CONTROLLED EXTRACTION OF ENERGY FROM NUCLEAR ISOMERS

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ABSTRACT

The Army must deploy increasingly powerful energy sources to support sustained operations anytime and anywhere in the world. Excited states of nuclei can store 5 orders of magnitude more energy than that stored in chemical bonds. Conventional nuclear storage systems such as radioisotopes cannot be turned on when the energy is required. This paper describes a switchable nuclear battery that may be formed from long-lived (ie. ~100 years) excited isomeric-states in the nucleus, which can then be triggered to release their energy on demand.

1. What are Isomers?

Isomers are excited-states of the nuclei that emit gamma radiations when de-excited. The energy stored in the individual nucleus can contain 100,000 times the energy of an individual chemical atom. [1] By analogy with chemical isomers, nuclear isomers are isotopes with the same number and constituents of neutrons and protons, but different physical configurations. When energy, stored in the isomer, is de-excited, gammas are released - no nuclei are split, no neutrons are released that can cause radioactive byproducts.

The four categories of isomers known to date are pictured in figure 1. The differences are characterized by changes of the physical configuration of the nucleus based on a) shape elongation, b) shape symmetry, c) spin traps and d) spin projections. The energy minima, that exists in each of the isomer modes, is the trap that characterizes these metastable states in the nuclear structure. These traps are ultimately responsible for the capability to switch the isomer on, creating the power on demand.

Regardless of the isomer type, the energy stored in the isomer resides in the internal motion of the nucleons. The release of the energy does not come from the binding energy, and therefore does not lead to fission, fusion, or transmutation. Therefore, the gamma emission constitutes a 'clean' form of release of nuclear energy. [3]

The path to developing this cleaner, high energy density isomeric source of energy involves a) creation of the isomeric metastable states in isotopes, b) techniques to externally trigger the isomer, releasing its stored energy into a lower energy state, and c) identification of efficient direct electrical conversion techniques for power and energy applications for the army.

2. What are the advantages of isomers over radio-isotopes?

Isomers are isotopes that can be turned on when needed, as energy on demand. By triggering the isomer with a low energy photon, the isomer switches to a lower energy state, liberating energy that can be converted directly to electricity. In some material cases (to be discussed below), a radioactive decay cycle also begins that gives off alphas, betas, or gammas, depending on the material in use.

Nuclear batteries are already in various stages of development in the marketplace. They are typically composed of small amounts (milligrams and micro-Curies) of high-energy-density radio-isotopes. These nuclear batteries can now be designed with isomeric materials. In this way, it would permit the power system to be turned on, on demand. After turn-on, these materials decay with a rate determined by the half-life of the ground-state isotope half-life. When long-shelf-life is important, isomers have great potential to fill this power on demand niche. Standard isotopes can have long half-lives, but are limited in applications were long shelf-life is required.

Another feature of the isomer, when compared to radio-isotopes, becomes apparent in the logistics of nuclear sources. Because the isomer source is not highly radioactive until it is triggered/turned-on, the transportation of these materials is significantly safer. This is a significant factor in overcoming the political "horrors" usually associated with using nuclear materials.

Applcations that require larger quantities of radio-isotopes than would be used in small nuclear batteries, would also become an option. Power supplies for base power concepts would be feasible, that could last for six months and then decay to low-level residual background. By transporting isomeric materials, (for use in larger power supplies than that

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of small nuclear batteries,) in the long-lived isomeric state, and then turning them on at the point of use, the transportation difficulties can be greatly reduced. By removing the heavy burdens usually associated with transporting radioactive materials, larger quantities of excited-state isomers can be transported and used in base power supply applications.

The energy density of most chemically stored energy is typically less than 1 kJ/g, while the energy density of nuclear materials is six orders of magnitude higher. The surface-to-volume ratio is large for smaller devices. For this reason, small nuclear battery designs have tended to focus on direct methods of energy conversion. For example, diodes have been constructed from silicon using a layer of P-type silicon adjacent to a layer of N-type silicon and a radioactive source placed on the top of the device. As the source decays, the energetic particles penetrate the surface and create electron-hole pairs in the vicinity of the P-N junction. This creates a potential across the junction, thus forming a battery.

Several commercial products [6, 7] are in developmental stages of progress. They promise to be useful in unattended sensor applications, both commercial and military. Nuclear batteries are capable of orders of magnitude greater energy density than chemical based energy stores and will last years or even decades. No power cords or transformers will be needed for the next generation of microelectronics in which voltage-matched supplies are built into electrical components.

3. Which isomers have the greatest potential for technology insertion?

Several isomers have been identified as potential candidates for long-shelf-life power on demand nuclear batteries [2]. These include ^{108}Ag, ^{166}Ho, ^{177}Lu, and ^{241}Am.[3] The more important figures of merit used in identifying isomers for long-lived energy storage include material creation/availability, energy stored per nucleus, amount and type of energy released during transition, and ease in triggering. Table 1 includes the values for these metrics. Each of the identified isomers, have characteristics identified that would lead us to examine further (through experimental verification) the triggering cross sections.

The only isomer that has been triggered to date is ^{180}Ta [4]. In this case the photon energy required to trigger was more than the energy released. This paper focuses on an experimental effort to trigger Lutecium. The triggering of isomeric Lutecium, with atomic number 177, has not yet been measured.

The most intense of the gamma lines emitted during the de-excitation of 177mLu are 413 keV and 319 keV. Amore complete list of the gammas emittied is shown in figure 3. These gammas can be converted directly to electricity, by successive Compton scattering collisions in a rad-hard solid-state diode structure such as silicon-carbide (SiC).

The list of isomers with halflife greater than 1 week exceeds 100 in number. The identification of isomers with greatest potential depends on weighting the factors discussed above (ie. availability, trigerability, energy stored, emission).

Several isomers are notable in that they bound the extremes as we know it today. These include the 180mTa isomer, extreme in its halflife of 10^{15} years. The isomer with the largest known energy density is 178mHf with an excess of 10^{9} J/g. And finally, but most abundant of all the known isomers is 242mAm.

The few eV difference in energy levels (see diagram in figure 4) suggest that 177mLu can be triggered by photons with less energy than will be released. This experiment, if successful, will be the first to measure an isomeric system that will serve as a source of stored energy and not an energy sink. No theoretical impediment exists to pumping out the stored energy [5]. There are several other possible energy storage isomer candidates that will be investigated in later experiments.

4. Experimental Approach

Bremsstrahlung radiation (320 kV max) from a continuous x-ray source (5 mA electron beam) with a flux of 10^{12} photons/sec has been focused on a target of 10^{13} nuclei of ^{177m}Lu for a period of two weeks. The experiment described here is commonly identified as a burn-up experiment, in that the target material is stimulated to trigger, consumed in the process, and the number of triggered events detected either. The detection of triggered events can occur during or after the irradiation event. In this ongoing burn-up experiment, measurements will be compared before and after irradiation. The number of nuclei de-excited (triggered) will be determined by measuring both the reduced activity of the excited isomeric state, and the increased activity in the ground state, using gamma spectroscopy. What is lost from the upper state should be detectable in the lower state. The expected result of the measurement of both upper and lower state gamma energies is pictorially shown in figure 2.

The 177mLu target is submitted to a gamma spectroscopy for a period of one month. The goal of the first phase of the experiment (see figure 2) is to determine that the significant gamma lines observed

are those of the 177mLu. In this way, the activity of the sample is evaluated, and the composition of the sample is identified (level of other isotopes present in target can be identified). In the second phase of the experiment (see figure 2), the target is irradiated. While measurements performed during the irradiation can suffer from signal-to-noise difficulties, beta decay from the single electron energy released may be detected. In the third phase, post irradiation measurements of the gamma spectrum can determine the activity remaining in the target, identifying the number of transitions induced by the photon irradiation.

The results of these experiments are expected to provide information necessary to understand the cross-section for triggering of these isomeric nuclear transitions. The goal of these experiments is to identify those isomers most useful as power and energy sources. We must continue to explore those isomers that most easily form long-lived isomeric states, which can be developed into more powerful and long-lived energy sources for sustained operations.

By obtaining targets of the most suitable isomeric isotopes, and measuring the cross sections for triggering, we hope to understand the underlying physical mechanisms.

5. Payoff to The Soldier

The energy density of radio-isotopes and isomers is easily six orders of magnitude greater than that of chemical energy storage system (ie. batteries and fuel cells) as depicted in figure 5. While chemically based systems are a necessary component to most systems, they are not sufficient to meet the growing needs of the Soldier.

Special forces carry approximately 120 pounds of equipment into and out of the field with them on 14-day missions. Batteries for radios and other gear account for 40 pounds of the weight.

Once the triggering cross sections are known to suitable accuracy, cost effective engineering approaches can be developed. Design can be developed for high-density-power sources for short-term (1 month to 1 year) applications such as unmanned aerial vehicle (UAV) power and base power, with require 1 kW to 100 kW levels. Alternately, designs for high-density-energy sources for long-term Soldier Power applications (nuclear batteries) can be specified.

References:

[1] P.Walker, G.Dracoulis, "Energy traps in atomic nuclei," Nature, 399, p.35-40

[2] J.J. Carroll, "An Experimental Perspective on Triggered Gamma Emission from Nuclear Isomers " Submitted to Laser Physics for Proceedings of 5th AFSOR Isomer Workshop: Hamburg ,Germany 2003.

[3] J.J. Carroll, S.A. Karamian, L.A. Rivlin, A.A Zadernovsky, "X-ray Driven Gamma Emission," Hyperfine Interactions, 135, p.3-50,2001

[4] D. Belic, C. Arlandini, J. Besserer, J. de Boer, J. J. Carroll, J. Enders, T. Hartmann, F. Käppeler, H. Kaiser, U. Kneissl, E. Kolbe, K. Langanke, M. Loewe, H. J. Maier, H. Maser, P. Mohr, P. von Neumann-Cosel, A. Nord, H. H. Pitz, A. Richter, M. Schumann, F.-K. Thielemann, S. Volz, and A. Zilges," Photo-induced depopulation of the $^{180}Ta^{m}$ isomer via low-lying intermediate states: Structure and astrophysical implications," Phys. Rev. C **65**, 035801 (2002) and C.B. Collins, C.D. Eberhard, J.W. Glesener, J.A. Anderson, "Depopulation of the isomeric state ^{180m}Ta by the reaction $^{180m}Ta(g,g')^{180}Ta$," Phys.Rev.C37, 5, p.2267

[5] B. Balko, J. Silk, D. Sparrow, "An examination of the possibility of controlled extraction of energy from nuclear isomers," Institute for Defense Analysis, AK-2-1894 -Briefing to Dr. George Ullrich, ODUSD(S&T), Sept 12 2002

[6] Trace Photonics, Ken Bower Pres. ; Alpha-V Inc., Ron DeFelice Pres. ; Nucell Technologies, Paul Brown Pres., Pat #4,835,433

[7] Brown, P., Solid Stat Isotopic Power Source for Computer Chips, 27th Intersociety Energy Conversion Engineering Conference, Aug 3-7, 1992, San Diego, CA, p.4345

[8] P.Walker, G.Dracoulis, Physics World, Feb. 1994, p.39

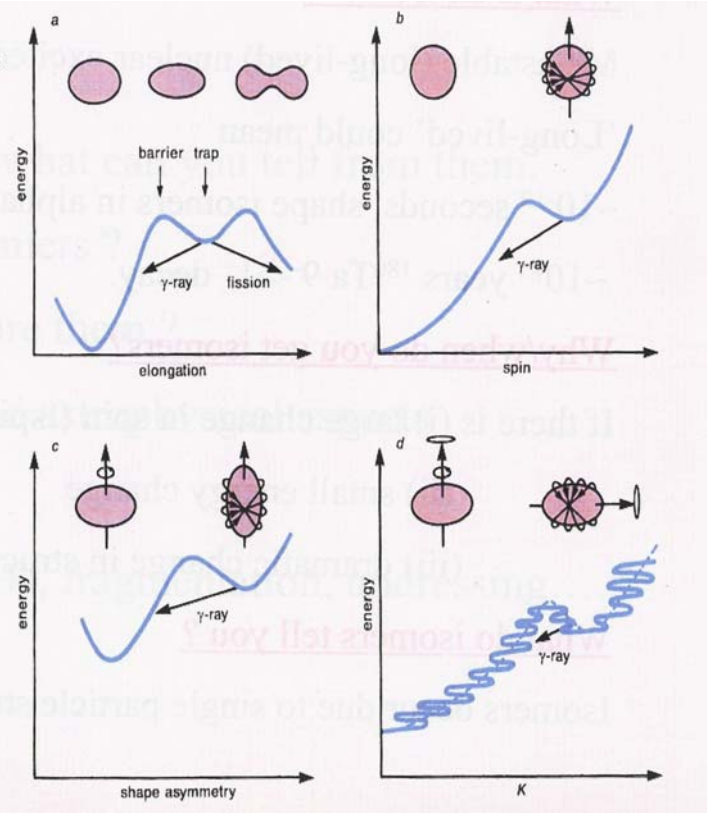


Figure 1. The excitation energy as a function of nuclear energetic variables is shown in the four configurations above. All isomers have in common the energy minima responsible for the changes of state that occur, which can be long-lived in some of the changes.

Relative figure-of-merit of selected isomers based on an equal weighting of various parameters.

Isomer	Prod. Factor	Storage factor			Triggering factor			Output factor	Overall Figure of Merit
	$\sigma_{(n,\gamma)}$ [b]	E_s [keV]	$T_{1/2}$ [y]		ICS_{trig} [eV b]	E_{trigg} [keV]		$<E_\gamma>$ [keV]	
$^{166}Ho^m$	3.5	6	1200	7,200	~ 1	257	0.004	263+	0.00005+
$^{177}Lu^m$	2.8	970	0.44	427	~ 10^4	< 100	100	230	0.06
$^{178}Hf^{m2}$	2×10^{-7}	2,446	31	75,800	< 3×10^4	~ 10	3,000	300	0.00003
$^{242}Am^m$	54	49	141	6,909	~ 100	4	25	53+	1+

Table 1. Figures of merit comparing four particular isomers with characteristics highly weighted in the areas of production, triggering cross section, and energy output characteristics.

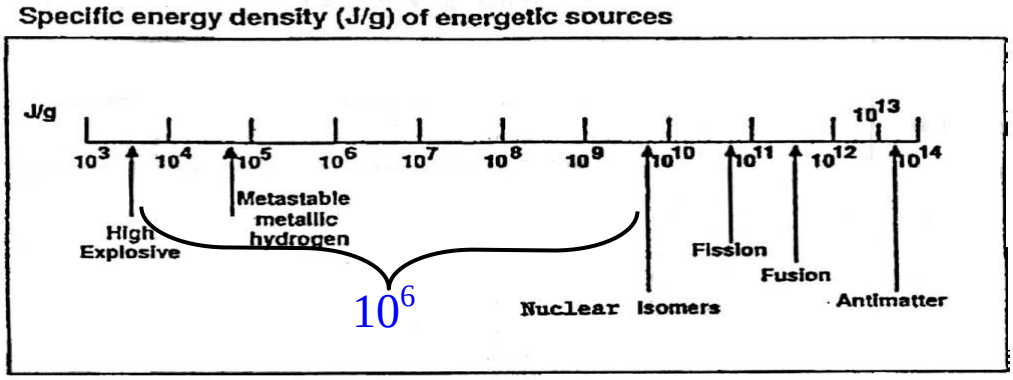
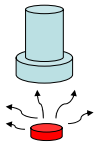
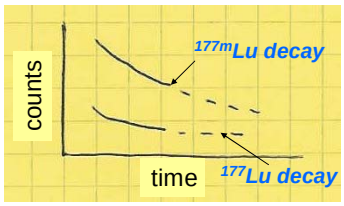


Figure 5. The energy density of nuclear sources exceeds that of chemical energy densities by more than six orders of magnitude.

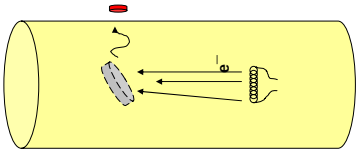
Burn-up Experiment

1. Pre-test gamma spectroscopy
   - Ortec γ detector

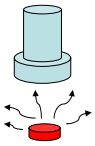




2. Radiate Target
   - 300 kV e-beam
   - 5 mA continuous



3. Post irradiation gamma spectroscopy
   - Increase on ground state
   - Decrease in isomer (excited) state



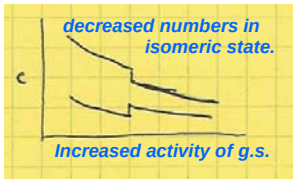


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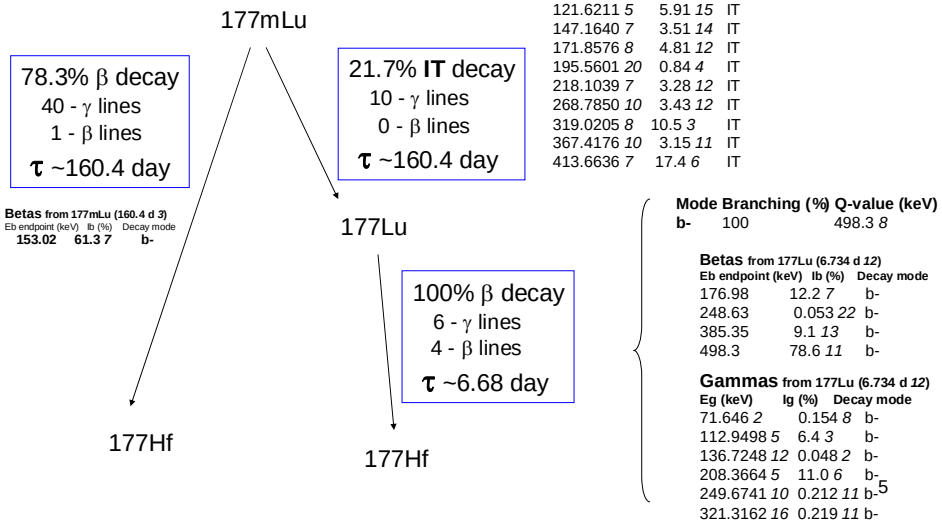
Figure 2. An unambiguous technique to determine triggering events, is shown above in a three step process. If sufficient number of events occur, a cross section for photon triggering can be calculated.

^{177m}Lu Decay Modes

- 2 decay modes



Betas from 177mLu (160.4 d *3*)

Eb endpoint (keV)	Ib (%)	Decay mode
153.02	61.3 *7*	b-

Gammas from 177mLu (160.4 d *3*)

Eg (keV)	Ig (%)	Decay mode
115.8682 *23*	0.65 *4*	IT
121.6211 *5*	5.91 *15*	IT
147.1640 *7*	3.51 *14*	IT
171.8576 *8*	4.81 *12*	IT
195.5601 *20*	0.84 *4*	IT
218.1039 *7*	3.28 *12*	IT
268.7850 *10*	3.43 *12*	IT
319.0205 *8*	10.5 *3*	IT
367.4176 *10*	3.15 *11*	IT
413.6636 *7*	17.4 *6*	IT

Mode	Branching (%)	Q-value (keV)
b-	100	498.3 *8*

Betas from 177Lu (6.734 d *12*)

Eb endpoint (keV)	Ib (%)	Decay mode
176.98	12.2 *7*	b-
248.63	0.053 *22*	b-
385.35	9.1 *13*	b-
498.3	78.6 *11*	b-

Gammas from 177Lu (6.734 d *12*)

Eg (keV)	Ig (%)	Decay mode
71.646 *2*	0.154 *8*	b-
112.9498 *5*	6.4 *3*	b-
136.7248 *12*	0.048 *2*	b-
208.3664 *5*	11.0 *6*	b-
249.6741 *10*	0.212 *11*	b-
321.3162 *16*	0.219 *11*	b-

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Figure 3. The isomer of Lutecium decays to the ground state of hafnium by two different paths. The isomeric decay mode occurs 21.7% of the time with a 10 unique gamma lines. The beta decay mode occurs 78.3% of the time with 40 gamma lines and one beta energy.

177Lu Energy Diagram

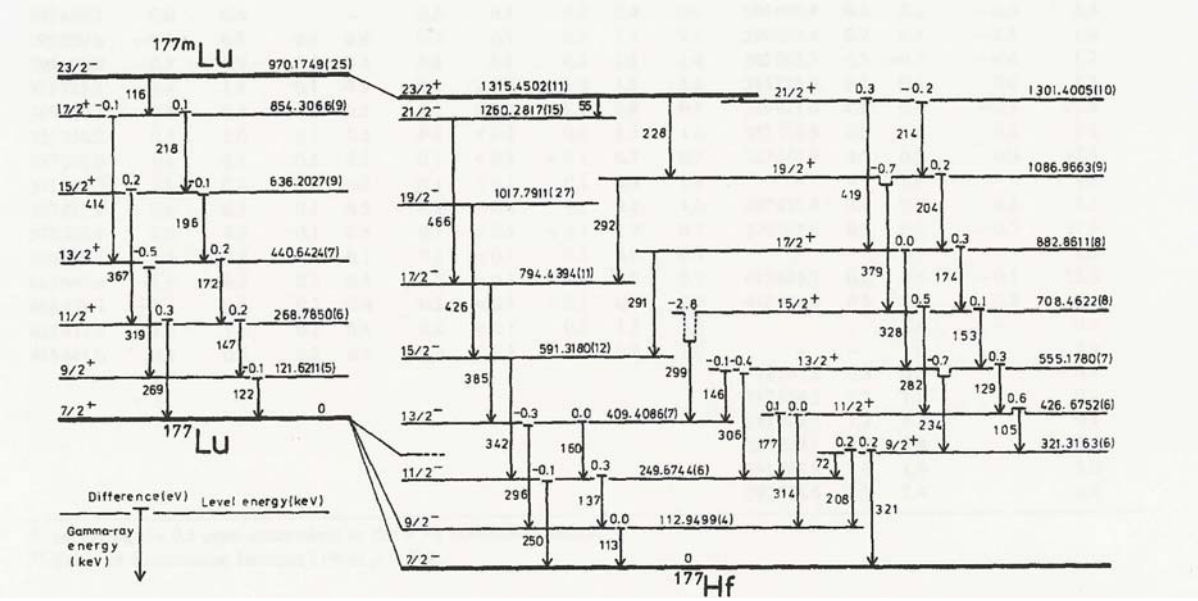

Figure 4. Decay Scheme of 177mLu. Shown are the level energies, spins, and level differences that created the released gamma energies.