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EVIDENCE OF THE ELECTRON-SCREENED OPPENHEIMER PHILLIPS REACTIONS ^{162}Er(d,n)^{163}Tm OR ^{162}Er(p,γ)^{163}Tm IN DEUTERATED MATERIALS SUBJECTED TO A LOW-ENERGY PHOTON BEAM

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Abstract

NASA Glenn Research Center (GRC) is investigating electron screened, enhanced nuclear reactions in deuterated materials exposed to photons with kinetic energies above and below the deuteron photo-dissociation energy in a stationary deuteron center-of-mass system using a repurposed medical linear accelerator (LINAC). The objective of the current work is to utilize a photon beam with energies around the deuterium photo-dissociation energy of 2.226MeV to induce possible reactions in deuterated materials and investigate the mechanisms producing these reactions.

Through these investigations, GRC has found that repeatable nuclear reactions have occurred resulting in nuclear products observed using high purity germanium (HPGe) gamma spectroscopy. Evidence of ^{162}Er(d,n)^{163}Tm or ^{162}Er(p,γ)^{163}Tm has been shown with the appearance of gamma peaks coinciding with ^{163}Tm from three samples containing deuterated erbium exposed to a photon beam in three separate experiments. These reactions may be the result of electron-screening and/or a variation of the Oppenheimer-Phillips reaction. This paper describes the theory behind the proposed reactions, the experiments conducted at GRC, and the experimental evidence of the suspected creation of the ^{163}Tm isotope.

THEORETICAL CONSIDERATIONS

Charged particle nuclear interactions, including nuclear fusion, require overcoming the Coulomb barrier. For example, the deuterium fusion reaction resulting in tritium and a proton, d(d,p)t, has a kinetic energy threshold of 10 keV. Yet, a number of accelerator experiments with deuterated metals have observed that the d(d,p)t reaction rates exceed the predicted rates using the theoretical cross-section and by measuring the rate of fusion proton production. Consequently, the electron-screening potential, U_e, has been experimentally measured at over twice the theoretical value resulting in theoretical reaction rate discrepancies in both astrophysical [1] and accelerator-driven nuclear phenomena [2] where for example, Czerski reports an U_e experimental value of 296±15eV vs. a theoretical value 133.8eV for palladium.

These experiments accelerated deuterons at a wide range of metal targets. It was presumed that metal target electrons provided the electron-screening allowing implanted target deuterons to fuse with the incoming deuterons. However, this interpretation ignores the additional free electrons produced as decelerating deuterons locally ionize metal atoms as Pines and Pines have noted [3].

Three of the leading hypotheses investigated which may lead to the found reaction products include:

- Enhanced electron-screening
- Mirror Oppenheimer-Phillips (O-P) reaction
- O-P reaction and then a subsequent proton capture

Electron-Screening

Electrons shield positive charges of reacting nuclei increasing Coulomb barrier penetrability and reducing the Gamow Factor. This reduction enhances nuclear interaction cross-sections thereby increasing nuclear reaction rates. Electron-screening has been observed with accelerated deuteron beams on metals [2] and alloys from aluminum to tungsten, resulting in a range of U_e from 13 eV to 2.1 keV [4]. The latter is especially important given the non-linear Gamow Factor and the threshold for deuterium-tritium fusion at 5 keV.

Oppenheimer-Phillips Reaction

The Oppenheimer-Phillips reaction [5] was initially proposed by Oppenheimer and Phillips to resolve an unexpected excess of protons vs. neutrons being “stripped off” of accelerated deuterons and consumed by a higher Z nucleus. Bethe [6] later commented upon the role of nuclear charge, Z, with Z>30, as being required for the O-P reaction. Some researchers [7] have concluded that deuteron accelerator experiments do not exhibit an excess of protons over neutrons, as in d(d,n)^{3}He, hence no O-P reactions. However, they reported electron-screening. Similarly, GRC may not have observed O-P reactions as the deuterium was stationary with insufficient deuteron acceleration to enable classic O-P reactions. Instead, a variation of the O-P reactions as described below may have occurred.

Enhanced Oppenheimer-Phillips Reaction Irradiating deuterated metal samples with x-rays or gamma rays increases the flux of energetic electrons and may cause a greater likelihood of the O-P reaction to happen, hence an enhanced O-P reaction. Here, an ionized target nucleus with Z > 30 polarizes the positive proton in the deuteron away from the positive target nucleus. Then, the neutron is ‘captured’ and the proton ‘ejected’. Neutron activation of deuterated samples has been observed in LINAC irradiation experiments [8] possibly due to this type of reaction.

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The postulated enhanced O-P reaction and energy levels of the participants in this reaction are indicated below, where M is the target nucleon, d is the deuteron, p is the proton and n is the neutron:

- ${}_{Z}^{A}M(d,p){}^{A+1}{}_{Z}M$
  - E.g. ${}_{68}^{162}Er(d,p){}_{68}^{163}Er$
- Expelled proton has up to > 6 MeV of kinetic energy
  - Deuteron: binding energy (B.E.) of ~2.2 MeV
  - Target nucleus: B.E. of ~8.8 MeV/nucleon
  - Leaves a difference of ≤6.6 MeV where up to 99% of that energy goes off with the ejected proton.

Mirror Oppenheimer-Phillips Reaction If sufficient electron-screening occurs, a Mirror O-P process may occur and the screened proton may be captured by the target nucleus ejecting the neutron instead of the proton. This suggests sufficient screening to prevent deuteron polarization.

The postulated Mirror O-P reaction and energy levels of the participants in this reaction are predicted below:

- ${}_{Z}^{A}M(d,n){}_{Z+1}^{A+1}M$
  - E.g. ${}_{68}^{162}Er(d,n){}_{69}^{163}Tm$
- Expelled neutron has > 6 MeV of kinetic energy
  - Deuteron: B.E. of ~2.2 MeV
  - Target nucleus: B.E. of ~8.8 MeV/nucleon
  - Leaves a difference of ≤6.6 MeV energy where 98% of that energy goes off with the ejected neutron. Note: the neutron leaves with less energy than the proton because of their mass difference.

EXPERIMENT DESCRIPTION

A LINAC Model LS200 manufactured by Varian was modified to expose the samples to photon energies of ≤2.4 MeV very close to its braking target as shown in Fig. 1. For this study, the specimens were positioned within approximately 7.4 mm (0.29 in.) from the exit plane of the tungsten-braking target. At 7.4 mm, it is estimated that the samples saw a radiation dosage of 2.4×10^{6} rad/min at ~2-MeV beam energy. An ionization gauge radiation detector was set up below the test samples at the isocenter ~100 cm from the braking target to monitor radiation levels emanating from the LINAC beam. The radiation level, reflected power, gun current (voltage), and target current (voltage) were recorded and used to monitor beam operation to ensure that the beam flux was not changing with time.

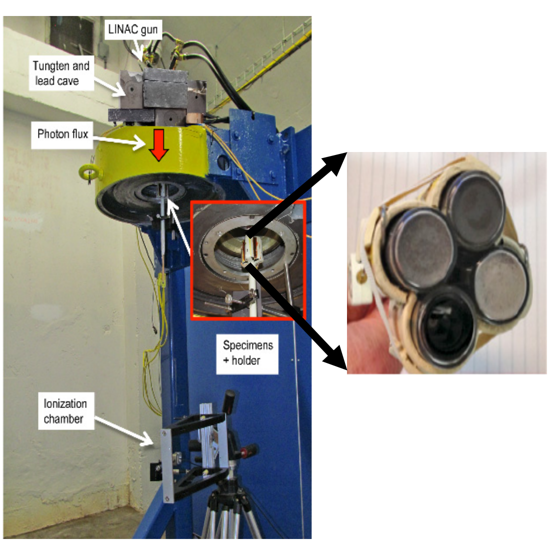

Figure 1: Photograph of LINAC and sample holder with test samples.

The samples exposed in this study were created with deuterium-loaded erbium (99% purity) metal ($ErD_{2.8}$) mixed with deuterated paraffin ($C_{36}D_{74}$) along with molybdenum to act as a neutron witness material. The samples were placed into glass vials and subsequently positioned in the exposure path of a modified industrial LINAC.

EXPERIMENTAL RESULTS

Before beam exposure, each as-received material was scanned for gamma spectral lines and none were found to have any activity different than standard background lines.

Samples were then exposed to the low-energy photon beam for ~4-6 hours. Gamma spectroscopy was used to measure nuclear activity in samples after exposure.

Gamma Spectroscopy Results

The resulting HPGe detector spectra were displayed and gamma peaks were analyzed with the PeakEasy software from Los Alamos National Laboratory [9]. Gamma peaks were checked also with the Lawrence Berkeley National Laboratory database [10], amongst other sources, to confirm identity of the radioisotopes. For every test sample, at least one 15-min duration gamma scan was performed. Follow-up scans were performed on several of the specimens to evaluate half-lives by examining the change in gamma activity levels. Figure 2 shows part of one of the gamma spectra collected showing x-ray lines consistent with ^{163}Tm (via possible O-P reactions with ^{162}Er), ^{163}Er (via neutron capture of ^{162}Er), and ^{165}Er (via neutron capture of ^{164}Er). The presence of ^{171}Er (via neutron capture) was also observed, but not shown in Fig. 2.

It is worth noting that the naturally occurring isotopes [11] of erbium and thulium are: ^{162}Er (0.14%), ^{164}Er (1.61%), ^{166}Er (33.60%), ^{167}Er (22.95%), ^{168}Er (26.80%), and ^{170}Er (14.90%); ^{169}Tm (100%). Any neutron capture of ^{166}Er or ^{167}Er would result in a stable isotope of erbium. Neutron capture of ^{168}Er would result in the production of ^{169}Er with a 9-day half-life, however, the gamma and x-ray

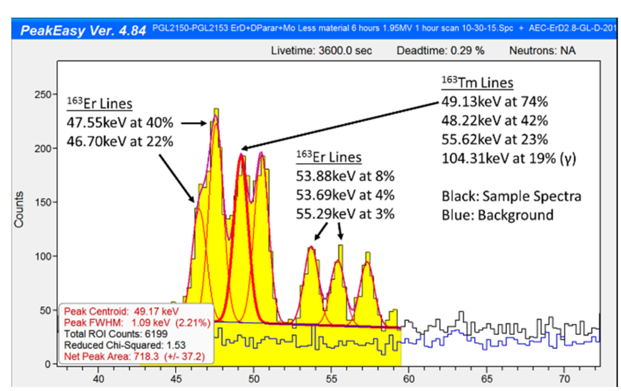

Figure 2: Gamma spectra of deuterated erbium sample.

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peaks of ^{169}Er are very weak and thus would not be seen in the gamma scans. Neutron capture of ^{169}Tm would result in ^{170}Tm with a half-life of 128.6 days and weak x-ray and gamma peaks which would be very difficult to detect.

Gamma peaks significantly above background from the collected gamma spectra were analyzed and separated into groups corresponding to the following radioisotopes: ^{163}Er, ^{171}Er, ^{99}Mo, ^{101}Mo, ^{99m}Tc, and ^{101}Tc. In addition, another radioisotope, ^{163}Tm, was identified consistent with either ^{162}Er(d,n)^{163}Tm or ^{162}Er(p,γ)^{163}Tm. Radioisotope identification was confirmed by identifying that the radioisotope's largest gamma or x-ray peak was present, determining that any subsequent peaks were present, and calculating the half-lives of each peak and comparing with published values.

Table 1 shows gamma results for an exposed sample (PGL2150-2153) containing $ErD_{2.8}$+$C_{36}D_{74}$+Mo and includes the corresponding net area counts, uncertainty, and full width half maximum (FWHM) energy for each gamma peak observed within PeakEasy. Gamma peaks indicating the presence of ^{99}Mo, ^{101}Mo, ^{99m}Tc, and ^{101}Tc were also observed but not reported here. The data included in Table 1 shows evidence of erbium and thulium radioisotopes.

Table 1: Gamma and X-ray Lines of ErD_3 Sample

Radio-isotopes identified	Accepted data per LBNL		Experimental data (60 min gamma scan)			
	Gamma & x-ray energy, keV	Intensity, percent	Centroid energy, keV	Net area counts	Count uncertainty, ±	FWHM
$^{163/165}$Er	46.70	22	46.49	486	34.3	1.09
$^{163/165}$Er	47.55	40	47.58	861	39.3	1.09
^{163}Tm	49.13	74	49.17	718	37.2	1.09
$^{163/165}$Er	53.88	8	53.70	342	31.2	1.09
^{163}Tm	55.62	23	55.41	284	30.0	1.09
^{163}Tm	104.31	19	104.49	25	16.3	0.44
^{171}Er	295.90	29	295.88	3184	58.9	1.09
^{171}Er	308.31	64	308.26	6653	82.7	1.08

Half-life Calculations

The half-life calculations of the ^{163}Tm and ^{163}Er radioisotopes identified in Table 1 were completed for three separate test samples and shown in Table 2. Percent differences from the published values were calculated as noted. Close agreement was found between the 49.13keV x-ray peak and the half-life for ^{163}Tm. The 104.31keV gamma peak was difficult to observe and therefore a half-life calculation was not possible. As noted in Table 2, the 47.55keV x-ray peak's half-life was somewhat higher than the published value of ^{163}Er for a difference of 13.33%. This higher half-life may be due to the known ^{163}Tm(β^+,γ)^{163}Er which would continue to produce ^{163}Er even after the LINAC was turned off.

Production Mechanisms

Mechanisms considered for creating ^{163}Tm include:

1. Neutron Capture of ^{162}Er and then decay to ^{163}Tm: ^{162}Er(n,γ)^{163}Er, however the subsequent decay would be $^{162}Er_{68}$(n,γ)$^{163}Er_{68}$(β^+,γ)$^{163}Ho_{67}$(β^+,γ)$^{163}Dy_{66}$ which is stable, hence there is no path to $^{163}Tm_{69}$.
2. Neutron capture of ^{169}Tm results in ^{170}Tm not ^{163}Tm.
3. Electron-screened, enhanced, Oppenheimer-Phillips reaction: ^{162}Er(d,p)^{163}Er then ^{162}Er(p,γ)^{163}Tm
   - Two Step cascade yields a fast proton then capture
   - JANIS cross-sections exist for (p,n) reaction on ^{166}Er, ^{167}Er, ^{168}Er and ^{170}Er, but no JANIS cross-sections exist for ^{162}Er(p,n)^{163}Tm nor ^{162}Er(p, γ)^{163}Tm.
   - Proton capture is documented by Ozkan, et al [12].
4. Electron-screened, enhanced, Mirror Oppenheimer-Phillips: ^{162}Er(d,n)^{163}Tm
   - Very unlikely that deuteron's proton would be captured over a neutron, however with enough electron-screening, the Coulomb barrier could be overcome.

Table 2: Half-life Determination for Radioisotopes Found

Radioisotope/ published ½ life			Observed x-ray peak (strongest)	Experiment average ½ life	Percent difference
^{163}Tm/ 109 min			49.13 keV		
	PGL2050 (Time between scans: 163 min)				
	Net area counts	Scan 1	1279		
		Scan 2	516		
	Calculated ½ life, min		123.9	124	
	PGL2150-2153 (Time between scans: 36 min)				
	Net area counts	Scan 1	1493		
		Scan 3	1052		
	Calculated ½ life, min		71.7	72	
	PGL2194-2197 (Time between scans: 57 min)				
	Net area counts	Scan 1	2569		
		Scan 4	1885		
	Calculated ½ life, min		127.8	128	
				108 min	**-0.92%**
^{163}Er/ 75 min			47.55 keV		
	PGL2050 (Time between scans: 163 min)				
	Net area counts	Scan 1	1883		
		Scan 2	588		
	Calculated ½ life, min		96.6	97	
	PGL2151-2153 (Time between scans: 17 min)				
	Net area counts	Scan 2	2042		
		Scan 3	1679		
	Calculated ½ life, min		60.3	60	
	PGL2194-2197 (Time between scans: 60 min)				
	Net area counts	Scan 5	2072		
		Scan 6	1351		
	Calculated ½ life, min		97.8	98	
				85 min	**+13.33%**

SUMMARY

NASA Glenn Research Center is investigating electron-screened, enhanced nuclear reactions by irradiating high density deuterated materials using electron LINAC generated bremsstrahlung gamma rays. Observed x-ray peaks and half-life calculations indicate ^{163}Tm was created in three separate experiments. Two possible routes to arrive at ^{163}Tm were postulated including; electron-screened enhanced Mirror O-P reaction of ^{162}Er(d,n)^{163}Tm or proton capture ^{162}Er(p,γ)^{163}Tm after electron-screened O-P reaction of ^{162}Er(d,p)^{163}Er. Confirmation of such reactions would be a significant contribution to the understanding of the Oppenheimer-Phillips process and electron-screening.

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