

Defense Threat Reduction Agency 8725 John J. Kingman Road, MS 6201 Fort Belvoir, VA 22060-6201



TECHNICAL REPORT

Accelerated Decay of Radioisotopes

Approved for public release, distribution is unlimited.

January 2013

DTRA MIPR 11-2362

Joseph W. Schumer et al.

Prepared by: Naval Research Laboratory 4555 Overlook Ave. SW Code 6770 Washington, DC 20375

DESTRUCTION NOTICE:

Destroy this report when it is no longer needed. Do not return to sender.

PLEASE NOTIFY THE DEFENSE THREAT REDUCTION AGENCY, ATTN: DTRIAC/ J-3 ONIUI , 8725 JOHN J. KINGMAN ROAD, MS-6201, FT BELVOIR, VA 22060-6201, IF YOUR ADDRESS IS INCORRECT, IF YOU WISH THAT IT BE DELETED FROM THE DISTRIBUTION LIST, OR IF THE ADDRESSEE IS NO LONGER EMPLOYED BY YOUR ORGANIZATION.

REPORT DOCUMENTATION PAGE					Form Approved OMB No. 0704-0188	
this burden to Department of Defense 4302. Respondents should be awar valid OMB control number. PLEASI	e that notwithstanding	arters Services, Directorate for any other provision of law, no pe OUR FORM TO THE ABOVE A	regarding this burden estimate o Information Operations and Repo	r any other aspect of this	arching existing data sources, gathering and maintaining the collection of information, including suggestions for reducing efferson Davis Highway, Suite 1204, Arlington, VA 22202- with a collection of information if it does not display a currently	
1. REPORT DATE (DD-MA		2. REPORT TYPE		3	. DATES COVERED (From - To)	
00-01-201	3		Technical		June 2011 – June 2012	
4. TITLE AND SUBTITLE				5	a. CONTRACT NUMBER	
Accelerated Decay of Radioisotopes					TRA MIPR 11-2362M	
					b. GRANT NUMBER	
				50	C. PROGRAM ELEMENT NUMBER	
6. AUTHOR(S) J.W. Schumer, B.V. V	Veber, J.C. Zi	er, J.P. Apruzese, J.	Seelv	50	d. PROJECT NUMBER	
J.W. Schumer, B.V. Weber, J.C. Zier, J.P. Apruzese, J. Seely					e. TASK NUMBER	
				51	. WORK UNIT NUMBER	
7. PERFORMING ORGANI Naval Research Labo 4555 Overlook Avenu Washington, D.C. 203	oratory, Code le SW	S) AND ADDRESS(ES) 6770		8.	PERFORMING ORGANIZATION REPORT NUMBER	
9. SPONSORING / MONITO Defense Threat Reduc 8725 John J. Kingman	tion Agency	NAME(S) AND ADDRE	ESS(ES)	10). SPONSOR/MONITOR'S ACRONYM(S)	
Ft. Belvoir, VA 22060 J9BAS/S. Peiris			11. SPONSOR/MONITOR'S REPORT NUMBER(S) DTRA-TR-12-74			
12. DISTRIBUTION / AVAIL Approved for public re						
13. SUPPLEMENTARY NO	TES					
14. ABSTRACT	1	. 1.0.7/ 1				
density is only a neces novel mechanisms for work was to theoretica ions or energetic neutr occurring with half –li radioactive decay rate	sary condition the rapid and lly, numerica ons. Radioiso ves up to billi	n for usefulness as a controllable –releas lly, and experimenta otopes were studied ons of years. The m	in energetic material se of nuclear energy ally quantify the cro which are energy-ri uclear reaction cross	l. Detailed here within radioise ss-sections or r ch (1e9 J/g) an s sections were	energies less than 1e4 J/g. Energy is the final report of a search for otopes. In particular, the focus of this nuclear reactions induced by heavy- d are more abundant or naturally quantified by changes in the normal orium ("enhanced secondary decay").	
Nuclear Physics Wa	celerators rm dense mat	ter	,			
16. SECURITY CLASSIFICATION OF: Unclassified			17. LIMITATION OF ABSTRACT SAR	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON J.W. Schumer	
	BSTRACT	c. THIS PAGE		21	19b. TELEPHONE NUMBER (include area	
Unclassified Un	classified	Unclassified		21	code) (202) 404-4359	
					Standard Form 298 (Rev. 8-98)	

Prescribed by ANSI Std. 239.18

CONVERSION TABLE

Conversion Factors for U.S. Customary to metric (SI) units of measurement.

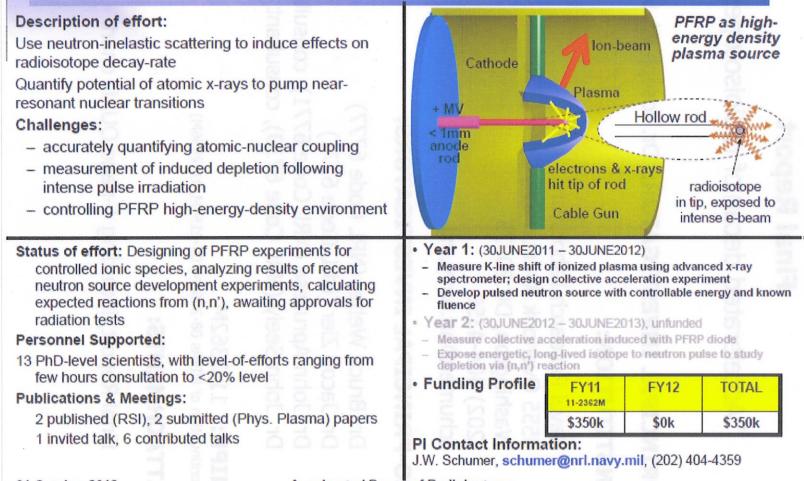
MUL	LIBL
-----	------

Y_____► BY _____► TO GET

TO GET 🗲	BY	DIVIDE
angstrom	1.000 000 x E -10	meters (m)
atmosphere (normal)	1.013 25 x E +2	kilo pascal (kPa)
bar	1.000 000 x E +2	kilo pascal (kPa)
barn	1.000 000 x E -28	meter ² (m ²)
British thermal unit (thermochemical)	1.054 350 x E +3	joule (J)
calorie (thermochemical)	4.184 000	joule (J)
cal (thermochemical/cm ²)	4.184 000 x E −2	mega joule/m² (MJ/m²)
curie	3.700 000 x E +1	*giga bacquerel (GBq)
degree (angle)	1.745 329 x E -2	radian (rad)
degree Fahrenheit	$t_{k} = (t^{\circ}f + 459.67)/1.8$	degree kelvin (K)
electron volt	1.602 19 x E -19	joule (J)
erg	1.000 000 x E -7	joule (J)
erg/second	1.000 000 x E -7	watt (W)
foot	3.048 000 x E -1	meter (m)
foot-pound-force	1.355 818	joule (J)
gallon (U.S. liquid)	3.785 412 x E -3	meter ³ (m ³)
inch	2.540 000 x E -2	meter (m)
jerk	1.000 000 x E +9	joule (J)
joule/kilogram (J/kg) radiation dose		
absorbed	1.000 000	Gray (Gy)
kilotons	4.183	terajoules
kip (1000 lbf)	4.448 222 x E +3	newton (N)
kip/inch ² (ksi)	6.894 757 x E +3	kilo pascal (kPa)
ktap	1.000 000 x E +2	newton-second/ m^2 (N-s/ m^2)
micron	1.000 000 x E -6	meter (m)
mil	2.540 000 x E -5	meter (m)
mile (international)	1.609 344 x E +3	meter (m)
ounce	2.834 952 x E -2	kilogram (kg)
pound-force (lbs avoirdupois)	4.448 222	newton (N)
pound-force inch	1.129 848 x E -1	newton-meter (N-m)
pound-force/inch	1.751 268 x E +2	newton/meter (N/m)
pound-force/foot ²	4.788 026 x E −2	kilo pascal (kPa)
pound-force/inch ² (psi)	6.894 757	kilo pascal (kPa)
pound-mass (lbm avoirdupois)	4.535 924 x E −1	kilogram (kg)
pound-mass-foot ² (moment of inertia)	4.214 O11 x E -2	kilogram-meter ² (kg-m ²)
pound-mass/foot ³	1.601 846 x E +1	kilogram-meter ³ (kg/m ³)
rad (radiation dose absorbed)	1.000 000 x E -2	**Gray (Gy)
roentgen	2.579 760 x E -4	coulomb/kilogram (C/kg)
shake	1.000 000 x E -8	second (s)
slug	1.459 390 x E +1	kilogram (kg)
torr (mm Hg, 0° C)	1.333 22 x E -1	kilo pascal (kPa)

*The bacquerel (Bq) is the SI unit of radioactivity; 1 Bq = 1 event/s. **The Gray (GY) is the SI unit of absorbed radiation.

"Accelerated decay of radioisotopes," Dr. J.W. Schumer, Naval Research Laboratory MIPR# 11-2362M



Extension to "Potential of Electron-Beam Ionization for the Accelerated Decay of Radioisotopes" Proposal# BRCALL07-N-2-0047 Accelerated Decay of Radioisotopes", MIPR 11-2362M

2 | Page

01 October 2012

Accelerated Decay of Radioisotopes

0

Objectives:

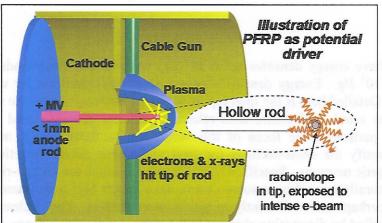
Radioisotopes have energy densities up to 10^9 J/g, whereas chemical-bonds typically store energies less than 10^4 J/g. Energy density is only a *necessary* condition for usefulness as an energetic material. Detailed here is the final report of work revolving around the search for novel mechanisms for the rapid- and controllable-release of nuclear energy stored in the form of radioisotopes. In particular, the focus of this work was to theoretically, numerically, and experimentally quantify the cross-sections (or total yields) for nuclear reactions induced by heavy-ions or energetic neutrons. Specifically, radioisotopes which are energy-rich (10^9 J/g), are more abundant (naturally-occurring in some cases), have longer half-lives (hundreds to billions of years), and are perhaps more efficiently triggered, were studied. The nuclear reaction crosssections were quantified by diagnosing changes in the normal radioactive decay-rate ("burn-up") or measuring the presence of a disturbed secular equilibrium ("enhanced secondary decay"). This work was a one-year extension of work performed under MIPRs# 08-2468M, 09-2156M, and 10-2956M entitled "Potential of electron-beam ionization for the accelerated decay of radioisotopes" based on lessons learned and novel new findings.

Summary of Work:

Funding for this one-year effort was received in late May 2011, with a period of performance from 30 June 2011 – 30 June 2012. This final report details work performed in this year, as well as referencing the 3-year DTRA6.1 effort from which it was spawned. Proposed was a two-year continued study of the (1) high-energy-density plasma environment produced within the plasma-filled rod-pinch diode and (2) the utilization of a pulsed neutron source to study inelastic neutron scattering to accelerate the decay of radioisotopes. Due to funding constraints, only one year's effort was pursued. To these ends, the following tasks were completed within the first year of a planned two-year program:

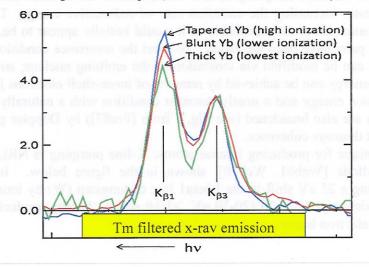
Task 1a (Atomic-nuclear resonance using PFRP plasma source): A potentially novel mechanism for nuclear pumping would employ an atomic-line resonantly matched in energy to the nuclear transition. Ideally, the pump K-line would induce the nucleus into a shorter-lived ground or excited state, increasing the emission rate of radioactive energy. The condition of atomic-nuclear resonance is not as improbable as it would initially appear to be, as a number of suitable atom-nuclei pairs have been identified. To meet the resonance condition, the atomic x-ray emission energy can be modified via ionization of the emitting nucleus; small (tens of eV) shifts in atomic line energy and a nearly-resonant condition with a naturally-narrow nuclear transition. The lines are also broadened (see Fig. 3 from [Pre87]) by Doppler processes, which both aids overlap but destroys coherence.

A potential technique for producing intense atomic K-line pumping is NRL's plasma-filled rod pinch (PRFP) diode [Web01, Web10], shown in the figure below. In this year, we successfully measuring a 25 eV shift in the Lbeta1 line of tungsten (W) by ionizing near soliddensity W to approximately $Z \sim +20$ (20-30 eV, >1/10 solid), thereby producing warm dense matter using intense electron beams.



Within the PFRP, high-power e-beams (500 kA, 1 MV, 50-ns) converge onto an mm-scale anode tip, generating a high-energy-density plasma (HEDP) environment (2.4 MJ/cm³, 1.6 TPa, 25 eV in tungsten) and an intense source of x-rays. X-ray emission from this highly-ionized plasma is affected by the HEDP environment and may allow efficient coupling of an atomic x-ray to a nuclear transition. Detailed basic research into the HEDP environment was pursued under the first 3-years of this project; we improved the fielding of a new x-ray diagnostic [Per10, Sel10] and ascertained the amount of x-ray line-shifting that may be obtained from the PFRP diode and its HEDP environment. Additionally, the experimental results agreed with analytic and numerical calculations of the PFRP plasma. Future numerical studies to study the coupling between the HEDP and nuclear mechanisms were not completed, as these were planned for Year 2.

Experimental determination of the plasma and x-ray environments with a PFRP diode on Gamble II were continued, using a combination of diagnostics available within Code 6770 and Code 7674. Our efforts using K-edge filters to measure small shifts in Kalpha lines are described in two papers (PER12a and PER12b). Within the anode tip, iridium (Ir) was substituted for the tungsten (W) in the anode rod to take advantage of novel diagnostic technique developed for this program. Given that the K-edge of Lu (lutetium) filter would partially-absorb the Kalpha emission from the Ir rod if the x-rays coming from the hot iridium plasma were upshifted as expected. Using this technique (details of which are the subject of refereed publication

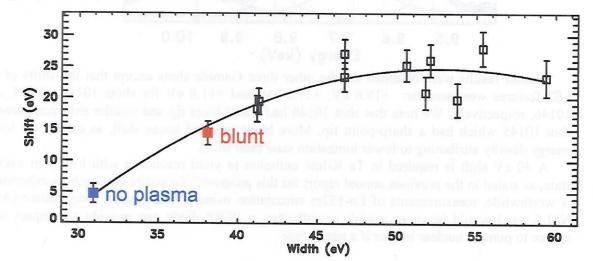


4|Page

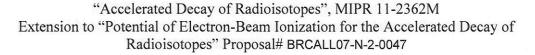
in draft now), a plasma temperature of about 16 eV was estimated. It was also observed that using a Yb (ytterbium) rod and Tm (thulium) filter the Kbetal line was attenuated with ionization as if due to a *down-shift* in x-ray energy, contrary to initial expectations. Theoretical work by Prof. W. Johnson of Notre Dame University was found to be in agreement with experiments.

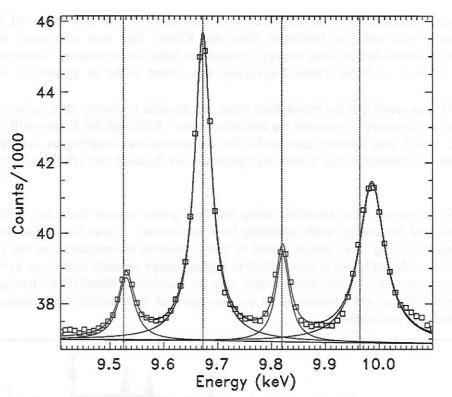
As the K-line shift was small and the broadening large, we decided to change approaches and attempt to use L-line spectroscopy to measure the ionization state. Although the K-line shift was difficult to measure, it still was present (and useful for atomic-nuclear coupling as described above). To obtain the confirmation that a shift was possible, we focused our efforts on L-line spectroscopy.

The tungsten (W) L spectra were recorded using the thin quartz crystal from the "EBIT" spectrometer and analyzed for energy shifts resulting from ionization. It was found that the L transition from the highest lying level which would be most sensitive to ionization, as the L β 2 transition at 9964 eV from the 4d5/2 level was shifted to higher energy on each shot by up to +28 eV with ±2 eV uncertainty. The largest energy shift was on Gamble II Shot#10145 having a sharp-point tip, with the lowest shift having blunt tip, as expected from initial assessments of energy density coupled into ionization.



The analysis of the spectrum from Gamble shot 10145, with the IP positioned near the RC, is shown below. The spectrum was summed and averaged in the vertical direction on the IP. The $L\beta$ -2 feature has a noticeable shift to higher energy. In the detailed spectrum, the shift of the $L\beta$ -2 feature from the 9964 eV tabulated energy is +22.0 eV; the average shift of the L\beta4, L\beta1, and L\beta3 transitions from the tabulated energies is 2.1 ± 2.5 eV. From the Table, the upper levels of these four transitions are L\beta4 (3p1/2), L\beta1 (3d3/2), L\beta3 (3p3/2), and L\beta2 (4d5/2). The L\beta2 transition is from the highest-lying level, is most sensitive to ionization, and has a significant shift to higher energy. The other three transitions are from lower lying levels, are less sensitive to ionization, and have no significant energy shift.





Similar results were obtained for the other three Gamble shots except that the shifts of the L β 2 features were smaller: +19.6 eV, +17.5 eV, and +11.8 eV for shots 10138, 10139, and 10146, respectively. We note that shot 10146 had a half-blunt tip and smaller shift compared to shot 10145 which had a sharp-point tip. More blunt tips had lower shift, as expect for lower energy density attributing to lower ionization state (and shift).

A 40 eV shift is required in Ta K-line emission to yield resonance with Eu-152m excited state, as stated in the previous annual report for this program. To assure that such an experiment is worthwhile, measurements of Eu-152m stimulation using a finely tuned x-ray source (APS, NSLS, etc.) would be more useful; nevertheless, a PFRP diode can provide a compact light source to pump a nuclear isomer if a need arose.

Task 1b (Use of PFRP for "collective acceleration")

"Collective acceleration" processes driven by the PFRP diode were numerically studied, as this phenomenon was discovered under the initial 3-year basic research effort [Sch10]. Nuclear reactions in pulsed-power ion-diodes are usually induced by proton- or deuteron-projectiles accelerated to high energy by the voltage across the anode-cathode (A-K) gap. Reactions for which the incident projectile has a larger atomic number (Z > 2) are inhibited by the Coulomb barrier and are not usually detected. However, during the 3-year DTRA 6.1 effort, anomalous heavy-ion acceleration mechanisms induced nuclear reactions during the operation of a plasmafilled rod-pinch (PFRP) diode fielded on the 2-MV Gamble II generator, some of which the required an energy threshold (Q_{value}) larger than the diode potential difference. Products of these nuclear reactions were observed by measuring the gamma-ray energies and half-lives in witness plates (i.e. experimental hardware designed to survive the electrical discharge and neatly

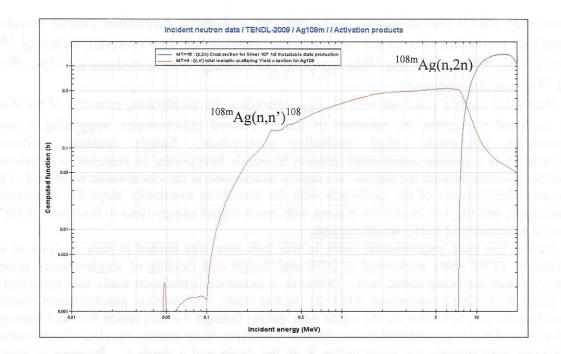
mount onto NRL's high-efficiency, high-count-rate high-purity germanium gamma detector). Significant production of a variety of short-lived radioisotopes were observed, including 38 K, 34m Cl, 43 Sc, 44m Sc, 48 V, and 52 Mn (up to 10⁸ per discharge). Weak evidence for 24 Na, 69 Ge, and 79 As was also observed.

Heavy-ion nuclear reactions with Q_{values} exceeding the accelerating potential of the diode are proposed to explain the presence of these observed radioisotopes, suggesting a novel acceleration mechanism called "collective acceleration". Simply speaking, collective acceleration is a plasma acceleration process in which heavy-ions, in response to relativistic electrons moving through the plasma, are rapidly accelerated in the electrostatic wakefield of the e-beam. Decoherence of the ion-bunch with the wavefront eventually stops the acceleration phenomena, and the ions depart the plasma with much higher energy (tens to hundreds of MeV) than the potential (1-2 MeV) would suggest.

During this year, experimental work in this task area was limited to data analysis of the handful of PFRP shots performed in 2010 and design and fielding of single-species plasma sources which are being tested now. However, a numerical/simulation study was presented at the IEEE ICOPS2012 conference [OTT12]. In this study, a cylindrical pinched-beam electron beam focuses to a tight spot on the anode axis, passes through the thin anode foil, and impinges into a beam dump region behind the anode foil. The beam dump usually consists of a cylindrical anode-can with roughly the same radius R as the diode and a depth L. Because of energy deposition from the intense electron beam, the interior surfaces of the anode-can are expected to be space-charge-limited emitters. Therefore the electron space charge in the anode-can will draw ions off these surfaces. As in the PFRP diode (which is a radially-convergent variant of this geometry), there is evidence of nuclear activation suggesting that ions are accelerated within this diode in excess of the diode voltage.

LSP particle-in-cell simulations were carried out to study the particle dynamics of the virtual cathode formation and to understand how some ions may be accelerated to high energy. It was found that if L is large compared to D, a virtual cathode forms in the anode-can, causing some electrons to return to the downstream endplate (anode foil). However, unstable field oscillations allow many electrons to move past the virtual cathode and propagate to the upstream endplate. Ions emitted from the interior surfaces of the anode-can are accelerated to high energy, providing partial charge- and current-neutralization of the electrons. Ions of approximately twice the diode accelerating voltage were produced in this geometry, with some ions exceeding that estimate if they surpassed the secondary virtual cathode. These would presumably be the source of the nuclear activation observed in various experiments. A paper detailing this work was recently submitted to Phys. Plasmas [RIC12].

¹⁰La using broad-band nautron sources was plaraed to be experimentally statical. Due of our and time to approve radiation safety perfoculs, only Let 176 experiments were permuted. The section cources would have been very sweldt for studying the (n.t.) reported in ¹⁰⁰ % producing a 2.4-min enhanced secondary decay with every depopulation. This is still plana. for future work if funding altimes: Details of this experiment are below. If the purity germentian (PPGe) gumma-my diagnostics were parately the induct activaly and allow an estimate of reaction rate work this perior in a labor. The male activaly and allow an estimate of reaction rate work this perior in a labor. The male activaly and allow an estimate of reaction rate work this perior in a labor. The male activaly and allow in testimate of reaction rate work first perior in a labor. The male activaly and allow in testimate of reaction rate work first perior in a labor. The male activaly and allow in testimate of reaction rate work in this perior in a labor. The male activation is a secondary decay.



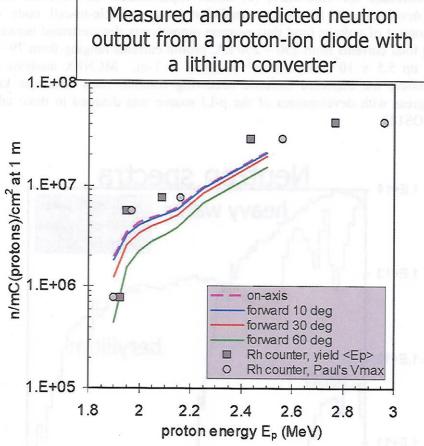
Task 2a (Development of intense pulsed neutron sources for (n,n') studies): The concept of exposing an energetic radioisotope to high-energy neutrons, potentially stimulating decay, was briefly studied. As the inelastic scattering (n,n') and high-energy (n,2n) reactions have reasonable cross-sections (up to a few barns) for neutron energies less than <20-MeV, these mechanisms were expected to be more fruitful than photon- or electron-based mechanisms. To study short-lived states induced by these neutrons, two sources of very intense, 0.1 - 10-MeV pulsed neutrons were demonstrated using the NRL Mercury generator at low-voltage (a 4-8 MV, 200-350 kA, 50-ns pulsed accelerator based on magnetically-insulated-transmission-line (MITL) with inductive voltage-adder (IVA) technology.

In September 2011, a low-energy neutron source was designed, developed, and fielded on Mercury based on the p-⁷Li ion beam-solid target reaction. The p-⁷Li is a convenient, controllable reaction providing neutrons of energies up to incident proton energy less the 1.9-MeV threshold, i.e. for a 2.4-MeV proton beam, a fairly-direction neutron source with energies up to 500-keV is generated. In December 2011, again using the Mercury generator but at the highest voltage (8-MV), a bremsstrahlung-based photoneutron source using a thick layer of heavy-water (D20) was designed and developed to produce a broad-band spectrum of neutrons up to 3 MeV. Details of both sources are below.

In late 2012, stimulated emission from energetic radioisotopes such as ^{108m}Ag, ¹⁵⁰Nd, and ¹⁷⁶Lu using broad-band neutron sources was planned to be experimentally studied. Due to long lead time to approve radiation safety protocols, only Lu-176 experiments were permitted. These neutron sources would have been very useful for studying the (n,n') reaction in ^{108m}Ag, producing a 2.4-min enhanced secondary decay with every depopulation. This is still planned for future work if funding allows. Details of this experiment are below.

High purity germanium (HPGe) gamma-ray diagnostics were used to quantify the induced activity and allow an estimate of reaction rate under this perturbing influence. The nuclear reaction cross-sections will be quantified by "burn-up" or "enhanced secondary decay". The

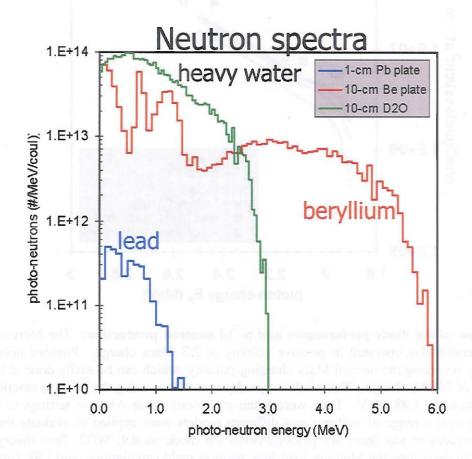
neutron energy spectrum needs to be controllable and varied to access reactions of interest (primarily (n,n') inelastic scattering). Theory and analysis using Monte Carlo codes (MCNPX) is still needed to benchmark to experimental data. Detailed results of the Lu activation experiment are shown below.



An overview of ion diode performance and p-⁷Li neutron production: The Mercury pulsed power generator was operated in positive polarity at 2/3 Marx charge. Positive polarity was achieved by reversing the normal Marx charging polarity, which can be easily done at the lower 50 kV (out of 75 kV) charge. The ion diode produced neutrons using the ⁷Li(p,n) reaction which has a threshold of 1.88 MeV. Data were taken at various diode AK gap settings to provided information over a range of voltages and different models were applied to evaluate the results. Analysis includes results from our pinched-beam ion diode model, MITL flow theory, circuit calculation to determine the Mercury load line, neutron yield calculations, and LSP simulations. Experimentally, the total/anode current I_a, the bound/cathode current I_c, and the ion current I_{ion} were measured using B-dot monitors, and the neutron yield was measured using rhodium activation counters and standardized bubble detectors.

Assessing the diode voltage was a key component of this experiment as the neutron spectrum and angular dependence is directly correlated to the ions with energies greater than 1.88-MeV. Normally, direct measurement of the voltage V in a high-voltage MITL is not possible, but measurement of I_a and I_c can be used to estimate V using MITL theory. However, with positive

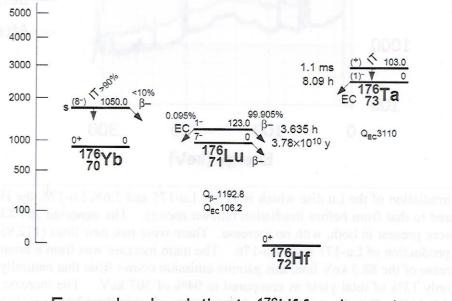
polarity layered MITL flow even this is problematic. Here, attempts to estimate the voltage were made in six ways: (a) assumed diode impedance scaling from pinched-beam ion diode model, (b) MITL theory, (c) generator load line/circuit model, (d) sharp-threshold of p-⁷Li reaction as single-point benchmark for 1.88-MeV, (e) direct experimental measurement using vacuum voltmeter (in development), (f) direct simulation using particle-in-cell code called LSP. Experimental control of voltage (and thus neutron output) was demonstrated between 1.0 - 3.0MV, producing total currents from 250 - 200 kA, proton currents ranging from 79 - 37 kA, and neutron fluxes up 5.5×10^7 neutrons/cm² in 50-ns at 1-m. MCNPX analysis needs to be performed to assess the expected inelastic scattering reaction rates based on known crosssections. Progress with development of the p-Li source was detailed in three talks [OTT10, RUZ12, and MOS12].



Development of intense photoneutron production: To achieve higher-neutron energies with a modicum of spectral control, a novel intense pulsed photoneutron-based source was designed, tested, and characterized. As the Mercury generator can produce a high-intensity bremsstrahlung pulse (8-MeV, 200-kA, 50-ns yielding >750 rad(CaF₂) at 1 m) which, when attenuated by a thick medium containing nuclei with a low (γ ,n) threshold, produced upwards of >10¹² neutrons in 50-ns with a survivable target allowing up to one shot per 10 minutes. Shown above are MCNP

models of expected neutron output when using heavy-water, beryllium, or lead as an attenuator/photoneutron converter with 8-MV Mercury. Heavy-water ($E_{threshold} = 2.2 \text{ MeV}$) was procured as the best conversion medium for a higher-energy pulsed neutron source, as the reaction is both prolific and energy-spectrum better suited for (n,n') reactions of interest. Details were presented in two talks in August 2012 [ZIE12, MIT12].

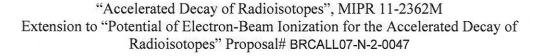
De-excitation of Lu-176 using (n,n') reaction: Using the intense pulsed 3-MeV photoneutron source, a sample of lutetium (Lu) was irradiated and measured. The goal was to stimulate the

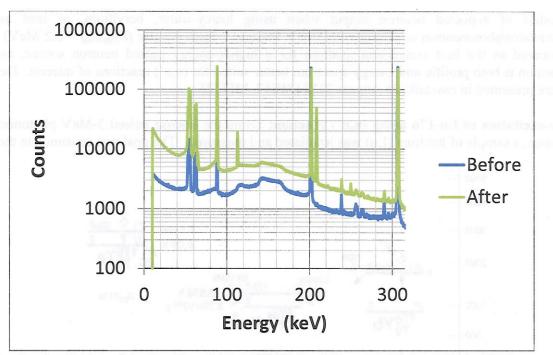


Energy levels relative to ¹⁷⁶Hf for elements with same mass number

long-lived (3.78e10 year half-life) ground state of Lu-176 into the isomer state of Lu-176m with a shorter half-life (3.635 hr) using neutrons and inelastic scattering (n,n') reaction. For each group of nuclei excited, the increased power output would be 1e14 times increase in power output. However, the conversion fraction would be expected to be many times less than unity.

chemical-bonds typically store energies around $10^{\circ} - 10^{\circ}$ Mg. Energy is related naturally as charged particles or photons via a long accay process, determining both power output and shelflife of the medium. For example, radiosotopes have been used for decades as a long-lived energy storage medium, powering devices ranging from smoke-detectors to smellites. Energy density is necessary for compactness, but not sufficient for true usefulness in many scenaries. Of particular interest, however, is the ability to efficiently ralease this stored coargy more quickly then the power decay process, and on-detaund.





Following irradiation of the Lu disc which is 97.4% Lu-175 and 2.6% Lu-176, the HPGe spectra was compared to that from before irradiation (shown above). The expected 201.83 and 306.78 keV lines were present in both, with no increase. There were two new lines (112.95 and 208.37 keV) from production of Lu-177 from Lu-176. The main increase was from a broad continuum and the increase of the 88.3 keV line; this gamma emission comes from that naturally existed Lu-176 but is only 13% of total yield as compared to 94% of 307 keV. The increase post-shot is 24x the 88.3 keV line (8.1e4 counts before, 1.9e6 counts after) in the 12-hour periods before and after the shot. Given exponential decay, approximately 2.1e8 isotopes of Lu-176m were generated via this experiment. A detailed analysis of this experiment will be documented in a future report.

DTRA relevance and potential applications - Many agencies within the Department of Defense (DoD) have recognized the need for advanced energetic materials (AEMs) and the required technologies to produce, control, and utilize them. AEMs include materials with nuclei in an excited state (radioisotopes) that may release this stored energy in a controllable and rapid fashion. These radioisotopes have energy densities on the order of $10^7 - 10^9$ J/g, whereas chemical-bonds typically store energies around $10^3 - 10^4$ J/g. Energy is released naturally as charged particles or photons via a long decay process, determining both power output and shelf-life of the medium. For example, radioisotopes have been used for decades as a long-lived energy storage medium, powering devices ranging from smoke-detectors to satellites. Energy density is necessary for compactness, but not sufficient for true usefulness in many scenarios. Of particular interest, however, is the ability to efficiently release this stored energy more quickly than the normal decay process, and on-demand.

Significance to the field - A variety of "accelerated depletion" techniques have been studied for the rapid release of stored nuclear energy, including the use of very intense, high-energy photon-, neutron-, and ion-beam sources. Unique to this study, however, is the consideration of a new-approach towards e-beam- and neutron-induced "triggering". The scope of this effort has been broadened to include other radioisotopes which are energy-rich (10^9 J/g) , are more abundant (naturally-occurring in some cases), have longer half-lives (hundreds to billions of years), and are perhaps more efficiently triggered. Of particular interest are both "electron-capture" radioisotopes which are directly affected by their atomic electron environment and radioisotopes with low-lying excited states which may be stimulated by a "pump" of sub-100 keV x-ray line radiation or warm neutrons.

Research Highlights: Research highlights for the 2008 - 2010 program were detailed in previous reports. Based on the past 12-months of funded activity, we have focused on neutron source characterization tasks and measurement of WDM in the PFRP diode. Several papers and conference presentations were made under this project. In addition, four notable inclusions to this section are:

- (1) Development of novel K-edge filtered x-ray spectrometer: although of limited utility as it requires a very subtle match between the rod material (e.g. Ir or Yb, in our case) and filter used over the image plate (e.g. Lu or Tm, in our case), the sensitivity of the technique cannot be called into question. With accurate knowledge of the x-ray line shapes, energy resolution down to a few eV for tens of keV x-rays is feasible. Work on this technique will continue, with a focus on continued theoretical support.
- (2) Measurement of 25 eV Lbeta1 shift in W and corroboration with previous HEDP assessments of the PFRP diode using XRD for temperature, radiography for density, and L-shift for ionization state provides a wealth of benchmarked data for warm dense matter studies of high-Z materials.
- (3) Demonstration of two intense pulsed neutron sources with controllable wide-band spectra: although the p-⁷Li and gamma-D reactions are very well studied and no new physics is uncovered here, use of these reactions with precise characterization in a pulsed power environment is noteworthy. High-fluence warm and hot neutron sources become a useful research tools in their own right. The <50 ns bremsstrahlung pulse gives a neutron production rate >1e20 neutrons/s during each pulse.
- (4) Production of Lu-176m isomer using intense pulsed neutrons was demonstrated. Approximately 2.1e8 isomers were generated from the ground state of Lu-176, and thereby transmuted to Hf-176 over ten hours. Stimulated decay using neutrons is more effective than that from electrons and photons in the experiments attempted thus far, but not efficient enough for DoD applications at present. Further research may be required.

Personnel Supported: A robust and diverse team of science professionals from various laboratories, universities, and industry have been assembled to support this effort, including:

Allen, Ray NRL 6771		PhD. Electrical Engineering	
Apruzese, John (PI)	NRL 6771/L3 Communications	PhD. Astrophysics	
Boyer, Craig	NRL 6777/L3 Communications	PhD. Physics	
Hinshelwood, David	NRL 6773	PhD. Physics	
Jackson, Stuart	NRL 6773	PhD Physics	
Mosher, David	NRL 6771/L3 Communications	PhD Physics	
Ottinger, Paul	NRL 6771/L3 Communications	PhD Physics	
Pereira, Nino	ARL/EcoPulse	PhD Physics	
Schumer, Joseph (PI)	NRL 6770	PhD Nuclear Engineering	
Seely, John (PI)	NRL 7674	PhD Physics	
Weber, Bruce (PI)	NRL 6773	PhD Physics	
Young, Frank	NRL 6773/L3 Communications	PhD Nuclear Physics	
Zier, Jacob NRL 6771		PhD Nuclear Engineering	

All members of this team worked part-time on this project (at most 20% FTE) with allowance of synergism with other projects for scheduled machine operations, diagnostics support, and computational modeling set-up successfully leveraged.

Publications & Technical Reports:

[PER12b] "Near-coincident K-line and K-edge energies as ionization diagnostics for some high atomic number plasmas," N.R. Pereira, B.V. Weber, D.G. Phipps, J.W. Schumer, J.F. Seely, J.J. Carroll, J.R. Vanhoy, K. Slabkowska, and M. Polasik, *submitted to Physics of Plasmas*, 2012.

[RIC12] "Acceleration of ions by an electron beam injected into a closed conducting cavity," A.S. Richardson, P.F. Ottinger, S.B. Swanekamp, and J.W. Schumer, *submitted to Physics of Plasmas*, 2012.

[PER12a] "~10 eV ionization shift in Ir Kalpha2 from a near-coincident Lu K-edge," N.R. Pereira, B.V. Weber, D. Phipps, J.W. Schumer, J.F. Seely, J.J. Carroll, J.R. VanHoy, K. Slabkowska, and M. Polasik, Rev. Sci. Instrum. **83**, 10E110, 2012.

[PER10a] "K-line spectra from tungsten heated by an intense pulsed electron beam," N.R. Pereira, B.V. Weber, J.P. Apruzese, J.W. Schumer, J.F. Seely, C.I. Szabo, C.N. Boyer, D. Mosher, S.J. Stephanakis, and L.T. Hudson, Rev. Sci. Instrum. **81** 10E302, 2010.

[SEL10b] "Spatial resolution of a hard x-ray CCD detector," John F. Seely, Nino R. Pereira, Bruce V. Weber, Joseph W. Schumer, John P. Apruzese, Lawrence T. Hudson, Csilla I. Szabo, Craig N. Boyer, and Scott Skirlo, Applied Optics **49** (23), 10 August 2010.

[SEL10a] "Lateral propagation of MeV electrons generated by femtosecond laser irradiation," J. F. Seely, C. I. Szabo, P. Audebert, E. Brambrink, E. Tabakhoff, and L. T. Hudson, Phys. Plasmas 17, 023102, 2010.

[LIT10] "Anomalous fluorescence line intensity in megavoltage bremsstrahlung," M.S. Litz, G. Merkel, N.R. Pereira, C.N. Boyer, G.E. Holland, J. W. Schumer, J.F. Seely, L.T. Hudson, and J.J. Carroll, Phys. Plasmas 17, 043302, 2010.

[SEM09] "Front and back side processed unintentionally doped GaAs Schottky detectors for Xray detection," F. Semendy, S. Singh, M. Litz, P. Wijewarnasuriya, K. Blaine, N.Dhar, Solid-State Electronics, Oct 2009.

[MOS09] "X-Ray Absorption and Scattering Issues for Rod-Pinch Radiographic Sources," D. Mosher, D.D. Hinshelwood, G. Cooperstein, B. Huhman, R.J. Allen, S.S. Lutz, M.J. Berninger, B.V. Oliver, S. Portillo, and T. Haines, in the Proceedings of the 2009 IEEE Pulsed Power Conference (Washington, DC, June 2009).

[APR09] "Feasibility of Self-Sustained Stimulated Excitation and Decay of Radioisotopes by Resonant Atomic K_{alpha} Radiation in a Two-Element Medium", J. P. Apruzese and J. W. Schumer, Pulsed Power Physics Branch Technote No. 2009-04, 3 March 2009.

[FER09] "Megavoltage bremsstrahlung end point voltage diagnostic," T. Feroli, M.S. Litz, G. Merkel, T. Smith, N. Pereira, J. Carroll, Rev. Sci. Instr. **80** (3), 034301, March 2009.

[WEB08a] "Radiographic Properties of Plasma-Filled Rod-Pinch Diodes," B.V. Weber, R.J. Allen, R.J. Commisso, G. Cooperstein, D.D. Hinshelwood, D. Mosher, D.P. Murphy, P.F. Ottinger, D.G. Phipps, J.W. Schumer, S.J. Stephanakis, S.B. Swanekamp, S. Pope, J. Threadgold, L. Biddle, S. Clough, A. Jones, M. Sinclair, D. Swatton, and T. Carden, IEEE Trans. Plasma Sci. <u>36</u>, 443 (2008).

Interactions/Transitions:

a) Participation/presentations at meetings, conferences, seminars, etc.:

[SEE12] "Tungsten L Transition Line Shapes and Energy Shifts Resulting from Ionization in Warm Dense Plasmas," John Seely, Bruce Weber, David Phipps, Nino Pereira, Uri Feldman, Lawrence Hudson, and Joseph Schumer, 16th International Workshop on Radiative Properties of Hot Dense Matter (RPHDM, Santa Barbara, CA), 5-9 November 2012.

[ZIE12] "Characterization of an Intense Pulsed Photoneutron Source for Active Detection," J.C. Zier, R.J. Allen, J.P. Apruzese, R.J. Commisso, D.D. Hinshelwood, A.L. Hutcheson, S.L. Jackson, L.J. Mitchell, D. Mosher, D.P. Murphy, D.G. Phipps, B.F. Phlips, J.W. Schumer, S.B. Swanekamp, R.S. Woolf, E.A. Wulf, and F.C. Young. 2012 Conference on the Application of Accelerators in Research and Industry (CAARI) (2012), invited.

[MIT12] "Measurements of Neutron Fluence from a Secondary Neutron Converter Irradiated with an 8 MeV Bremsstrahlung Pulsed Power Accelerator Beam," L.J. Mitchell, A.L. Hutcheson, B.F. Phlips, E.A. Wulf, C.S. Gwon, J.C. Zier, S.L. Jackson, J.W. Schumer, R.J.

Commisso, R.S. Woolf, F.C. Young, and L. Jackson. 2012 Conference on the Application of Accelerators in Research and Industry (CAARI) (2012).

[MOS12] "Prompt Sub-MeV Neutron Production from the 7Li(p,n)7Be Reaction on Mercury," D. Mosher, J.P. Apruzese, R.J. Commisso, D.D. Hinshelwood, S.L. Jackson, J.W. Schumer, F.C. Young, J.C. Zier, J. O'Malley, C. Clemett, M. Ellis, P.N. Martin, A. Thandi, J.R. Threadgold, L. Hutcheson, L.J. Mitchell, B.F. Phlips, R.S. Woolf, and E.A. Wulf. *2012 IEEE International Conference on Plasma Science* (Edinburgh, UK, July 2012).

[OTT12] "Virtual Cathode Ion Acceleration behind the Anode Foil of a Pinched-Beam Ion Diode," P.F. Ottinger, A.S. Richardson, S.B. Swanekamp, and J.W. Schumer. 2012 IEEE International Conference on Plasma Science (Edinburgh, UK, July 2012).

[PER12] "Near-coincident K-line and K-edge energies as ionization diagnostic for some high atomic number plasmas," N.R. Pereira, B.V. Weber, D.G. Phipps, J.W. Schumer, J. Vanhoy, J.J. Carroll, and M. Polasik. *19th Topical Conference High-Temperature Plasma Diagnostics* (2012).

[RUB12] "Modelling of the 7Li(p,n)7Be Neutron Yield from Mercury using GEANT-4 and LSP," M. Rubery, J. Threadgold, J. O'Malley, C. Clemett, M. Ellis, P.N. Martin, A. Thandi, J.C. Zier, S.L. Jackson, D.D. Hinshelwood, D. Mosher, R.J. Allen, J.P. Apruzese, R.J. Commisso, D.G. Phipps D.P. Murphy, J.W. Schumer, B.V. Weber, F.C. Young, A. Hutchinson, L. Mitchell, B. Phlips, E. Wulf, and R. Woolf. 2012 IEEE International Conference on Plasma Science (Edinburgh, UK, July 2012).

[SCH10a] "Evidence of Heavy-Ion Reactions from Intense Pulsed Warm, Dense Plasmas," J.W. Schumer, F.C. Young, B.V. Weber, S.L. Jackson, C.N. Boyer, D. Mosher, and S.J. Stephanakis, 2010 IEEE International Conference on Plasma Science (Norfolk, VA, June 2010), p. 384 and 2010 CAARI (Ft. Worth, TX, August 2010).

[WEB10] "K-Shell Spectra of Warm, Dense Plasmas Produced by Intense Pulsed Electron Beams," B.V. Weber, J.P. Apruzese, C.N. Boyer, D. Mosher, J. W. Schumer, J.F. Seely, S.J. Stephanakis, C.J. Szabo, N.R. Pereira, and L.T. Hudson, 2010 IEEE International Conference on Plasma Science (Norfolk, VA, June 2010), p. 227.

[OTT10] "Voltage and Ion Current Measurements for an Ion Diode Driven by Mercury in Positive Polarity with Layered MITL Flow," P.F. Ottinger, R.J. Allen, J.P. Apruzese, D.D. Hinshelwood, S.L. Jackson, D. Murphy, D. Phipps, J.W. Schumer, B.V. Weber, and F.C. Young. 2010 IEEE International Conference on Plasma Science (Norfolk, VA, June 2010), p. 385.

[PER10b] "K-Line Spectra of Warm, Dense Plasmas Produced with Intense Pulsed Electron Beams," N.R. Pereira, B.V. Weber, J.P. Apruzese, D. Mosher, J.W. Schumer, J.F. Seely, C.I. Szabo, C.N. Boyer, S.J. Stephanakis, L.T. Hudson, 18th High-Temperature Plasma Diagnostics Conference (Wildwood, NJ, May 2010).

[WEB09] "HEDP Production from Plasma-Filled Rod-Pinch Diodes," B.V. Weber, R.J. Allen, J.P. Apruzese, R.J. Commisso, G. Cooperstein, D.D. Hinshelwood, D. Mosher, D.P. Murphy, P.F. Ottinger, D.G. Phipps, J.W. Schumer, S.J. Stephanakis, and S.B. Swanekamp. *DOE ReNeW HEDLP Workshop* (Rockville, MD, November 15-18, 2009).

[PER09b] "Anomalous fluorescence line intensity in megavoltage bremsstrahlung," N. Pereira, M. Litz, G. Merkel, J. Schumer, J. Seely, J. Carroll, Bull. Am. Phys. Soc. 54 (15), (2009).

[SCH09a] "Basic research towards use of radioisotopes as an energetic material," J.W. Schumer et al., Energetic Materials Intelligence Seminar (invited), Aberdeen Proving Ground, May 2009.

[SCH09b,c] "Potential for Electron-Beam Ionization for Accelerated Decay of Radioisotopes," J.W. Schumer, J.P. Apruzese, B.V. Weber, J.F. Seely, M.S. Litz, G. Merkel, and N. Pereira, 2009 Winter Colloquium on Physics of Quantum Electronics (Snowbird, Utah, January 2009) and Army Research Laboratory Radioisotope Battery Workshop (Adelphi, MD, February 2009).

[WEB08b] "High Energy-Density Plasma Production from Plasma-Filled Rod-Pinch Diodes," J.W. Schumer, B.V. Weber, D. Mosher, and J.P. Apruzese, Bull. Am. Phys. Soc. 53 (5), 89 (2008).

[LIT08] "Investigations toward the possible use of energy from isomeric nuclei," M. Litz, G. Merkel, N. Pereira, J. Carroll, Bull. Am. Phys. Soc. <u>53</u> (5), 103 (2008).

[BAL08] "Preliminary Tests of Induced Depletion of ^{108m}Ag Using Bremsstrahlung Radiation," T. Balint, G. Trees, I. Mills, M. Ragan, N. Caldwell, T. Harle, R. Gurney, M. Litz, G. Merkel, N. Pereira, M. Helba, H. Roberts, J. Schumer, S. Karamian, J. Carroll, Bull. Am. Phys. Soc. <u>53</u>(3), 2008 (Youngstown, OH).

[SEL08] "Hard X-Ray Inner-Shell Spectra Produced by Intense Picosecond Laser Pulses and Recorded by Cauchois Type Spectrometers," J. Seely, G. Holland, C. Szabo, L. Hudson, A. Henins, P. Audebert, S. Bastini-Ceccotti, and E. Tabakhoff, 21st Intl. Conf. on X-ray and Inner Shell Processes, June 2008.

b) Consultative and advisory functions to other laboratories and agencies and other DoD laboratories. Consulted with DTRA program managers and directors in related 6.2 efforts (various times and locations). Continued discussions of current and future research with Jason Aquino & Ed Scholz of SciTor Corporation (who are working in conjunction with Dr. George Ullrich/SAIC) in the collection of information for DASD/Forces Transformation and Resources (Mr. Mark Gunzinger's office) and OSD/Policy with regard to various nuclear technology options (July 2008 – August 2009). Attended Energetic Materials Intelligence Seminar (McLean, VA, May 2008 and Aberdeen Proving Ground, May 2009) and discussed research with interested parties in attendance. Discussed topics of accelerated decay of radioisotopes with colleagues at Youngstown State University (Dr. Jeff Carroll), Max Planck

Institute for Nuclear Physics (Dr. Adriana Palffy), Institute for Defense Analysis (Dr. Jim Silk), and served on Army Research Laboratory review panel for radioisotope batteries.

c) Transitions. None.

d) New discoveries, inventions, or patent disclosures: None.

e) *Honors/Awards* (not received during this reporting period): Dr. John Apruzese, Fellow of the American Physical Society (APS) and Dr. John Seely, Fellow of the American Physical Society (APS) and Optical Society of America.

[L1103] "Investigations toward the possible tractof energy from isometric model," M. Lizz, G. Atakel N. Freiz, J. Commun. Nerkel N. Freiz, J. Commun. Nerkel N. Freiz, 10 (2008).

[BAL08] Productory Tests of Induced Depletion of ^{15m}Ag Using Branestrahlung Rediction, T. Bullist, G. Treis, I. Mills, M. Ragan, N. Caldwell, T. Hacle, R. Gurney, M. Litz, D. Merkel, N. Pereiro, M. Hichit, H. Raberts, J. Scittamer, S. Kargming, J. Carroll, Bull. Nu-Proc. Soc. 53 (2), 2008 (Young nuwn, OH).

[MIL08] "Last X-tiny Unrer-Shell Spectra Produced by Internet Picturezond Laser Poiston and Recorded by Cauchers Type Spectrovision, J. Scoly, G. Holland, C. Szabo, L. Budsan, A. Perrins, P. Andebert, S. Busini-Ceccoti, and E. Lubakhoff, 21ⁿ Intl. Conf. on X-ray and lower Soft Processes, June 2008.

b) Consultative and advisory functions to order laboratories and operated and after 1967 (observations). Consulted with OTRA program managers and directors in related 6.2 cilorts (various directors and locations). Continued discussions of current and future research with Lason Aquine & Ed Scholz of Sciffer Corporating (who are coping in conjunction with Dr. George Ultrait/SAIC) in the collection of infimumion for DASDForces Fransformation and the conjunction with Dr. George Research (Markes) (Various De Corporating (who are coping in conjunction with Dr. George Ultrait/SAIC) in the collection of infimumion for DASDForces Fransformation and technology optimes (July 2008 – August 2009). Attended Energetic Materials Intelligence discussed for the order of the discussed in the discussed of the discussed for the discussed of the discussed for the discussed for the discussed of the discussed for the discused for the discussed for the discussed for the dis

DEPARTMENT OF DEFENSE

DEFENSE TECHNICAL INFORMATION CENTER 8725 JOHN J. KINGMAN ROAD, SUITE 0944 FT. BELVOIR, VA 22060-6201 ATTN: DTIC/OCA

DEPARTMENT OF DEFENSE CONTRACTORS

EXELIS, INC. 1680 TEXAS STREET, SE KIRTLAND AFB, NM 87117-5669 ATTN: DTRIAC