Gamma Emission Evaluation in Tungsten Irradiated By Low Energy Deuterium Ions

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ABSTRACT

An analysis of tungsten and tantalum foils before, during and after deuterium discharge with the gamma/x-ray spectrometry is described. An increase in light isotopes in tungsten cathodes during and after deuterium discharge was identified using gamma/x-ray spectrometry. The comparison of thermal ionization mass-spectrometry (TIMS) data and a series of energy peaks in gamma-spectra is evidence that the series of peaks observed in gamma spectra belong to the following isotopes:

$^{169}_{70}Yb$; $^{170}_{72}Hf$; $^{171m}_{70}Yb$; $^{172}_{72}Hf$; $^{178}_{70}Yb$; $^{180}_{70}Yb$; $^{180m}_{72}Hf$.

Correlation of TIMS and Gamma spectrometry data lead to the assumption that the appearance of light isotopes in the tungsten was the effect of the initiation of the low energy decay process with deuterium discharge.

1. Introduction

Weak gamma-emission, short-term neutron bursts, neutron and gamma energy-spectra were measured. This data were published in the previous papers [1-2]. Neutrons with energy up to 17 MeV are detected. The intensity ratio of neutron groups (2.45 MeV / 14 MeV) shown the anomalous type of nuclear reaction [1-2]. X-ray film placed in contact with Pd, Ti and Ag were found to be blackened after irradiation with deuterium discharge [3-6]. Blackening of x-ray films placed inside and outside of the discharge chamber with stainless steel wall was observed [6]. In agreement with the radiography, radioactive isotopes corresponding to different energy levels were detected on Pd cathode after deuterium discharge: both high-energy and low-energy components [3].

A change in the level of uranium radioactivity after deuterium and hydrogen discharge was found [7-9]. The observed alpha emission increased by a factor of four after deuterium discharge for 500 hours, on both the irradiated and unirradiated sides. The increase of beta and gamma emission intensity was less than the alpha increase. The comparison of the intensity of energy peaks in gamma spectra allow to see that the intensity ratio (cps) of thorium and uranium in energy peaks was changed [10].

The appearance of increased light isotopes in tungsten and tantalum after exposure to deuterium glow discharge (DGD), measured using x-ray/gamma spectrometry are presented in this article.

2. Methods

2.1 Experimental method

Deuterium discharge was the experimental method. The experimental setup is shown in Fig. 1.

The x-ray/gamma-ray emission was measured with CdTe XR 100T detector (Amptec). The energy peaks in x-ray/gamma spectra of W and Ta before, during and after irradiation in deuterium discharge was estimated. X-ray/gamma emission intensity for the different deuterium discharge parameters was analyzed in counts per second (cps). Gamma spectra during deuterium discharge were measured through a double quartz tube. The average intensity of x-ray and gamma ray was estimated as the total counts per time of spectrum recording. Figure 1 shows the location of the CdTe detector (it is the instrument marked “1”) during deuterium discharge measurements.

Figure 1. The gas glow discharge apparatus with CdTe detector.

2.2. Analytical Method

X-ray/gamma spectrometry was used as the analytical method. The distance between the Be window of detector and W foil during deuterium discharge was ~50 mm (6 mm thickness of double quartz tube + 3 mm layer of water cooling + 40 mm deuterium gas at ~5 Torr of pressure).

The tungsten foil was located in contact with Beryllium window on the distance ~1 mm during gamma/x-ray spectrometry after deuterium discharge exposure.

The analyzed foils had the diameter ~20 mm, and the ion-irradiated zone had a ~12 mm diameter.

2.2.1. Characterization of CdTe detector for the x-ray and gamma ray spectrometry

The XR-100T-CdTe is capable of detecting energies from a few keV to several hundred keV. This system has a thermoelectric cooler with feedback.

Figure 2a. X-ray and gamma ray detector CdTe XR 100T detector (Amptec)

The PX4 is a component in the complete signal processing chain of the nuclear instrumentation system. It replaces many different components in a traditional instrumentation system: the shaping amplifier, the multichannel analyzer, logic devices, high voltage power supplies, and several auxiliary components. The PX4 digitizes the preamplifier output, applies real-time digital processing to the signal, detects the peak amplitude (digitally), and bins this value in its histogramming memory, generating an energy spectrum.

The MultiChannel Analyzer (MCA) gives the number of events with the corresponding peak value.

The CdTe detector of 1 mm thickness has 100% efficiency from 8 keV to 60 keV.

Fluctuations are observed as rise time variations of the voltage step at the output of the charge sensitive preamplifier. As a result, the acquired spectra suffer from increase background counts and degraded energy resolution. Cooling the FET reduces its leakage current and increases the transconductance, which in turn reduce the electronic noise of the system.

The model XR-100T-CdTe is a new high performance X-Ray and gamma ray detector, preamplifier, and cooler system using a $3 \times 3 \times 1$ mm^3 Cadmium Telluride (CdTe) diode detector mounted on a thermoelectric cooler. Also, the input FET and feedback components to the Amptek A250 charge sensitive preamp are mounted on the cooler. The internal components are kept at approximately -30°C, and can be monitored by a temperature sensitive integrated circuit.

The calibration CdTe detector of soft radiation (diameter 9 mm and thickness 1 mm) was made. The efficiency of the CdTe detector is shown below in Fig. 2(b and c) and Table 1. An estimation of efficiency was made with Monte-Carlo calculation for isotropic source for case of the foils in contact with Beryllium window of CdTe detector in Fig.2c and Table 1.

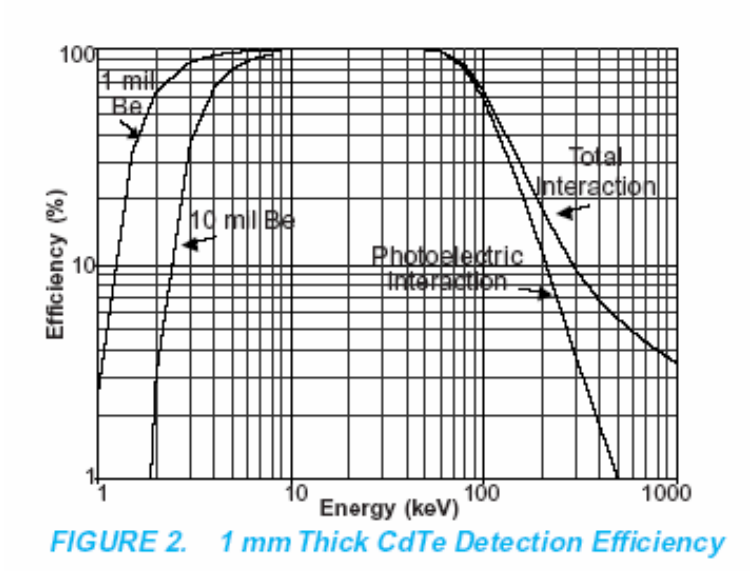


FIGURE 2. 1 mm Thick CdTe Detection Efficiency

Figure 2b. Efficiency of CdTe detector with 250 micrometers thick of Be window.

Table 1. The CdTe detector efficiency with foils in contact with the beryllium window of detector

E,keV	75	100	125	150	175	200	250	300	350	400	450	500
Eff %	48.06	42.0	33.12	25.87	20.08	15.8	9.55	6.0	4.43	3.2	2.34	2.0

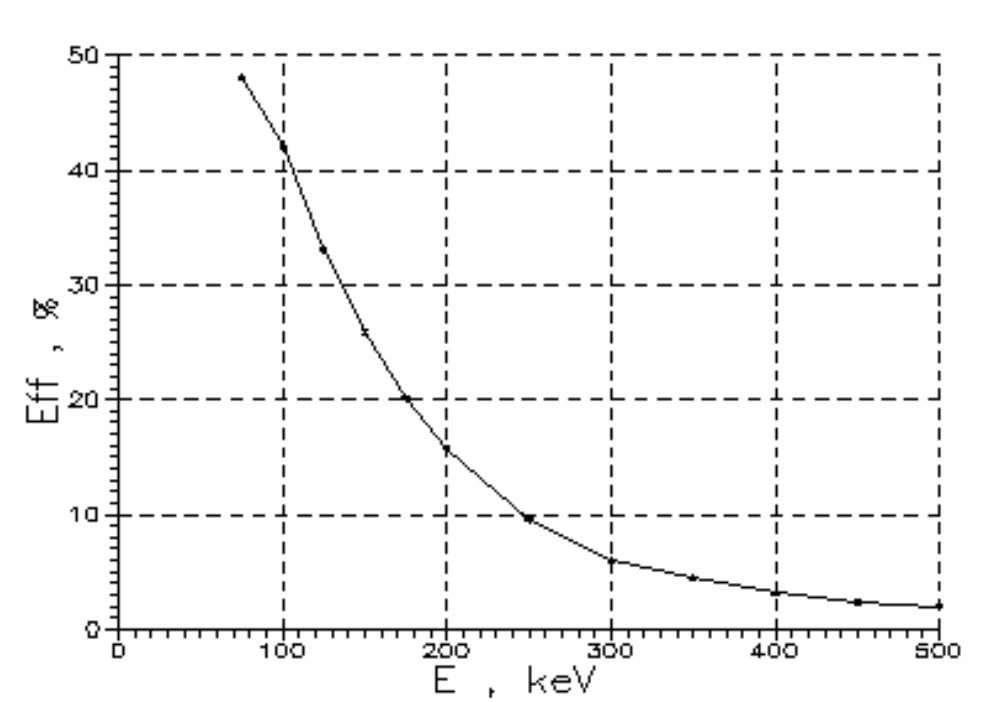


Figure 2c. The efficiency of CdTe detector for case of foils contact with Beryllium window.

2.3. Estimation of Gamma/ X-ray intensity.

An estimation of Gamma/ X-ray intensity as the value of energy peaks was performed. The value is measured in counts per second (cps).

3. Experimental results.

Gamma emission before, during and after Deuterium Discharge (DGD) experiments with W and Ta was measured.

Five series of the gamma emission measurements using gamma/x ray CdTe XR 100T detector were carried out:

1- During DGD experiments outside of double quartz walls;
2- After DGD experiments outside of double quartz walls;
3- W foils after DGD experiments, placed in contact with Be window of gamma/x ray CdTe detector on ~1 mm distance;
4- W foils before experiments
5- Background

It was shown that the gamma/x-ray spectra were reproducible under the same experimental conditions for short time intervals. A comparison of energy peaks in gamma

spectra of tungsten and tantalum after GDG showed repeatability of energy peaks in spectra of foils of various experiments during and after GDG experiments. A series of the main energy peaks of isotopes were found with mass-spectrometry and gamma-spectrometry data (See “Transmutation in Tungsten Irradiated by Low Energy Deuterium Ions” in these proceedings.). The peak values were estimated with two methods: gamma spectrometry data; and the gamma energy listed in a table of isotope decay data [11].

The intensity of gamma emission for the different experiments depended of exposure dose, current density and other experimental parameters.

More intensive energy peaks, reproduced in different experiments, were identified according to the table of isotope decay data [11].

Gamma spectra after experiment allowed us to surmise that gamma/X-ray emission continues after stopping the experiment.

X-ray and gamma-ray spectra for W and Ta during and after irradiation in the deuterium discharge with using Gamma/X-ray CdTe detector is presented in Figures 4 - 6.

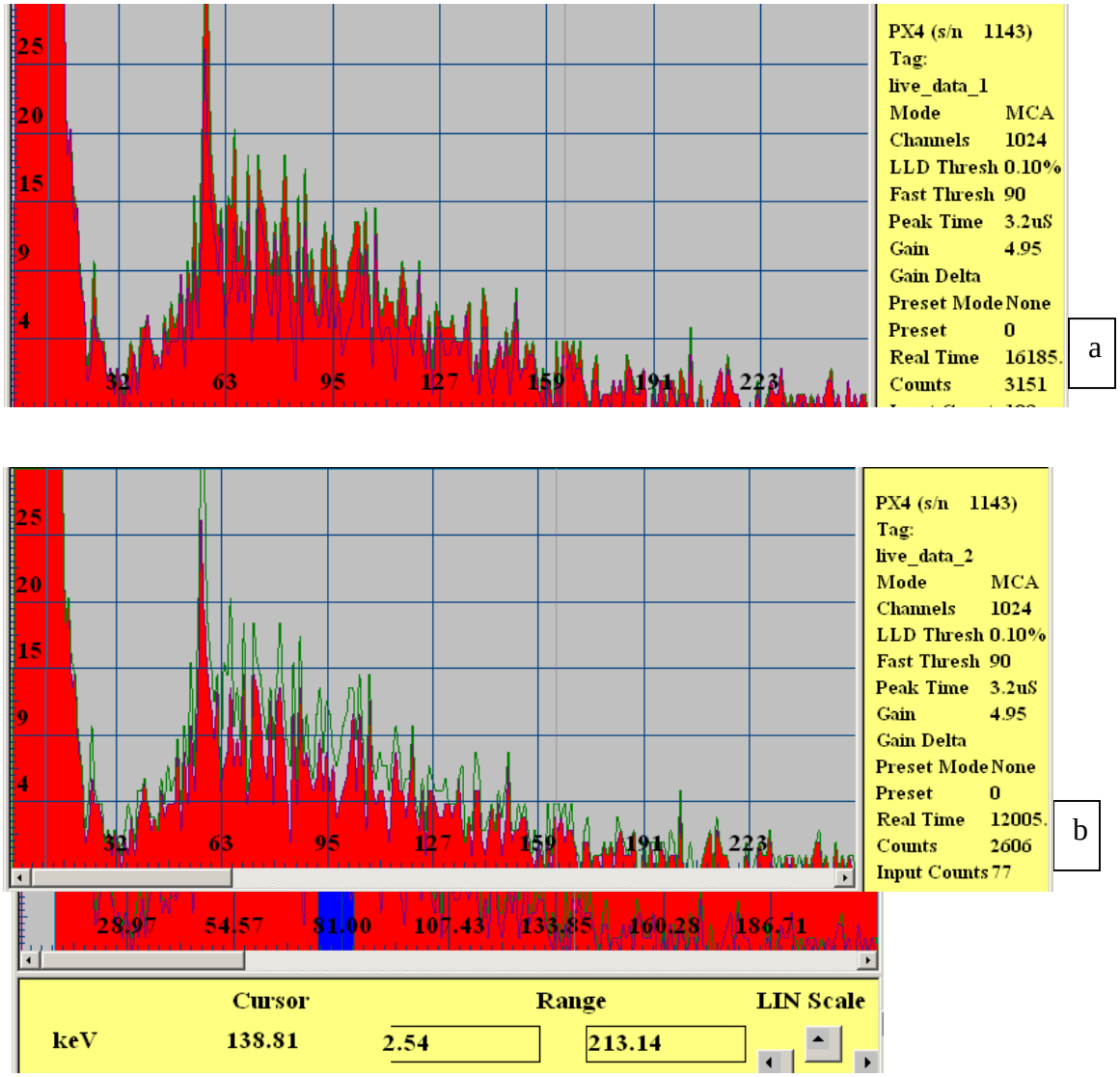


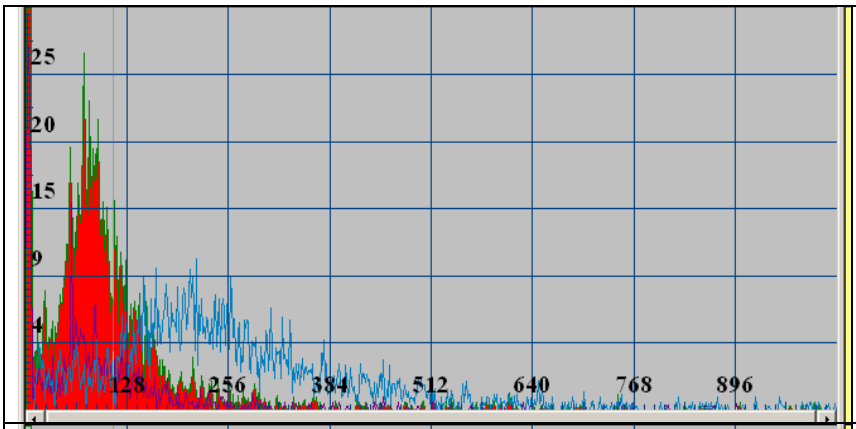
Figure 4. Gamma spectra of tungsten after glow discharge.

Figure 4 includes the x-ray/gamma-ray spectra of W foil (#1817) after deuterium discharge (**a** and **b**) and **c** – the energy in keV.

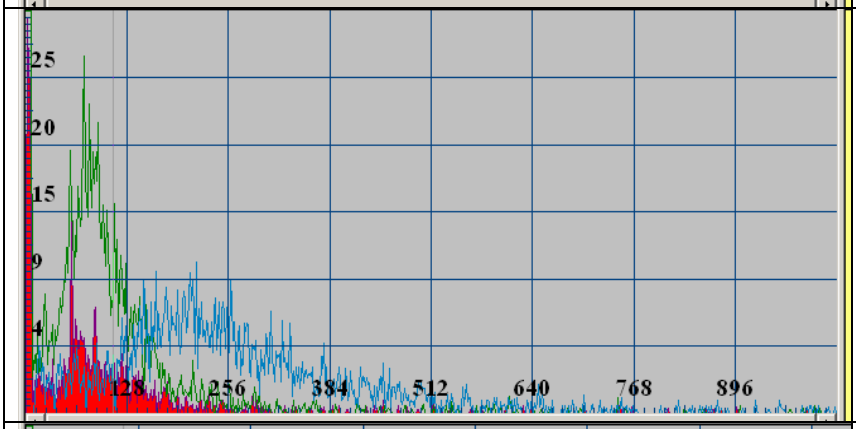
- **a.** 16000sec after Deuterium Discharge; average rate ~0.195 cps;
- **b**. 12000sec after Deuterium Discharge; average rate ~0.217 cps (background's intensity is 0.09± 0.05 cps).

Figure 4 (**a** and **b**) illustrates the reproducibility of the main energy peaks for the different periods (12000 seconds and 16000 seconds). The energy and intensity of the

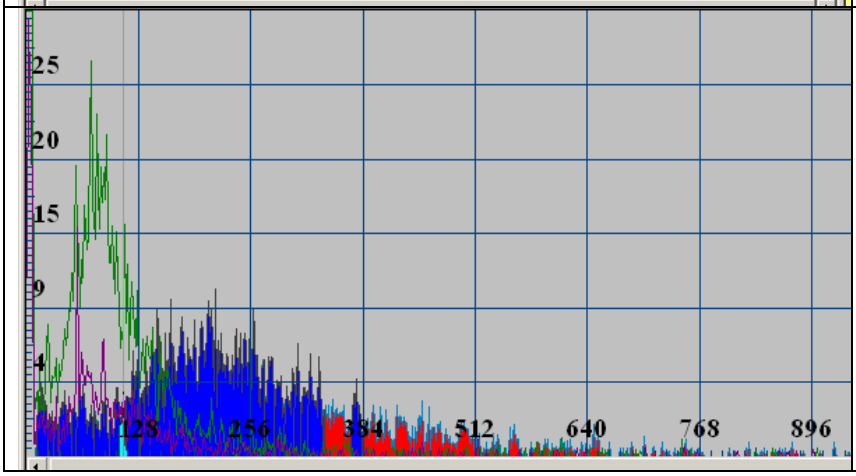
peaks have good reproducibility. The energy peaks of more intensive isotopes in different experiments were reproduced using gamma/x-ray CdTe detector and identified with using an Internet Table of Radioactive Isotopes [11].



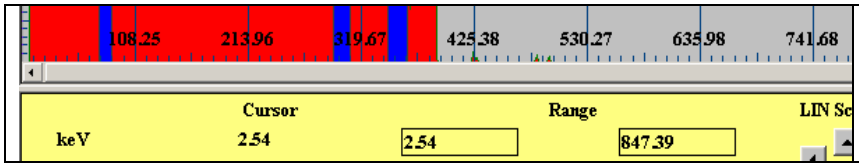
a) Tungsten **during deuterium discharge,** – red square-zone with **peak energy 63.8 keV**, 20 000 seconds time recording, 10000 counts, **average rate of counts ~0.5 cps**



b) Tungsten **after deuterium discharge (** red square-zone) in contact 8251 sec, 773 counts, rate= **0.12 cps**; main energy peaks **~50, 63.8 and ~73,6 keV**



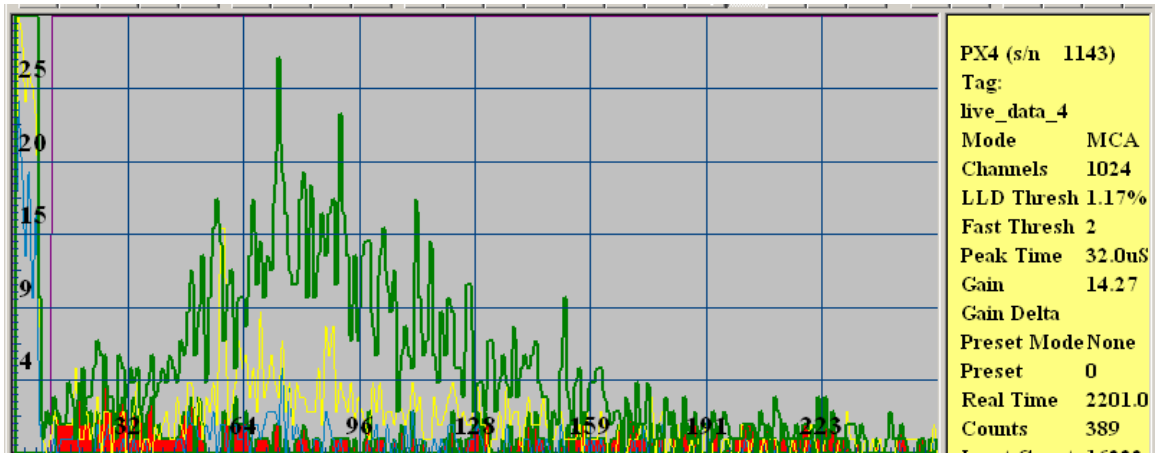
c) Unirradiated tungsten in contact with detector; blue spectrum - **background; time recording –** 20 000 sec., 2093 counts, rate= ~**0.1cps**; region of maximum energy ~**125-215 ke**V



d) Energy in **KeV**

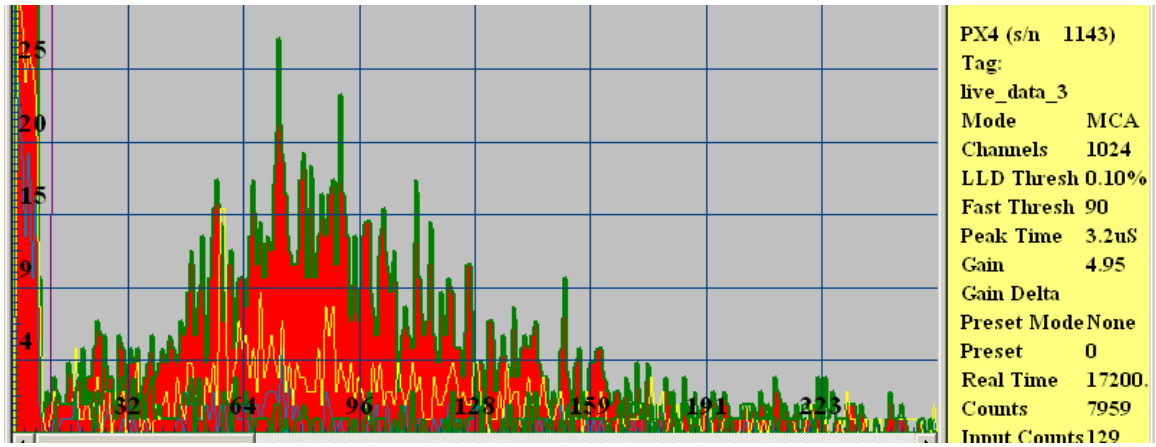
Figure 5. The spectra of x-ray/gamma emission of tungsten #1820 during deuterium discharge (a), after deuterium glow discharge (b) and unirradiated tungsten.

Examples of W foil spectra during of Deuterium discharge.

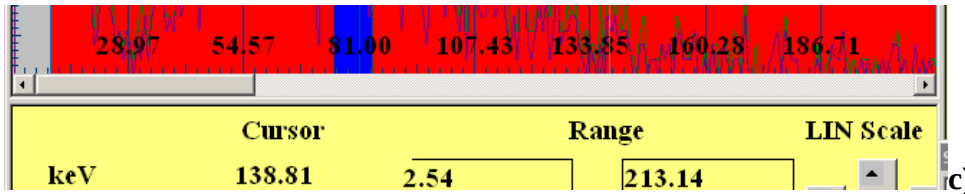


a)

Figure 6 a. Time recording =2200 sec., counts = 389, intensity = 0.17 cps (background intensity = 0.09± 0.05 cps)



b)



c)

Figure 6b. Time recording =17200 sec., counts = 7959, average intensity = 0.46cps; c) –scale in keV.

Figure 6a shows the first stage of deuterium bombardment without significant effects in intensity gamma emission (red zone). Figure 6b illustrates the interval from 4 keV to 200 keV of energy with energy peaks reproduced for the different time duration of recording (yellow and green contour).

The gamma/X-ray intensity of emission for the different spectra was analyzed. The maximums with high intensity of gamma emission are presented on Fig. 7. Peak time is 4.77(7) h in intensity (Table 4) and the peak intensity is 3.5 cps for an experiment lasting ~5.5 hours.

The intensity of gamma emission depended on experimental time, dose, current and other parameters. Gamma/x-ray emission continued after stopping the experiment.

3. Correlation of TIMS and Gamma spectrometry data

3.1. Comparison of main isotopes was observed using two different methods (mass spectrometry and Gamma spectrometry).

The definition of the probable isotopes according to energy peaks in gamma/x-ray spectra and comparison with isotope mass peaks intensity in mass-spectra was performed.

Isotopes of lighter elements during and after exposure in deuterium discharge were selected using the block of energy peaks for each isotope (Table of Radioactive isotopes) and mass spectrometry data.

Table 2. ENERGY PEAKS FROM TUNGSTEN AND TANTALUM AFTER DEUTERIUM GLOW DISCHARGE IN THE GAMMA SPECTRA USING X - ray /Gamma CdTe DETECTOR (for the first group of isotopes)

W	W	W	Ta	Isotope	Eγ keV	Half -life	Decay mode	I γ(%)	Mass*, TIMS
1817	1820	1818	1824						
kev, after									
	20,7	20,7	20,7±1	$^{169}Yb_{70}$	20,75	32d	ε	0,19	169
42±1	43	42	42,18	$^{169}Yb_{70}$	42,76	32d	ε	0,25	169
50±1	51,2	50,44	51	$^{169}Yb_{70}$	51,1	32d	ε	0,018	169
63±1	63	62,83	63,5±0,5	$^{169}Yb_{70}$	63,12	32d	ε	44,2	169

19±1	19,89	19,06	19.1	^{171m}Yb	19,39	5,25 ms	IT	14,8	171
22,5±1	23,19	23,19	23.2	$^{172}_{72}Hf$	23.4	1,87 y	ε		172
24±1	24	24,02	24,84	$^{172}_{72}Hf$	23,93	1,87 y	ε	20,3	172
60±1	60,5	60,35	60,5±0,5	$^{172}_{72}Hf$	60,65	1.87 y	ε	1,1	172
67±1	63	62,35	67.5	$^{172}_{72}Hf$	67,3	1,87 y	ε	5,3	172
91±1	91±1	91,74	91	$^{172}_{72}Hf$	91,3	1,87 y	ε	0,11	172
115±1	114	114,03	114,03	$^{172}_{72}Hf$	114,06	1,87 y	ε	2,6	172
115±1		115	115.6	$^{172}_{72}Hf$	116,1	1,87 y	ε	0,034	172
119±1	119	118,99	119,8	$^{172}_{72}Hf$	119	1,87 y	ε		172
129,03	129	127,25	127.5	$^{172}_{72}Hf$	127,9	1,87 y	ε	1,46	172
42±1	43	42	42,18	$^{178}_{70}Yb$	42,4	74m	β-	6,7	178
13±1	14,1	14,1	13,3	$^{180}_{70}Yb$	13,9	2,4m	β-		180
57±1	58,7	57,05	57,88	$^{180m}_{72}Hf$	57.555	5.5h	IT	48.0	180m

*Mass, measured with TIMS.

- Nine peaks corresponding to $^{172}_{72}Hf$.
- The isotope with IT 99.7%, β-0.3% and T1/2=5.47 h was suggested as $^{180m}Hf_{72,}$ taking into account results from Table 4.

Table 3. PEAK ENERGY FROM TUNGSTEN AND TANTALUM AFTER DEUTERIUM GLOW DISCHARGE IN THE GAMMA SPECTRA USING X - ray /Gamma CdTe DETECTOR (for the second group of isotopes)

W 1817	W 1820	W 1818	Ta 1824	Isotope	Isotope's Eγ, keV	Half-life	Decay mode	I γ(%)	Mass**, TIMS
Kev, contact*									
1	2	3	4	5	6	7	8	9	10
45±1	45,10	44,46	46,46	$^{170}_{72}Hf$	44,52	16,01 h	$\varepsilon+\beta^+$	0,32	170
55,4	55,5	55,4	55,4	$^{170}_{72}Hf$	55,2	16,01 h	$\varepsilon+\beta^+$	1.1	170
99,9	99,0	100,8	100,8	$^{170}_{72}Hf$	99,93	16,01h	$\varepsilon+\beta^+$	2	170
113,2	113,3	113,2	113,2	$^{170}_{72}Hf$	113,9	16,01h	$\varepsilon+\beta^+$	0.18	170
115,6	115,0	115,7	115.6	$^{170}_{72}Hf$	115,5	16,01h	$\varepsilon+\beta^+$	0,2	170
133,0	132,0	132,2	132,7	$^{170}_{72}Hf$	132,2	16,01h	$\varepsilon+\beta^+$	0,044	170
138,81	138,5	138,0	138,8	$^{170}_{72}Hf$	139,2	16,01h	$\varepsilon+\beta^+$	0,018	170

- These gamma energy peaks were observed for different W and Ta foils. In other words, the same isotopes are created under different conditions at deuterium discharge and after the experiment was stopped.
- Contact* - foil after exposure at deuterium discharge was located in contact with the detector's Be window during gamma spectrometry.
- ** Column 10 includes the mass measured with TIMS.
- Seven peaks corresponded to $^{170}_{72}Hf$.

Based on a comparison of these data, we suppose the gamma spectra peaks observed belong to the following isotopes: $^{169}_{70}Yb$; $^{170}_{72}Hf$; $^{171m}_{70}Yb$; $^{172}_{72}Hf$; $^{178}_{70}Yb$; $^{180}_{70}Yb$; $^{180m}_{72}Hf$.

We can see the same isotopes in Table 2 and Table 3, formed in W and Ta under the different experimental conditions after deuterium discharge.

Correlation of TIMS and Gamma spectrometry data leads to the assumption that the heavy isotopes decay under low energy impact.

Main isotopes Hf and Yb in different experiments with W and Ta foils for various time intervals were observed.

3.2. Estimation of Gamma/ X-ray emission intensity (value of energy peaks) and mass spectrometry data were performed

The maximum emission intensity was 3.5 cps after an experiment lasting ~5.5 hours. The intensity, estimated as integral counts of full duration of spectrum recording per second, designated as average intensity, was 0.5 cps. It is five times more than background. Background = 0.09±0.005 cps.

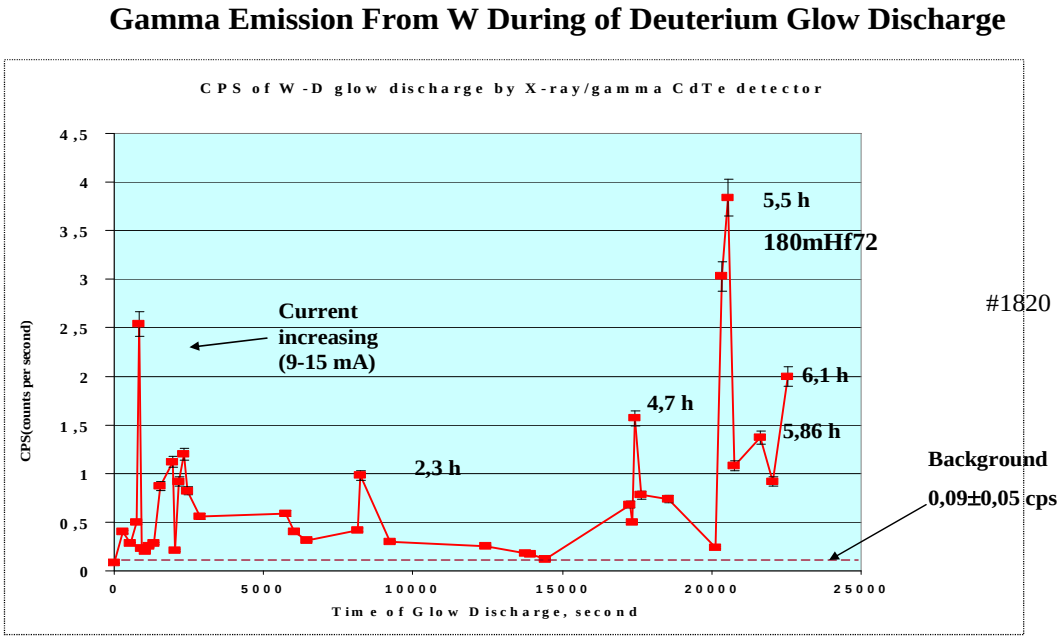


Figure 7. The intensity of gamma emission in CPS vs. experimental time during Deuterium discharge.

The majority of the isotopes were identified by the main energy peaks for X-ray and gamma-ray relating to masses, which were found by the peaks of intensity measured in counts per second (cps) (Fig. 7) and by mass-spectrometry analyses. Some repeatable measurements of peak time are included in Table 4.

Table 4. INTENSITY PEAKS VS. PEAK TIME IN GAMMA SPECTROMETRY MEASUREMENT

1817 Cps	Peak Time after, min	1818 Cps	Peak Time after, min	1819 Cps	Time Peak after, min	1820 Cps	Peak Time after, min	1820 Cps	Peak Time during, min	Average peak time, min	Suggested Isotope (half life)
1*	**2***	**3***	**4***	**5***	**6***	**7***	**8***	**9****	**10****	**11**	**12**
		0.16	***13***			**0.48**	***14.3***	**2.54**	***14.7***	***14.5±0.2***	$^{182}_{77}Ir$ (15m)
8	**24.5**	**0.5**	***25***								
1.28	***28***	**0.44**	***28***	**0.91**	***~30***	**0.37**	***27.6***		***27.6***	***29±1***	$^{190}_{74}W$ (30m)
		0,5	**35**	**0.96**	**34.16**			**1.12**	**35**	***34.7±0.5***	$^{175m}_{74}W$ (35m)
31.4	**43**			**1.57**	**40**			**1.20**	**44.6**	***42.5±2.5***	
		0.21	**51.7**	**0.82**	**52**			**0.56**	**50**	***51±1***	$^{167}_{71}Lu$ (51.5m)

- *Column 1-8 – the measurements of gamma emission after deuterium discharge in contact of foil with detector (a ~10 minute pause after GDG ended was taken into account).
- ** Column 9-10 - the measurements of gamma emission for contact of detector with double quartz tube. It means that CdTe detector was on the ~50 mm distance from discharge zone during Deuterium Discharge and measurements were made through double quartz tube.
- Background was 0.09 ± 0.006 cps for the most measurements.
- The counts and time at seconds in Table 4 were written down by hand. It means that the intensity in cps and peak time could not be measured very exactly.

The isotopes that presumably caused the most intensive gamma emission, corresponding to maximums of gamma intensity in Fig. 7, are listed in column 12 of Tables 4 and 5.

Some time peaks with maximal intensity are presented in Table 5 for experiments with W foils.

Table 5

Cps	**Peak Time in Experiments**	**Suggested Isotope(half life)**
1.6	**6.66 m**	${}^{172}_{74}W$ (6.6m); ${}^{168m}_{71}Lu$ (6.7 m); ${}^{166}_{72}Hf$ (6.77m)
1.14	**10.8 m**	${}^{189}_{74}W$ (10.7 m); ${}^{165}_{71}Lu$ (10.74 m)
0,52	**61.5 m**	${}^{182}_{72}Hf$ (61.5m)
1.0	**74 m**	${}^{178}_{70}Yb$ (74m)
0.32	**2 h**	${}^{177}_{70}Yb$ (1.9 h)
0.68	**4.8 h**	${}^{192}_{80}Hg$ (4.85h)
1.57	**5.5 h**	${}^{180m}_{72}Hf$ **(5.5h)**
0.24	**21.6 h**	${}^{180m}_{72}Os$ (22.1 h)

Mass defect, spin and parity were included for the following formula:

$$ {}^{182}_{74}W + {}^{2}_{1}D \rightarrow {}^{178}_{72}Hf + {}^{178}_{70}Yb + {}^{12}_{5}B \sim +1.22MeB $$

-48 (0+) +13(1+) = -49.7(0+) +13.37(1+) ~1.22 MeV

Tungsten is supposed to be used in the first wall of a thermonuclear plasma reactor. Therefore researchers responsible for stability of the first wall should be aware of the formation of new groups of radioactive isotopes under irradiation by low-energy deuterium ions.

Conclusion

1. X-ray/gamma emission occurs *during* deuterium discharge experiments under 1000 ± 200 Voltage and *after stopping* of these experiments from the foils irradiated by deuterium ions.
2. The same isotopes are found from experiments under different conditions, and both during and after deuterium discharge.

3. The several x-ray and gamma of peaks and radiation intensity (cps) have been chosen for each prospective isotope $^{169}_{70}Yb$; $^{170}_{72}Hf$; $^{171m}_{70}Yb$; $^{172}_{72}Hf$; $^{178}_{70}Yb$; $^{180}_{70}Yb$; $^{180m}_{72}Hf$.
4. The *comparison of thermoionization mass-spectrometry and x-ray/gamma* emission data allowed us to suppose that the peaks observed in gamma spectra belong to the following isotopes: $^{169}_{70}Yb$; $^{170}_{72}Hf$; $^{171m}_{70}Yb$; $^{172}_{72}Hf$; $^{178}_{70}Yb$; $^{180}_{70}Yb$; $^{180m}_{72}Hf$.
5. Correlation of TIMS and Gamma spectrometry data leads to the assumption that *the appearance of light isotopes in the tungsten and tantalum was the result (effect) of the stimulation (initiation)* by low energy deuterium discharge.
6. The formation of new groups of radioactive isotopes under irradiation by low-energy ions of deuterium must be taken into account in order to estimate of operational properties of the first wall of a thermonuclear reactor.

Acknowledgments

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