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DECAY OF TUNGSTEN UNDER LOW- ENERGY DEUTERIUM DISCHARGE AND CREATION OF MORE LIGHT ISOTOPES

8 Workshop on Anomalies in Hydrogen/Deuterium Loaded metals, Catania, 13-18 October, 2007

1. CREATION OF MORE LIGHT ELEMENTS IN TUNGSTEN IRRADIATED BY LOW-ENERGY DEUTERIUM IONS.

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CONTENT : RESULTS OF THE THERMOIONISATION MASS-SPECTROMETRY OF TUNGSTEN AND TANTALUMAFTER DEUTERIUM DISCHARGE

1. **Tungsten Isotopes Decay In More Light Elements After Deuterium Discharge.**
   Last results of experiments with refractory metals (W, Ta) during and after Deuterium Discharge are presented.

1.1. Two series of experiments were fulfilled:

First set–"right away"- W foil was analyzed with thermoionization mass-spectrometer (TIMS) - every 15 minutes after experiments in deuterium discharge.

Second set - W foils were analyzed with TIMS ~ after 3, 4, 5 months.

1. 2. Comparison of original W (and Ta) foil mass spectra with mass spectra after deuterium bombardment was carry out.

1.3. Calibration and reproducibility of mass spectra were realized.

1.4. The increasing of the peak magnitudes in mass spectra for separate isotopes with masses more light than W isotopes by factor 5 – 1000 was observed.

METHODS:

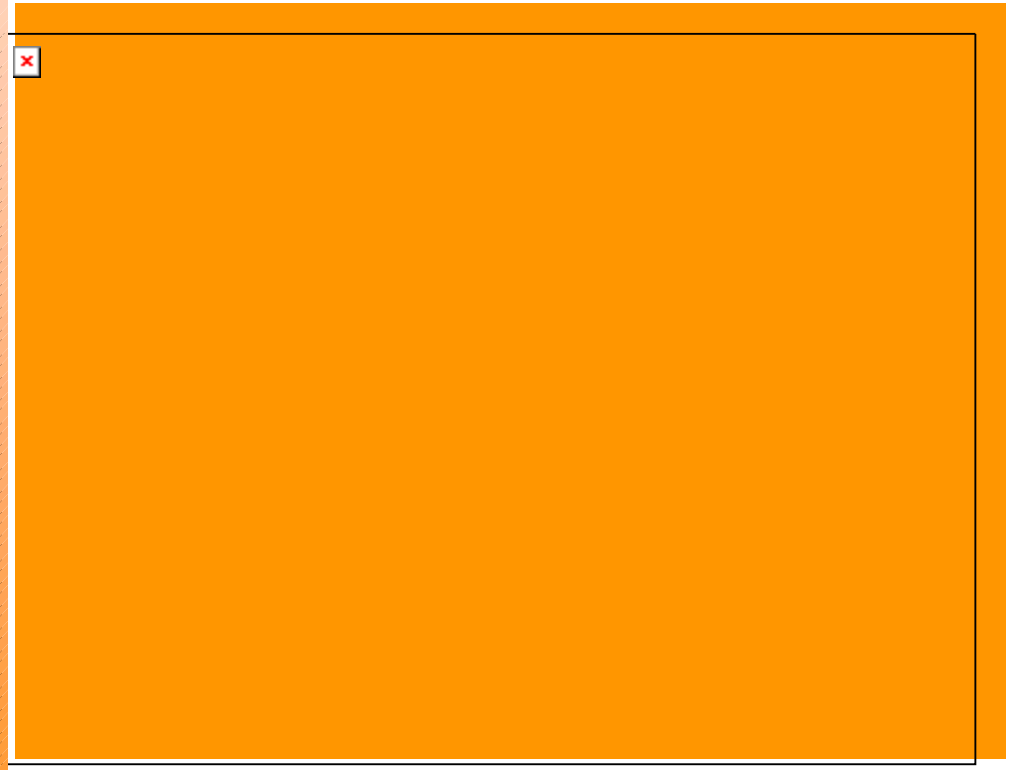
1. Deuterium discharge was used as experimental method .

2. Thermoionization mass-spectrometry method was used as analytical method.

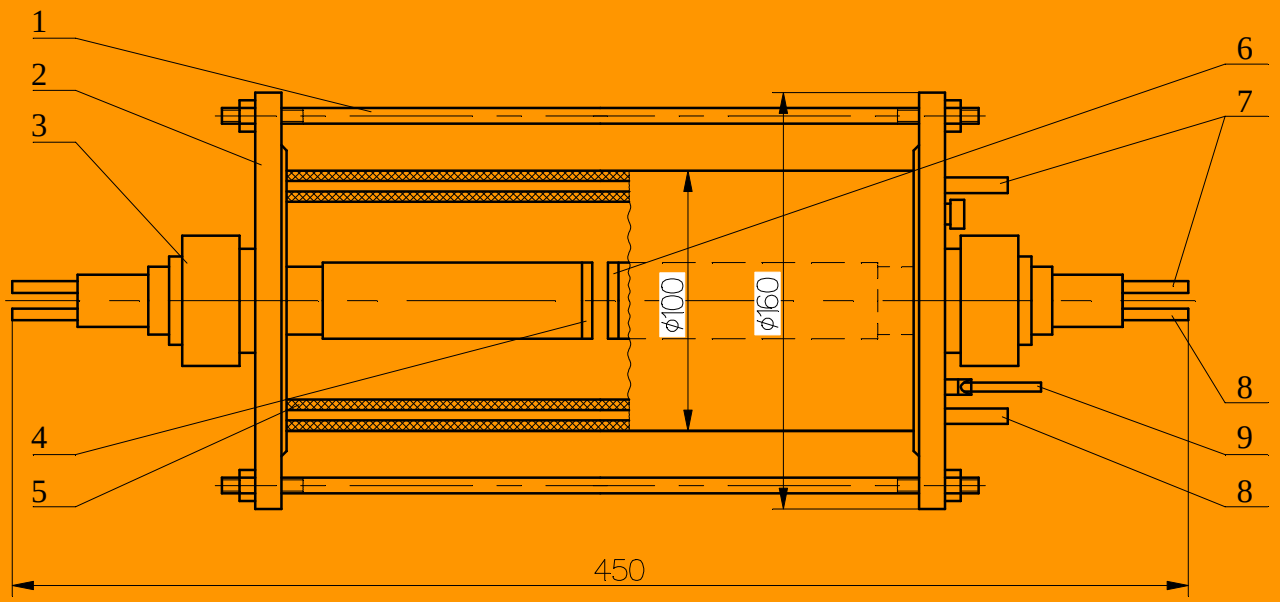
3. Estimation of isotopes intensity in mass spectra under thermoionozation mass-spectrometry (TIMS) is done in CPS (counts per second)

GLOW DISCHARGE INSTALLATION

- Glow Discharge Installation was made with using of double quartz tubes with cooling water between these quartz tubes.
- The sequence of operations before discharge experiments was following:
  - Vacuum degassing into 10^{-3} Torr,
  - Deuterium loading ~3-10 Torr.
- Molybdenum was used as Anode
- W foil was placed on Cathode.
- W foil had ~ 100 microns thickness and ~20 mm diameter.
- Anode and Cathode were cooled by flow water.



Glow Discharge Installation



1-tie, 2-flange, 3 - input of power, 4 – anode, 5–double quartz tube, 6 – cathode, 7,8 - input and output of the cooling water (anode, cathode and space between double quartz tube),

TUNGSTEN FOILS AFTER IRRADIATION

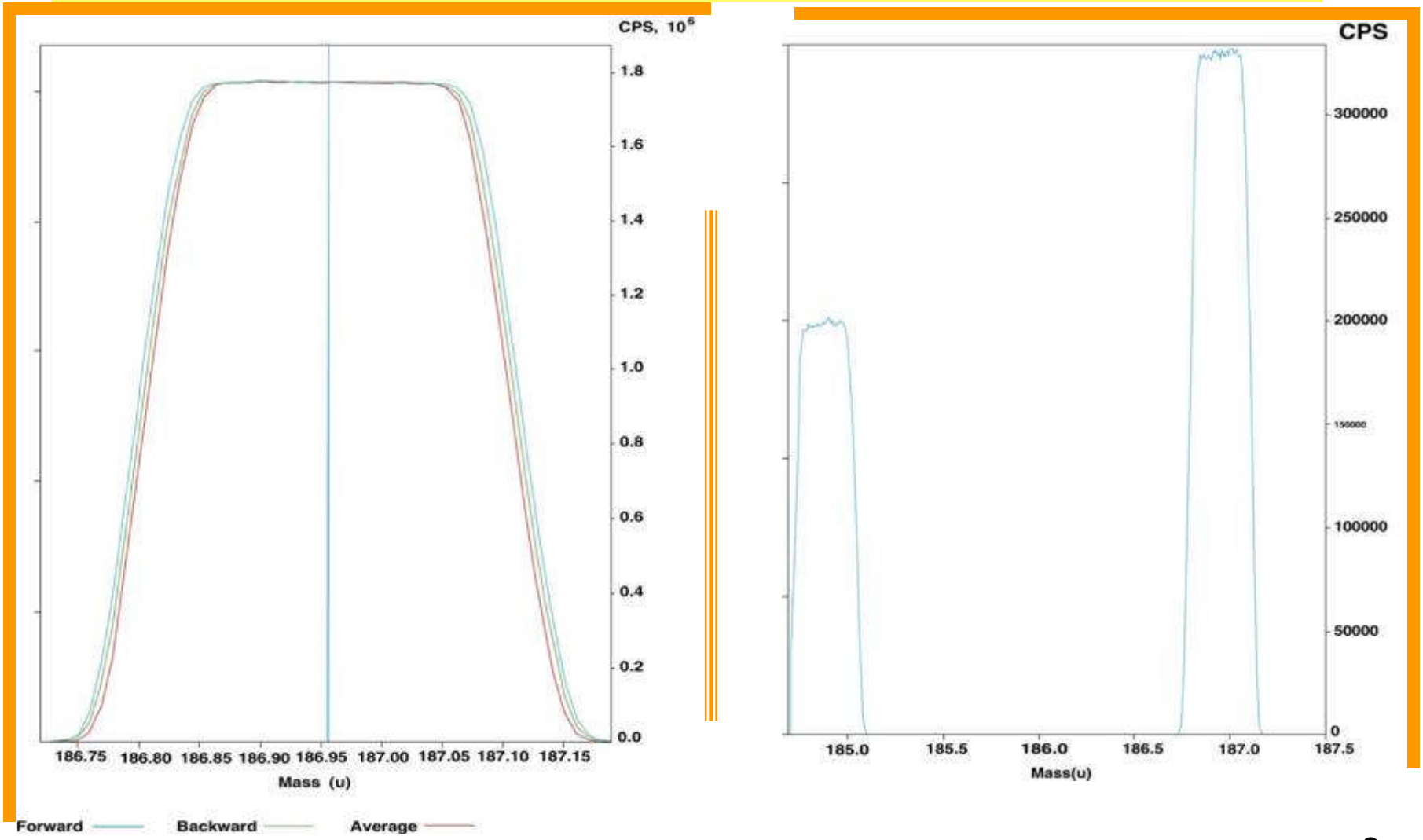
GAMMA/ X- RAY DETECTOR (CdTe)

Location of CdTe detector

The distance between Be window of detector and W foil during deuterium discharge is about ~ 50 mm (6mm is the thickness of double quartz tube + 3 mm layer of cooling flow water + 40 mm Deuterium gas pressure ~ 5Torr); W foil was located in contact with Be window on distance ~1mm after deuterium discharge.

Calibration of TIMS spectra on Re cathode

Example of precise definition of isotope mass:
left specter 186,95 mass - Re; right specter – Re185 and Re 187



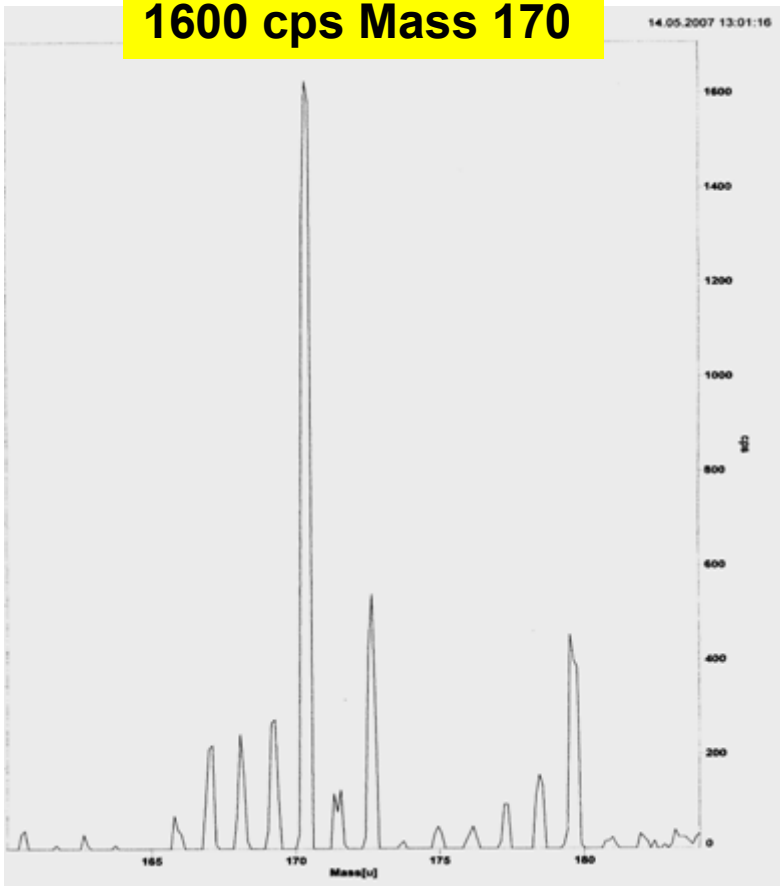
CHARACTERIZATION OF TIMS ANALYZES:

- The analysis was performed using «Finnigan» MAT-262 and "Triton" mass - spectrometers in range of masses 3-210. The main estimation was main in 166-206 range of masses.
- Temperature of foils analyzed was ~ 1800ºC
- The analyzed tungsten strip had the width ~ 1 mm, the length ~ 20 mm and the thickness ~ 100 microns. The strip was cut off from the central part of W foil, irradiated by Deuterons.
- Analyzing zone included the unirradiated part of W foil as well, which led to reduction of lighter isotopes contribution.
- The spectra of minimal intensity (CPS) were removed from the table of TIMS data.
- The data regarding to mass numbers 185 and 187 corresponding to Rhenium (Re) isotopes were removed from the table data as well, due to Re usage for TIMS device as a cathode.
- The cathode had two W foils layers.
- Side of foil irradiated with deuterium ions was analyzed.

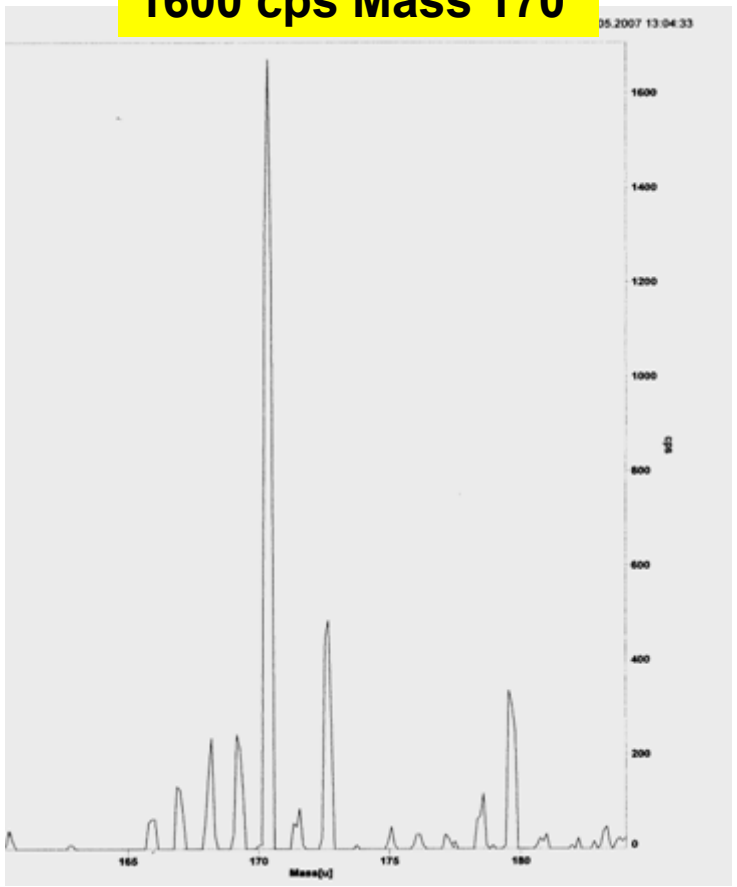
TIMS Spectra of the Same W Foil after Deuterium Discharge.

Time Interval Between the Right and Left Spectra was about ~ 3 Minutes.
(The main isotopes spectra are similar.)

14.05.07 13:01
1600 cps Mass 170

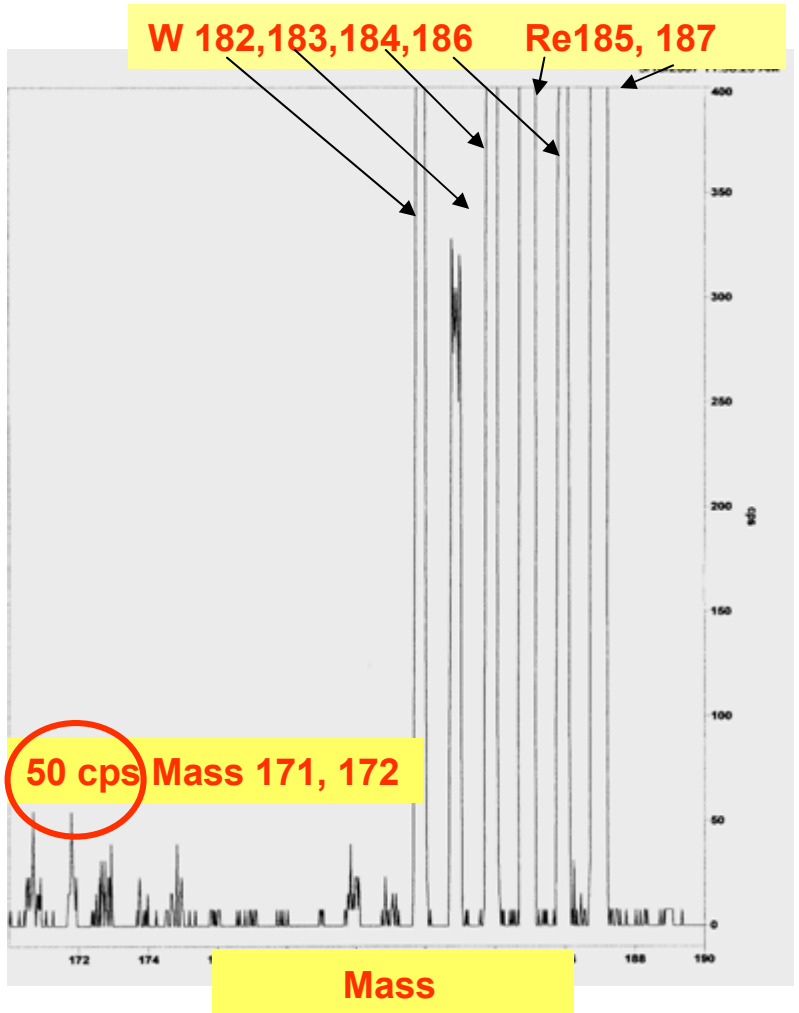


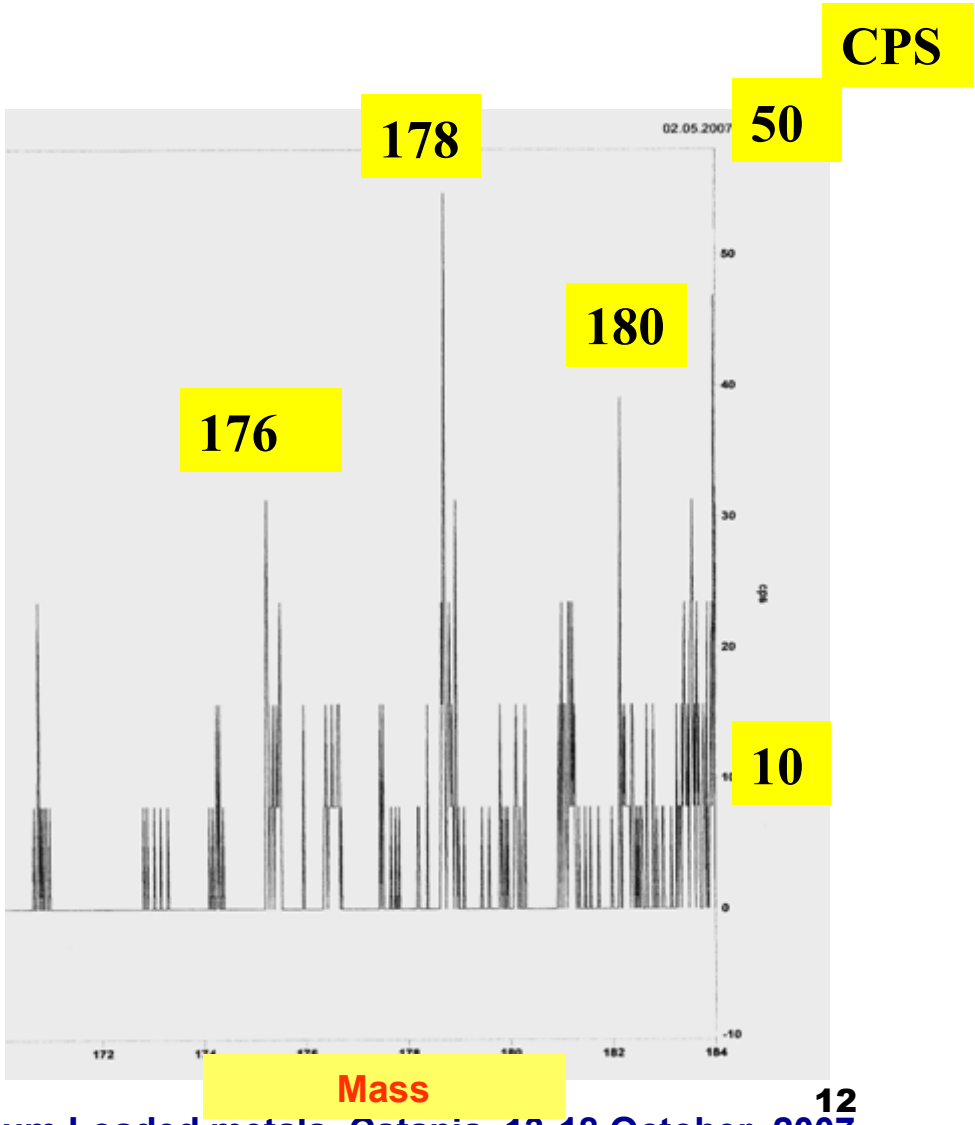
14.05.07 13:04
1600 cps Mass 170



Mass spectrum of original W in the 170-182 mass range (CPS)

The intensity of isotopes with mass numbers 170-190 was 10- 50 cps.

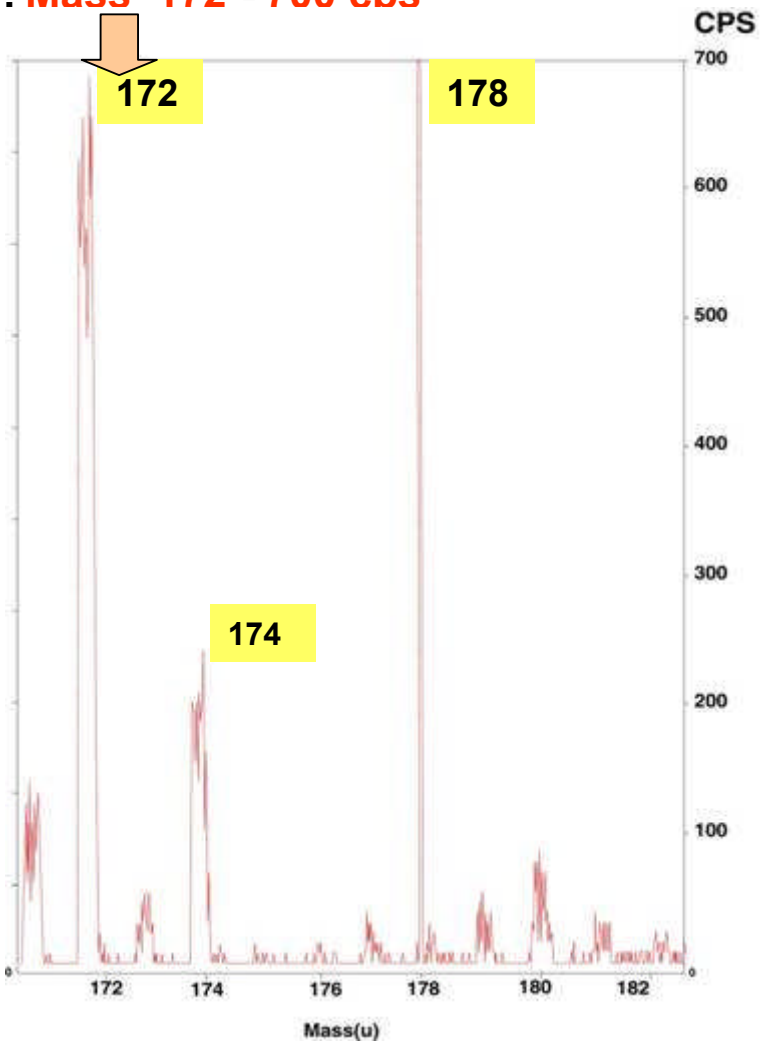




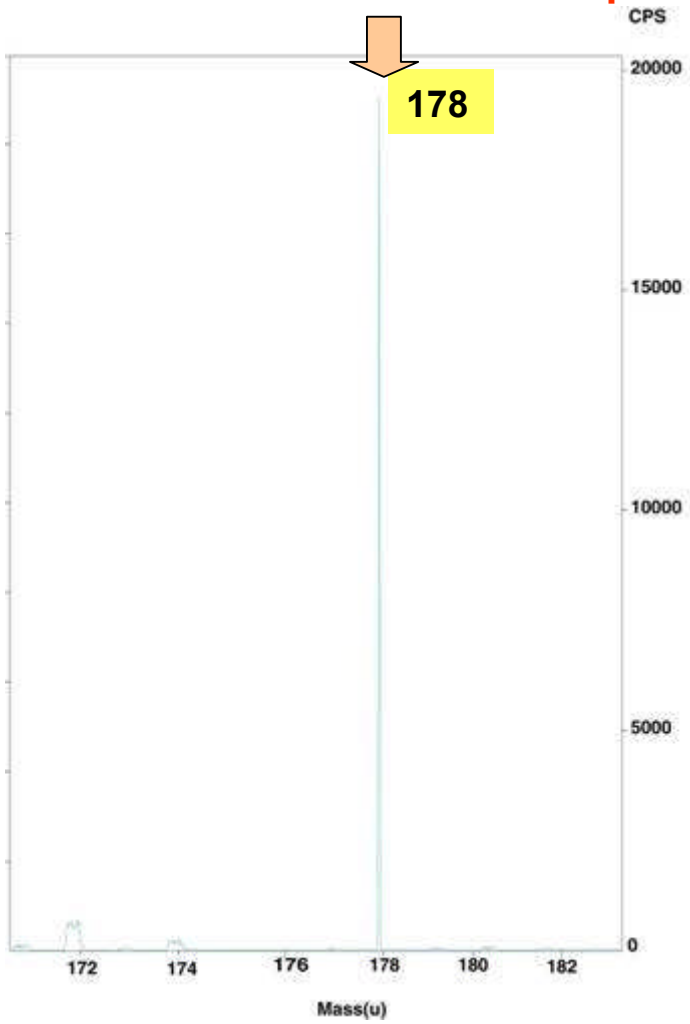
Mass spectra of W (1820-23.04.07) after Deuterium discharge for the same foil.

The intensity of 172 and 178 masses increased into 12 and 400 times, accordingly, after discharge

1. Mass 172 - 700 cps

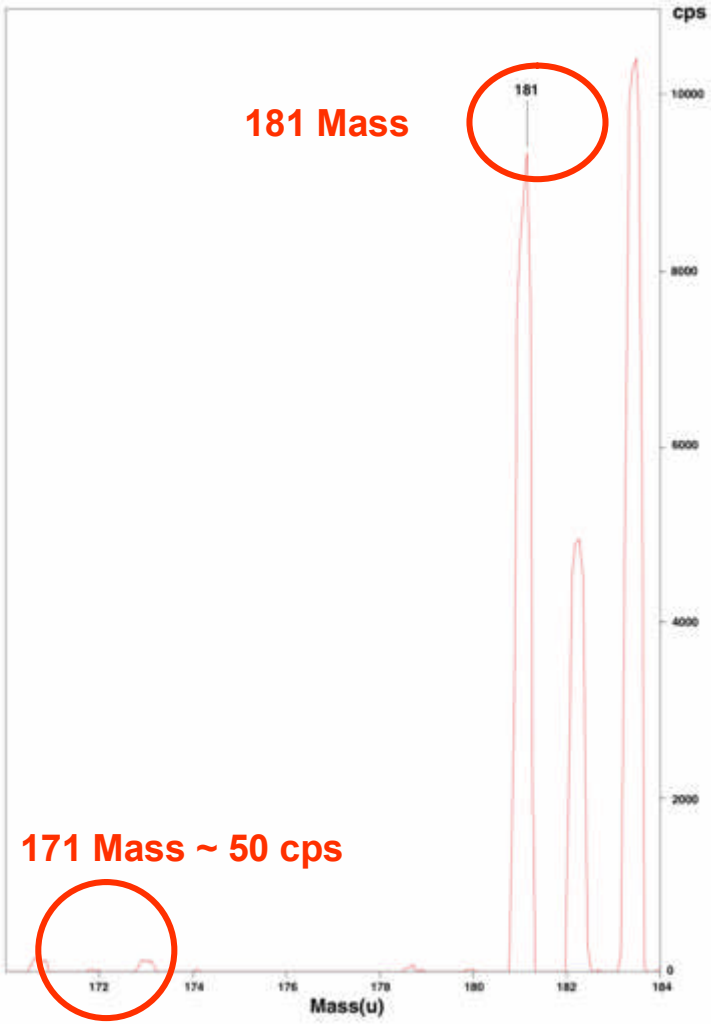


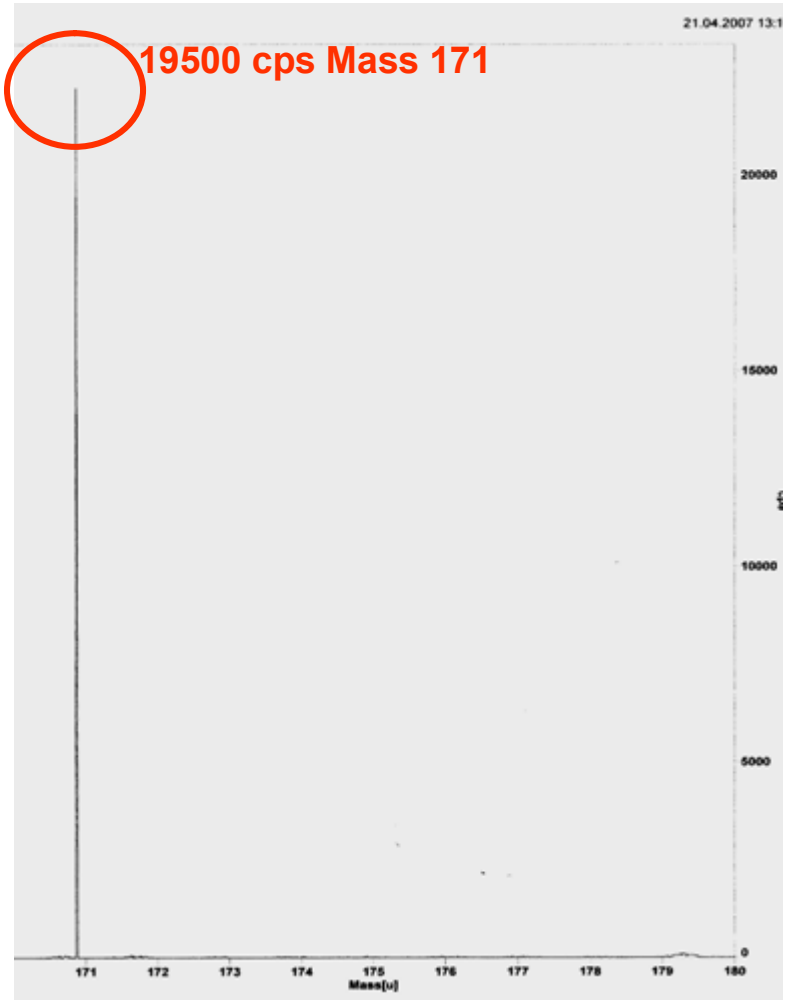
2- Mass 178 - 20000 cps



The creation of more light isotopes in Ta. Ta before (left) and after (right) the Deuterium Discharge. The intensity increasing is ~2000 times

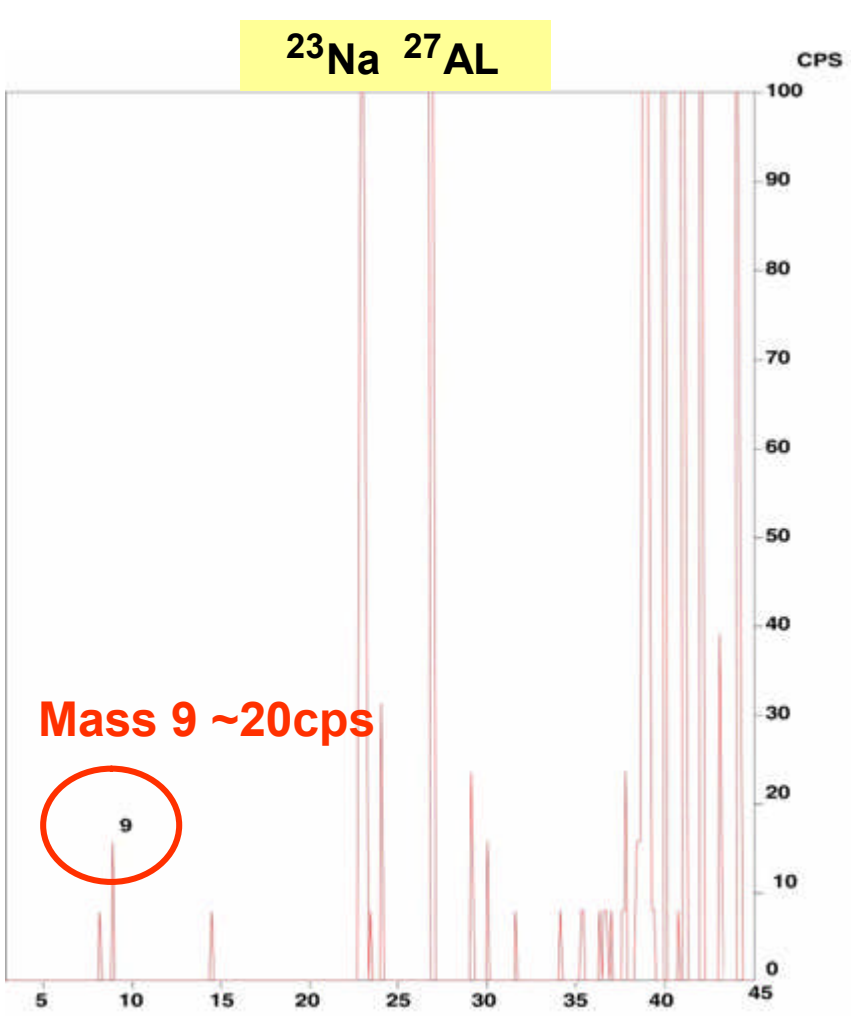
Before: intensity of 171 mass =50cps **After: intensity of 171 mass =19500cps**

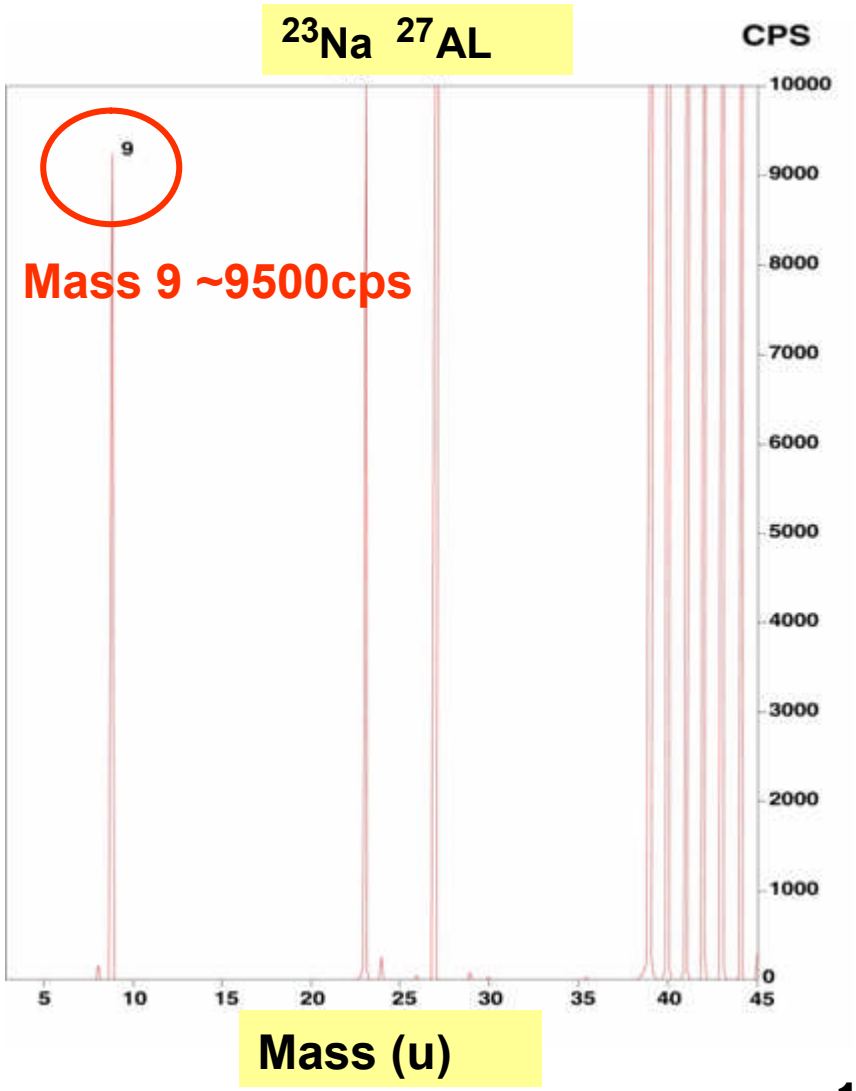




Mass 9 in Ta before (left) and after (right) of the Deuterium Discharge

The intensity increasing of mass 9 is ~ 475 times.





TUNGSTEN, IRRADIATED BY DEUTERIUM AND ANALYZED BY TERMO-IONIZATION MASS-SPECTROMETRY METHOD (COUNTS PER SECOND – CPS)

Time* / Mass	84*	101*	137*	1062*	1073*	1133*	1150*	**
168			40	30	60	2000	30	10 ±10
170			40	55	50	1600	100	5 ±5
171			60	95	100	100	70	5 ±5
172			70	100	100	200	100	0
173			80	75	70	300	100	15 ±15
174			30	55	60	200	100	5 ±5
175			40	55	70	40	85	5 ±5
176			40	55	40	95	75	5 ±5
177			40	55	40	10	100	10 ±10
180		70	10	45	100	20	30	25 ±5
181		100	10	30	40	50		5 ±5
189	70		20	30	10		50	5 ±5
193	60		20	30	10		0	5 ±5
194	70		40	65	0		10	10 ±10

Set 1. This demonstrates that decay of the heavier isotopes into the lighter ones goes on after stopping of the experiment.

*Minutes after experiment

**intensity of the original W in cps

TUNGSTEN AFTER DEUTERIUM DISCHARGE ANALYSED BY TIMS (CPS)

set 2

# exper	1817				1820					1821	Original
Date	16.3.7	16.3.7	19.3.7	20.3.07	21.4.7	21.4.7	23.4.7	14.5.7	14.05.07	20.3.7	16.3.7,2.5.07 2.3.7
Mass	3 months after experimenr				4 months after			5 months after			
1	2	3	4	5	6	7	8	9	10	11	12
168		0			**235**	**200**	75		**130**		30±10
169		25			**475**	**500**	85		**243**		30±10
170		70			**600**	**600**			**243**		30±10
171	40	70	40	45	**950**	**950**	**150**	**140**	**1670**	25	35±10
172	80	80	55	55	**5000**	**6000**	**700**	15	40	65	20±10
173	**400**	**400**	**300**	**300**	**200**	**200**	50	40	**488**	**200**	25±10
174	45	50	25	30	**1600**	**1615**	**230**	8	0	46	**15±10**
175	**125**	**170**	**75**	**80**	15		15	35	**300**	70	20±5
176*	8	8	8	8	30		15	50	0		20±5
177	8	8	8	0	30		40	**130**	35		8±1
178	15	8	0	8	50		**19500**	20	30		8±1
179	0	8	0	8	70		60	**220**	**100**		30±10
180	25	15	8	0			80	**480**	**320**		20±5
181			0	**120**			40	**1000**			30±5

***Assuming, that mass 176 corresponds to $^{176m}Yb_{70}$, whose half-life is – 11,4 s (IT), that could explain the possible cause why the isotope's intensity has not been detected.**

Result on mass-spectrometry data

- Characterization of refractory metal (W, Ta) after Deuterium Discharge confirmed that heavy isotopes decay occurs with thermal ionization mass spectrometry.
- The group of more light mass isotopes 169, 170, 171, 178, 180 with higher intensity was find during thermal ionization mass spectrometry after deuterium discharge.
- Isotopes with more light masses (comparing to W isotopes masses) have been creating for at least 3-5 months after the exposure to the Deuterium Discharge. The observed increasing of the separate more light isotopes were into 5 – 1000 times (from 5-50 cps into 100-20000 cps).

2. GAMMA/X RAY EMISSION OF TUNGSTEN CATHODES BEFORE, DURING AND AFTER DEUTERIUM DISCHARGE

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Gamma/X-ray emission of W and Ta during and after irradiation in deuterium discharge with gamma/x-ray CdTe XR 100T detector (Amptec)

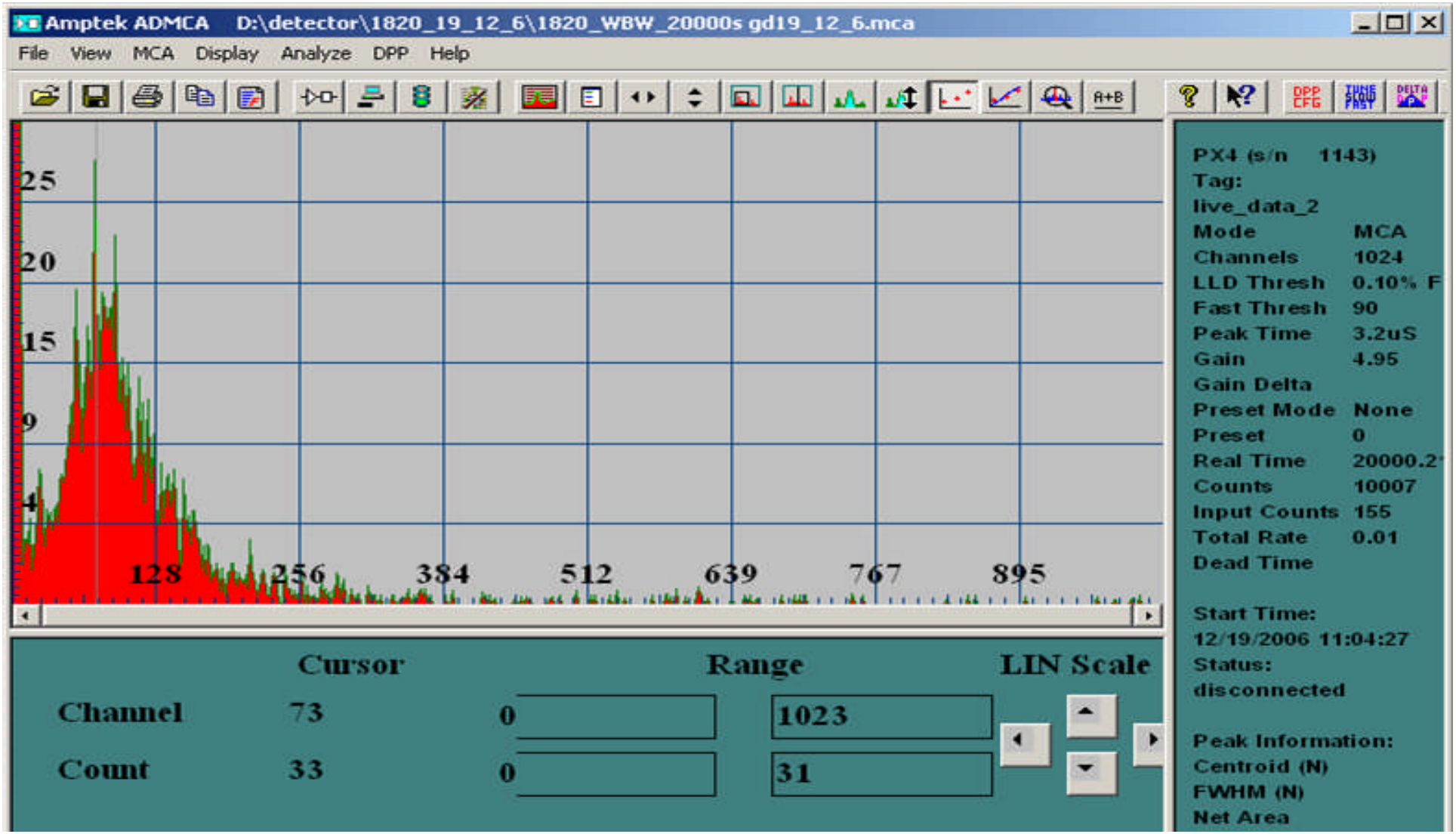
1. Gamma/ X ray intensity of emission for the different specimens was analyzed.
2. The intensity of Gamma emission depended on experimental time, dose, current and other parameters.
3. Main energy peaks of more intensive isotopes in different experiments were reproduced and identified using gamma/x ray spectra.

Correlation of TIMS and Gamma spectrometry data

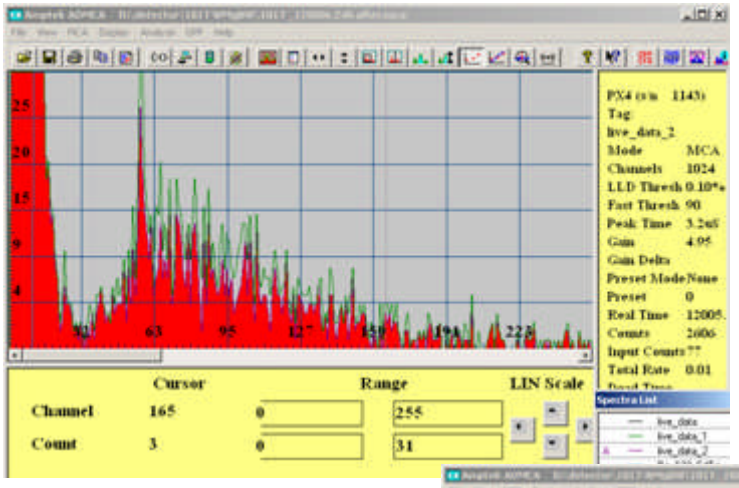
1.Estimation of Gamma/ X-ray emission intensity (value of energy peaks) and mass spectrometry data were fulfilled in CPS .
2.Comparison of main isotopes was observed using two different methods (mass spectrometry and Gamma spectrometry).

Spectrum Of Gamma/X Ray CdTe XR 100T Detector

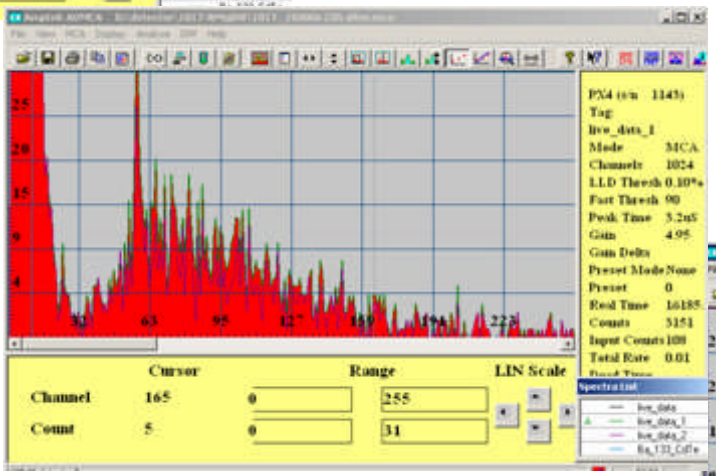
 # 1820 (~5,5 hours Δ rate= 3,5cps); Average rate = 0.5 cps; Background =0.09±0.005 cps



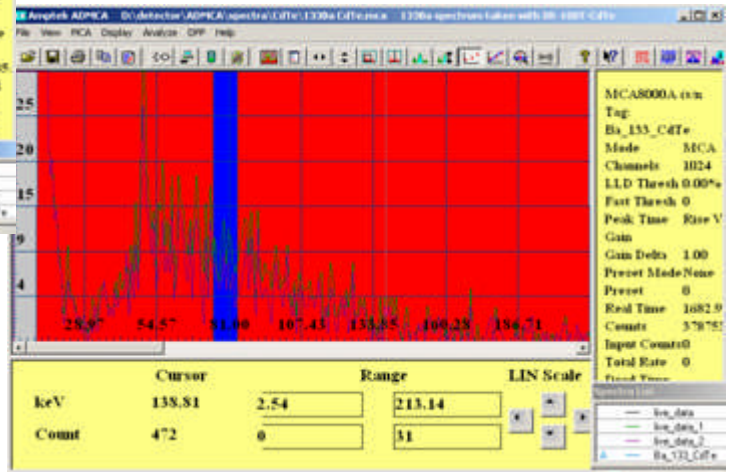
GAMMMA SPECTRA OF TUNGSTEN AFTER GLOW DISCHARGE



After Glow Discharge
12000seconds

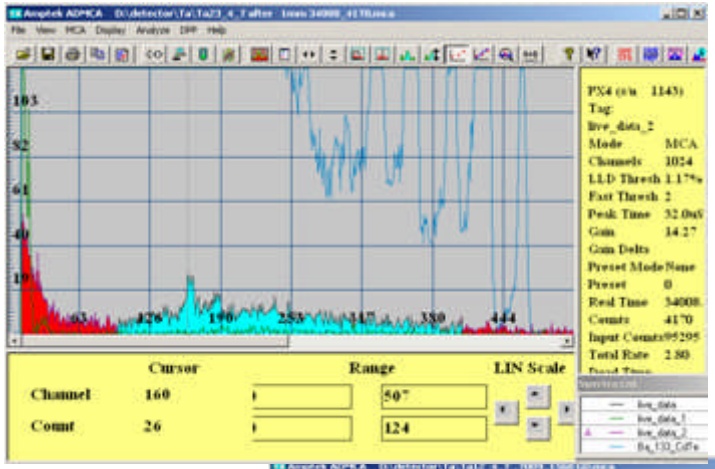


Afer Glow Discharge
16000seconds

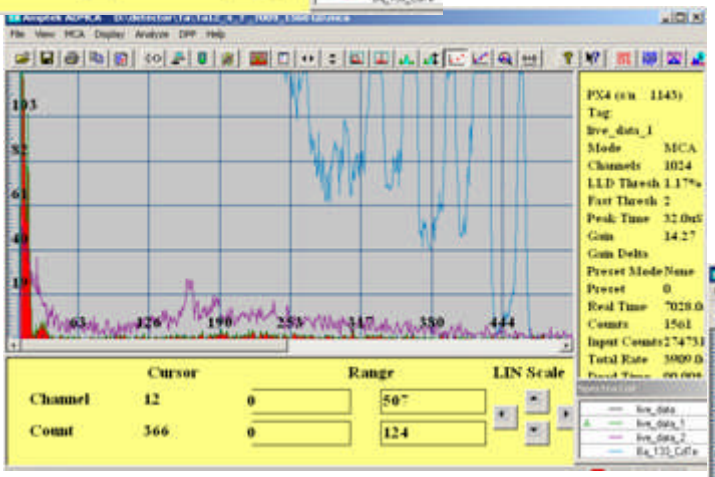


Calibration is in keV

The example of Ta gamma spectra obtained with CdTe XR 100T Detector:

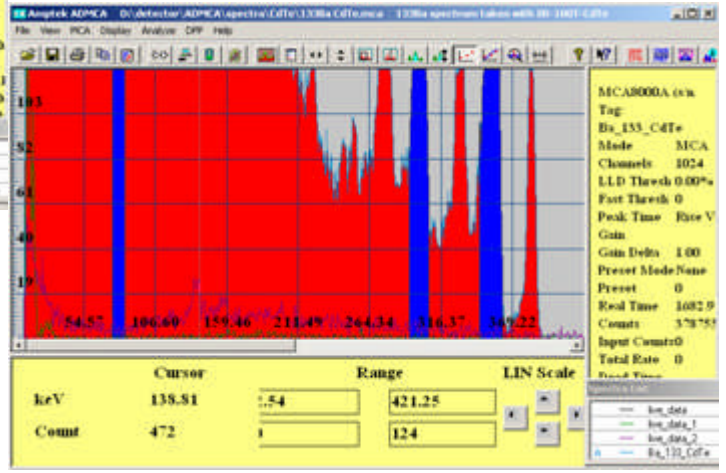


1. While Glow Discharge: 5060 counts made during 7000 seconds, average intensity =0.72 cps



2.After Glow Discharge. Distance between Be window of detector and W foil is ~1 mm. 41700 counts made during 34000 seconds, average rate =1.23 cps

3. Calibration spectrum 133Ba. Data are in keV.



CORRELATION OF GAMMA SPECTROMETRY AND MASS-SPECTROMETRY DATA

(DEFINITION OF ISOTOPES ACCORDING to ENERGY PEAKS IN GAMMA SPECTRA AND COMPARISON WITH ISOTOPE MASS PEAKS IN MASS-SPECTRA)

GAMMA EMISSION FROM TUNGSTEN AFTER DEUTERIUM GLOW DISCHARGE
using Gamma/X ray CdTe DETECTOR

W	W	W	Ta	Isotope	Eγ keV	Half-life	Decay mode	I γ(%)	Mass
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	5,25ms	IT	14,8	171
22,5±1	23,19	23,19	23.2	$^{172}Hf_{72}$	23.4	1,87 y	ε		172
24±1	24	24,02	24,84	$^{172}Hf_{72}$	23,93	1,87 y	ε	20,3	172
60±1	60,5	60,35	60,5±0,5	$^{172}Hf_{72}$	60,65	1.87 y	ε	1,1	172
67±1	63	62,35	67.5	$^{172}Hf_{72}$	67,3	1,87 y	ε	5,3	172
91±1	91±1	91,74	91	$^{172}Hf_{72}$	91,3	1,87 y	ε	0,11	172
115±1	114	114,03	114,03	$^{172}Hf_{72}$	114,06	1,87 y	ε	2,6	172
115±1		115	115.6	$^{172}Hf_{72}$	116,1	1,87 y	ε	0,034	172
119±1	119	118,99	119,8	$^{172}Hf_{72}$	119	1,87 y	ε		172
129,03	129	127,25	127.5	$^{172}Hf_{72}$	127,9	1,87 y	ε	1,46	172
42±1	43	42	42,18	$^{178}Yb_{70}$	42,4	74m	β-	6,7	178
13±1	14,1	14,1	13,3	$^{180}Yb_{70}$	13,9	2,4m	β-		180
57±1	58,7	57,05	57,88	$^{180m}Hf_{72}$	57.555	5.5h	IT	48.0	180m

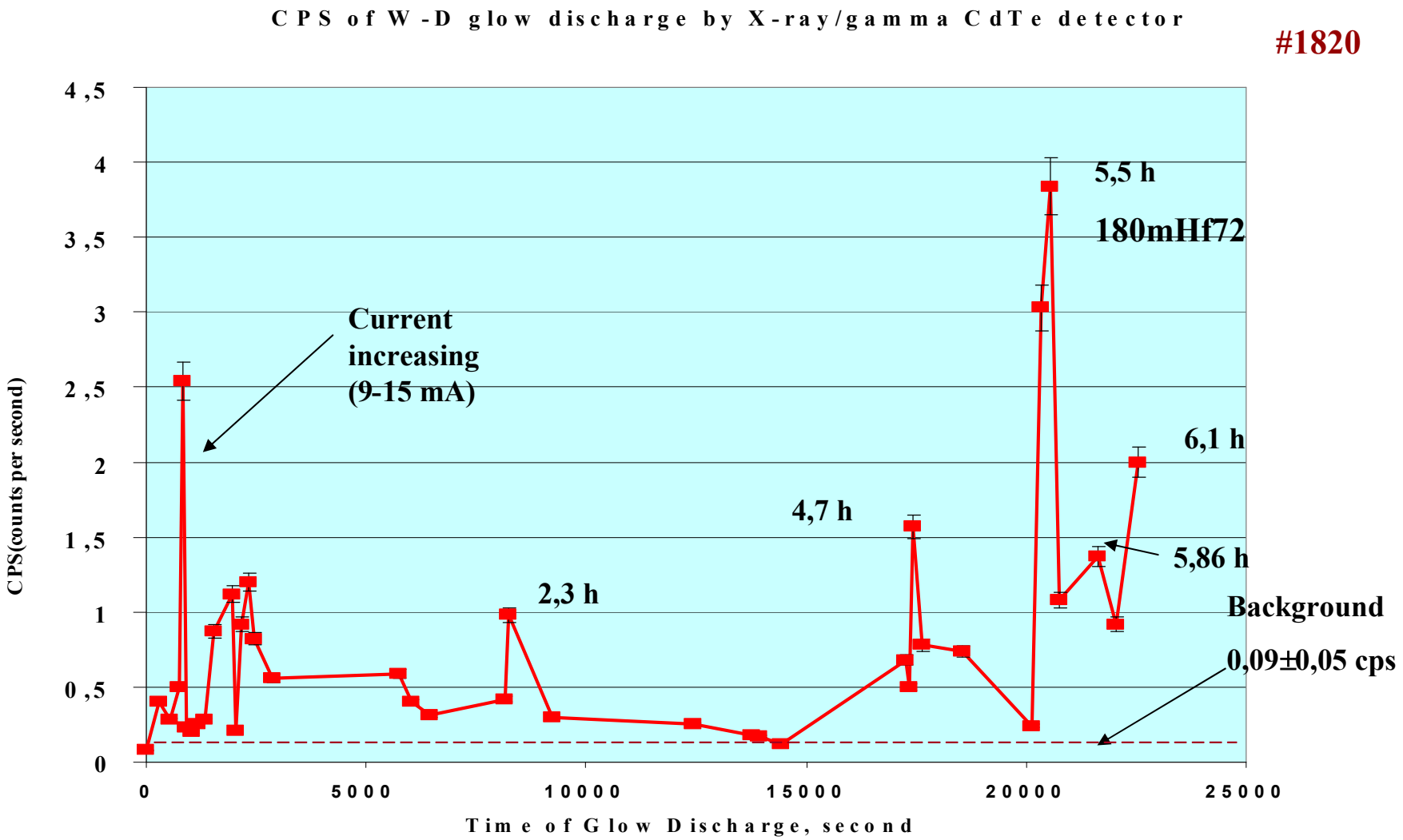
Nine peaks correspond with $^{172}Hf_{72}$ $^{180m}Hf_{72}$ IT 99,7%, β-0.3% T1/2=5.47 h

Gamma /X ray Emission from Tungsten after Deuterium Discharge (CdTe detector)

W 1817 kev, contact*	W 1820 kev, contact	W 1818 kev, contact	Ta 1824 kev, contact	Isotope	Isotope's Eγ, keV	Half-life	Decay mode	I γ(%)	Mass, TIMS
1	2	3	4	5	6	7	8	9	10
45±1	45,1	44,46	46,46	$^{170}Hf_{72}$	44,52	16,01 h	$\varepsilon+\beta^+$	0,32	170
55,4	54,57	55,4	55,43	$^{170}Hf_{72}$	55,2	16,01 h	$\varepsilon+\beta^+$	1.1	170
99,99	99	100,82	100,8	$^{170}Hf_{72}$	99,93	16,01h	$\varepsilon+\beta^+$	2	170
113,21	113,3	113,2	113,2	$^{170}Hf_{72}$	113,9	16,01h	$\varepsilon+\beta^+$	0.18	170
115,69	115,	115,7	115.6	$^{170}Hf_{72}$	115,5	16,01h	$\varepsilon+\beta^+$	0,2	170
133	132	132,2	132.7	$^{170}Hf_{72}$	132,2	16,01h	$\varepsilon+\beta^+$	0,044	170
138,81	138,5	138	138,8	$^{170}Hf_{72}$	139,2	16,01h	$\varepsilon+\beta^+$	0,018	170

- These gamma energy peaks were observed for the different W and Ta foils. It means – the same isotopes are created for the different conditions at deuterium discharge.
- Contact* - it means that foil after exposure was located in contact with detector's Be window.
- Seven peaks correspond with $^{170}Hf_{72}$.

Intensity of Gamma/x-ray Emission From W During the Deuterium Glow Discharge (cps)



Defect of mass, spin and parity

-48 (0+) +13(1+) = -52(0+) +2,45(0+) +7,3 (1/2+) + 8,07(1/2+) + ~1 MeV

$$^{182}_{74}W + ^{2}_{1}D \rightarrow ^{178}_{72}Hf + ^{4}_{2}He + ^{1}_{1}p + ^{1}_{0}n + \sim 1MeV$$

PEAKS CORRELATION FOR VARIOUS EXPERIMENTS

(COUNT PER SECOND DURING AND AFTER DEUTERIUM GLOW DISCHARGE)

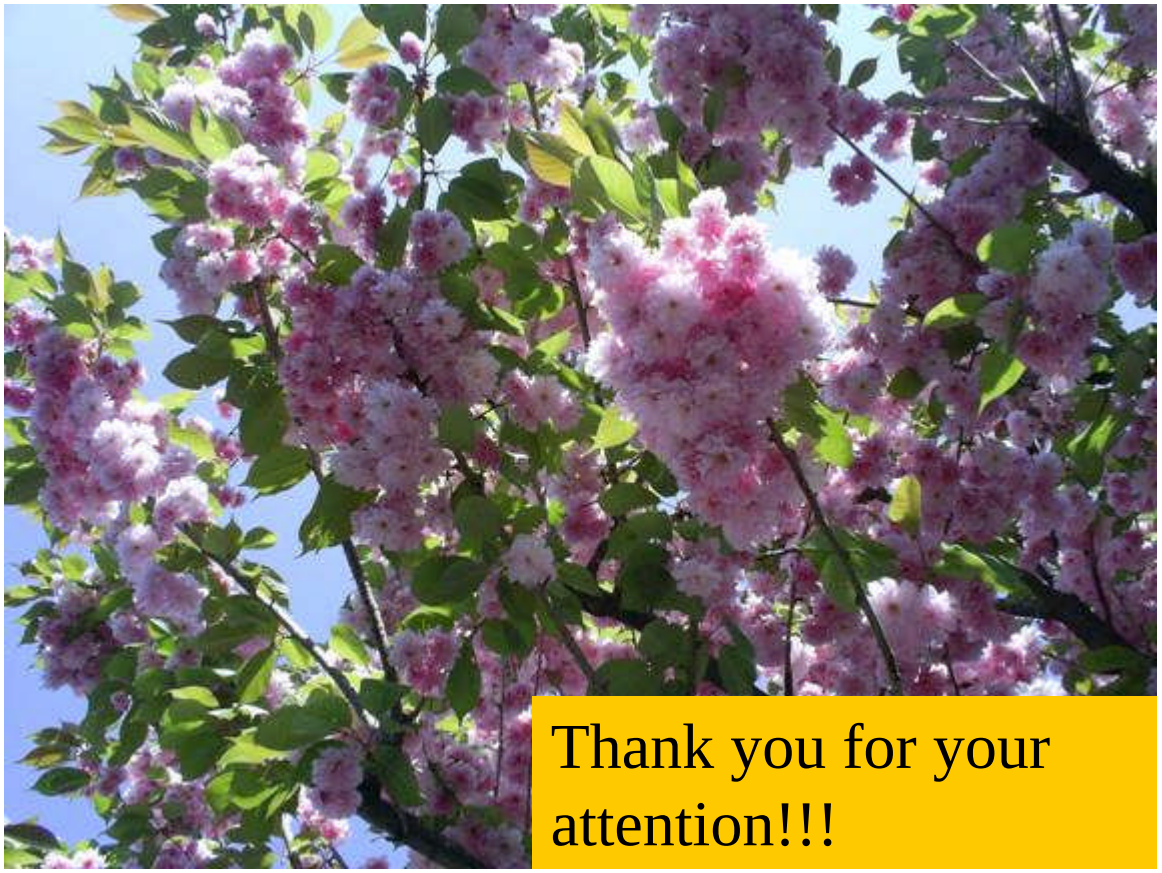
(gamma emission detected with CdTe XR100T) Background 0.09±0.006 cps

1817 Cps	Peak Time after	1818 Cps	Peak Time after	1819 Cps	Time Peak after	1820 Cps	Peak Time after	1820 Cps	Peak Time During
1*	2*	3*	4*	5*	6*	7*	8*	9**	10**
								0.4	10
8	24,5m	0,16	13m			0,48	14,3 m	2,54	14,7
1,28	28m	0,44	28m			0,37	27,6 m		27,6m
31,4	43m	0,5	35(31.7)m					1.12	35m
0,52	63	0.21	51,7m(52)	0,82	52m			0,56	50m
									1,66h
									2.29h
						0,32	2 h		2.32h
								0,68	4h55m
								1,57	5h15m
								8,13	6,05h
		0,24	21,55h					1,08	6,1
		0,7	46,13h					1,37	6,3 h

•Column 1-8 –the measurements were made in contact of foil after exposure with Be window of detector; ** 8-9 - the measurements were made in contact Be window of detector with quartz tube (~50 mm from discharge zone) during Deuterium Discharge.

CONCLUSION

- Recent characterization of refractory metal (W, Ta) during and after Deuterium Discharge confirmed that heavy isotopes decay occurs by gamma/x-ray spectrometry and thermal ionization mass spectrometry.
- The group of more light mass isotopes 169, 170, 171, 178, 180 with higher intensity was find during thermal ionization mass spectrometry after deuterium discharge.
- Isotopes with more light masses (comparing to W isotopes masses) have been creating for at least 3-5 months after the exposure to the Deuterium Discharge. The observed increasing of the separate more light isotopes were into 5 – 1000 times (from 5-50 cps into 100-20000 cps).
- Gamma/x-ray emission after stopping of experiment still takes place.
- The comparison of these data allows to suppose that the gamma spectra peaks observed belong to the following isotopes : $^{169}Yb_{70}$; $^{171m}Yb_{70}$; $^{172}Hf_{72}$; $^{178}Yb_{70}$; $^{180}Yb_{70}$; $^{180m}Hf_{72}$; $^{170}Hf_{72}$. The same isotopes are created for the different conditions after deuterium discharge
- Correlation of TIMS and Gamma spectrometry data leads to the assumption that the heavy isotopes decay under low energy impact.



Thank you for your attention!!!