

**NUCLEAR REACTIONS EXCITED
BY RECOIL PROTONS ON A NUCLEAR REACTOR**

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ABSTRACT. The nuclear reactions excited by recoil protons and of the detection possibility of the various chemical elements with the use of these secondary nucleus reactions were investigated. The recoil protons are produced on a nuclear reactor in the result of (n, p) inelastic and elastic scattering interaction of fast neutrons with nuclei of hydrogen. It is well known that the share of fast neutrons in energetic spectrum of reactor's neutrons in comparison with the share of thermal neutrons is small. Consequently, the share of recoil protons produced in the result of fast neutron interaction with nuclei of light elements, capable to cause the nuclear reactions, is also small. des, due to Coulomb barrier of nuclei the recoil protons can cause the nuclear reactions only on nuclei of light and some middle elements. Our studies show that observable yields have radio nuclides excited in the result of nuclear reactions on Li, B, O, V and Cu. Our experimental results have demonstrated that the proton activation analysis based on the application of secondary nuclear reactions is useful technique to determine large contents of various light and medium chemical elements. Detection limits for studied chemical elements are estimated better than 10 ppm.

INTRODUCTION

The determination of lithium, oxygen and other light chemical elements in microgram level in various modern materials is of importance problem of analytical science. Such problems are solved with use of some variants of nuclear methods of the analysis. There are several non-traditional reactor activation analysis techniques to solve such problems which have been developed and applied in various fields of semiconductor industry, biology, geology [1]. In recent years these techniques obtained the name of charged particles activation analysis based on the use of the nuclear reactor (NRCPAA).

We distinguished two possibilities of the application of a nuclear reactor as charged particles source. During last years the possibilities of NRCPAA were investigated intensively and some our results were applied to determine of light elements contents [1-3].

Purpose of this presentation is a study of nuclear reactions excited by recoil protons and of the detection possibility of the various chemical elements with the use of these secondary nucleus reactions.

EXPERIMENTAL TILE

We investigated the yields of radio nuclides produced by secondary nuclear reactions with recoil protons on chemical elements with $Z \leq 29$. On these elements the nuclear reaction (p, n) is excited, but on boron also (p, α) and (p, α n). Nuclear data for reactions and radio nuclides are listed in the Table 1.

Table 1- Nuclear -physics characteristics of radio nuclides produced by secondary nuclear reactions with recoil protons on chemical elements with $Z \leq 29$

Chemical elements	Radio nuclides	$T_{1/2}$	E_γ , key, I_γ (%)
Li	^7Be	53,61 d	477,5(10,3) *
B	^7Be	53,61 d	477,5(10,3)
N	^{11}C	20,34 m	511(199,52)
O	^{18}F	1,83 h	511(1993,4)
S	$^{34\text{m}}\text{Cl}$	31,99 m	3305 (11,6) 2928(48,4), 11744(14,1) 640(0,48) 511(120) 145,7(35,8)
Ti	^{48}V	16,18 d	2421,7(5-3) 2375,6(0,010) 2240 (2,4) 1312(98) 983,3(100), 944,3(8) 511(99,6)
V	^{51}Cr	27,8 d	320(9,63)
Cr	^{52}Mn	5,60 d	1645(0,04), 1434,4(100), 1333,8(5,1\), 1247(4,7), 935(94), 848,4(3,2), 744,2(85), 511(55)
	^{54}Mn	312,5 d	834,81(99,978)
Cu	^{65}Zn	245,7 d	511(2,82), 1115,45(50,6)

*) γ -quant used in estimated process

Specimen's preparation

The methods of samples preparations are several differ for studied chemical elements.

Oxygen To study the possibility of the determination of oxygen by means of nuclear reaction $^{18}\text{O}(p,n)^{18}\text{F}$ we have used distilled water of of 1 ml volume flooded in polyethylene tubes (3 pieces). Each tube has wrapped the filter paper and has put in container for irradiation.

Sulphur First way comprised of following action: The polyethylene has fused with crystalline sulphur in mass correlation 201 mg: 63 mg under corresponding temperature. Viscous mass was carefully mixed by means of glass stick and have left to cool off. Second way comprised of following action: The polyfoam of 150 mg mass have dissolved in acetone. Crystalline sulphur of 50 mg mass has melted on weak fire under corresponding temperature. Viscous mass was carefully mixed by means of glass stick and have placed in dry closet for removing the acetone. The received samples have packed in polyethylene package and have put in Al-container for the further irradiation.

We have chosen only one way of the samples preparation for all metallic chemical elements. The powder specimens were fixed in some hydrogen - containing material. As such material we have chosen various organic compounds - polystyrene, the polyfoam or epoxy. In some cases we have chosen the polyfoam dissolved in organic solvent (for example, in toluene). The investigated powdery samples are carefully mixed in this viscous liquid. The weight of the polyfoam must be approximately in 3 times more than weight of investigated samples. So on preparation of one sample we took approximately 150 mg polyfoam. The prepared sample flooded in cup Petri and left to dry during full day. The dried samples present itself flexible film which then packed in polyethylene bags for irradiation.

The samples placed in quartz ampoule of 20 mm diameter was sealed and surrendered on irradiation in Al-container. Time of the irradiation has formed 24 hours, cooling time was 7 days. The experiments have shown that such ways of the sample preparation and their irradiation on a nuclear reactor not always give the satisfactory results. In some cases the samples are charred. Besides sometimes quartz ampoule even is break up due to the accumulation of gas produced in result of the decomposition of organic materials. For this reason there is need of searching for other material steadfast for radiation heating. The high - molecularly compounds - the resin, the epoxies answer this requirement.

Sample preparation with using the epoxy was conducted as follows. The resin divorced with using the solvent in proportions 5:1. As it was mentioned above, weight "proton -creating" material must be in 3-4 times more weight of investigated sample. Since weight of sample was 30-50 mg we took 200-300 mg of this viscous mixture and fixed him with the investigated sample.

In Table 2 are brought chemical elements and the list of the irradiated compounds.

Table 2. Chemical compounds used for investigation

Investigation chemical element	Irradiated chemical element compound	Weight sample, mg
Li	LiOH	$m_1=32,4; m_2=48,5; m_3=48,0$
B	H_3BO_3	$m_1=44,5; m_2=48,0; m_3=50,0$
V	V_2O_5	$m_1=46,5; m_2=50,0; m_3=50,0$
Ti	TiO_2	$m_1=32,4; m_2=48,0; m_3=48,5$
Cu	$CuCl_2 \cdot 2H_2O$	$m_1=44,1; m_2=48,0; m_3=48,0$

For preparing of sample we have weighted out the compounds on electronic weight in Radio - Analytical Centre of INP of the Republic of Uzbekistan. We took three samples of 30-50 mg. weight each. Probes on glass substrate were mixed with reconnoitred epoxy. After carefully mixing the samples were left for full hardening during 8-10 hours.

The solidified samples left on substrates were put in polyethylene bags which were folded in Al-foils, numbered and sealed in quartz ampoule. There

were irradiated not more than 10 samples in single quartz ampoule of 25 mm diameter. The quartz ampoule was placed in Al-container for irradiation in channel of the nuclear reactor.

Irradiation sample

The samples of the oxygen and sulphur were irradiated in ninth channel of the nuclear reactor of INP in current of 3 minutes. After irradiation the samples were carried in detection room for measurement of their radio activities. The metal samples were irradiated in neutron flux during 20 hours. The flux density of thermal neutrons was $7.10^{13} \text{ cm}^{-2} \text{ s}^{-1}$. The cooling time of irradiated samples depends on half life of radio nuclides and is changed from 2 to 7 days. The cooled samples were carried from "hot cameras" in boxes for unpacking samples where quartz ampoules were split and samples were placed in separate polyethylene bags for measurement of radioactivity.

Measurement of saturated radioactivity of samples

The saturated radioactivity of irradiated samples was measured on semiconductor detector REGE of company CANBERRA. Time of the measurement was changed from 200 to 1000 and more seconds that depends on value of radioactivity. Since the radionuclide ^{18}F is pure positron emitter the measurement its radioactivity was realized using γ - quantum with energy equal 0.511 MeV. It is known that row of other radio nuclides emits also 0.511 MeV γ -quantum. So the identification of radionuclide ^{18}F we realized by decomposition of the decay crooked on components. We began the measurement of the radioactivity of irradiated distilled water in glass cup having disposed it directly on surface of the detector. The measurement lasted during uptime at each minute on several seconds. The measured results were inflicted on half logarithmic scale paper for building of the decay crooked of radionuclides. It is discovered that the decay crooked consists of amount of long lived components - $^{18}\text{F}, ^{13}\text{N}$. The last radionuclide can be formed on reaction $^{13}\text{C}(p,n)^{13}\text{N}$ and $^{16}\text{O}(p,\alpha)^{13}\text{N}$.

The measurement of radioactivity of other samples conducted on stand installed on different distances from surface of the detector. The value of activity of radio nuclides from the other elements was found with using of photo peaks of γ - quantum marked in italics (Table 1).

YIELDS OF RADIONUCLIDES

For choice of the optimum conditions of the analysis and the estimation of analytical parameters of methods the experimental data - the yield of radionuclide or the cross section of the nuclear reaction is necessary. However, in practice of activation analysis it is more comfortable to deal the yield of radionuclide than the cross section of the nuclear reaction.

Yield of radionuclide is defined by simple expression:

$$Y = \frac{A_0}{m \left(\frac{1 - e^{-\lambda t_{\text{irr}}}}{\lambda} \right)} \quad (1)$$

Where,

Y- Yield of radionuclide;

m- Mass of irradiated chemical element;

t_{irr} - Irradiation time;

A_0 - Initial activity of radionuclide ($t_{\text{cool}} = 0$);

λ - The decay constant;

Activity brought about unit of the mass, we found as

$$A = \frac{A_0}{m} \quad (2)$$

In Tables 3-6 are presented the experimental data obtained on this presentation which has the dimensionalities:

I_0 - intensity of γ -quant area, imp/s;

A_0 -Activity, kBq;

A- Brought activity, kBq/g;

Y- Yield of radionuclide, kBq/g.h;

Table 3. Experimental data for nuclear reaction ${}^7\text{Li}(p,n){}^7\text{Be}$.

N_{sample}	$T_{\text{cool}}, \text{Days}; I, \text{imp/s}$				I_0	A_0	A	Y
	17	39	66	95				
4	3,126	2,274	2,389	0,996	4,18	0,21	22,1	22,0
5	4,029	2,558	1,822	1,421	5,60	0,28	19,8	19,7
6	3,255	2,517	1,716	1,080	4,10	0,21	14,6	14,5
Average	3,47 ±	2,45 ±	1,98 ±	1,17 ±	4,62 ± 0,62	0,23 ±	18,8 ±	18,7 ±
	0,37	0,51	0,28	0,17	4,38 ± 0,56	0,03	2,8	2,8

Table 4. Experimental data for nuclear reactions ${}^{10}\text{B}(p,\alpha){}^7\text{Be} + {}^{11}\text{B}(p,\alpha n){}^7\text{Be}$

N_{sample}	$T_{\text{cool}}, \text{Days}; I, \text{imp/s}$				I_0	A_0	A	Y
	10	38	66	95				
7	0.92	0.82	0.64	0.30	0.98	49	6.20	6.19
8	1.00	0.71	0.51	0.26	1.20	60	7.05	7.04
9	0.77	0.55	0.31	0.16	0.87	44	4.96	4.95
Average	0.90	0.69	0.49	0.24	1.01 ± 0,12	51 ± 6	6.07	6.06
	± 0.08	± 0.09	± 0.12	± 0.06	1,10 ± 0.12		± 0.89	± 0.88

Table 5. Experimental data for nucleus reaction $^{51}\text{V}(p, n)^{51}\text{Cr}$

N_{sample}	$T_{\text{cool}} = 118 \text{ Days}; I, \text{imp/s}$	I_0	A_0	A	Y
16	2.02+0.01	38.11	1.7	63.2	52.4
17	4.68+0.04	88.30	4.0	138.4	114.9
18	4.76+0.07	89.81	4.1	141.8	117,7
Average				114.5 ± 27,3	95.0 ± 28.4

Table 6. Experimental data for nucleus reaction $^{65}\text{Cu}(p, n)^{65}\text{Zn}$

N_{sample}	$T_{\text{cool}} = 118 \text{ Days}; I, \text{imp/s}$	I_0	A_0	A	Y
13	7.98+0.02	11.1	1.85	111.4	95.8
14	15.11+0.07	21.0	3.50	194.4	167.2
15	12.91+0.06	18.0	3.00	166.6	143.3
Average				157.4 ± 30,6	135.4 ± 26.4

We have not been able to obtain the experimental data for nucleus reaction $^{34}\text{S}(p, n)^{34}\text{Cl}$ due to following reasons. The threshold of this reaction is 6.5 MeV that excites on isotope ^{34}S with little abundance (4.22 %). As we have mentioned above, the lot of recoil protons with energy above 6 MeV in spectrum protons is small. Besides, half-life of ^{34}Cl is relatively short ($T_{1/2} = 31.99$ minutes, $E_p = 3305(11,6), 2928(48,4), 1174,4(14,1), 511(120), 145,7(35,8)$ keV). Therefore the main radioactivity lost before detection.

We evaluated the yield of fluor-18 produced by nuclear reaction $^{18}\text{O}(p, n)^{18}\text{F}$ equal 11.6 kBq/g.h

DISCUSSION OF RESULTS

To estimate the analytical possibility of the NRC PAA we have compared our experimental data with data obtained with using of the cyclotron [4]. These data are listed in Table 7. The comparison is possible only conditionally since the data obtained on cyclotron corresponds the case when small surface layer of sample is irradiated. Consequently, the irradiated mass of sample is small. In the case of nuclear reactor the mass of irradiated sample can be significant. For this reason we have been able to compare only brought values of yields.

Our data for lithium is smaller in 200 times than such data obtained with using of a cyclotron. The real spectrum of irradiated sample of lithium for LD gives the value approximately equal $9 \cdot 10^{-5}$ that is also smaller in 200 times

than LD for lithium for the case of a cyclotron. Our data for boron is smaller in 80 times than such data obtained with using of a cyclotron. LD is $7 \cdot 10^{-4}$ g.

Table 7 - Comparative data for two ways of activation

Nuclear reaction	Nuclear reactor		Cyclotron ($E_p = 6$ MeV[4])	
	Y, kBq/(g.h)	LD	Y, kBq/(μ A.h)	LD [4]
$^{18}O(p,n)^{18}F$	11.6		1950	
$^7Li(p,n)^7Be$	18.7 ± 2.8	$10^{-3} - 10^{-4}\%$	3260	$10^{-6} - 10^{-5}\%$
$^{10}B(p,\alpha)^7Be$ $^{11}B, p,\alpha)^{10}Be$	6.06 ± 0.88	$10^{-3} - 10^{-4}\%$	470	$10^{-5} - 10^{-4}\%$
$^{51}V(p,n)^{51}Cr$	95.0 ± 28.4		970	
$^{65}Cu(p,n)^{65}Zn$	135.4 ± 26.0		28	

Our data for vanadium is smaller in 10 times than the data obtained on cyclotron, but for copper our data turned out to be even above approximately in 4 times. These are a last data need to check again. Such high yield can be conditioned due to the impurity of chromium in irradiated sample because the nuclear reaction of capture of thermal neutrons is possible which can produce same radionuclide as in the case of recoil protons. We do not bring the data for titanium, chromium, calcium, scandium and others since they are found on stage of the study.

It is need to note that the proposed method can be used simultaneously with NAA. The irradiation of investigated sample in proton - forming shell and without allows using as reactions with neutrons, so with recoil protons that in turn allows to increase the circle of simultaneously determined elements. Besides, the possibility for expansion of the number of radio nuclides produced on a nuclear reactor (for instance, berillium-7) appears.

CONCLUSION

Our studies show that the yields for radio nuclides produced in result of collision of recoil protons with Li, B, O, V and Cu are sufficiently high. The values of yields are in range from 6.06 for boron till 135.4 kBq/(g.h) for copper.

Our experimental results demonstrate that the proton activation analysis based on the application of secondary nuclear reactions is useful technique to determine large contents of some light and medium chemical elements. LD for studied chemical elements are estimated better than 10 ppm.

The possibility for expansion of the number of radio nuclides produced on a nuclear reactor appears.

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