Production of a ^{131m}Xe calibration source for Cyclotron Radiation Emission Spectroscopy and use with Project 8 setup

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This paper outlines the procedure and findings of a project aimed at producing a source of radioactive Xenon. The project was successful in producing a source compatible with the Project 8 setup at the University of Washington. The source had a pressure on the order of 10^{-4} Torr, and we achieved a rate of capture of radioactive Xe of 50%.

Motivation

Project 8 measures the energy of electrons by recording their frequency within a magnetic field via a process known as Cyclotron Radiation Emission Spectroscopy (CRES). The first stage of the Project 8 research has focused on using CRES on gaseous sources to determine the energy of single electrons. Thus far, the project has been successful in determining the energy of electrons between 18 keV and 32 keV with high precision.[1] The ability of the Project 8 experiment to view β decay spectra with such high resolution at these relatively low energies proposes a solution to some shortcomings of the current ^{6}He experimental setup at CENPA.

The purpose of this investigation was to create a gaseous source that produces electrons of higher energy. This source would be connected to the Project 8 setup in order to test the apparatus against higher energy electrons to investigate the potential for using CRES to measure the β decay spectrum of ^{6}He.

This task called for a gaseous source that fulfilled several requirements. First, the source needed to emit monoenergetic electrons. Second, it needed to have a half-life long enough to allow for production in one location and transportation to another before connection to the apparatus: a half-life of more than one day. Third, the source needed to emit electrons with energies in the 30 keV range as well as above 100 keV, to allow for comparison against previous Project 8 results, and to test the system at a higher energy. Finally, the source needed to fulfill the above qualifications at a pressure compatible with the Project 8 apparatus. In the investigation, a pressure on the order of 10^{-6} Torr was attempted. Based on these specifications, we chose ^{131}Xe as the element to use for the investigation.

Production of ^{131m}Xe

Radioactive ^{131}I undergoes gamma decay to produce ^{131}Xe. Approximately 1% of these decays result in a metastable state of ^{131}Xe, referred to as ^{131m}Xe, which has a half-life of 11.84 days.[2,3] As metastable ^{131}Xe decays, it releases conversion electrons. Conversion electrons are created when the photon emitted from the nucleus interacts with one of the electrons in orbit, transferring its energy to the electron and causing the electron to be emitted in place of the photon. This process is shown in Figure 1. In certain situations, ^{131m}Xe decay produces an Auger electron. This occurs when the conversion electron interacts with the electron that fills the orbital hole it caused upon emission. The energies of the conversion electrons and Auger electrons emitted from ^{131m}Xe are shown in Table 1.

Traditionally, radioactive Xenon is produced by neutron irradiation. However, this process generates sources at a pressure greater than 1 atmosphere, which is unsuitable for our purposes. The main challenge we faced was how to isolate the ^{131}Xe from other products generated during the creation of the source in order to maintain a low pressure.

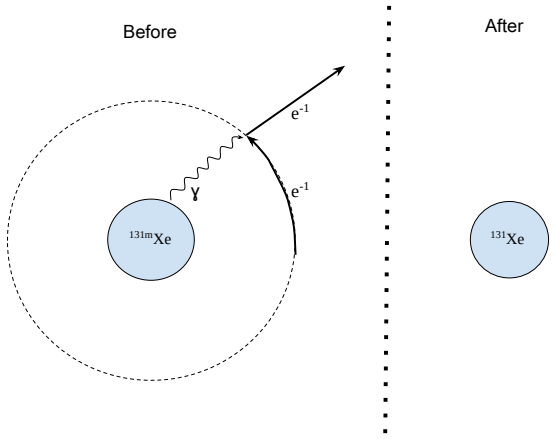

Figure 1. Conversion electron release during gamma decay of ^{131m}Xe.

Shell	Binding Energy	$E_{electron}$ (keV)	Fraction (%)
K	34.9	129	60.7
L-M	3.9	"160"	37.44
Auger Electron		25	4.4

Table 1. Conversion electron energies of ^{131m}Xe decay.

Procedure

A schematic of the experimental setup to produce the ^{131m}Xe source is shown in Figure 2. After assembling the system, the roughing pump and turbo pump were turned on. The system was left pumping overnight with all valves (A, B, and C) open to establish a baseline pressure. We achieved a vacuum of about $1x10^{-6}$ Torr. The following day, valve A was closed, trapping the ^{131m}Xe and ^{131}Xe produced during the decay of the ^{131}I sample within the ^{131}I cell. After five days, valve C was closed and valve A was opened.

^{131}Xe condenses at a temperature of 150K at atmospheric pressure. A copper cooling rod was clamped onto the copper cell and submerged in liquid Nitrogen, causing ^{131m}Xe and ^{131}Xe to condense on the walls of the copper cell. Once the cell was cooled, valve B was closed, the pumps were turned off, and the copper cell was disconnected.

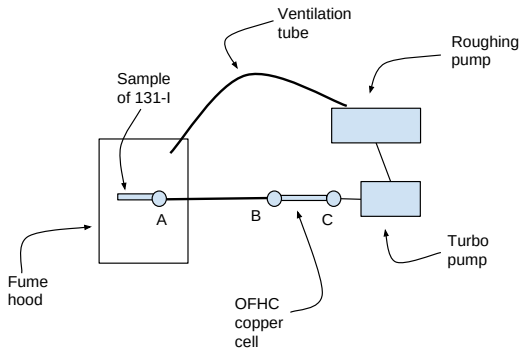

Figure 2. ^{131m}Xe sample capture set-up

In order to determine the pressure inside the copper cell, it was connected to a larger test chamber under high vacuum. A schematic of this setup is shown in Figure 3. The system had been pumping for several days to establish a baseline pressure with valve D open before the copper cell was attached. During this time, the test chamber was wrapped in heat tape, insulated with aluminum foil, and baked. We achieved a pressure of $1x10^{-6}$ Torr in the test chamber. The heat tape was unplugged and the system was allowed to cool to room temperature before connecting the cell containing the ^{131m}Xe was attached. Then, the copper cell was connected and to the system and valve D was closed. Valve B was then opened and the contents of the copper cell were released into the test chamber. The spike in the pressure reading of the test chamber corresponded to the pressure within the copper cell.

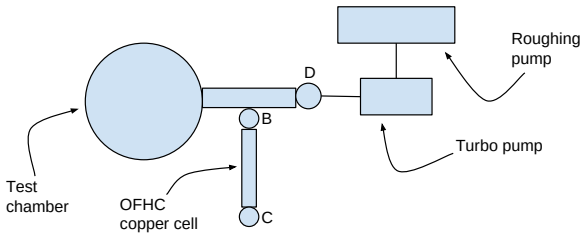

Figure 3. Pressure test set-up

Determination of ^{131}I activity

On June 13, 2016, a sample of the ^{131}I to be used in our investigation was placed in a Ge gamma detector in the Radiation Oncology

department of the University of Washington Medical Center. The activity of the ^{131}I source was calculated using Equation 1.

$$\frac{dN}{dt}_{measured} = \eta \frac{dN}{dt}(Fraction)$$

Equation 1

In Equation 1, $\frac{dN}{dt}_{measured}$ represents the number of counts per second measured at a given energy level, the $Fraction$ factor is the probability that the released gamma particle will have that energy, and η represents the efficiency of the Radiation Oncology department's Ge detector at that energy. η was calculated using the equation $\eta = \eta_{Ba} e^{-\mu x}$. Here, η_{Ba} is the efficiency of the Radiation Oncology department's Ge detector for the ^{133}Ba source at the chosen energy level, μ is the attenuation due to the copper cell, and x is the thickness of the walls of the copper cell, which in our investigation was 0.18 cm.

The energy of the particle released during the decay of ^{131}I to 131Xe is about 364.5 keV, close to the ^{133}Ba decay energy of 356 keV. The calculations for the efficiency of the detector were therefore done using data from that energy level. η_{Ba} at 356 keV was determined to be 0.0397%, and was calculated by multiplying the measured activity at that energy, 9.328 Bq; and the probability of decay at that energy, 62.05%.

For a measured activity of 1317.723 Bq at 364 keV, a probability of this energy decay of 86%, and η evaluated as described above, the activity of Iodine on June 13 was calculated to be 4.42 x 10^6 Bq.

Creation of ^{131}I source for experiment

A Silver Nitride zeolite was included in the cell containing the ^{131}I sample, as well as in the tube between valves A and B in Figure 2. The zeolite was intended to absorb the ^{131}I and other materials present in the system but leave the ^{131}Xe largely unaffected.[4] This served to decrease the impurities within our source, and to contain the ^{131}I within the fume hood.

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Determination of number of ^{131}Xe atoms expected in cell

The activity of ^{131}I on the day of our experiment was calculated using Equation 2, where $\frac{dN_{June\ 13}}{dt}$ is the activity of ^{131}I on June 13; λ is $\frac{\ln(2)}{8.0252}$, 8.0252 being the half-life of ^{131}I; and t is the elapsed time in days between June 13 and the day of the ^{131m}Xe sample extraction. An activity of 6.06 x 10^5 Bq was calculated for the ^{131}I source.

$$\frac{dN}{dt} = \frac{dN_{June\ 13}}{dt} e^{-\lambda t}$$

Equation 2

Calculating the expected number of ^{131}Xe atoms present within our OFHC copper cell was made more complicated by the fact that the ^{131}I source's activity was changing throughout the decay process. The activity of the ^{131}I source at a given time t can be described by Equation 3, where $\frac{dN_{0_I}}{dt}$ is the initial activity of ^{131}I that was measured on June 13.

$$\frac{dN_I}{dt} = e^{-\lambda_I t} \frac{dN_{0_I}}{dt}$$

Equation 3

The activity of ^{131m}Xe at a given time t is given by equation 4, where N_{xe} is the number of ^{131}Xe atoms.

$$\frac{dN_{Xe}}{dt} = 0.01 \frac{dN_I}{dt} - \lambda_{Xe} N_{xe}$$

Equation 4

Substituting Equation 3 for $\frac{dN_I}{dt}$, this equation becomes equation 5.

$$\frac{dN_{Xe}}{dt} = 0.01 e^{-\lambda_I t} \frac{dN_{0_I}}{dt} - \lambda_{Xe} N_{xe}$$

Equation 5

In order to solve Equation 5, a solution of the following form is proposed in Equation 6.

$$N_{Xe} = A e^{-\lambda_I t} + B e^{-\lambda_{Xe} t}$$

Equation 6

The first derivative of this proposed solution is shown in Equation 7.

$$\frac{dN_{Xe}}{dt} = -\lambda_I A e^{-\lambda_I t} - \lambda_{Xe} B e^{-\lambda_{Xe} t}$$

Equation 7

Substituting Equation 6 for N_{Xe} in Equation 5, and setting Equation 5 equal to Equation 7, one can solve for A, as follows.

$$-\lambda_I A e^{-\lambda_I t} - \lambda_{Xe} B e^{-\lambda_{Xe} t} = 0.01 e^{-\lambda_I t} \frac{dN_{0_I}}{dt} - \lambda_{Xe}(A e^{-\lambda_I t} + B e^{-\lambda_{Xe} t})$$

$$A = \frac{0.01 \frac{dN_{0I}}{dt}}{\lambda_{Xe} - \lambda_I}$$

This leads to a simplified version of Equation 7, shown in Equation 8.

$$N_{Xe} = \frac{0.01 \frac{dN_{0I}}{dt}}{\lambda_{Xe} - \lambda_I} e^{-\lambda_I t} + B e^{-\lambda_{Xe} t}$$

Equation 8

Assuming that at time $t = 0$, there were zero atoms of ^{131}Xe inside the cell, $B = -A$, and Equation 8 becomes Equation 9.

$$N_{Xe} = \frac{0.01 \frac{dN_{0I}}{dt}}{\lambda_{Xe} - \lambda_I} (e^{-\lambda_I t} - e^{-\lambda_{Xe} t})$$

Equation 9

The ^{131}I sample decayed for a period of five days before it was released into the copper cell. Using Equation 9, I calculated that 1.823 x 10^{11} atoms of ^{131}Xe should have been produced.

Determination of expected pressure based on expected values

Given a cell volume of 4.90 cm^3, and assuming that all of the decays were captured by the copper cell, the pressure in the cell was can be calculated using the Ideal Gas Law. I calculated an expected pressure within the cell of 1.170 x 10^{-8} Torr. The test chamber was approximately 3000 times larger than the copper cell, meaning that the cell's pressure was expected to register as mid 10^{-12} Torr within the test chamber.

Measurement of ^{131m}Xe activity

Before connection with the test chamber, the ^{131m}Xe sample was placed in the Ge detector in room 106 at CENPA. η was again calculated using the equation $\eta = \eta_{Ba} e^{-\mu x}$. In this case, η_{Ba} is the efficiency of the Ge detector in CENPA room 106 and was calculated to be 0.908%. This calculation is shown in Appendix A. This yields a value of 0.793% for η. Using Equation 1, with a measured activity of 0.955 Bq and a fraction of 1.95%, the activity of the cell at 164 keV was measured to be 617.721 Bq.

This activity indicates that there were 9.117 x 10^8 ^{131m}Xe atoms in the cell. However, this measurement only reflects the 1% of atoms that are in the metastable state, so the total number of ^{131}Xe atoms in the cell was 9.117 x 10^{10}.

Determination of expected pressure based on measured values

Using the ideal gas law and the same specifications as the calculation of expected pressure based on expected values, the pressure in the cell due to the ^{131}Xe atoms should have been 5.851 x 10^{-7} Torr.

Conclusions

When the sample of ^{131m}Xe was connected to the test chamber, a pressure spike of 1.0 x 10^{-6} Torr was seen. This translates to a pressure inside the cell on the order of 10^{-4} Torr. This pressure is higher than the expected value, indicating that the cell contained material other than the ^{131}Xe. Although the pressure within the cell was found to be higher than the expected value, it would still be compatible with the Project 8 setup.

We expected to produce 1.823 x 10^{11} atoms of ^{131}Xe, but we measured only 9.117 x 10^{10} atoms. This means that we captured 50% of the produced ^{131}Xe atoms inside the copper cell. In order to

improve our cooling system to maximize the number of atoms that are captured in the cell in the future, we are planning to improve the fit of the cooling rod around the copper cell, which would cool the cell more effectively.

Appendix A: Efficiency of Ge Detector in CENPA Room 106

In order to understand how effective our system was at capturing the ^{131m}Xe and ^{131}Xe, the cell was placed in the Ge detector in room 106 at CENPA and the activity at 164 keV was measured. The efficiency of the detector needed to be calculated at many different points within the detector cavity, since the cell extended several inches on either side of the detector. The calculation was done using equation 3. $\frac{dN}{dt}_{measured}$ is the activity registered by the detector at the given energy level, $\frac{dN}{dt}$ is the ^{133}Ba activity calculated for the day of testing based on the printed activity on the source's packaging, and $Fraction$ is the probability that a decay will occur at the given energy level.

$$\eta_{Barium} = \frac{\frac{dN}{dt}_{measured}}{\frac{dN}{dt}(Fraction)}$$

Equation 3

The efficiency curve calculated at various points within the detector cavity is shown in Figure 4.

The average value of these efficiencies was used in the calculation of the activity of ^{131m}Xe within the cell. This value is 0.908%.

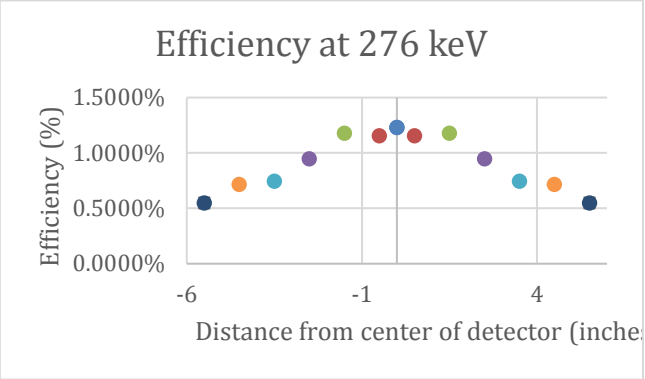

Figure 4. Efficiency of Ge detector at CENPA vs. distance from center of detector

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[1] Project 8: Neutrino Mass Experiment, "About Project 8," http://www.project8.org/about.html

[2] Yu. Khazov, I. Mitropolsky, A. Rodionov, Brookhaven National Laboratory, Nuclear Data Sheets 107, 2715 (2006)

[3] Yu. Khazov, I. Mitropolsky, A. Rodionov, Brookhaven National Laboratory, Nuclear Data Sheets 107, 2715 (2006)

[4] Wang, Wei-Hsung, PhD thesis, Purdue University. 1-3, 19-22 (2000).