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Changes in Isotopic and Elemental Composition of Substance in Nickel-Hydrogen Heat Generators

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Abstract—Results of the isotopic and elemental composition analysis of fuel and matter near the active zone of four nickel-hydrogen reactors before and after experiments with the integral excess energy up to 790 MJ are presented. No significant changes in the isotopic composition of nickel and lithium were observed. A significant increase in the concentration of impurities of a number of nuclides has been observed not only in fuel, but also in structural elements adjacent to the active zones of the reactors.

After publication of the report about Andrea Rossi high-temperature heat generator test in Lugano [1], [2], many attempts were made to create similar devices [3]. In some of them, heat generation significantly exceeding the energy consumption were shown. Excess heat release many times exceeds the potential of chemical reactions and is comparable to the energy release in nuclear reactions, although it is not accompanied by harmful radiation and radioactivity. But the nature of this surprising effect remains unclear. The study of elemental and isotopic changes in the operation of reactors is of paramount importance for clarifying the nature of this effect. This paper provides information on the results of the analysis of changes in fuel and in structural materials that occurred in several nickel-hydrogen reactors created by our team.

I. ESTIMATION OF POSSIBLE CHANGES IN THE ISOTOPE COMPOSITION OF FUEL

We can estimate the possible changes in the isotopic composition of the fuel assuming that the excess heat release occurs as a result of nuclear transmutations in accordance with the law of conservation of energy. For example, in nickel, containing hydrogen, a nuclear reaction may occur

$$^{58}Ni + ^{1}H + 2e^{-} \rightarrow ^{59}Co + 2\nu_e + 10,32MeV$$
 (1)

Since 1 MJ is equal to $6.3 \cdot 10^{18}$ MeV, about $6 \cdot 10^{17}$ nickel nuclei (0.00006 g) are consumed as a result of this reaction to release 1 MJ of energy, and the same amount of cobalt

is formed. Nickel-hydrogen reactors usually contain about 1 g of fuel. It is quite possible, using modern technology, to detect 0.00006 g of cobalt in 1 g nickel (0.006%).

It is more difficult to detect changes in the isotopic ratios. Conventional mass spectral analyzers allow one to capture changes in isotopic ratios of elements of the order of 1%. It is not possible to detect a change of the order of 0.01% that occurs when 1 MJ of energy released into 1 g of fuel as a result of the reaction (1). To reduce the content of the isotope 58 Ni by 1%, an excess energy of the order of 100 MJ is necessary.

If the fuel contains lithium, a nuclear reaction is possible

$$^{7}Li + {}^{1}H \rightarrow 2^{4}He + 17,35MeV$$
 (2)

As a result of this reaction, in a mixture of lithium isotopes (the natural mixture contains 92.6% of ⁷Li and 7.4% of ⁶Li) the content of ⁷Li decreases and, accordingly, the content of ⁶Li increases. Suppose that all excess energy release is associated with reaction (2). To release 1 MJ of energy, $4 \cdot 10^{17}$ ⁷Li nuclei are required ($4.2 \cdot 10^{-6}$ g). A typical reactor with fuel mixture of lithium-aluminum hydride and nickel contains about 0.02 g of ⁷Li. Therefore, when 1 MJ is released, only 0.02% ⁷Li is removed. It is almost impossible to detect such a change. With the release of 1000 MJ of energy, 20% of ⁷Li is removed. This leads to an increase in the content of ⁶Li from 7.4 to 10%. This change is quite possible to detect, although not easy because of the small mass of the material available for analysis.

Thus, the appearance of isotopes that are absent in the initial fuel can be detected with excess energy of the order of 1 MJ per gram of fuel. To reliably detect changes in the ratios of isotopes in elements that originally are a part of fuel, excess energy exceed 100 MJ/g is required. It is natural that a thorough analysis of the fuel of the GS3 reactor made by Alan Goldwater did not reveal any noticeable isotope changes, since the excess energy production in it was about 50 MJ/g [3], [4]. The excess energy production in the high-temperature heat-generator of Rossi, according to [1], was 5800 MJ/g. This is quite sufficient for radical changes in the isotopic composition of both nickel and lithium. Further, we will present the results of an analysis

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of isotope changes in fuel and in structural materials that occurred in several nickel-hydrogen reactors created in our laboratory.

II. Reactor AP2

Reactor AP2 [5] was charged with a fuel mixture of 640 mg Ni + 60 mg LiAlH₄. It worked from March 16 until March 22, 2015, and produced about 150 MJ of excess heat.



Figure 1. AP2 reactor.

Analyzes of fuel before and after the experiment were made using several methods in different organizations. The analysis of the elemental composition using an electron scanning microscope was made at the Prokhorov General Physics Institute, Russian Academy of Sciences and All-Russian Research Institute of Experimental Physics (VNIIEF, Sarov). Two fractions confidently differ in the fuel mixture measured before experiment: gray crystals and white granules. Gray crystals mainly contain Al, O, and Cl. White granules consist of nickel with a small admixture of iron, aluminum and oxygen. In the fuel after the experiment, white molten and gray slag-like structures are visible. White structures contain mainly nickel with an admixture of Fe, Al, Cr, Mn, Si and O. Slag-like structures consist mainly of Al and O.

Analysis of the elemental composition of the fuel before and after experiment using laser atomic emission spectrometer was made at the Kurnakov Institute of General and Inorganic Chemistry, Russian Academy of Sciences. It showed that the content of K and Cr increased tens of times after the experiment. The content of Si, Na, Mg, Ca, Ti, and V increased manifold. The content of Al, Ni, Cl, Mn, Cu, Zn decreased. It should be noted, that this method of analysis, as well as analysis using a scanning electron microscope, provides information on the atomic composition only on the surface of the test substance.

Analysis of the isotope composition of the fuel before and after the experiment in AP2 reactor was made using ICP-MS method, which gives information on the isotopic composition on average over the sample. Such analysis was made in Vernadsky Institute of Geochemistry and Analytical Chemistry of Russian Academy of Sciences. Total content of aluminum and lithium after the experiment decreased, while the relative content of ⁶Li increased slightly. However this increase (by 0.5%) fall within the range of possible measurement error. There are no significant changes in the isotopic composition of nickel.

The analysis of AP2 reactor fuel by the ICP-MS method was also made at Uppsala University (Sweden). The results of these measurements are shown in Table I.

Table I Analysis of AP2 reactor fuel by the ICP-MS method at Uppsala University (Sweden).

%	⁶ Li	$^{7}\mathrm{Li}$	⁵⁸ Ni	60 Ni	61 Ni	62 Ni	64 Ni
	7,4	92,6	68,1	26,2	1,14	3,63	0,93
	15,4	84,6	63,4	27,6	1,3	5,2	2,5
	7,6	92,4	68,0	26,2	$1,\!14$	3,71	0,93

According to these measurements, the relative content of ⁶Li in the sample of spent fuel has more than doubled. Quite noticeable changes have occurred also in the ratio of nickel isotopes. These results differ from the results obtained in Vernadsky Institute of Geochemistry and Analytical Chemistry RAS. This difference can be explained, perhaps, by the unevenness of the changes in the sample volume. It should be noted that reliable results for lithium are difficult to obtain because of a very low concentration of lithium in spent fuel (<0.01%).

III. REACTOR PROTOK-6

In experiments with devices that claim to obtain heat in an amount exceeding the energy consumed, it is very important to measure the heat released with the greatest possible accuracy. Taking this into account, a series of experiments was carried out in our laboratory using a calorimeter with flowing water, which makes it possible to measure the heat dissipation power with an error of less than 3%. One of the tested reactors, 'Protok-6', worked continuously with this calorimeter from April 11 to May 29, 2016, with the release of excess heat power from 20 to 65 watts. The integrated excess energy in this reaction is about 100 MJ [6]. Unlike previous designs having an external heater, this reactor had a heater, made out of tungsten wire, located inside a sealed ceramic tube. The fuel (1.8 g of nickel powder mixed with 0.2 g of lithium)aluminum hydride) was located in a ceramic tube wrapped in a tungsten heater. The tube with the heater was in a hermetically sealed ceramic pipe of larger diameter.

After the experiment was over, the reactor was opened (Fig. 3). It was found that the inner surface of the outer tube near the heater was covered with lumpy gray glassy coating. The physical configuration of the inner tube and the heater winding was preserved. However, the changes inside were significant: a vitreous mass with inclusions of metal balls measuring about 0.1 mm was formed. Several balls had a diameter of up to 1 mm. At the ends of the filling, the fuel took the form of a sintered mass containing small metal balls. Furthermore, powder from the inner tube poured out. Using a magnet, a fraction was extracted



Figure 2. Scheme of the 'Protok-6' reactor .



Figure 3. Reactor 'Protok-6' after opening.

from this powder consisting of filaments with a transverse dimension of about 0.1 mm and length up to 5 mm.

Several samples were subjected to mass-spectroscopic analysis at Vernadsky Institute of Geochemistry and Analytical Chemistry RAS using the ICP-MS method. The following were investigated: the initial fuel mixture, the metal ball from the spent fuel, the fuel at the edge of the filling, the substance accumulated between the inner and outer tubes, and the coating on the inner surface of the outer tube. Due to the large amount of information received, it is not possible to present it completely. Partially, the results of the analysis are shown in Table II. In addition to the data for samples recovered from the reactor after its operation, information is given on the content of isotopes in the fuel, as well as in the ceramic and tungsten wire, before the experiment. This information is important, since the appearance of new elements can be associated not with transmutations, but with migration from structural materials, which is quite possible at high temperatures. Unfortunately, the ICP-MS method can not determine the content of isotopes with masses of 1-5, 12-22, 32, including isotopes of carbon, oxygen, nitrogen, fluorine and sulfur.

The obvious result is an increase in the content of many nuclides in comparison with their content in the initial fuel and structural materials. The exception is lithium (decrease in about 100 times) and aluminum in fuel (decrease in more than 10 times). We note a particularly great increase in the presence of boron, iron, gallium, cerium, zirconium, strontium and bismuth. The most significant anomalies are found in the powder accumulated in the space between the inner and outer tubes. Especially great amount of ¹⁴⁰Ce appeared: 6.3% (in the initial fuel <0.0001%). A significant amount of tungsten found in the samples after being inside the reactor is probably due to the migration of this element from the incandescent tungsten coil.

The investigation of possible changes in the isotopic composition of lithium and nickel is of great interest. Unfortunately, the very low content of lithium in the samples after experiment did not allow us to make reliable measurements. The results obtained for nickel are presented in the Table III. Since the data on 64 Ni is unreliable due to the uncontrolled additive of 64 Zn, when compiling the table, the value from the reference book [7] was used for the 64 Ni fraction. Since this fraction is small, such an assumption can change the fractions of the remaining isotopes only slightly.

It can be seen that the data for the various samples studied differ somewhat from the natural ratio [7], but differ insignificantly between different measurements. A noticeable increase in the 62 Ni fraction, due to a decrease in the fraction of the remaining isotopes, that was found in the experiment in Lugano [1], [2] was not observed in any of the samples studied. It is possible that the effect is not visible due to the fact that the excess energy production



Figure 4. Reactor VV3 and spent fuel extracted from it.

in Lugano experiment was 60 times greater than in the described one.

IV. REACTOR VV3

Reactor VV3 (Fig. 4) differs from the 'Protok 6' reactor with a different heater design and the absence of a calorimeter. A mixture of nickel powder with lithium aluminum hydride weighing 1.5 g was used as fuel. The fuel contained pieces of tungsten wire with a total mass of 0.77 g. The reactor operated from June 14 to July 24, 2016, producing surplus power up to 330 W. A total of 790 MJ of excess heat was generated.

After the experiment was finished, the spent fuel, which looked like a drop-shaped ingot, was extracted from it. It was analyzed at Vernadsky Institute of Geochemistry and Analytical Chemistry RAS using the ICP-MS method, with separate analyzes of the surface and deeper layers. Partially the results of the analysis are shown in Table IV. In addition to the data for the samples recovered from the reactor after its operation, information is given on the content of isotopes in the initial fuel, including tungsten wires embedded in it.

It can be seen that the isotopic composition of fuel as a result experiment has changed noticeably. The content of boron, copper, cerium and silver increased significantly.

Just as in the above-described reactors, the data on isotopic composition of Ni samples, although slightly different from the natural ratio, differ insignificantly between each other (Table V).

V. Reactor KV3

KV3 reactor (Fig. 5) operated from December 20, 2016 until January 31, 2017, with an excess power of 100-200 watts. The integrated excess energy during the whole operating time of KV3 reactor is about 400 MJ. The main difference from the previous reactors is that it was loaded with 1.8 grams of nickel powder without an admixture of lithium aluminum hydride. Saturation with hydrogen was carried out by keeping it in hydrogen gas. In addition, unlike the above-described reactors, it had not a ceramic but a quartz outer tube. The heater was made not from pure tungsten, but from a tungsten-rhenium alloy.



Figure 5. Reactor KV3 at the beginning of work.



Figure 6. Reactor KV3 opened after ending of experiment.

The ICP-MS analysis by Vernadsky Institute of Geochemistry and Analytical Chemistry RAS was used to investigate: fuel and structural materials prior to operation of the reactor, as well as fuel in the central zone and near the edge, powder from the space between the inner and outer tubes, and structural materials after work of the reactor. The results are partially shown in Table VI.

Just as in the reactors Protok 6 and VV3, a lot of tungsten appeared in the space between the inner and outer tubes. In addition to tungsten, a lot of iron, sodium, potassium, nickel, silicon, calcium, scandium and a number of other elements have accumulated there.

Comparing fuel before and after the experiment, one can see a decrease in the content of sodium, potassium and iron. Attention is drawn to the appearance of a significant amount of copper.

A lot of tungsten and rhenium appeared in the inner ceramic tube with fuel, which was wrapped with a heater. Table VII shows nuclides, the relative content of which in the ceramic tube has increased more than 10-fold.

 $\begin{tabular}{ll} Table II \\ Relative content of isotopes (atomic \%) in fuel and near the core of the 'Protok-6' reactor before and after reactor operation. Isotopes with a content> 0.1\% are shown. \\ \end{tabular}$

Before reactor operation									
71;		74	231	Ceran	7.02	$23 N_{\odot}$	vv wi	.re 5.27	-
$23 N_{\odot}$	1	,14	241	Na Ma	1,05	24 M	r.	0.25	
1Na	1	,90 19	25	Ma	1,01	27 A 1	5	0,25	
27 A1	3	,12 63	26	Ma	0,23 0.28	²⁹ Si		1.88	
²⁹ Si	1	$04^{,00}$	27	41	65.05	^{31}P		0.18	
³⁹ K	1	.60	29	Si	1.55	³⁹ K		6.09	
44 Ca	0	.28	31	5	0.16	^{44}Ca		1.06	
^{45}Sc	Ő	,22	39]	K	8,36	^{45}Sc		0.80	
^{51}V	0	,68	44(Ca	0,94	$^{54}\mathrm{Cr}$		0,40	
$^{53}\mathrm{Cr}$	0	,22	45 9	Sc	$0,\!61$	56 Fe		10,46	
^{55}Mn	0	,17	48	Гi,	0,15	^{182}W	I	18,50	
56 Fe	0	,99	⁵⁴ (Cr	0,41	^{183}W	V	9,52	
58 Fe,Ni	5	5,91	56]	Fe	10,00	^{184}W	7	$21,\!48$	
60 Ni	2	3,58	58]	Fe,Ni	$0,\!15$	^{186}W	V,Os	21,29	
61 Ni	1	,10	89	Y	0,25	^{200}H	g	0,20	
62 Ni	3	,63	907	Zr	$0,\!44$	^{202}H	g	0,21	
⁶⁴ Ni,Zn	1	,24	92 9	Sr,Mo	0,16	^{198}H	g	0,21	
⁶⁶ Zn	0	,16	94 6	Sr,Mo	0,16				
$^{68}Zn_{70-}$	0	,12	138	Ba,Ce	0,33				
⁷⁹ Br	0	,13	200	Pb	$0,\!13$				
⁸¹ Br	0	,12	200	Pb	0,29				
¹³⁸ Ba,L	a,Ce 0	,25							
200 Pb 207 Pl	0	,32							
208 Pb	0	,25							
PD	0	,09 Aft	er re	eactor	operatio	n			_
Metal		Coa	ating	g on	S	ubstan	ce ap	peared	
ball in sp	ent	inne	r su	rface		betw	een ii	nner	
fuel		of or	ıter	tube		and or	uter t	ubes	
¹¹ B	0.19	^{23}Na	ı	1,56	¹¹ B	0,44	⁷⁵ A	s	0,43
23 Na	5,07	$^{24}\mathrm{M}$	g	1,16	23 Na	14,70	^{76}G	e,Se	0,16
^{24}Mg	0,21	$^{25}\mathrm{M}$	g	0,15	^{24}Mg	0,82	$^{77}\mathrm{Se}$	e	$0,\!17$
^{27}Al	0,22	^{26}M	g	$0,\!17$	^{26}Mg	0,15	^{79}B	r	0,97
²⁹ Si	3,94	^{27}Al		0,23	^{27}Al	0,92	^{81}B	r	$1,\!03$
^{31}P	0,14	²⁹ Si		0,77	29 Si	9,37	90 Z	r	$0,\!16$
³⁹ K	3,51	³⁹ K		0,86	³¹ P	0,32	¹¹⁵ I	n,Sn	0,26
43 Ca	0,14	44 Ca	a	0,71	³⁹ K	9,89	120 0	Sn,Te	$0,\!12$
⁴⁴ Ca	1,08	⁴⁵ Sc		$0,\!24$	43 Ca	0,35	¹²		$0,\!15$
⁴⁵ Sc	0,91	51 V		0,10	⁴⁴ Ca	2,15	140	3a,La,C	e0,36
51 V 52 G	1,56	53 Ci		0,57	⁴⁰ Sc	1,95	140	Je	6,54
⁵² Cr 53C	0,14	54 Ci		0,10	⁴⁰ Ti,C	a0,13	182	Je,Nd	0,85
54 Cr	0,51	56 D		1,17	52 C	6,08	183	W	3,50
55 M.	0,40	57 Fe		19,10	53 Cm	0,48	184	WOa	1,11
56Fo	0,14 7.96	- ге 58 г.	NI:	0,40 20 91	54 C π	2,07 0.61	186 1	WO_{2}	4,09 200
$_{57}^{Fe}$	1,30	59 C	;,⊥NI ⊃	o∡,o1 0 /0	55Mn	0.01	206 т	Ph	0.91
⁵⁸ Fe Ni	0,10 45.07	60 Ni	,	13.03	56_{Fo}	0,28 6.48	207	Ph	0,21 0.10
⁶⁰ Ni	19.81	61Ni		10,90 0.68	57 Fe	0,40 0.18	208	Ph	0,13 0.49
⁶¹ Ni	0.86	^{62}N		2.10	58 Fe.N	18.25	1		0,10
⁶² Ni	2,97	^{64}Ni	,Zn	5,06	⁶⁰ Ni	3,30			
$^{63}\mathrm{Cu}$	0,14	66 Zr	í	2,88	61 Ni	0,15			
64 Ni,Zn	1,62	$^{67}\mathrm{Zr}$	ı	0,47	$^{62}\mathrm{Ni}$	0,54			
⁶⁶ Zn	0,52	$^{68}\mathrm{Zr}$	ı	2,02	$^{63}\mathrm{Cu}$	0,17			
68 Zn	0,40	88 Sr		0,11	⁶⁴ Ni,Z	n1,48			
^{75}As	0,15	115 I	n,Sn	$0,\!13$	66 Zn	0,81			
$^{79}\mathrm{Br}$	0,35	^{140}C	Ċe	$0,\!37$	⁶⁷ Zn	$0,\!15$			
⁸¹ Br	0,36	^{182}V	V	$2,\!81$	68 Zn	$0,\!63$			
¹³⁸ Ba,La,C	e0,14	183V	V	$1,\!54$					
¹⁸⁴ W,Os	0,12	¹⁸⁴ V	V,Os	3,52					
^{∠08} Pb	0,17	186 V	V,Os	3,24					

 $\begin{tabular}{ll} Table III \\ The ratio of nickel isotopes in fuel and near the core of the 'Protok-6' reactor before and after the experiment. \end{tabular}$

%	⁵⁸ Ni	60 Ni	⁶¹ Ni	⁶² Ni	⁶⁴ Ni
Initial fuel	65,78	27,74	1,29	4,28	0,91
Metal ball	65,00	28,57	1,24	4,29	0,91
Fuel at edge	65,58	27,88	1,27	4,36	0,91
Coating on ce-	65,32	28,16	1,37	4,24	0,91
ramics					
Powder	66,74	26,71	1,23	4,41	0,91
between tubes					
Natural ratio	68,27	26,1	$1,\!13$	$3,\!59$	0,91

 $\begin{array}{c} \mbox{Table IV}\\ \mbox{Isotope content (atomic \%) in the reactor fuel 'VV3'}\\ \mbox{before and after reactor operation. Isotopes with a}\\ \mbox{content> } 0.1\% \mbox{ are shown.} \end{array}$

_								
	Before		After					
				Sur	face		Deep	
	Initial						layer	
_	Fuel							
	²³ Na	2,61	^{10}B	$0,\!15$	^{75}As	$0,\!17$	^{10}B	$0,\!14$
	^{24}Mg	$0,\!15$	^{11}B	$0,\!67$	$^{79}\mathrm{Br}$	0,11	^{11}B	$0,\!66$
	²⁷ Al	2,93	^{24}Mg	1,50	^{81}Br	0,21	23 Na	3,72
	²⁹ Si	1,22	^{25}Mg	$0,\!12$	88 Sr	0,23	^{27}Al	1,02
	³⁹ K	2,52	^{26}Mg	0,27	90 Zr	0,20	^{28}Si	0,25
	^{44}Ca	0,44	^{27}Al	$1,\!14$	^{107}Ag	$1,\!13$	^{51}V	2,14
	^{45}Sc	0,34	²⁸ Si	0,54	^{109}Ag	2,01	^{52}Cr	0,30
	^{51}V	0,55	^{44}Ca	0,16	^{127}I	$0,\!43$	^{53}Cr	0,73
-	^{53}Cr	0,18	^{45}Sc	0,18	$^{140}\mathrm{Ce}$	0,72	^{54}Cr	0,21
	^{55}Mn	0,14	^{51}V	2,41	^{182}W	3,34	56 Fe	2,55
	56 Fe	2,92	^{52}Cr	0,31	^{183}W	1,61	⁵⁸ Fe,Ni	44,47
	⁵⁸ Fe,Ni	45,09	^{53}Cr	0,81	$^{184}W,Os$	3,39	⁶⁰ Ni	18,23
	⁶⁰ Ni	19,01	56 Fe	0,42	185 Re	0,26	⁶¹ Ni	0,87
	⁶¹ Ni	0,88	⁵⁸ Fe,Ni	46,08	¹⁸⁶ W,Os	2,91	⁶² Ni	2,83
-	⁶² Ni	2,93	⁶⁰ Ni	19,34	¹⁸⁷ Re,Os	\$0,60	⁶⁴ Ni,Zn	0,87
	⁶⁴ Ni,Zn	1,00	⁶¹ Ni	0,96	²⁰⁶ Pb	0,18	^{75}As	0,12
	⁶⁶ Zn	0,13	⁶² Ni	3,02	207 Pb	0.13	$^{79}\mathrm{Br}$	0.19
	⁶⁸ Zn	0,10	^{63}Cu	0.39	208 Pb	0.34	$^{81}\mathrm{Br}$	0.19
	$^{79}\mathrm{Br}$	0,10	⁶⁴ Ni,Zn	1,31		,	90 Zr	0.39
	7 Li	0,60	^{65}Cu	0,16			92 Sr,Mo	0.14
	$^{81}\mathrm{Br}$	0.10	66 Zn	0.37			94 Sr.Mo	0.16
	¹³⁸ Ba,La,Ce	0,20	68 Zn	0,20			^{107}Ag	1.37
	^{182}W	3.73		,			109 Ag	1.39
	^{183}W	1.92					140 Ce	0.78
	^{184}W	4.33					142 Ce.Nd	0.10
	186 W.Os	4.29					^{182}W	3.49
	²⁰⁶ Pb	0.26					^{183}W	1.90
	²⁰⁷ Pb	0.20					184 W.Os	4.17
	²⁰⁸ Pb	0.56					185 Re	0.38
		-,					186 W.Os	3.72
							¹⁸⁷ Be Os	0.60
							²⁰⁸ Pb	0.17
_							10	0,11

Table V The ratio of Nickel isotopes in fuel VV3 before and after reactor operation.

%	⁵⁸ Ni	⁶⁰ Ni	⁶¹ Ni	⁶² Ni	⁶⁴ Ni
Initial fuel	65,93	27,98	$1,\!19$	$3,\!98$	0,91
Surface	65,79	$27,\!61$	1,37	4,31	0,91
Deep layer	66, 36	27,20	1,29	4,23	0,91
Natural ratio	68,27	26,10	$1,\!13$	$3,\!59$	0,91

Table VI

Isotope content (atomic %) in fuel and near the active zone of the KV3 reactor before and after reactor operation. Isotopes with a content >0.1% are shown.

Boforo

т	T ::: 1 C 1		Delore			TT			
- 0222	nitial f	uel	Ceramics			Heater wire			
²³ Na		0,33	² Al	88,15	5 23	'Na	$0,\!47$		
³⁹ K		0,38	²³ Na	1,98	29	'Si	$0,\!27$		
56 Fe		$0,\!45$	^{24}Mg 0,82		36	γĸ	$0,\!40$		
58 Fe,	Ni	64, 49	^{25}Mg 0,12		44	$^{\rm l}Ca$	$0,\!12$		
60 Ni		27,63	²⁶ Mg 0.14		56	⁵ Fe	0,21		
61 Ni		1.18	29 Si	0.37	58	Fe,Ni	0.12		
⁶² Ni 3,88		3.88	^{39}K	2.10	18	^{32}W	20.24		
64 Ni	Zn	1.21	^{44}Ca	0.21	18	^{33}W	11.02		
	511	-,	47 Ti	0.88	18	^{34}W Os	24.39		
			⁴⁸ Ti	0.18	18	⁸⁵ Be	6 97		
			54Cr	0,10	18	$^{36}WO_{c}$	0,01		
			56 E	2 17	18	³⁷ Pa Oa	11.95		
			58 E	3,17 N: 0.99	19	⁸ 11. Dt	11,60		
			60 NC	NI 0,28	20	°Hg,Pt	0,12		
			80 N1	0,11	20	^{oo} Hg	0,14		
			128 m	0,13	20	/ ² Hg	0,13		
			138 Ba	a,Ce 0,17	20	°Pb	$0,\!18$	_	
Fuel			Cora	mics		Su	hetanc	0	
contr	al		Cera	annes		ou an	Deared	1	
centra						botw	oon in	nor	
20110	,					and o	utor ti	ibos	
$23 N_{\odot}$	0.12	11 D	0.12	64 N; 7n	0.22	23 Mo	uter tt	5 52	
31 _D	0,13 0.11	23 No	15.61	667n	0,22	$24 M_{\odot}$		0,00	
г 391/2	0,11	24 M.m	10,01 1.06	76 C a S a	0,11	27 A 1		0,00	
56 D	0,14	25 M	1,00	88g	0,20	29 C		0,52	
58 Fe	0,23	26 Mg	0,13	so Sr	0,21	-* S1 31 D		1,42	
60 Fe,N1	65,39	20 Mg	0,24	90 Y	0,22	30 r		0,16	
61 N1	26,15	21 Al	6,05	⁹⁰ Zr	$0,\!42$	³³ K		6,93	
⁰¹ Ni	1,18	²⁹ Si	4,26	⁹² Sr,Mo	0,22	⁴⁴ Ca		$0,\!88$	
⁶² Ni	3,99	³⁹ K	15,26	⁹⁴ Sr,Mo	$0,\!17$	45 Sc		$0,\!82$	
⁶³ Cu	0,84	43 Ca	0,26	¹⁰⁹ Ag	$0,\!10$	^{54}Cr		0,26	
⁶⁴ Ni,Zn	$1,\!14$	^{44}Ca	3,15	^{127}I	0,16	⁵⁶ Fe		7,04	
65 Cu	$0,\!42$	^{45}Sc	2,04	138 Ba,Ce	$_{\pm 0,39}$	58 Fe,N	Ji	$1,\!80$	
		⁴⁸ Ti,	0,23	^{182}W	4,32	⁶⁰ Ni		0,74	
		^{51}V	0,22	^{183}W	2,35	⁶² Ni		0,10	
		^{54}Cr	0,96	^{184}W	5.01	^{63}Cu		0.13	
		^{55}Mn	0.10	185 Re	5.95	⁶⁴ Ni.Z	'n	0.15	
		56 Fe	21.14	186 W.Os	4.77	^{182}W		18.29	
		57Fe	0.15	²⁰³ Tl	0.15	^{183}W		10.44	
		58 Fe Ni	0.91	206 Ph	0.15	184 W		21 36	
		60 Ni	0.37	207 Ph	0.15	186 117	\bigcap_{α}	21,00	
		63 C u	0,37	208 DL	0,10	198 LL	US	20,90	
		υ	0,14	20011	0,40	ng		0,17	
				202 Hg	0,20				
				208 DI	0,18				
				²⁰⁸ Pb	$0,\!15$				

Table VII

Before

Relative content of nuclides (atomic %) in the ceramic tube before and after the operation of the KV3 reactor. Nuclides are shown whose content has increased more than 10 times.

After

	Defore	111001	THICT / DCI
^{10}B	0,0008	0,0318	41,8
^{11}B	0,0054	0,1277	23,4
²⁹ Si	0,3709	4,2603	11,5
43 Ca	0,0158	0,2638	16,7
^{44}Ca	0,2123	3.1461	14.8
^{45}Sc	0,0507	2,0384	40,2
⁴⁶ Ti.	0.0074	0.0836	11.3
51 V	0.0028	0.2151	78.0
^{53}Cr	0.0057	0.0753	13.3
⁶⁴ Ni Zn	0.0186	0.2224	12.0
⁶⁶ Zn	0.0099	0.1102	11.1
⁶⁷ Zn	0.0014	0.0211	15.0
68 Zn	0.0080	0.0808	10.1
^{72}Ge	0,0001	0.0037	27.2
75 As	0,0001	0.0138	102.2
76 Co So	0,0001	0,0106	102,2 17.2
775	0,0113	0,1970	82.2
78Se Kr	0,0001	0,0000	10.7
79 Br	0,0028	0,0542	20.3
81 Br	0,0020	0,0500	10.6
83 Kr	0,0040	0,0190	13,0 13.7
107 A g	0,0001	0,0009	13,7
109 A g	0,0007	0,0803	13,0
113 Cd In	0,0071	0,1020	14,5 12.7
^{114}Cd Sp	0,0001	0,0003	11.0
116Cd Sp	0,0000	0,0004 0.0275	12.8
117Sn	0,0022	0,0275	12,8
118 Sn	0,0011	0,0123	17.0
119 Sn	0,0024	0,0422	11,5 11.7
120 Sn To	0,0014	0,0105	10.5
119 Sn	0,0034	0,0070	13,5 11.7
122 Te	0,0014	0,0103	15.0
127 I	0,0007	0,0101	15,0 25,7
128 To	0,0002	0,1003	20,1
124 To	0,0002	0,0040	11 4
130 To	0,0008	0,0092	11,4 16.7
182 W	0,0000	4 3168	10,7 567.8
183117	0,0070	2 2 4 8 0	671.7
184 W	0,0055	2,3489	658.8
185 Ro	0,0070	5,0087	058,8
$186WO_{C}$	0,0000	0,9409 4 7749	5276
198Hg	0,0089	4,7740	007,0 038 5
199 L a	0,0001	0,0321	230,5
200 Lg	0,0007	0,0248	128.0
202 L	0,0004	0,0500	100,9
203 TI	0,0005	0,0000	101.2
204 Ph Ha	0,0010	0,1490	101,2
го,пд	0,0010	0,0101	10,0

It can be seen that in addition to tungsten and rhenium, the appearance of which can be explained by its migration from the heater coil, the boron content in the ceramic tube greatly increased, as well as nuclides with atomic masses of 43-53, 64-83, 107-130, 198-208.

Table VIII shows the results of an analysis of the ratio of nickel isotopes in fuel, as well as in the surrounding ceramic and in the substance accumulated between the inner and outer tubes, before and after the reactor operation. When analyzing the isotope composition, in order to avoid the errors associated with the registration of 64 Zn, the 64 Ni share was taken from the reference book [7].

It can be seen that the isotopic composition of nickel in fuel before and after the experiment remained practically unchanged. Some differences are noticeable in the results obtained for the ceramic tube and the substance between the tubes. But these results can not be considered accurate, since the concentration of nickel in the samples studied is not high enough for reliable analysis.

In addition to the Vernadsky Institute of Geochemistry and Analytical Chemistry RAS, the analysis of KV3 fuel before and after the experiment, as well as the substance from the space between the inner and outer tubes, was made by the research company Coolescence LLC, Boulder, Colorado, USA. EDS analyzes were performed using an electronic scanning microscope, as well as analyzes using the ICP-MS method. These studies confirmed the insignificant changes in the isotopic composition of the fuel, the appearance in the fuel of about 1% of copper and the

After/before

Table VIII The ratio of Nickel isotopes in fuel and near the core of the KV3 reactor before and after reactor operation.

%	58 Ni	60 Ni	61 Ni	62 Ni	64 Ni
Initial fuel	65,93	27,98	1,19	$3,\!98$	0,91
Fuel after work	65,74	28, 17	1,20	3,98	0,91
Substance be-	66, 66	27,33	1,30	3,79	0,91
tween tubes					
Ceramics	$67,\!65$	27, 37	0,82	3,26	0,91
Natural ratio	68,27	$26,\!10$	$1,\!13$	3,59	0,91

presence of many nuclides in the substance from the space between the tubes.

VI. DISCUSSION

Thus, a significant change in the nuclide composition as a result of the operation of the investigated nickelhydrogen reactors occurs not only in the fuel, but also in the ceramics surrounding the reactor core. In addition, a substance containing sodium, potassium, silicon, iron, boron, calcium, zinc and many other elements accumulates in the cavity between the inner and outer tubes. Especially great amount of tungsten appeared. It is reasonable to assume that the source of tungsten is the hot spiral of the heater. The most understandable mechanism of substance migration is evaporation in places with high temperature and condensation in less heated places. As the measurements show, the temperature of the heater wire reaches 1700°C. But even at this temperature, the density of tungsten vapor ($<10^{-10}$ Pa) is too low for such mechanism to work with a noticeable intensity. Obviously, more complex physicochemical processes take place with the participation of hydrogen and other reagents that may be present in the reactor. It is possible that a number of other elements appear as a result of migration from structural materials, since sodium, potassium, silicon, calcium, iron and a number of other elements are contained in appreciable quantities in the heater wire, thermocouples and in ceramics. However, there are some elements (cobalt, cerium, gallium, germanium, arsenic, selenium, cadmium, tellurium) that appeared in significant quantities, which are practically not presented in the initial fuel and structural materials. This indicates the possibility of their appearance as a result of nuclear transmutations. For example, cerium can be a product of the fission of tungsten

$$^{182}W \rightarrow ^{140}Ce + ^{42}Ca + 4e^- + 4\tilde{\nu}_e + 76.04MeV$$
 (3)

Attention is drawn to the appearance of a significant amount of copper in the fuel of the KV3 reactor (0.84% 63 Cu and 0.42% 65 Cu) with a total mass of about 20 mg. It can be assumed that this is due to the course of nuclear reactions given below:

$${}^{62}Ni + {}^{1}H \rightarrow {}^{63}Cu + 6,125MeV$$

 ${}^{64}Ni + {}^{1}H \rightarrow {}^{65}Cu + 7,450MeV$ (4)

About 200 MJ are released as a result of the reactions (4), when 20 mg of copper is formed. This energy release does not contradict the total excess heat release in the KV3 reactor (about 400 MJ). In addition to heat generation, the appearance of such a quantity of copper should cause a decrease in the relative content of 62 Ni by 0.8% and 64 Ni by 0.4%. The data presented in Table VIII does not show such changes. It should be noted that the predicted changes lie within the limits of a possible measurement error, and the 64 Ni content is generally difficult to measure reliably due to uncontrolled additions of 64 Zn.

VII. CONCLUSION

1. The isotopic and elemental composition of the substance in four nickel-hydrogen reactors of various designs with an excess energy output from 100 to 790 MJ has been analyzed. Not only the changes in fuel, but also the materials adjacent to the active zone have been investigated. In addition, the composition of the substance accumulating in the cavity of the reactor near the active zone has been studied.

2. There were no significant changes in the isotopic composition of nickel and lithium, except for the analysis of the fuel of the AP2 reactor at Uppsala University (Sweden).

3. A significant increase in the concentration of impurities of a number of nuclides has been detected not only in fuel, but also in structural elements adjacent to the active zones of the reactors. In addition to tungsten and rhenium, the appearance of which can be explained by migration from the heater coil, the content of boron increased greatly, as well as nuclides with atomic masses of 43-53, 64-83, 107-130, 198-208.

4. In the substance that was found in the cavity of the reactor near the active zone, in addition to tungsten, a lot of iron, sodium, potassium, nickel, silicon, calcium, scandium and a number of other elements accumulated.

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