Measurement of an Upper Limit of Fission Energy Release in HOLOG using a Germanium Gamma-Ray Detector

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Summary

An upper limit of less than 4 mg TNT equivalent fission energy release from the HOLOG experiment was determined using a germanium γ -ray detector to measure the ratio of selected fission-product and plutonium γ rays. Only three hours of γ -ray data collected immediately after the zero-time were analyzed to calculate the above limit. We found no peaks corresponding to the $^{97}\text{Zr} - ^{97}\text{Nb}$ fission product pair at the gamma-ray energies of $E_\gamma = 743$ keV and $E_\gamma = 658$ keV, respectively. No information on the plutonium isotopic ratios is revealed because γ -ray peaks in the energy region below 100 keV are not observed due to the high absorption in the containment barrier. The measurement is relatively easy to perform and is not subject to false-positive results because specific fission product and plutonium γ ray energies need to be detected.

Measurement of the spectrum of gamma-rays emitted by plutonium and its fission products, if any, from a subcritical experiment (e.g., HOLOG) can be used to inexpensively and reliably obtain a limit of the plutonium fission efficiency (i.e., the fraction of plutonium that fissioned). In this study, we apply such a measurement through the HOLOG containment barrier. The actual quantity we measured is the ratio of fission product γ -rays to plutonium γ -rays. Knowing the time of the experiment, start and stop time of the measurement, and documented fission product yields¹ (e.g., the fraction of fissions that result in an atom of ^{97}Zr), one can convert this ratio to plutonium fission efficiency. However, one needs to make an assumption that the fission is uniformly mixed throughout the plutonium. To the extent that the fission to plutonium ratio varies, the measurement will be less accurate because the detector only “sees” the γ -rays from any fission products and plutonium that are closest to it. It is possible to take data at several positions outside the containment barrier to check for uniformity although for HOLOG a single detector and single counting position were used. The total fission energy release can then be estimated from the measured fission efficiency knowing the mass of plutonium used in the experiments.

One can select from a large number of possible fission products for this measurement. The best choices have a high fission product yield, half-lives comparable to a realistic measurement time soon after the zero time, and have high-energy high-abundance γ -rays per decay. Some choices which meet these criteria are the ^{97}Zr ($t_{1/2}=16.8$ h, $E_{\gamma}=743$ keV) and ^{97}Nb ($t_{1/2}=1.2$ h, $E_{\gamma}=658$ keV) pair

and the ^{140}Ba ($t_{1/2}=12.75$ d, $E_{\gamma}=1510$ keV) and ^{140}La ($t_{1/2}=1.68$ d, $E_{\gamma}=1597$ keV) pair. The mass-97 and mass-140 chains each have high fission branches (~5%). The mass-97 chain half-lives are short enough to permit a high fraction of the decays to be observed but also long enough to allow measurements if data accumulation could not start until reentry of the tunnel was allowed. For these reasons, and to limit additional costs from a more complicated data analysis, we chose to simply use a measurement of the ^{97}Nb - ^{97}Zr γ rays to determine a limit for the plutonium fission efficiency. Gamma rays from these nuclides are normalized to one of the two strongest ^{239}Pu intrinsic γ -rays in the 400-keV energy region, either the 375-keV or 414-keV ^{239}Pu γ -rays.

A high purity germanium (HPGe) detector with a γ -ray detection efficiency of 70% (compared to a 3"x3" sodium iodide (NaI) detector) was used to measure the γ -rays up to 3 MeV from both the ^{239}Pu and the fission products. (NaI and other kinds of γ -ray detectors do not provide sufficient energy resolution.) This detector, surrounded by a 1/4" thick Pb collimator, was placed facing the containment barrier that consists of 1" thick steel and 12" thick concrete. In order to increase the ^{239}Pu detection efficiency after the experiment, we placed the detector as close to ground level as possible assuming that the Pu concentration in the chamber would be highest at the chamber floor, as shown in Figure 1. Signals from the detector were amplified via standard electronics and recorded in a PC-based data acquisition system. (Figure 2) We also programmed the data acquisition software to perform hour-by-

hour histogram dumping so that data can be segmented into one-hour intervals.

Figure 1. The Ge detector was placed as close to the ground level as possible.

Figure 2. A PC laptop based data acquisition system was placed next to the sign-in board outside the zero room.

We collected thirty-six hours of γ -ray background data before the plutonium was moved into the zero room, fifteen hours of γ -ray data after the plutonium was moved into the zero room, and sixty-seven hours of γ -ray data after the zero time. A 15-hour γ -ray spectrum (Figure 3) taken before any Pu was placed in the zero room

was normalized to 3 hours for comparison with the first three hours of data collected after zero time (Figure 4). Figure 5 shows an expanded view of the ^{97}Zr and ^{97}Nb γ -ray energy peak positions from the same three-hour spectrum shown in Figure 4. In all of the collected spectra, the 100-keV region is completely attenuated due to the containment barrier. Therefore, no information on the isotopic composition of Pu that was used in the experiment can be deduced. The strong 375- and 414- keV γ -ray peaks in Figure 4 demonstrate that the Pu indeed scattered all over the zero room after the blast.

Figure 3. Background γ -ray spectrum (collected for 15 hours and normalized to 3 hours for comparison with the 3-hour post-experiment spectrum in Figure 4) before the plutonium was moved into the zero room.

Figure 4. Three-hour γ -ray spectrum collected immediately after the zero time.

Figure 5. Expanded view of the same three-hour γ -ray spectrum shown in Figure 4 showing the expected γ -ray energy peak positions of ^{97}Zr and ^{97}Nb .

Spectra were analyzed using an graphic PC version of GAMANL,² one of the long integrated spectra collected after the zero time was analyzed using MGAH³ to obtain the combined transmission and the

detector efficiency. Prior to the experiment, we have also performed Monte Carlo calculations of the combined transmission and detector efficiency. The simulation results are in good agreement with results from the MGAHI fitting within 10%.

Results from GAMANL show that an integrated $1627 \pm 8\%$ counts under the 375-keV ^{239}Pu peak in the three-hour spectrum that were collected after the zero time. We have also found no evidence for a peak at either the 658- and 743-keV ^{97}Zr and ^{97}Nb peak energies in the same spectrum (expanded region is shown in Figure 5). To estimate upper limits of counts at 658- and 743- keV peaks with backgrounds, we used standard statistical methods⁴ to estimate counts in Gaussian peaks with fixed full-width-half-maximum energy resolution and found upper 3σ limits (99.74% confidence level) of 66 counts and 66 counts for the 658- and 743-keV peaks, respectively. The combined transmission and detector efficiency obtained from the MGAHI fitting is 4.2×10^{-7} for γ -ray energy of 375 keV and 1.5×10^{-6} for gamma-ray energy of 743 keV.

One milligram of TNT equivalent fission energy release will produce 1.4×10^{11} fissions for $n + ^{239}\text{Pu}$. By folding this number with the combined transmission and detector efficiency, the branching of the $n + ^{239}\text{Pu}$ to $^{97}\text{Zr} - ^{97}\text{Nb}$ pair, the half-lives of the pair to the total collecting time, and finally, normalized with respect to the ratio of ^{239}Pu viewed by the germanium detector to the total ^{239}Pu used in the experiment. We obtain an upper limit of 4 mg of TNT equivalent fission energy release from the three-hour spectrum collected immediately after the zero time. We predicted⁵ similar sensitivity prior to the measurement. Our current calculated sensitivity could

be improved by at least a factor of 10 if we can 1) employ shorter data dumping time intervals or collect the data in list mode and 2) analyze the spectra in detail for multiple fission products, especially those with shorter half-lives when the counting takes place through zero time. Then it is possible to select fission products with half-lives of minutes which would completely decay during a short counting time (20% of the ^{97}Nb decayed during the 3-hour counting interval used to calculate the fission efficiency) as well as correspondingly reducing the background. However, because the experimental objectives were met, a more complicated analysis that would result in higher expenses was not performed.

In summary, we have proved that a single shielded HPGe detector outside the containment barrier of a subcritical experiment can be used to measure the plutonium fission efficiency and total fission yield from HOLOG. By carefully choosing the detection geometry, the data acquisition sequence, and the fission product, we are able to achieve a detection sensitivity of better than 4 mg of TNT equivalent fission energy release. To apply this measurement technique to future subcritical experiments, we note that if the containment barriers are substantially thicker (e.g., REBOUND), a re-entrant tube or other means would be needed to prevent increased γ -ray absorption. The measurement is relatively easy to perform and is not subject to false-positive results because specific fission product and plutonium γ ray energies are measured.

¹ T.R. England and B.F. Rider, "Evaluation and Compilation of Fission Products Yields", ENDF-349, LA-XXXX-MS, 1993

² R. Gunnink, J.B. Niday, UCRL-51060, 1997. Graphics and PC port was made by T.F. Wang.

³ MGAHI is currently under development by T.F. Wang. This is a high energy only (> 200 keV) version of MGA . The MGA was developed by R. Gunnink, and Wayne D. Ruhter, UCRL-LR-103220, 1990.

⁴ W.T. Eadie, D. Drijard, F.E. James, M. Roos, B. Sadout "Statistical methods in experimental Physics" pp. 272. North-Holland-Publishing, 1971.

⁵ "Subcritical Experiments," Jason Report, JSR-97-300, MTTRE Corporation, Jan. 21, 1997.