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Evaluation of ^{166m}Ho: In Search of Photonuclear Cross Sections and Trigger-Level Paths

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Abstract—Part of the Army Research Laboratory's battery program is research in the viability of nuclear batteries. Particularly attractive is a nuclear battery that can be stored without loss of energy, and then turned on by some external signal. This paper describes the progress toward that goal.

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INTRODUCTION

The Army Research Laboratory (ARL) is actively exploring advanced isotope batteries. With a suitable radioactive power source, such batteries could enable sensors to operate unattended in remote locations for as long as the isotope's radioactive half-life. Isomers are particularly interesting for such batteries, because external irradiation of an isomer with *x* rays might take the nucleus from the relatively stable isomeric level to another level that is less stable and decays faster. With the proper half-lives of the isomer, such a battery could be inactive for a long time, until it is turned on by irradiation. We started our exploration of isomers with ^{166m}Ho, since this isomer would be desirable for a nuclear battery if it could be activated by X rays.

A standard chemical cell consists of two reactive chemicals separated by a thin layer. Likewise, a cell in an isotope battery has two layers—one contains the radioisotope power source, and the other converts the radiation to electricity. A good converter is silicon carbide (SiC), a wide-bandgap semiconductor. When the voltage is too low for the intended application, the cells can be stacked in series, as in a conventional battery. Figure 1 shows what this might look like.

Nuclear batteries are enticing, mainly because the energy and the energy density of the active material can be very high, up to 100000 times higher than in chemical batteries. A a nucleus can easily contain 100 keV or more, while the typical chemical energy stored in an atom is 1 eV. However, in a practical isotope battery, the bulk of the mass is not the active material, but in accessory elements such as the radiation-to-electrical converter and the radiation shielding. Therefore, a comparison between nuclear and chemical batteries should be done on the basis of a conceptually complete design, explicitly containing the accessories that are known as the balance of the system (BOS).

Radioactive thermal generators (RTG) are ~200 W batteries designed to provide power to satellites far from the Sun [1]. They almost always combine alpha emitters with thermoelectric energy (TE) converters. The most common isotope used in RTGs is plutonium-238. More than two dozen of these RTGs have been sent up in the last 35 years. However, the radiological concerns in using alpha emitters make them unacceptable for Army applications. For this reason, our most interesting radioactive power source is based on beta emitters.

Small batteries for Army applications that are based on long-lived power are ideal for unattended sensors. High-energy density materials would allow sensors to operate for long periods of time, avoiding maintenance and upkeep. This is important for instrumentation of various types, e.g., those placed in remote spots or embedded in structural components (helicopter blades, I-beams, brick walls, etc.), to keep track of the mate-

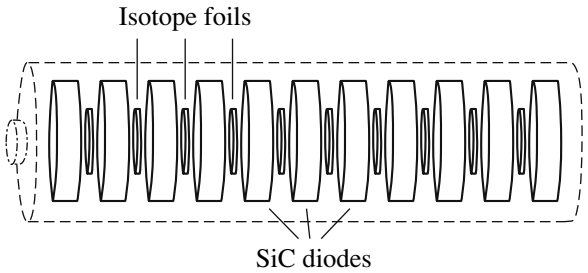

Fig. 1. Schematic of a conceptual nuclear battery in AA format.

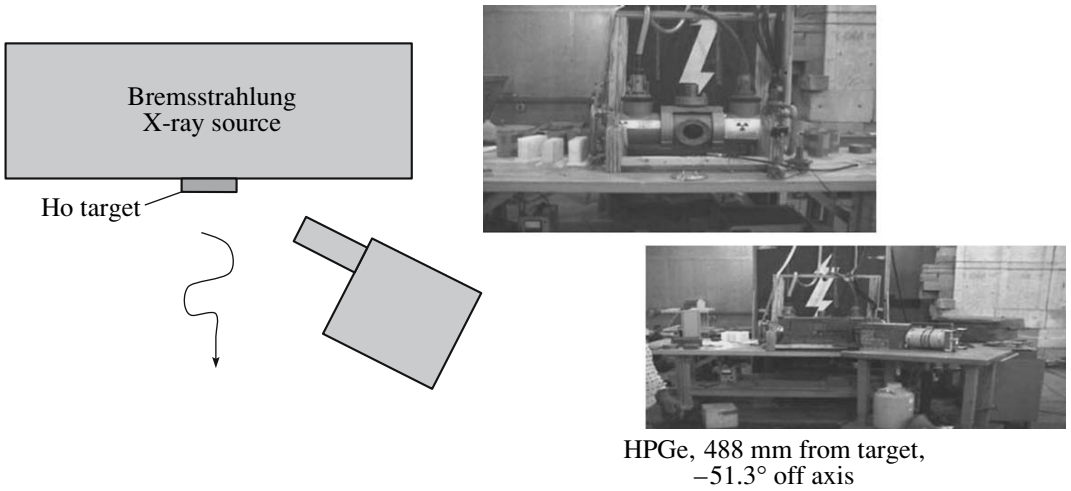

Fig. 2. Experimental setup.

rial's integrity. High-energy density isotope-based materials, coupled with semiconductors for direct-energy conversion (DEC) would make these types of sensors practical. The potential simplicity and efficiency of DEC, and the possibility of turning the battery on with X rays from an outside source, are two characteristics of these small batteries that make them worth pursuing.

DEC is a natural match to beta emitters, and the approach is especially favorable when the beta particle has a relatively low energy. Low-energy electrons produce low-energy bremsstrahlung, which is easily shielded. Moreover, a parametric study [2] of the energy deposition in SiC for different beta energies shows that lower energy betas deposit a higher percentage of their energy into the SiC DEC. Tritium is a prime example. The mean beta-particle energy is 5.7 keV, the maximum is 18.6 keV, and various types of tritium batteries already exist. The battery's shelf life is limited by the isotope's half-life, which is 12 years for tritium. Conceptually more attractive for a beta battery would be isotopes with a longer half-life, especially if this isotope were to be an isomer whose decay rate could be increased by an external radiation source. This makes the isomer ^{166m}Ho an interesting candidate for an isotope battery.

The half-life of ^{166m}Ho is 1200 years, corresponding to a 1/e decay time of 4×10^{10} s. Deexcitation by external radiation gives the unstable ground state of holmium, ^{166}Ho, whose half-life is slightly longer than a day (26.8 h or almost 10^5 s). A conceptual Ho-powered battery would be minimally radioactive ($\ll$10 μCi) for many centuries, until the ^{166m}Ho nuclei are activated. Activation would increase the isomer's radioactive decay rate by the ratio of the decay times. The battery's power would increase by a factor of 4×10^5 if all of the isomeric nuclei could be activated, and the battery would provide all of its power in a day or so.

Of course, the power increase in an isomeric battery is less than the ratio of the half-lives when the external radiation activates only a fraction of the ^{166m}Ho nuclei. However, even if only 1% of the nuclei could be activated, the result would still be a 4000-fold increase in power. The initial measurements in this paper are intended to find the requirements to activate ^{166m}Ho with external radiation.

EXPERIMENTAL PROCEDURE

For the present measurements on ^{166m}Ho, we followed last year's procedure on the activation of ^{177m}Lu [3]. Figure 2 shows the radiation source, with the target placed in front of the source window, and the location of the high-purity germanium γ-ray spectrograph that is the primary diagnostic. The X-ray source at this time is the Norelco MG-300, a 300 kV continuous X-ray generator with a 10-mA DC current. The γ detector is an HPGe Ortec model GMX-20P. The Ge crystal is 51.1 mm in diameter and 52.6 mm in length, with a 0.5-mm Be window.

To date, this setup has not shown evidence that ^{166m}Ho can be activated by X rays. New lines at 54.2, 72.9, 136.6, and 257.0 keV should follow the excitation of ^{166m}Ho by X rays around 263 keV. [4, 5] A simplified decay scheme for the ^{166m}Ho nucleus is shown in Fig. 3, but we did not see these new γ lines. The null result is disappointing, but could have been expected since our 300 kV bremsstrahlung source produces relatively few X rays whose energy match the 263 keV level [6, 7]. We are presently trying to install a 2 MV X-ray source to provide harder X rays that could excite higher energy intermediate states through which the isomer could deexcite to the ground state more easily than through

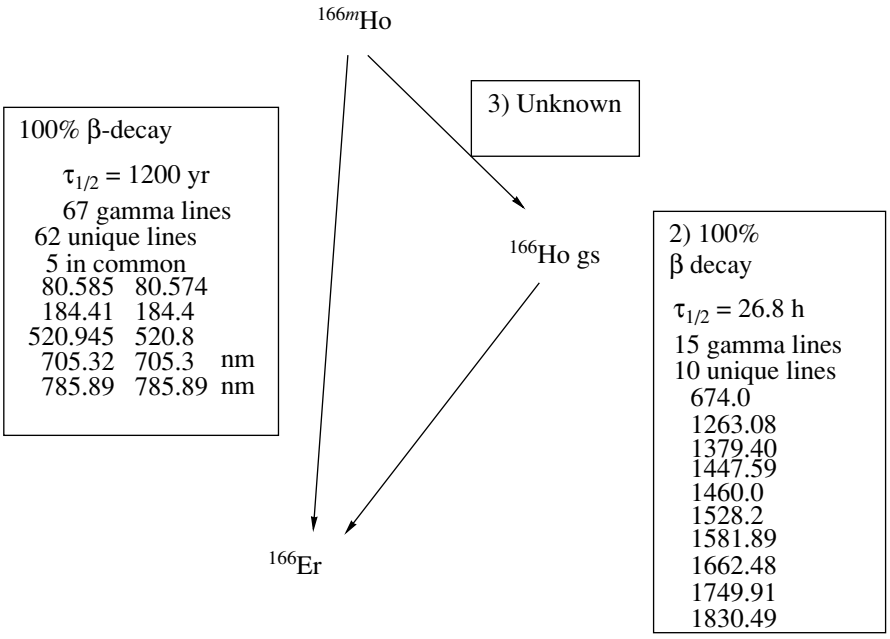

Fig. 3. Decay path of ^{166m}Ho.

resonant photoexcitation. In addition, we intend to quantitatively estimate the conditions under which we might expect to see the desired effects.

When the radiation-produced isotope cannot be seen directly, isomer researchers sometimes try to find whether the number of decays coming out of the isomer sample has decreased. A decrease in the number of energetic isomers due to irradiation would, indeed, reduce the sample's radioactivity, and vice versa: fewer post-irradiation decays might be caused by irradiation. The evocative name for this is burn-up. In addition to the direct decrease in decays from the isomeric state, ^{166m}Ho can decay into the ground state first before decaying into erbium (^{166}Er).

Initially, we had inferred some tantalizing suggestions of such an effect from an initial look at the data; but, closer scrutiny later on has convinced us that we should have attributed the effect to a change in the temperature of the instrumentation. We are presently improving the setup to avoid this and other spurious effects.

Burn-up can be detected in several ways depending on the isotope. For ^{166m}Ho, the first and most obvious approach is to look for any new lines known from the natural decay from the unstable ^{166m}Ho ground state to the ^{166}Er, as previously mentioned. These lines would appear in the spectrum immediately after irradiation, in the first few half-lives of the ^{166}Ho ground state. New lines in a gamma spectrum taken over three half-lives, or ~96 h, directly after irradiation would then indicate the conversion of some isomeric nuclei to the ground state, and the liberation of energy from the isomer. Our standard procedure is to irradiate for 39 h, long enough to establish equilibrium between excitation and decay, with the only source we have available at this time, the Norelco MG-300. It produces 3×10^{12} photons/s after its intrinsic filtering, quoted as 0.2 mm copper.

Figure 4 shows the isomeric sample's gamma spectrum. It has 84 gamma lines. Of these, 68 come from the decay of ^{166m}Ho. Of the remaining 16 lines, 11 are attributed to ^{154m}Tb, a minor contaminant in the sample that remained after chemical separation. The remaining 5 low-intensity lines are, as yet, unidentified background. The measurement contains 16384 channels with a resolution of 1.16 keV per energy bin. This spectrum is identical to that of the ^{166m}Ho sample before irradiation. New lines did not appear. Hence, triggering was not positively identified by this measurement.

A second way to identify triggered isomer decay after irradiation is to compare the number of gamma photons from the decay of ^{166m}Ho before and after irradiation. Fewer decays from ^{166m}Ho are consistent with fewer isomeric nuclei, with the difference indicating the amount of triggering. The difference was monitored by counting the number of photons in the two lines with the highest count rate in the in spectrum (in Fig. 4, the 80 keV line and the 180 keV line). A set of spectra was accumulated each hour for 96 h (4 days), or more than three 26.9-h half-lives of the Ho ground state. Within the statistical error associated with the measurement system, we could not detect a difference.

Post-processing provides the total number of counts under an individual line after removal of the background. Figure 5 shows one result for the 184.41 keV

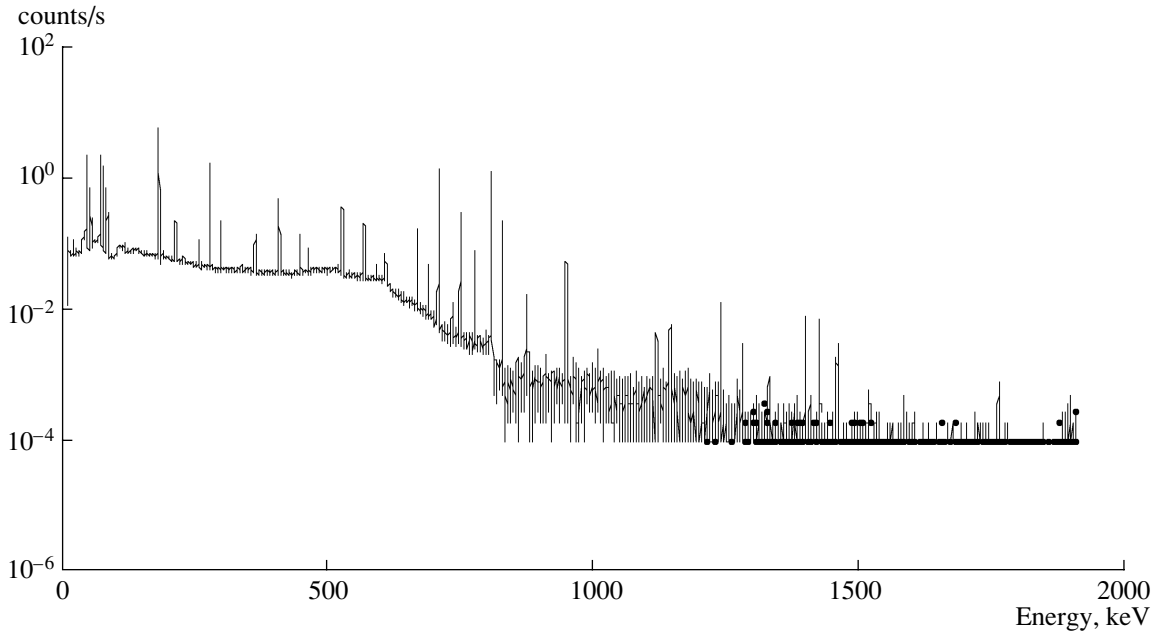

Fig. 4. ^{166m}Ho spectrum from the HPGe detector.

line. On the right-hand side in the figure lies the average number of counts per second over each hourly period for the sample before irradiation. The count rate for this sample is a constant (5.2 counts per second) to within the statistical error for the initial period; but, in the middle of the second day of counting, something must have happened. Suddenly, the number of counts drops 0.13 counts per second or 2%, to another constant value around 5.08 counts per second. Standard analysis provides a linear fit to the data, with the slope of the line corresponding to a decay time of ~10^7 s, much longer than ^{166m}Ho's decay time (~4×10^{10} s).

Initially, we had interpreted the faster decrease in the count rate to burn-up, but when the presentation had already been nearly finalized and the figures could no longer be redone, we realized that this interpretation was in error. To be true to the presentation, this paper maintains the original figures and discusses the considerations that led us to modify our initial conclusions.

The right-hand side of the figure provides the number of counts after irradiation. Now, the count rate is seemingly erratic. Initially, the count rate is fairly stable and about the same as for the unirradiated sample; but, after the first day, it drops by about 3%, followed by another 3% about 48 h after the start of the measurement, after which it recovers, stabilizes for a 12-h period, and then drops again to a stable value for the remainder of the measurement. The problem with the erratic count rate was flagged while preparing the presentation, long after the measurement had been completed. Although no written record is available, we suspect that the decrease in the count rate after 48 h correlates with refilling the high-purity germanium detector with liquid nitrogen. At the time, we recognized that refilling the spectrometer was undesirable, and the spectrometer is now equipped with a larger liquid nitrogen reservoir that enables undisturbed counting over the entire period. But, we have not yet had a chance to redo the measurement.

The variable count rate in the older premodification measurement corroborates the need to improve the spectrometer and the analysis procedures, as we have already done. The more stable count rate of the irradiated sample corroborates this conclusion. Count rate stability improves when the liquid nitrogen reservoir is large enough for the entire run. This same topic came up in the discussion following the presentation at the conference, and confirmed our conclusions.

CONCLUSIONS

With the equipment we have had available to date, it was not possible to see a change in the radioactive decay rate of our ^{166m}Ho sample, which might signal a decrease in the number of isomeric holmium nuclei due to deexcitation of the isomer or the creation of a short-lived isotope. Over the next few years, we intend to use higher energy X rays (up to 2 MeV) that might access higher energy gateway states, and to install more precise diagnostics.

The null results to date are disappointing, but are still valuable, because they imply an upper limit for the triggering cross section, 0.2 bar, over the photon energy range in the present experiment. This integrated cross

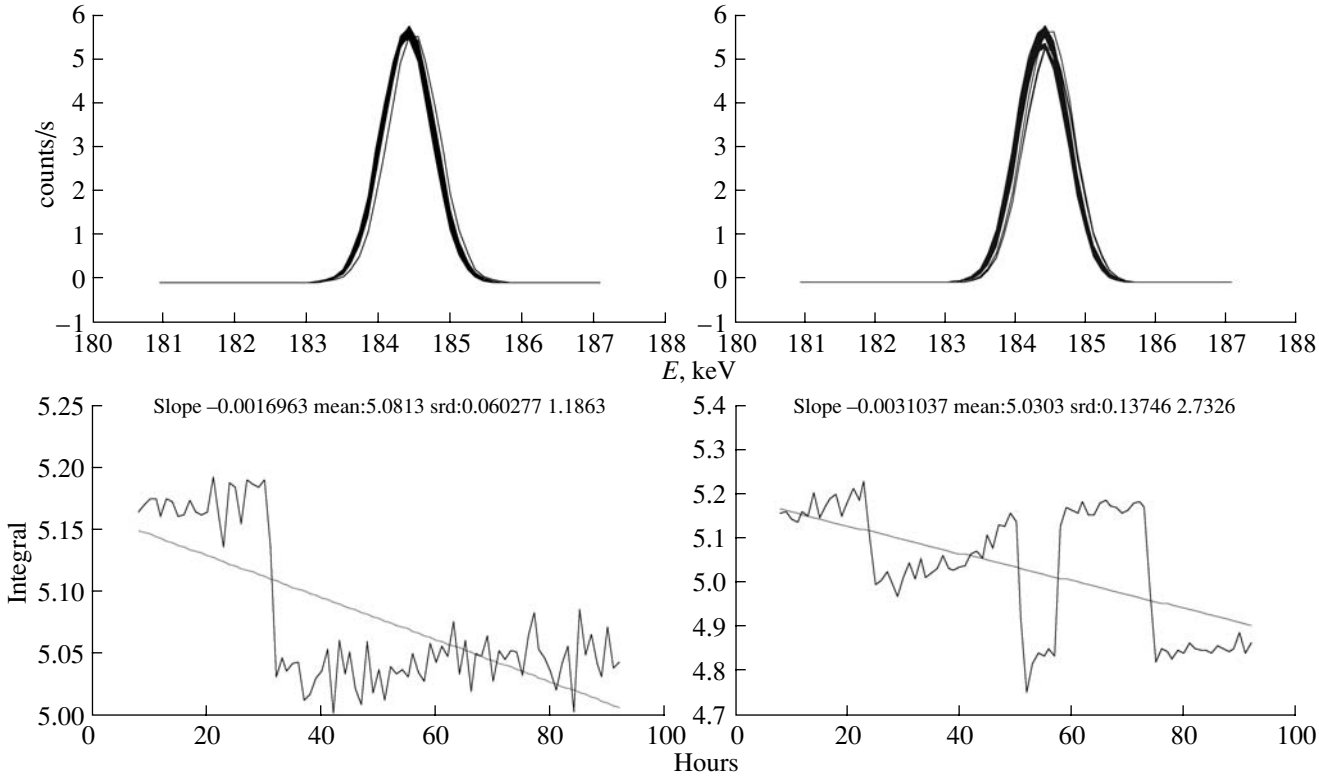

Fig. 5. Hourly count in the 184.41 keV line over ~90 h before irradiation (left). The same γ line after a 39 h bremsstrahlung irradiation with 3×10^{12} photons/s/cm^2 (right).

section can, then, be used in an evaluation of the possible utility of isomeric energy. The cross-section estimate and its practical consequences should be available at next year's conference. While calculating this estimate, we uncovered an additional problem, this time in the radiation source: its measured output is three times lower than expected from computations. Therefore, we prefer to defer a detailed presentation of the results until this issue is resolved.

Although we could not measure the photonuclear cross section in this experiment, it does provide an upper limit to the integrated triggering cross section as follows. The number of isomer atoms burned-up during irradiation (N_b) is equal to the number of isomer atoms initially present in the sample (N_i), times the flux, ϕ (photons/cm^2), times the cross section σ (cm^2) for triggering the isomers:

$$N_b = N_i \phi \sigma. \quad (1)$$

Given the number of atoms in the target (~10^{16}) and the activity of the target, 7.5 μCi, this corresponds to 2.775×10^5 Bq (decays per second). Both before and after irradiation, we measure 26.2 ± 0.2 counts/s, corresponding to a ~0.7% counting efficiency, which is consistent with the counting system's geometry. The decay rate is, then, $2.775 \times 10^5 \pm 2134$ Bq. Since the number of burned-up nuclei N_b cannot be larger than that which corresponds to the relative error, 2134 counts/s transitioned atoms, the cross section is at most 2×10^{-25} cm^2 or ~0.2 bar. Photonuclear cross sections are typically much smaller. For now, the sole conclusion is that, the random error in our measurement system sets an upper limit to the cross section for the induced decay of ^{166m}Ho isomer with a 300 kV endpoint bremsstrahlung at 0.2 bar. This cross section can then be used in a systematic evaluation of externally excited nuclear batteries—something we intend to do later on.

Our plan to decrease the error associated with these measurements includes (i) moving the detection system to a room where the temperature and humidity is controlled much better than can be controlled in the radiation test cell, and (ii) complete daily measurements with known calibration sources to identify variations in the measurement system in order to document any other systematic errors that may be present. In reducing the measurement fluctuation, it is hoped that we will be able to measure the smaller cross sections anticipated from photonuclear cross sections.

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