

Towards the Direct Observation of Xenon-135 Poisoning in a Zero-Power Reactor via Gamma Spectroscopy

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Abstract—Xenon-135 (Xe-135) is a high-yield fission product with high neutron-capture cross section. It is a commonly encountered reactor reactivity poison. Direct observation of Xe in reactors typically relies on off-gas measurement techniques, which can be challenging or costly to implement and often require high-flux reactors. Gamma spectroscopy of irradiated U or Pu samples provides an alternative method for obtaining fission yields and population estimates. When predicting the Xe populations of a given reactor, we rely on calculations and models to estimate the instantaneous reactivity worth and post-shutdown dynamics of Iodine-135 (I-135) and Xe-135 decay that leads to the so-called ‘Xenon pit’. In this work, developing towards a direct estimate of the total Xe-135 population in an operating reactor, we measured the gamma-ray emissions of Xe-135 and I-135 emanating from the CROCUS zero-power reactor using a high-purity germanium detector in an irradiation channel. CROCUS is a uranium fueled, light water moderated, zero-power reactor operated by the Laboratory of Reactor Physics and Systems Behavior at EPFL, Switzerland. We placed an Ortec GEM-15180-P detector in the irradiation beam port of CROCUS at roughly 4.3 meters from the core center. We measured the gamma ray spectra using a CAEN DS5730 digitizer to obtain gamma-ray time stamps and pulses for the start-up, at-power operation for 1 hour at 20 W, and subsequent shutdown of the reactor for 100 hours. At power, the spectrum is dominated by the gamma ray emission from fission and neutron capture in water and aluminum. Following shutdown, the spectrum transforms, displaying a dense set of characteristic line emissions. To observe Xe-135, we used its 250 keV emission, whilst for I-135 we observed the 1260 keV emission. We binned the spectra in time for 60-minute intervals to accumulate sufficient counts under the photopeak of interest. The time dependent Xe-135 population was observed to follow the expected ‘Xenon pit’ shape, i.e. we were able to directly observe the Xe-135 and I-135 populations in the reactor. These preliminary data are currently only processed into count rates over time. Future work includes determining the detector efficiency in counts per emission in the fuel to finally estimate the whole-core Xe-135 and I-135 populations. Given the published IRPhE geometry of CROCUS, this dataset holds potential as a validation benchmark for reactor poison prediction models.

Index Terms—Gamma spectroscopy, Nuclear Research Reactor, Xenon-135.

I. INTRODUCTION

NEUTRON-induced fission produces prompt neutrons that, given suitable reactor geometry, can sustain a chain reaction, whose rate is controlled by inserting neutron absorbers—control rods or soluble poisons—to maintain safe, stable operation. In addition to engineered poisons, the fission product ¹³⁵Xe (thermal absorption cross section $\sim 2.6 \times 10^6$ b), also generated via decay of the fission product ¹³⁵I, exerts a strong, time-dependent reactivity feedback that gives rise to the so-called “xenon pit” or “iodine pit” following power changes [1], [2].

Conventional reactor neutron detectors are used to infer ¹³⁵Xe levels indirectly via calculations, whereas in-situ gamma-ray spectroscopy can identify the 249.79 keV line of ¹³⁵Xe directly. High-purity germanium (HPGe) detectors fulfill the energy resolution requirements to detect this emission, but require fast digitizers with pile-up rejection and operation in low-flux (zero-power) environments to avoid damage and saturation [3], [4]. Recent experiments using HPGe detectors in low-power reactors include measurements at LR-0 [5] and CROCUS [6]. In both, the integral gamma ray spectra of long multi-hour measurements were analyzed.

Herein, we report time-resolved HPGe measurements of ¹³⁵Xe and ¹³⁵I after shutdown of the CROCUS zero-power reactor after a 20 Wh irradiation. Using fast digitizer electronics, we achieve $\sim 2 \times 10^4$ cps immediately after shutdown, yielding sufficient statistics to track xenon dynamics. In Section II we detail the setup and analysis and in Section III we present the results.

II. METHODS

A. Decay-Chain Kinetics

After shutdown of the reactor, the iodine ($I(t)$) and xenon ($X(t)$) populations follow these equations:

$$I(t) = I_0 e^{-\lambda_I t}, \quad (1)$$

$$X(t) = \frac{\lambda_I I_0}{\lambda_X - \lambda_I} (e^{-\lambda_I t} - e^{-\lambda_X t}) + X_0 e^{-\lambda_X t}, \quad (2)$$

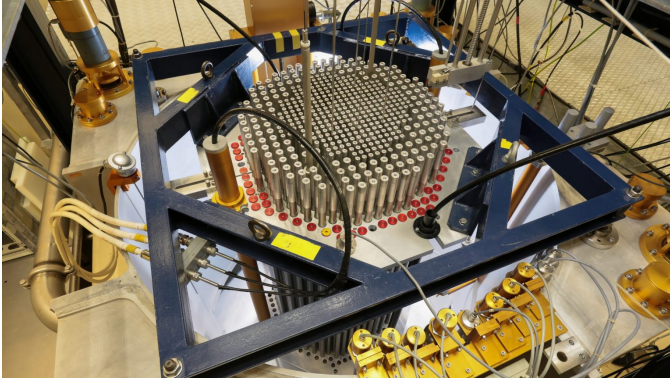


Fig. 1. Picture of the CROCUS zero-power research reactor, uranium fueled and water moderated with a maximum power of 100 W [14].

where I_0 , X_0 are the populations of ^{135}I and ^{135}Xe at the moment of shutdown [7]. As ^{135}Xe is produced by ^{135}I even after shutdown, the resulting transient equilibrium leads to an increase in population at first for ca. 11 hours and then to a decay.

B. CROCUS Reactor

Measurements were carried out on the EPFL CROCUS reactor (100 W, max. thermal flux $10^9 \text{ n/cm}^2/\text{s}$), a light-water-moderated, zero-power research facility documented in the IRPhE handbook [8] (see Figure 1). CROCUS has been extensively studied via other gamma-ray experiments [9]–[11], yet to date no time-resolved gamma spectroscopy was attempted. The core ($\varnothing 58 \text{ cm} \times 100 \text{ cm}$) contains 336 UO_2 rods (1.8 wt.% ^{235}U , 12.6 mm diameter, 1.8 cm pitch) and up to 180 U-metal rods (0.9 wt.% ^{235}U , 17 mm diameter, 2.9 cm pitch) in aluminum cladding. The active height is 100 cm; the core sits in a 130 cm-diam. aluminum tank (1.2 cm wall). Reactivity is adjusted via water-level control with 0.1 mm precision (0.4 pcm). The neutron flux at the detector location was estimated to be $\sim 10^4 \text{ n/cm}^2/\text{s}$, which should not significantly damage the detector [12], [13].

C. Detector and Data Acquisition

We used an Ortec GEM-15180-P coaxial HPGe (57.2 mm x 50 mm) in PopTop configuration, operated at 3.6 kV with 0.235 % resolution (FWHM) at 662 keV. Signals were preamplified by a Canberra DSA-1000 and digitized by a CAEN DS5730 (DPP-PHA firmware) in list mode, with readout via USB and CoMPASS software [15], [16]. The detector was placed 4.3 m from core center, 1 m behind the beam-channel opening [17], see Figure 2. A 100 keV lower-level discriminator suppressed low-energy background, yielding $\sim 2 \times 10^4 \text{ cps}$ immediately after reactor shutdown.

D. Energy Calibration and Peak Fitting

Energy was calibrated assuming a linear relation,

$$E = Ax + B,$$

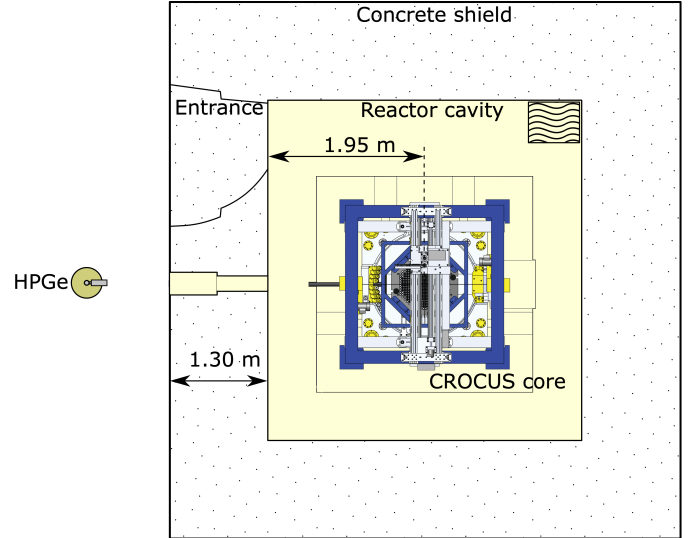


Fig. 2. Top view of the experimental setup: The HPGe was set at ca. 1 meter from the external surface of the reactor shielding, a total of 4.25 meters from the reactor core center.

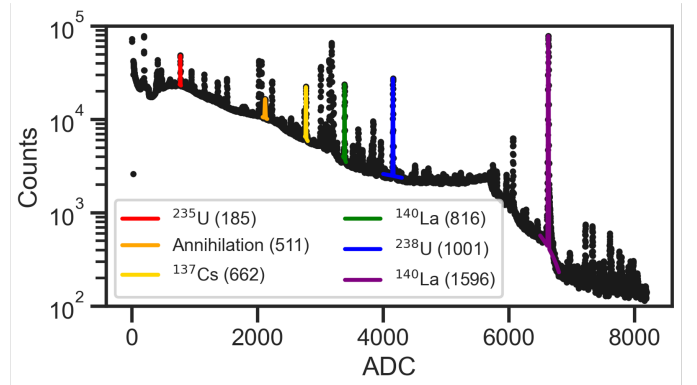


Fig. 3. Energy calibration of the gamma spectrum was achieved by using prominent emission lines identified in previous experiments [6].

where x is the ADC channel. Six photopeaks between 185 and 1600 keV (see Figure 3) were fitted with

$$G(E) = A \exp\left[-\frac{(E-\mu)^2}{2\sigma^2}\right] + aE + b$$

using trust-region nonlinear least squares. The fitted μ was then used for linear fitting across the ADC range. The calibration residuals have $\text{RMSE} \leq 0.4 \text{ keV}$, confirming a linear response in the examined region.

E. Spectrum treatment

Each gamma ray pulse was registered with a time stamp and a pulse integral. The histograms for time-dependent analysis were created with a 0.3 keV bin width and 1-hour wide time bins. The thus created gamma ray spectra were processed with a SNIP-type algorithm [18] to remove the background under photopeaks (see Figure 4). The peaks of interest (249.8 keV for Xe-135, 1260.5 keV for I-135) were then integrated via trapezoidal sum of the histogram in a three sigma range. For the peaks of interest herein, no neighboring peaks or overlapping peaks of concern were encountered.

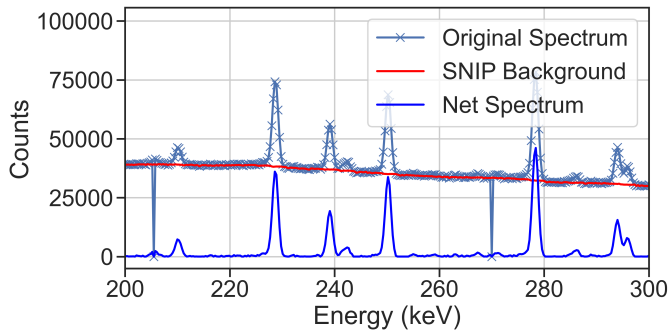


Fig. 4. Example of the gamma spectrum background removal via SNIP algorithm [18].

III. RESULTS

In Figure 5 we display examples of the observed gamma ray spectra for different times after shutdown of the reactor. In the first hour immediately after shutdown the spectrum is dominated by delayed neutron-induced fission gamma rays and the 2.22 MeV emission from $H(n,\gamma)$ and its single- and double-escape peaks. It is worth noting that 24 hours after shutdown, the count rate is higher than in the first hour. This is due to the fact that in the first 7 hours, the count rate was so high, that pile-up rejection and dead-time dominated and most pulses remained unregistered. In order to measure isotopes with short half-lives, one would need to operate at even lower reactor powers prior to shutdown, or change the shielding geometry.

Following the post-processing outlined in Section 2, we show in Figure 6 the time-dependent evolution of the Xe-135 and I-135 peaks. The expected 'xenon pit' shape, peaking at roughly 11 hours is observed, whilst ^{135}I decays exponentially with the expected half life of 6.57 h.

IV. CONCLUSION

In this work, we present the first analysis of time-dependent HPGe measurements of the CROCUS zero-power reactor immediately after shutdown until 90 hours thereafter. We specifically investigated the emissions of Xe-135 and I-135 at 249.8 keV and 1260.5 keV, respectively, to directly observe their populations in the reactor. Due to the high gamma-ray flux immediately after shutdown after a 1 at 20 W critical operation, a quantitative estimate of the populations was only possible after some 7 hours after shutdown. The Xe-135 count rates follow the expected transient equilibrium behavior. This observation is a first step towards the determination of the reactor-wide Xe-135 population to provide a new validation dataset for reactor poison predictors and other reactor physics codes.

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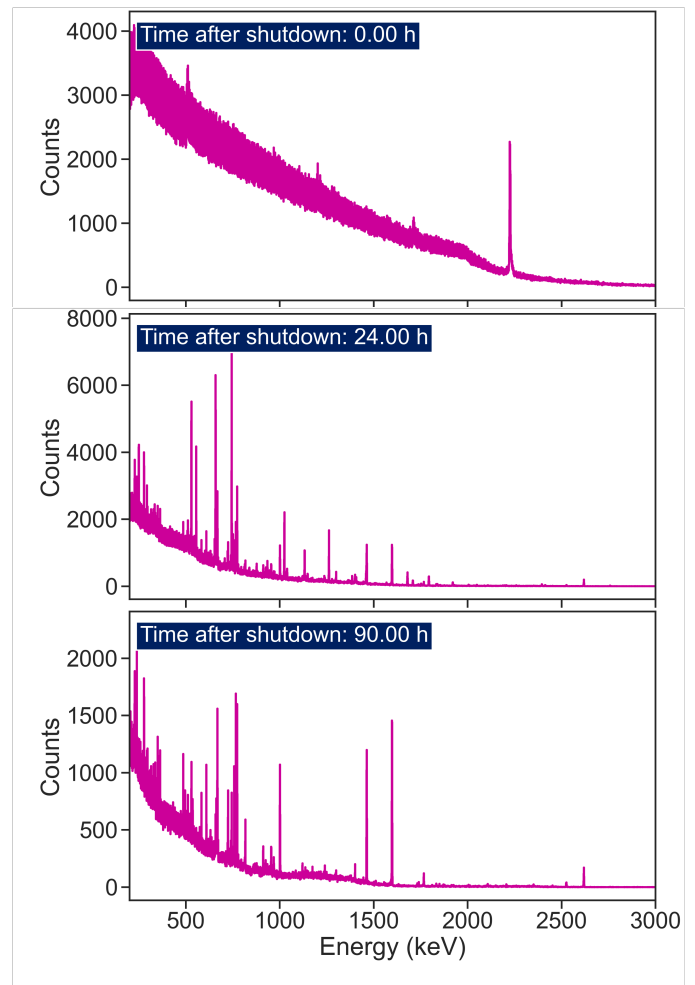


Fig. 5. Examples of measured gamma ray spectra over one hour. **Top:** Right after shutdown. **Middle:** 24 hours after shutdown. **Bottom:** 90 hours after shutdown.

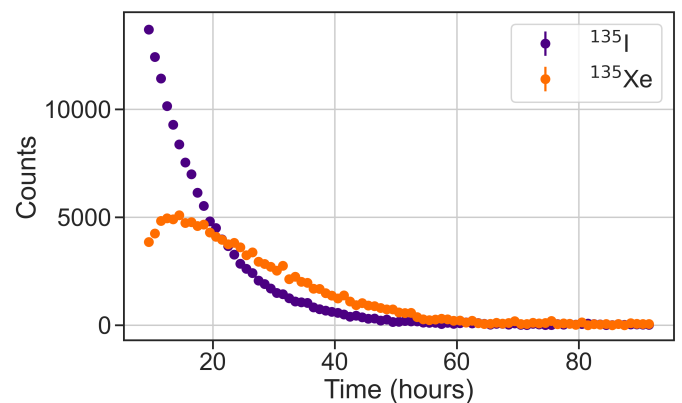


Fig. 6. First evaluation of the count rates of the 249.8 keV and the 1260.5 keV peak time evolutions.

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