Transmutation in Tungsten Irradiated By Low Energy Deuterium Ions

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ABSTRACT

Analysis of tungsten and tantalum foils after deuterium discharge with the Thermal Ionization Mass Spectrometry (TIMS) method is described. Tungsten isotopes transmutation in elements lighter than tungsten was observed. Higher intensity of mass numbers 169, 170, 171, 178 and 180 was found. The mass spectra peak magnitudes for isotopes lighter than W increased by factors ranging from 5 to 400. The increase was from 5-50 cps in the original foils, to 100-20000 cps after deuterium discharge. Possible explanations for the reactions are suggested.

1. Introduction

In our previous papers [1-5] we described changing element and isotope structure in palladium and titanium. Weak gamma-emission, brief neutron bursts [6-7], changes of surface structure and of the elemental and isotopic composition of palladium cathode were observed. The increase in impurities in palladium, which were not detected in the original material, ranged from 0.5% to 5% (an increase of 100-10000 times).

Partial, local blackening of x-ray films placed in contact with Pd, Ti and Ag foils after irradiation by deuterium discharge was observed [3, 7, 8]. Local blackening of x-ray films placed both inside and outside of the discharge chamber stainless steel wall was observed [3, 9]. Based on the results of radiography analysis, the various energies of radioactive isotopes on the Pd cathode after deuterium discharge included high-energy and low-energy components [3]. Observed effects were explained by a fusion-fission reaction on the cathode; an interaction of a palladium lattice with deuterium and by the subsequent decay of more light elements. The majority of the elements found after deuterium discharge were in local zones (hot spots) [1, 2, 3, 8].

Groups of elements such as Sc, Ti, V; Ag, Cd, In; P, Cl, Br, Ge, As, Kr, Sr, Y, Ru, Xe were found in the Pd after an irradiation by ions of all types (D, H, Ar, Ar + Xe), but within various content. After irradiation in Ar, the impurity elements in the Pd samples increased by a factor of 2 or 3 with H bombardment, and by a factor or 10 with D bombardment [9].

Considerable changes in the isotope ratios for $^{10}B/^{11}B$; $^{12}C/^{13}C$; $^{60}Ni/^{61}Ni/^{62}Ni$; $^{40}Ca/^{44}Ca$ and $^{90}Zr/^{91}Zr$ were observed with different mass-spectrometry methods. See Ref. 6. Changes in the isotope ratios of $^{109}Ag/^{107}Ag$ from the ratio of 1/1 in initial Pd up to 3/1 and 9/1 after gas discharge exposure was described in Refs. 5 and 9.

Changes in uranium alpha, beta and gamma emission after deuterium and hydrogen discharge was found for both sides of the uranium (the irradiated and un-irradiated sides) [10, 11].

Two series of experiments were performed. The first set produces immediate results. A W foil was analyzed with Thermal Ionization Mass Spectrometry (TIMS) in 15-minute intervals after deuterium discharge. In the second set of experiments W foils were analyzed with TIMS 3, 4 and 5 months after deuterium discharge.

Increases by factors ranging from 5 to 400 in the peak magnitudes for different isotopes with masses lighter than W were observed.

2. Methods

2.1. Experimental Method

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

Figure 1. Glow discharge apparatus.

The glow discharge apparatus was made with double quartz tubes (as the wall of the discharge chamber) with cooling water between these quartz tubes.

The sequence of operations before discharge experiments was as follows: vacuum degassing to 10^{-3} Torr, followed by deuterium loading to between 3 and 10 Torr.

Molybdenum was used as the anode. Tungsten foil was placed on the cathode. The foil ~100 microns thick and ~20 mm in diameter. The anode and cathode were cooled by flowing water.

2.2. Analytical Method.

Thermal Ionization Mass Spectrometry (TIMS) was used to estimate isotope (ions) intensity, which is measured in counts per second (CPS).

2.2.1. Characterization of TIMS Analysis

The analysis was performed using MAT-262 and a mass spectrometer (Thermo Scientific, Finnigan TRITON). The most detailed analyses were performed in the range of mass numbers 166-206. The temperature between Re ionizer and analyzed foils 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 or Ta foils, irradiated by deuterons. The analyzed zone included the unirradiated part of W foil as well as irradiated parts, which led to a reduced contribution by more light isotopes. 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, because Re was used as the cathode. The cathode had two W foils layers. The side of the foil irradiated with deuterium ions was analyzed.

Figure 2 illustrates the TIMS spectra calibration on Re mass. An example of a precise definition of isotope mass is shown in the left spectrum, where we can see that 186.95 mass is ^{187}Re estimated very exactly. Peaks of $^{\mathbf{185}}$Re and of $^{\mathbf{187}}$Re isotopes are presented in the spectrum on the right.

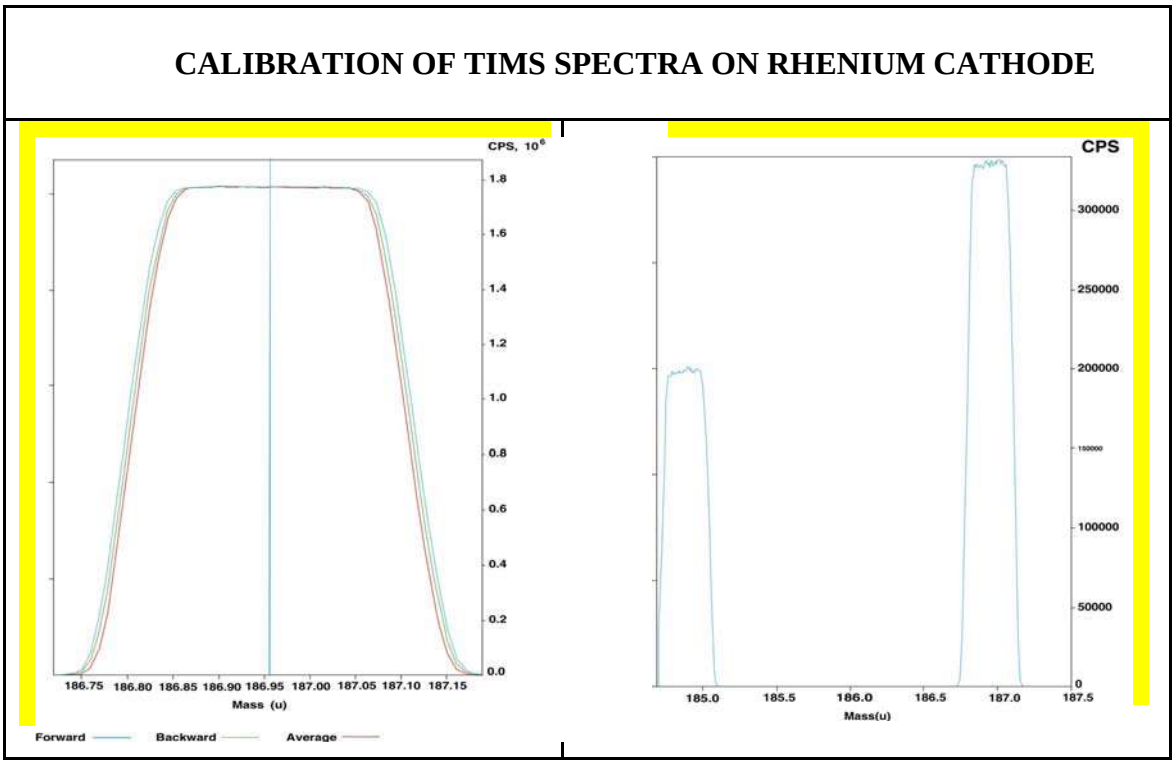

Figure 2. Calibration of TIMS spectra on isotopes of Rhenium.

TIMS spectra measurements have good reproducibility. It should be noted that calibration on Re was performed before each investigation of new foil. The good reproducibility remained for short time intervals between scanned spectra.

The significant difference in isotopes intensity was found for the different conditions of experiments: especially, for dose of exposure and time interval after the experiment stopped. The confirmation of spectra reproducibility is presented on the Fig. 3.

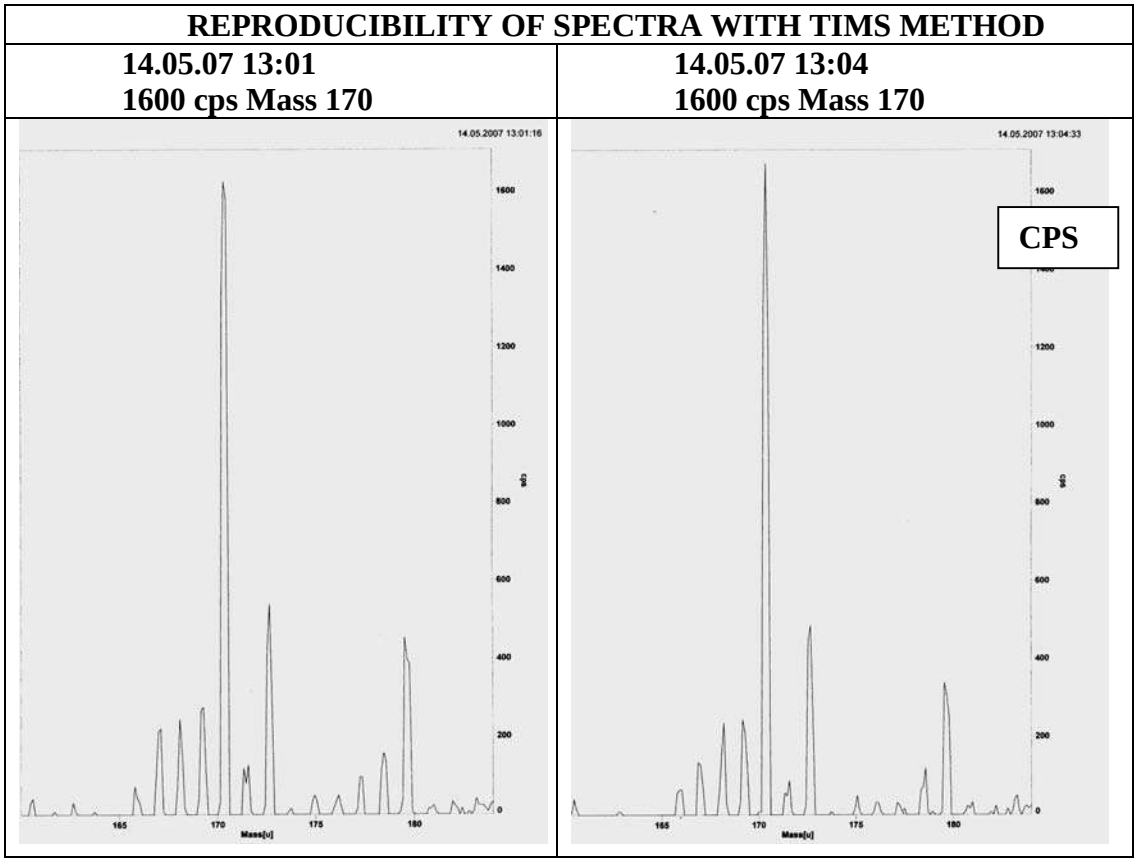

Figure 3. TIMS spectra of the same W foil with time interval between scans of about 3 minutes.

In Fig. 3, the time interval between the right and left Spectra was ~3 minutes. It is possible to estimate that the intensity of main isotopes is closed for the both spectra. The main isotopes and their intensity are proportional to each other.

3. Experiments

As noted above, two series of experiments were performed: 15 minutes after irradiation, and 3, 4 and 5 months after experiments, both using TIMS. Both W and Ta samples were irradiated.

Tables 1 and 2 compare mass spectra data from the original unused W and W after deuterium bombardment. The main isotopes changes in the different experiments with W and Ta foils for various time intervals were investigated in detail for the mass range 166-210. An increase in isotopes with masses lighter than tungsten by factors ranging from 5 to 400 was found. The analysis temperature was ~1800°C.

A mass spectrum of original W in the 170-200 mass range is shown in Fig. 4. It shows the intensity ranged from 10 to 30 cps.

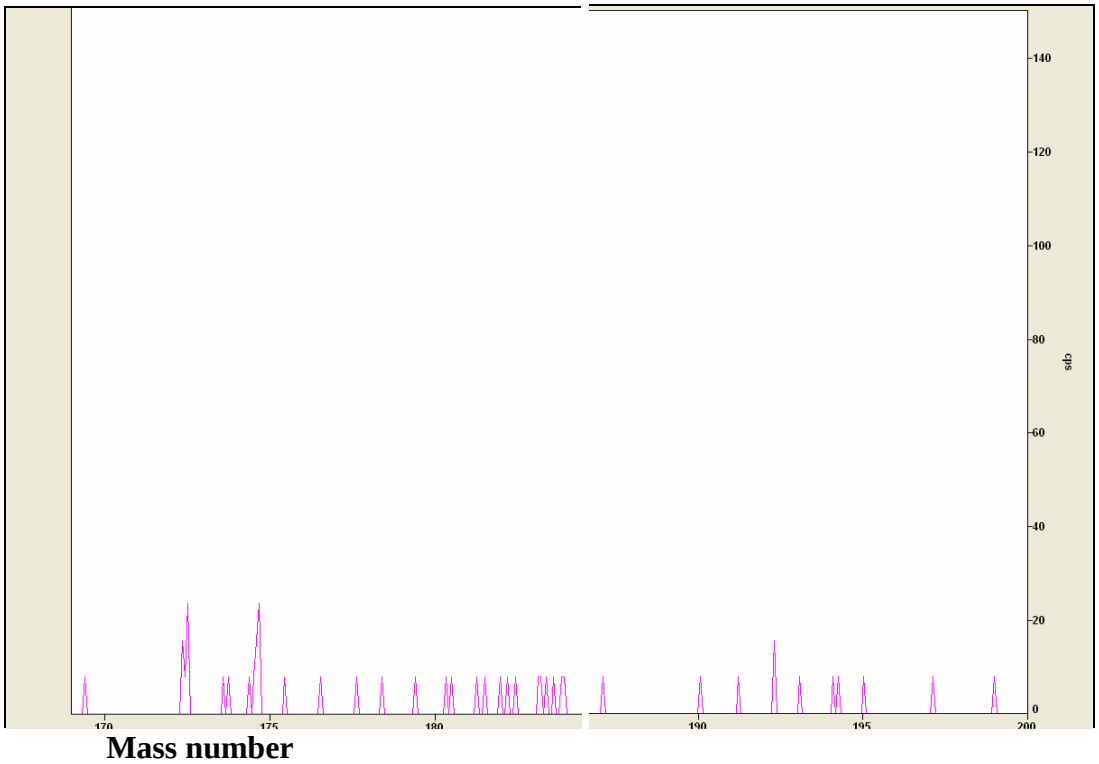

Figure 4. Mass spectra of original W in the 170-200 range of masses (CPS).

A mass spectrum of the W after deuterium discharge for the mass range of 170-182 for the same foil with two minutes intervals is shown in Fig. 5. Mass 172 is ~700 cps on the left spectrum, and mass 178 is ~20000 cps on the right spectrum. The intensity of these isotopes was 10 - 50 cps before experiments for similar scanning parameters. The increase of masses 172 and 178 intensity in the same tungsten foil was from 12 to 400 times, respectively.

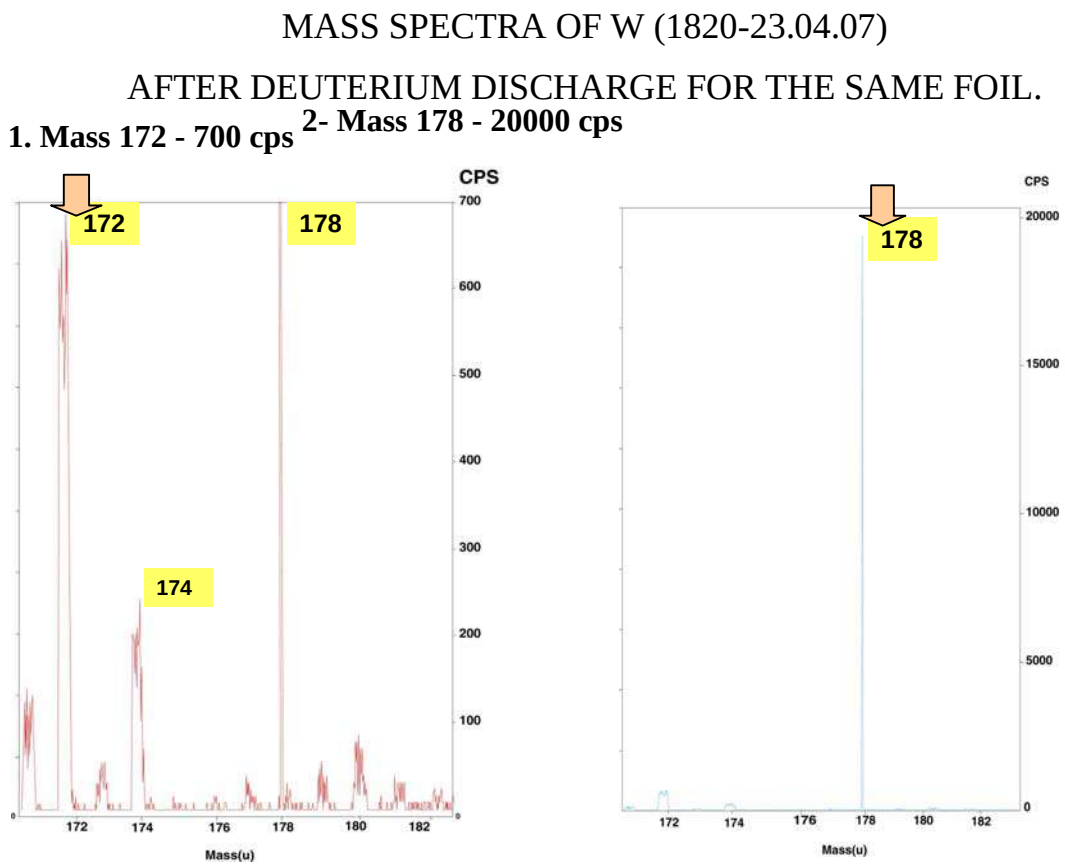

Figure 5. Mass spectra of W #1820 through ~ 4 months after Deuterium discharge stop.

THE CREATION of MORE LIGHT ISOTOPES in Ta

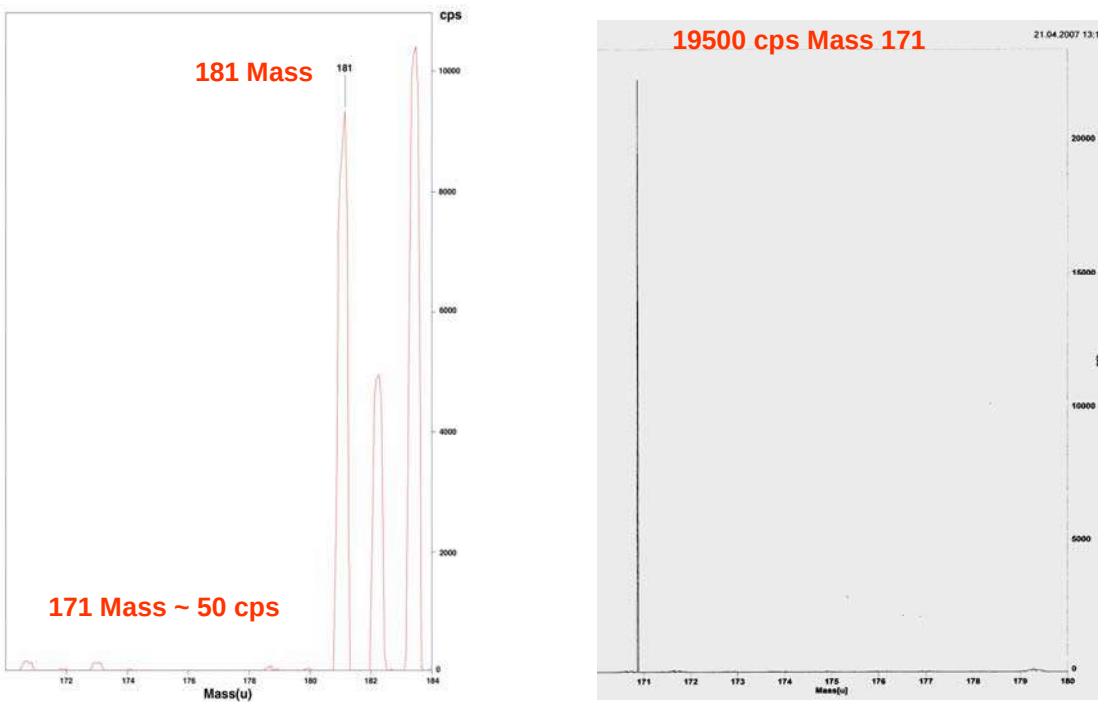

Figure 6. Ta before (left) and after (right) the Deuterium Discharge.

Tantalum before (left) and after (right) deuterium discharge are compared in Fig. 6. The intensity of mass 171 was ~50 cps before the experiment, and after the experiment it reached 19500 cps, increasing by a factor of ~390.

4. RESULTS

Table 1

INTENSITY OF THE MORE LIGHT ISOTOPES IN TUNGSTEN FOILS, IRRADIATED BY DEUTERIUM AND ANALYZED WITH THERMO - IONIZATION MASS-SPECTROMETRY (IN COUNTS PER SECOND - CPS) * Set 1**

Time,min* _____ Mass	84*	101*	137*	1062*	1073*	1133*	1150*	**Original W
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

*First line shows the time in minutes after experiment.
**Last column is intensity of isotopes in the original W.
***Set 1- The first analysis was carried out in 45 minutes after experiment stop. Farther the each analysis was fulfilled every 15 minutes during ~3 hours and then analysis was made in ~10 hours every 15 minutes during few hours.

Table 1 shows data from the first set of the experiments with W, showing that transmutation of heavier isotopes into lighter ones continues after the experiment is stopped. Increases from ~20 to ~200 times for different isotopes were found.

An analysis of tungsten foils irradiated with different doses of deuterium discharge performed 3, 4 and 5 months after irradiation is presented in Table 2.

Table 2

INTENSITY OF THE MORE LIGHT ISOTOPES IN TUNGSTEN FOILS, IRRADIATED WITH DEUTERIUM AND ANALYZED BY THERMO- IONIZATION MASS- SPECTROMETRY (IN COUNTS PER SECOND - CPS)*** Set 2

# exper	#1817				#1820					#1821	Original
Date Mass	16.03.07 *	16.03.07 *	19.03.07 *	20.03.07 *	21.04.07 **	21.04.07 **	23.04.07 **	14.05.07 ***	14.05.07 ***	20.3.07 ***	****
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

Columns 2-5, 11 – results of analysis after 3 months after experiments.
Columns 6-8 – results of analysis after 4 months after experiments.
Columns 9-10 – results of analysis after 5 months after experiments.
Columns 12****- average CPS of isotopes in the original spectra for 3-5 analyzed tungsten foils during all months.

5. DISCUSSION

Mass spectroscopy results include the following:

- Tungsten transmutation of heavy isotopes into lighter elements after exposure to deuterium discharge was confirmed.
- The group of lighter isotopes with mass numbers 169, 170, 171, 178, 180 had high intensity after deuterium discharge soon after the experiment, and also 3, 4 and 5 months later.
- Isotopes with lighter masses (compared to W isotopes) continued forming for at least 3-5 months after the exposure to the deuterium discharge. The observed increase of different light isotopes was into 5 - 400 times (from 5-50 cps in the original W up to 100-20000 cps after experiments).
- Experiment with tantalum was carried out to strengthen understanding of the possible reaction. Tantalum was selected because it has only one stable isotope, with mass number 181. As can be seen in Figs. 6 and 7, the increase in intensity of only of one isotope with mass number 171, and a decrease in the single stable isotope of Ta was found.

The intensity increasing of mass 9 in Ta after deuterium discharge.

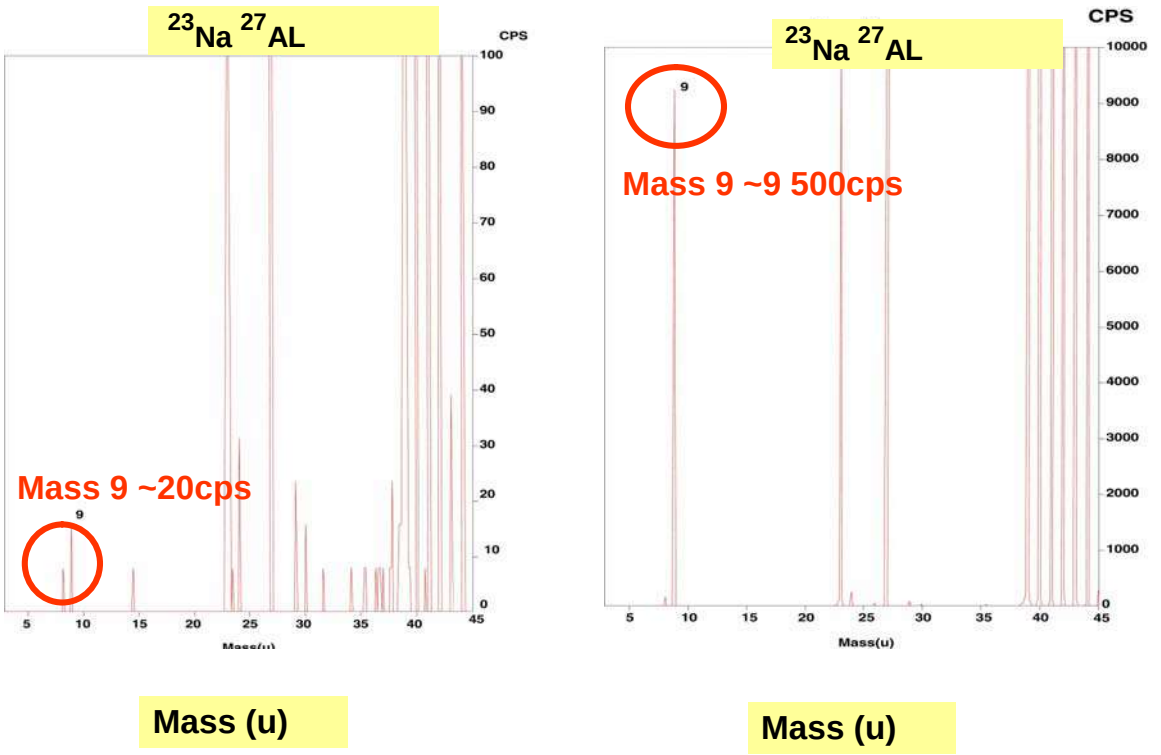

Figure 7. Significant increase of 9 mass in Ta spectra observing after deuterium discharge.

Figure 7 shows mass 9 in Ta before (left) and after (right) of the deuterium discharge. Mass 9 increased from 20 to 9500 cps, a factor of ~475.

This observation suggests the possibility of the following reactions:

$^{2}_{1}D + {}^{181}_{73}Ta \rightarrow {}^{171}_{72}Hf + {}^{10}_{2}He^{*}$ **+2n;**

(1+)+ (7/2+) → (7/2+)+ (0+) +2(1/2+) - spin, parity;

(+13 MeV) +(-48.44 MeV) → (-55.4 MeV) + (+48.8 MeV)+ (+16.14 MeV)+ **84.90 MeV** ;

$^{10}_{2}He^{*} \rightarrow \mathbf{n} + {}^{9}_{2}He^{*}$

(0+) → (1/2+) + (1/2-)

(+48.8 MeV) → (+8.07 MeV) + (40.94 MeV) +0.7 MeV

$^{9}_{2}He^{*} \rightarrow {}^{9}_{3}Li^{*} \rightarrow$ 178ms: $\beta^{-} \rightarrow {}_{4}\mathbf{Be}^{9}$

This is only one variant of the possible reaction. Nevertheless, an alternative hypothesis might better reflect the details of this complex process. The polyneutron theory of transmutation, suggested by John Fisher [13], may be applicable. It is necessary to pay more attention to the electro-magnetic excitation for stimulation of this process. We noted this early [9], and Peter Hagelstein published it in one of his papers [14]. The cluster decay and the neutrino and dineutrons participation in this process may also take place [15].

More research is called for. However this will require additional funding for new applications and new, detailed knowledge. Even though our research has been fruitful, a lack of funding has meant that we cannot produce detailed data, which means we cannot get more funding – a vicious circle.

Conclusion

1. The isotopes with masses 169, 170, 171, 178, 180, 181 (less than W and Ta isotopes) after deuterium glow discharge were found in W and Ta by TIMS.
2. The isotopic changes continue to occur at least 3 - 5 months after glow discharge exposure. The separate isotopes with masses less than W and Ta isotopes increased by factors ranging from 5 to 1000 times.
3. The comparison of mass spectra with gamma spectra points to the existence of the following isotopes: ${}^{169}_{70}Yb$; ${}^{170}_{72}Hf$; ${}^{171m}_{70}Yb$; ${}^{172}_{72}Hf$; ${}^{178}_{70}Yb$; ${}^{180}_{70}Yb$; ${}^{180m}_{72}Hf$.

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