produces a Compton electron that is tallied in coincidence with the resulting lower energy γ-ray. According to the definition of Compton scattering, the resulting electron energy plus the resulting γ-energy should sum to 661.7 keV, leading to a diagonal curve in the β-γ coincidence tally.

In order to efficiently reproduce this reaction, we assume that full energy deposition of the Compton electrons occurs in the β-cell.[6] This assumption allows us to model the counts in the β-cell using a point source of 661.7 keV and the counts in the NaI(Tl) detector using a surface source of varying energy at the interface of the β-cell and the NaI(Tl) detector. Here, the γ-counts were recorded for source energies from 190–660 keV at 10 keV intervals because the minimum energy for a γ-ray in this experiment is 184 keV and corresponds to a scattering angle of 180°. Following with conservation of energy for the scattering event, the maximum energy for the Compton electron is ~478 keV.

This data is used to build a β-γ coincidence array where the initial values are zero except on the rows corresponding to the γ-source energies listed above.

Table 1. Dominant decay modes of the radioxenon isotopes of interest[4]

Decay mode	^{131m}Xe	^{133}Xe	^{133m}Xe	^{135}Xe
Half-life (days)	11.84	5.24	2.19	0.38
Primary γ-ray energy (keV)	163.90	81.00	233.20	249.80
γ-Ray abundance (%)	1.96	37.00	10.30	90.00
X-ray energy (keV)	30.00	31.00	30.00	31.00
X-ray abundance (%)	34.30	48.90	45.70	5.30
β-Particle endpoint energy (keV)		346.00		905.00
β-Particle abundance (%)		99.00		96.00
Conversion electron energy (keV)	129.40	45.00	198.70	214.00
Conversion electron abundance (%)	61.60	54.00	63.50	5.70

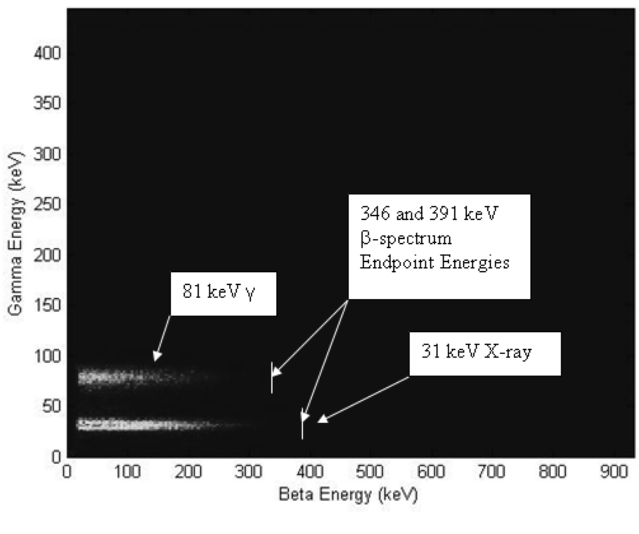

Fig. 1. β-γ coincidence data produced by the ARSA detector. This is a sample consisting of mostly ^{133}Xe

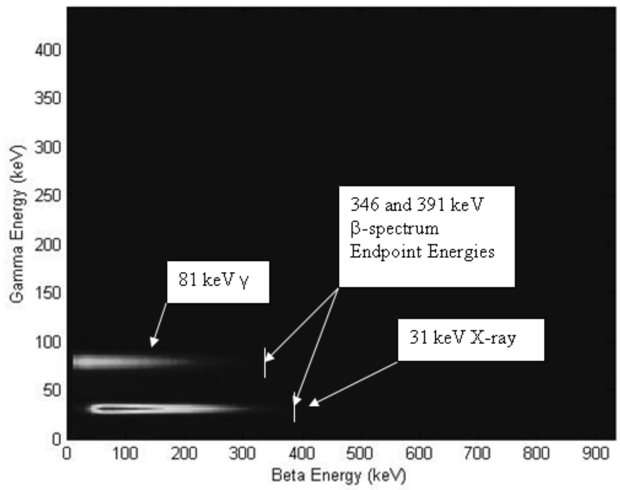

Fig. 2. β-γ coincidence data for ^{133}Xe produced through MCNPX simulation. Note the endpoint of the β-spectrum coincident with the 31 keV X-ray is approximately 45 keV greater than that coincident with the 81 keV γ-ray

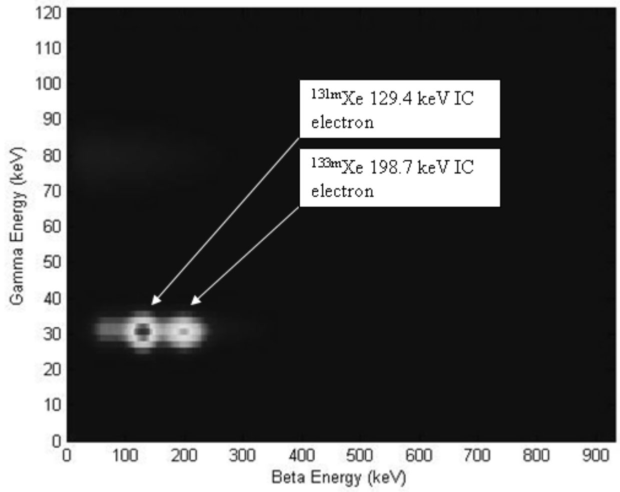

Fig. 3. β-γ coincidence data for ^{131m}Xe and ^{133m}Xe with a low level ^{133}Xe background produced through MCNPX simulation

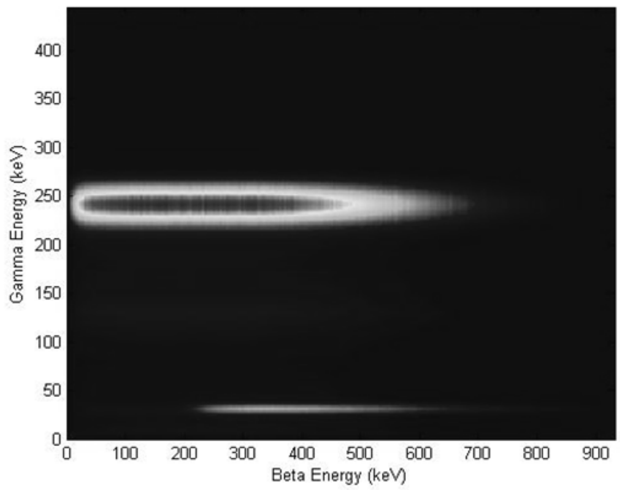

Fig. 4. β-γ coincidence data for ^{135}Xe produced through MCNPX simulation

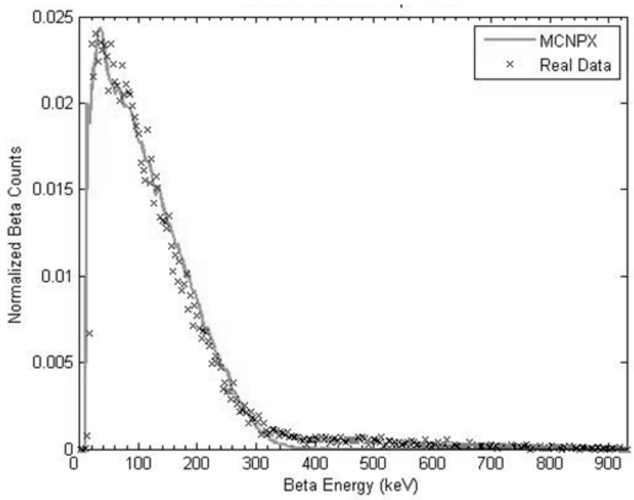

Fig. 5. Plot of the β-spectra in coincidence with the 81 keV γ-ray of ^{133}Xe from the real sample and MCNPX model

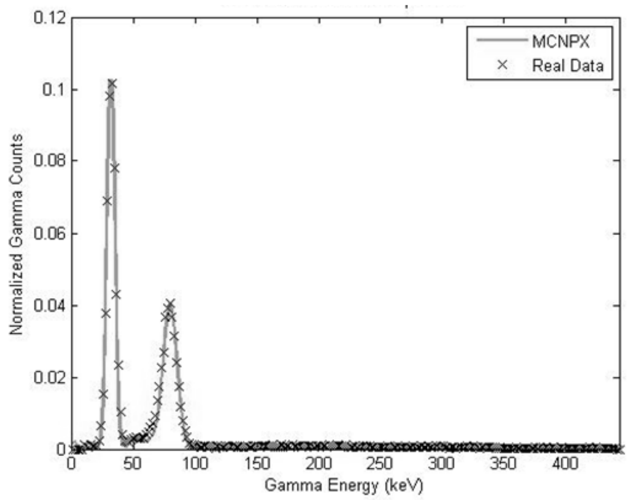

Fig. 6. Plot of the γ-spectra in coincidence with β-particles from the decay of ^{133}Xe

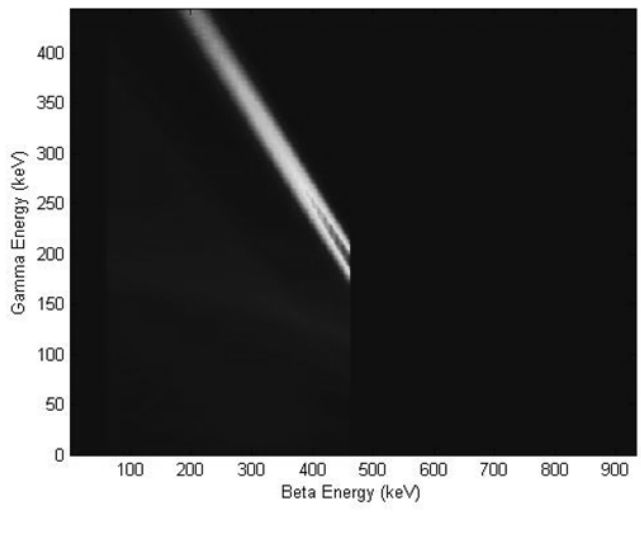

Fig. 7. β-γ coincidence spectrum of the ^{137}Cs calibration source produced through MCNPX simulation

Those rows are filled with the tally from the corresponding γ-source energy multiplied by the tally from the β-cell using the 661.7 keV point source. The rest of the values in the array are filled using linear interpolation across the array and the result is shown in Fig. 7.

Results and discussion

Comparing Figs 1 and 2 shows the model of the decay of ^{133}Xe to be a good representation of the coincident spectrum, including the capability to recreate the 45 keV shift in the β-spectrum coincident with the 30 keV γ-ray. This is evident from the higher energy endpoint of that β-spectrum.

This work shows the capability to correctly model the β-γ coincidence detection of radioxenon isotopes in the ARSA detector and produce accurate data sets for the β-γ coincidence spectra of individual isotopes of xenon. Further modeling is required to accurately predict the spectra produced by the decay of ^{135}Xe due to Compton scattering in the β-cell. This problem is more complicated than the Compton scattering of ^{137}Cs γ-rays because the Compton electron is coincident on the β-cell with the non-monoenergetic β from the decay of ^{135}Xe.

Conclusions

Future work will include purchase of a β-γ coincidence detector similar to what is utilized in an ARSA or SAUNA[6,7] system. We will also construct equipment for creating and separating radioxenons. The radioxenon will be produced through irradiation of uranium in the 1.1 MW TRIGA Mark II research reactor at The University of Texas at Austin. The samples produced will provide greater activity samples for counting in the detector which will give better counting statistics. The goal will be the development of an accurate and reliable deconvolution tool for determining isotopic abundances of radioxenon for use in support of the CTBT.

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