MEASUREMENT OF THE REACTION RATES IN 232Th SAMPLES IRRADIATED BY 4 GeV DEUTERONS AND SECONDARY NEUTRONS

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This work is done in the framework of the international collaboration

Beginning from the 90s of the XX century all over the world the research of accelerator driven sub-critical systems is being actively done with the aim of practical application in order to receipt nuclear power Accelerator Driven Subcritical systems and transmutation of long-living fission products and actinides.

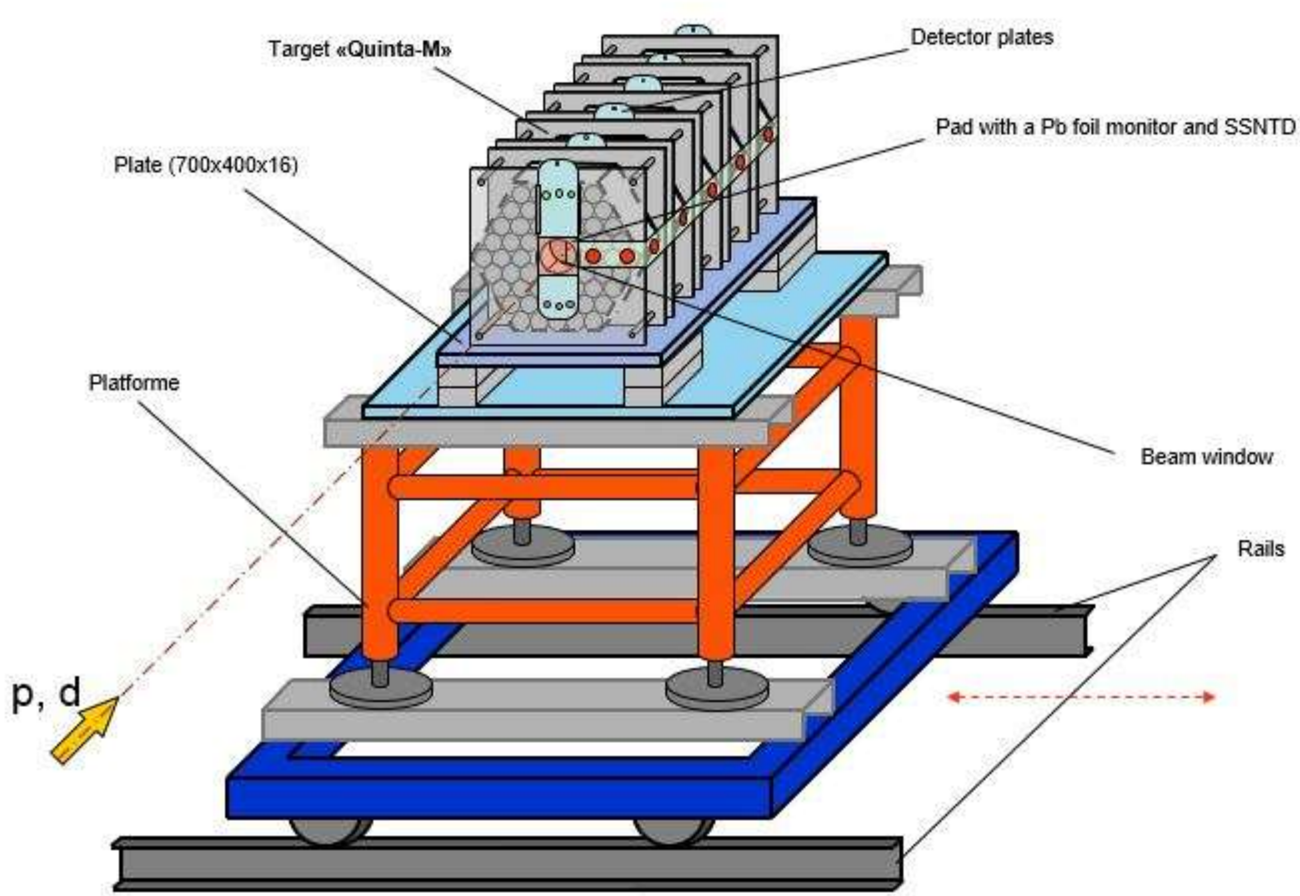
In connection with the observed data on neutron reaction rates particularly those about nucleuses ^{232}Th are quite appropriate and useful.

This work focuses on the formation of a hard neutron spectrum inside a spallation target. It is expected that the fast neutrons can more effectively “ burn “ natural (depleted) uranium and thorium in order to produce energy. Simultaneously, long-lived components of the spent fuel of nuclear power plants can be incinerated by fast neutrons with higher efficiency.

In recent years, such studies have been done and are being held on the Nuclotron accelerator VBLHEP JINR by the collaboration “Energy plus Transmutation of radioactive waste”. As a part of this program, a large number of experiments have been carried out with the subcritical spallation uranium target “QUINTA”.

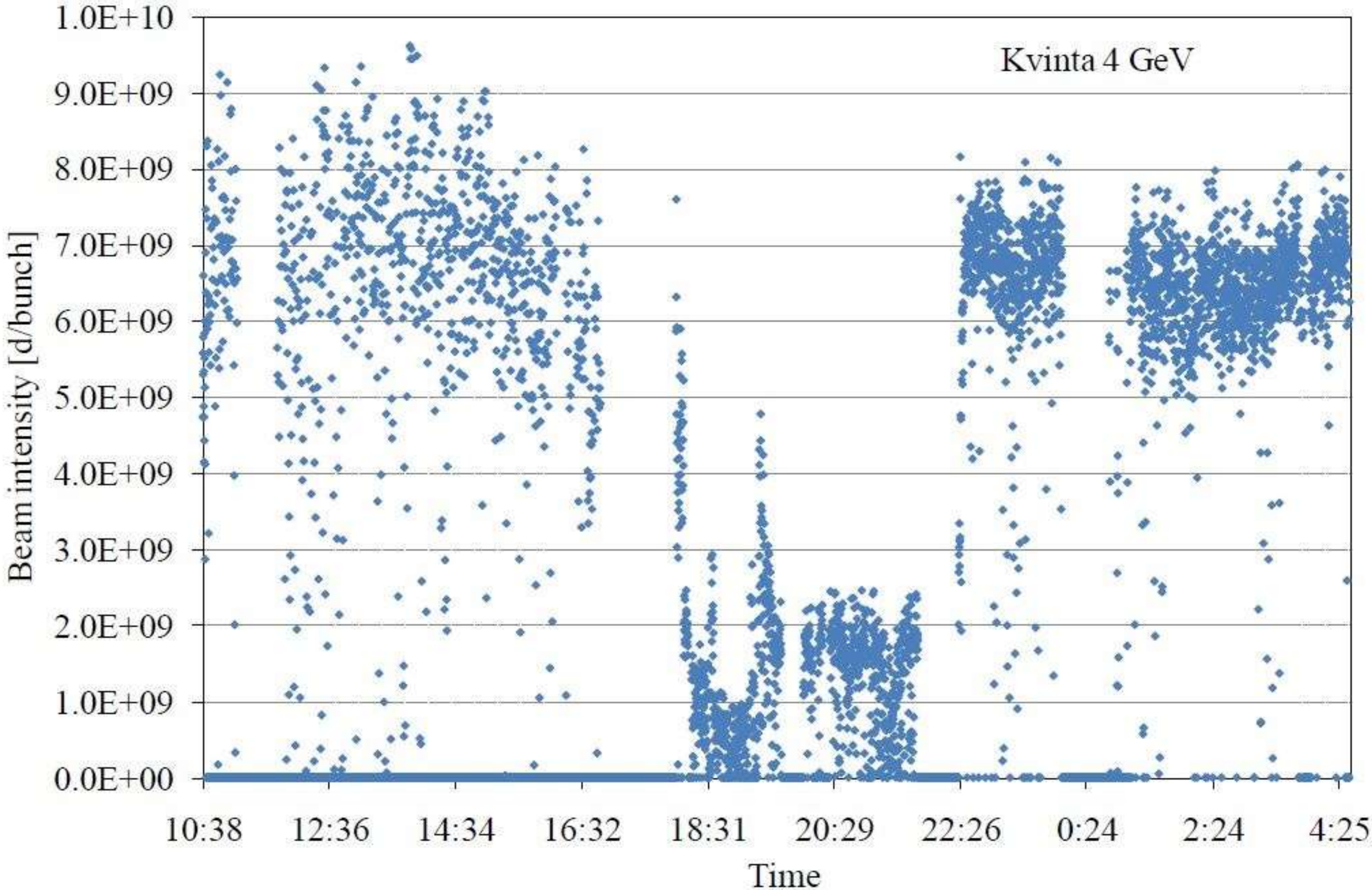
The experiment was performed at the "*QUINTA*" (uranium target assembly, weight of 512 kg, consisting of 5 separate sections) on the deuteron beam (4 GeV) of the Nuclotron accelerator VBLHEP JINR.

QUINTA-M setup layout at the irradiation position



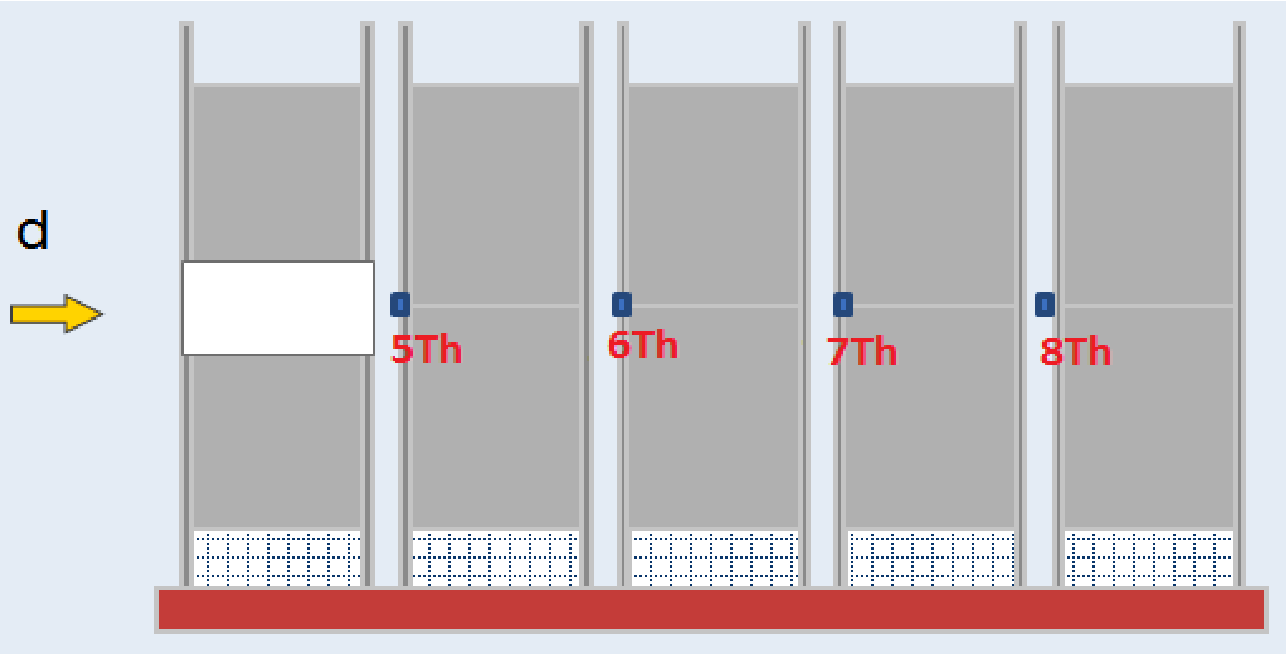
- SSNTD and AD positions at the QUINTA-M target surface

Target *QUINTA*



The course of irradiation during 4 GeV deuteron run on Kvinta.

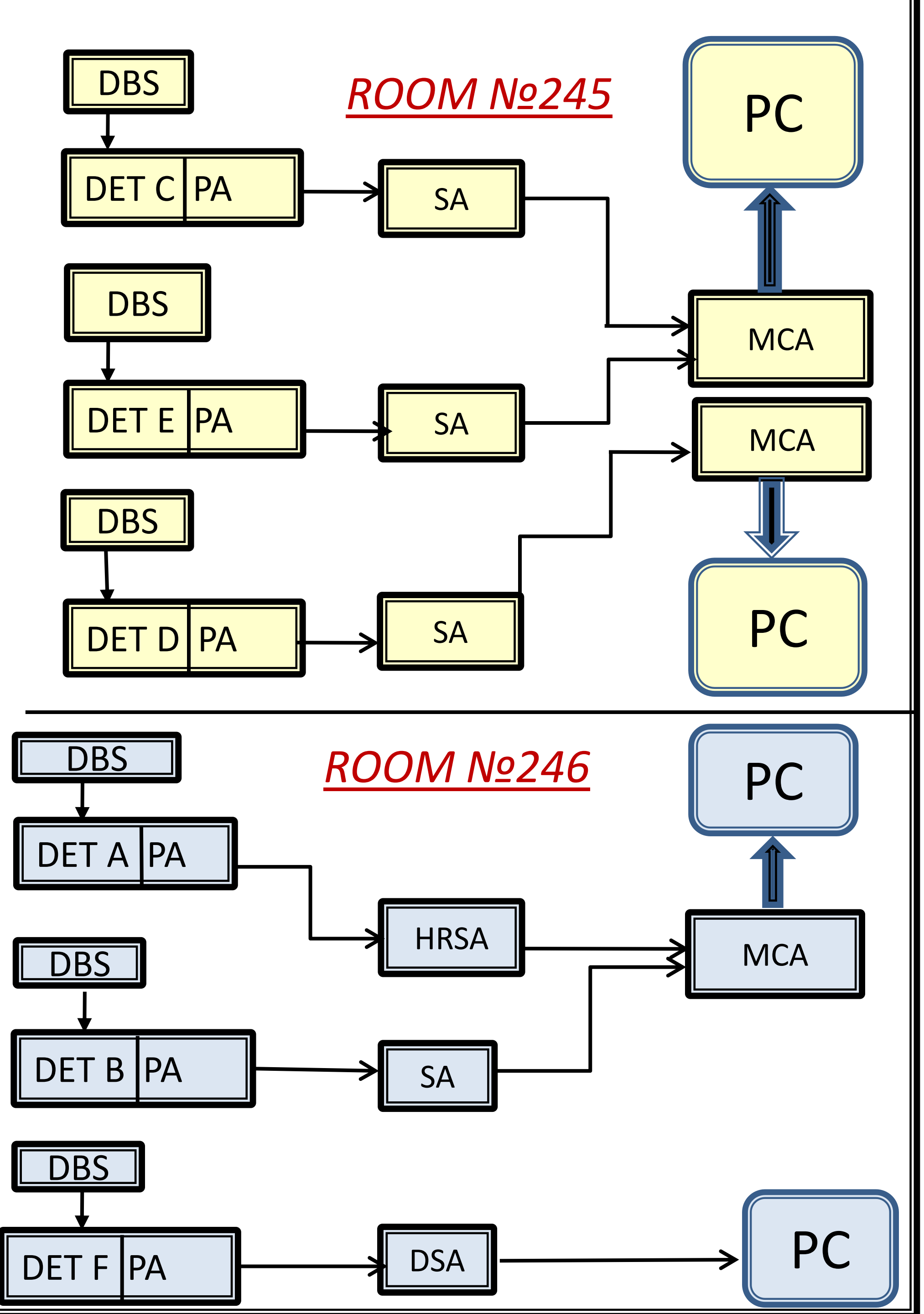
The total flux of deuterons on the target was **1.41E+13** particles during **1157 min.** irradiation. The samples of 232Th were placed inside the uranium assembly at the deuteron-beam.



The position of investigated ^{232}Th samples.

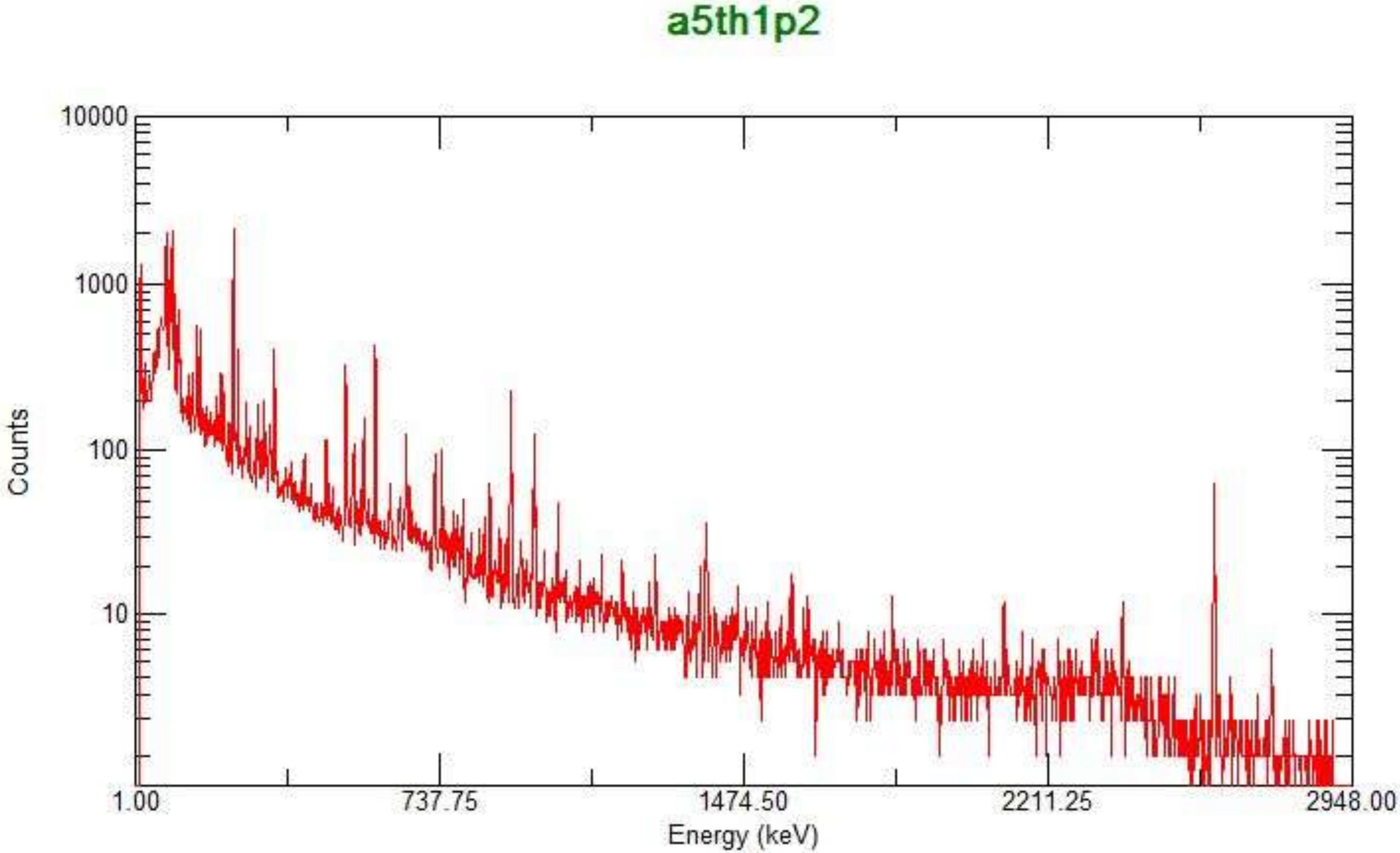
After irradiation, the sample was taken to the spectrometric complex YASNAPP-2 in the JINR LNP, where the spectra of γ-radiation were measured using HPGe-detectors.

Firm detector	Resolution on line ^{60}Co 1332(keV)	Relative effciency(%)
A(canberra)	1.9	18
B(ortec)	1.8	26
C(ortec)	1.8	32.9
D(ortec)	335eV(on line 5.9 keV)	-
E(ortec)	1.9	28.3
F(canberra)	1.8	34.7



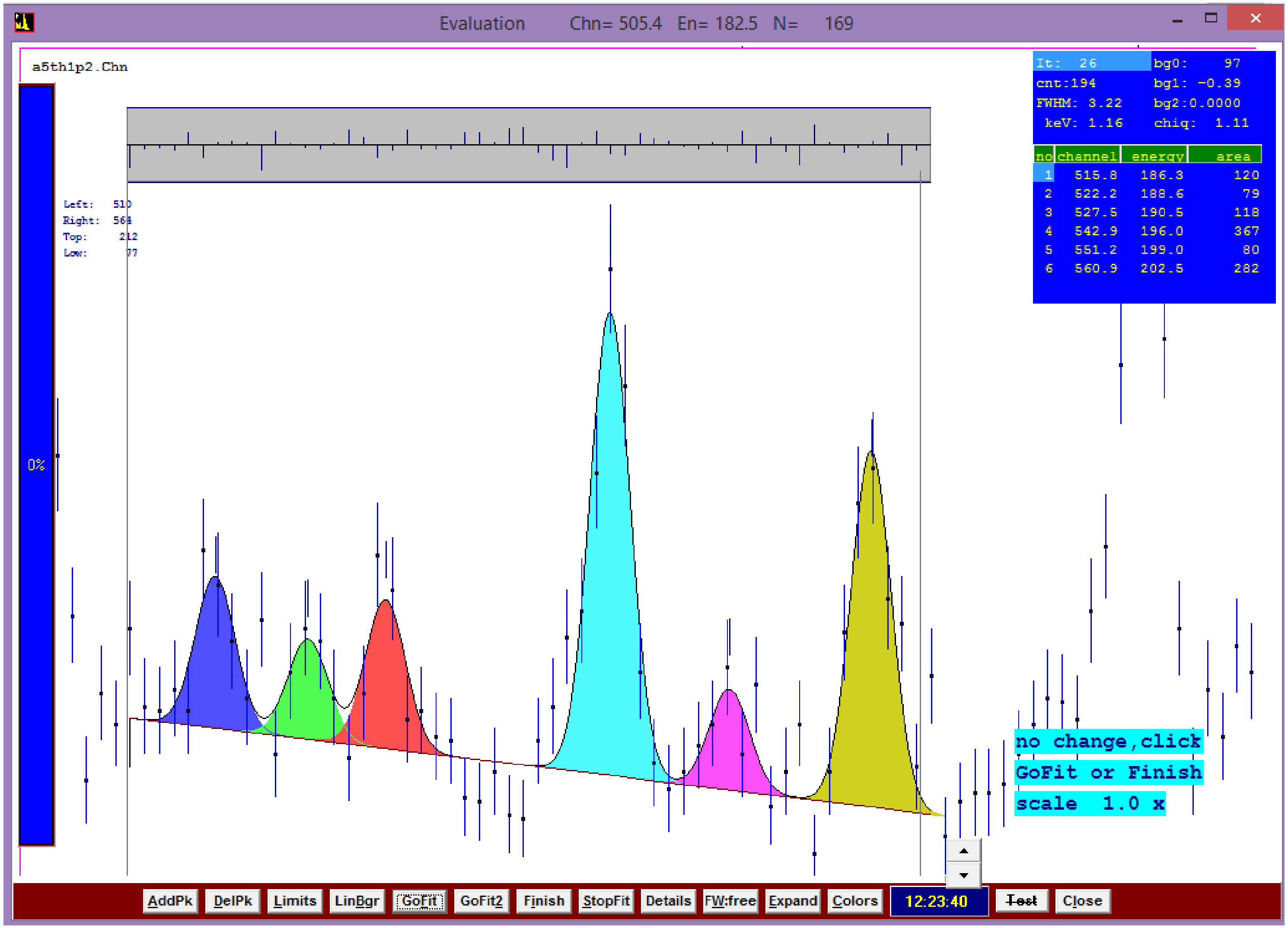
High-Rate Spectroscopy with a HPGe Detectors for Gamma Rays or for X-Rays

ABBREVIATION	DENOMITION	FIRM, MODEL
DBS	Detector Bias Supply	
DET C	HPGe Detector	ORTEC GMX-20190
DET E	HPGe Detector	ORTEC GMX-30
DET D	KX HPGe Detector	ORTEC GeLP-36360/13
DET A	HPGe Detector	CANBERRA GR-1819
DET B	HPGe Detector	ORTEC GMX-23200
DET F	HPGe Detector	CANBERRA
PA	Preamplifiers and HV Filters	
MCA	Multichanell Analyzer	ORTEC 919, 921
DSA	Digital Spectrum Analyzer	Canberra DSA-1000
HRSA	High-Rate Amplifier	ORTEC 973
SA	Spectroscopy Amplifier	CANBERRA 2024,2026,2020

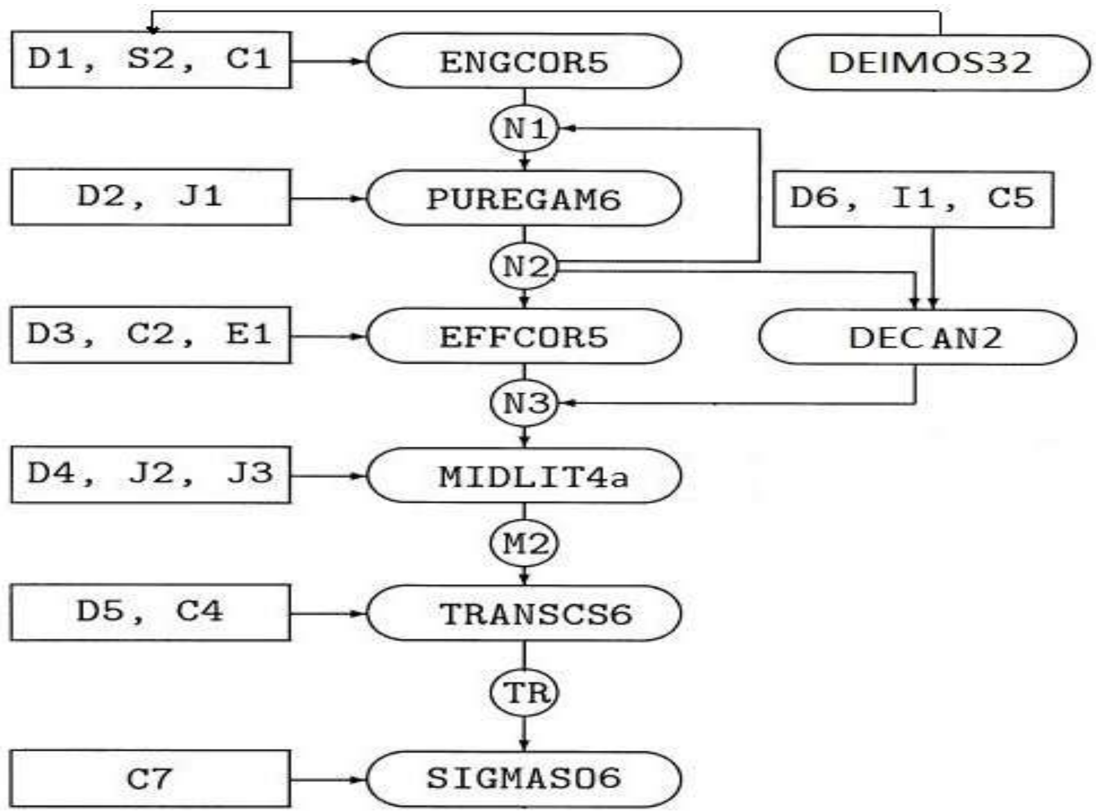


An example of the γ-radiation spectrum of the ^{232}Th sample.

Measurements of the sample were performed repeatedly over various time intervals (from 10 minutes to several days.) Spectra were processed using the DEIMOS32 code.



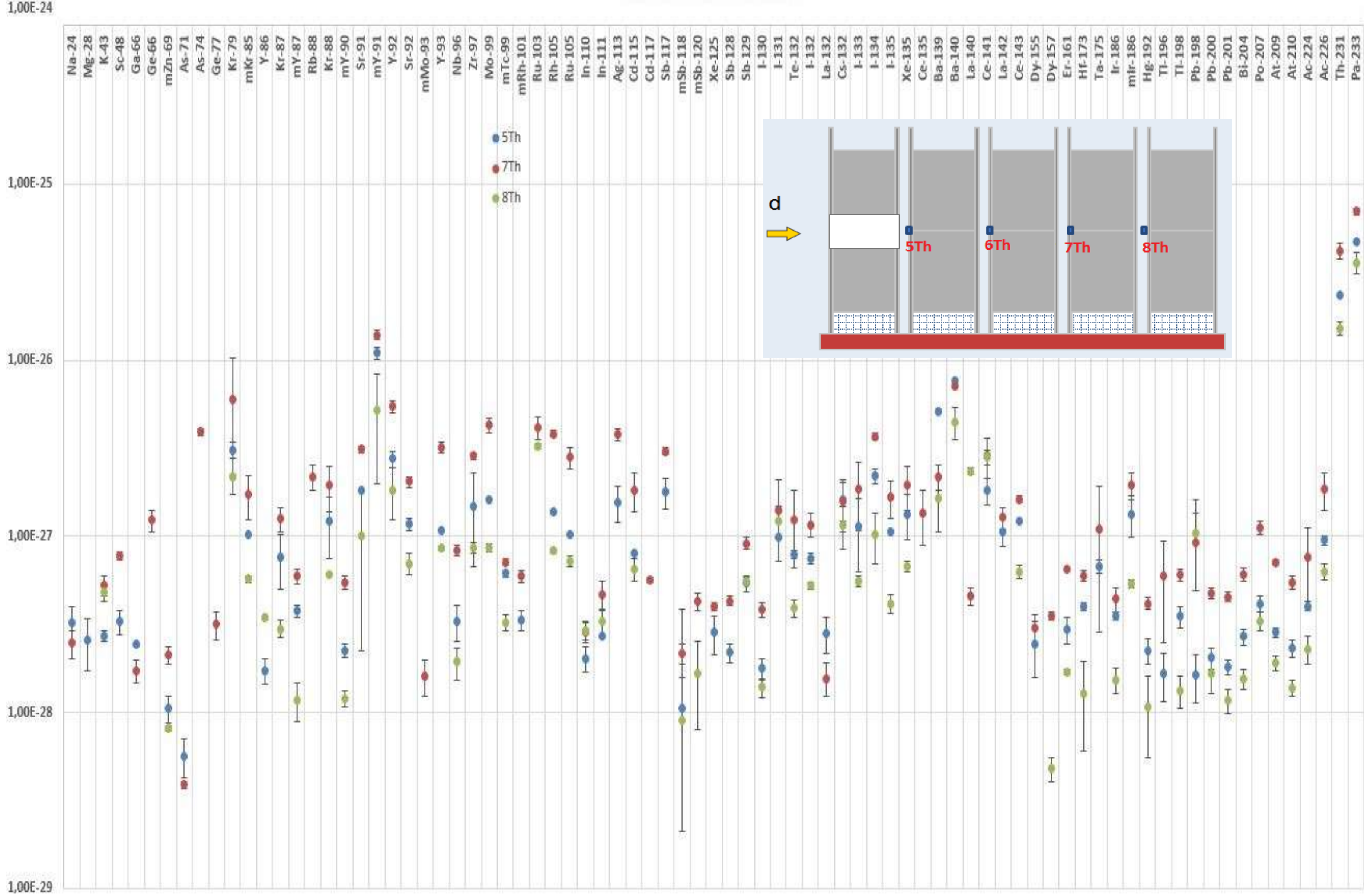
Identification of the nuclei formed by reactions of deuterons and secondary neutrons with ^{232}Th was carried out using the data published in and software package



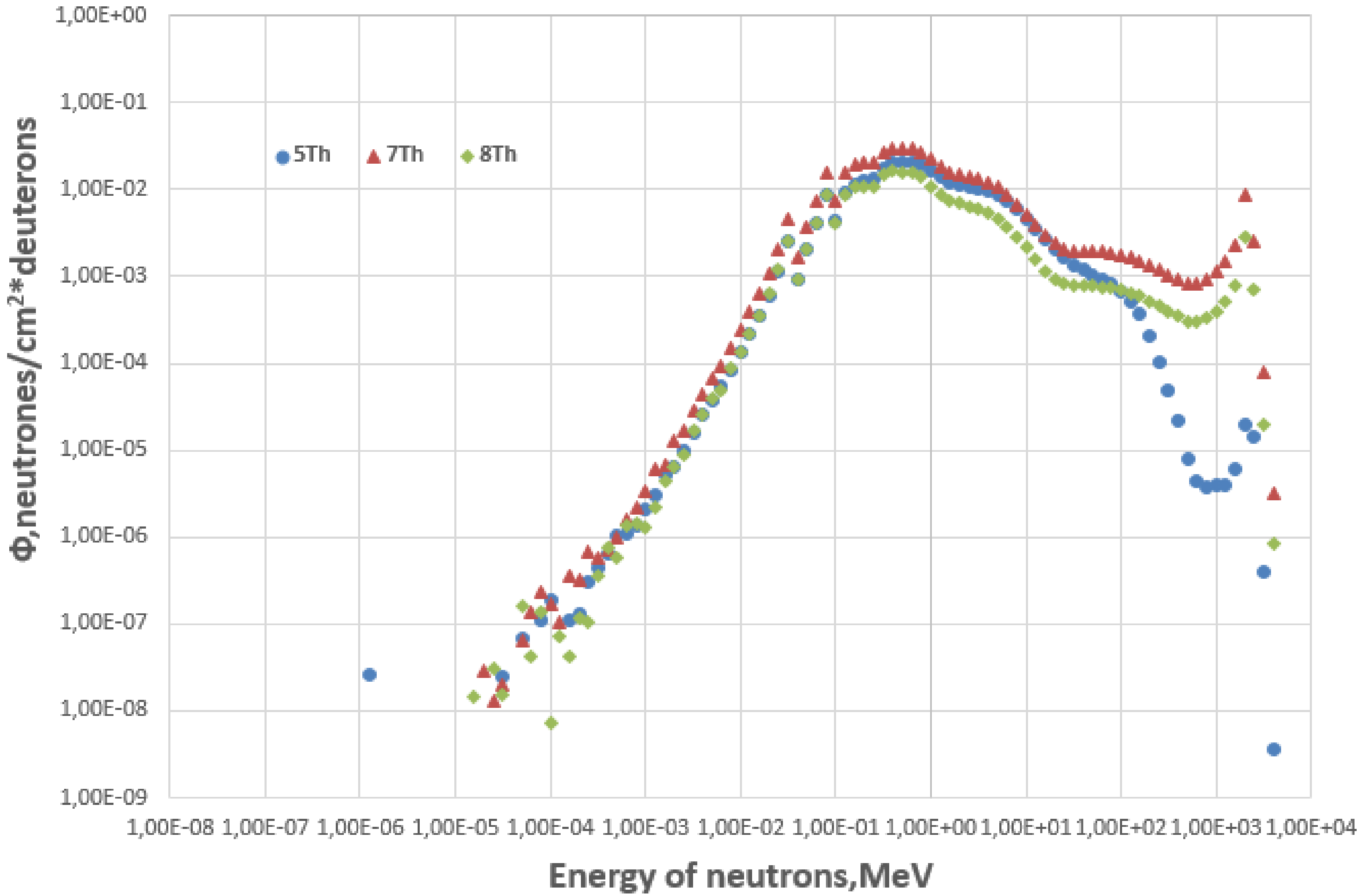
A detailed cascade of codes has been used for the energy calibration, subtraction of background gamma-ray lines and single and double escape peaks, efficiency calibration and determination of experimental half-lives for the identification of several hundreds of gamma-ray lines. Various isotopes and fission fragments are assigned only when energy, half-life, and intensity of peaks match with the values available in the literature.

The reaction rate $R(A_r, Z_r)$ is defined as the number of produced residual nuclei $Q(A_r, Z_r)$ per one atom in the sample N_t and one incident deuteron per second N_d according to the following equation:

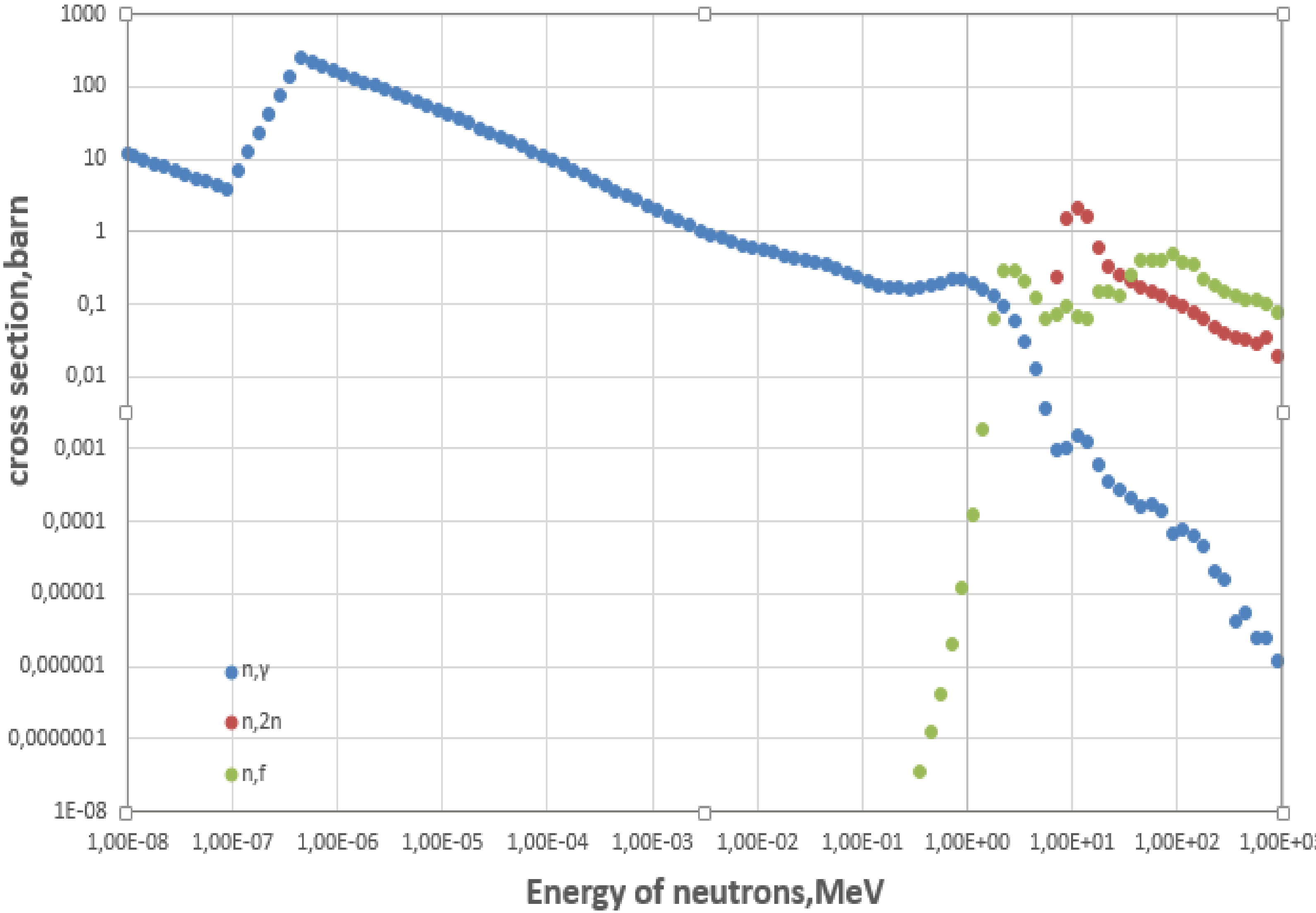
$$\boldsymbol{R(A_r, Z_r) = \frac{Q(A_r, Z_r)}{N_t N_d}}$$



Results of analysis of *γ*-ray spectra of 232Th after irradiation by secondary neutrons from *Ed* = 4 GeV.

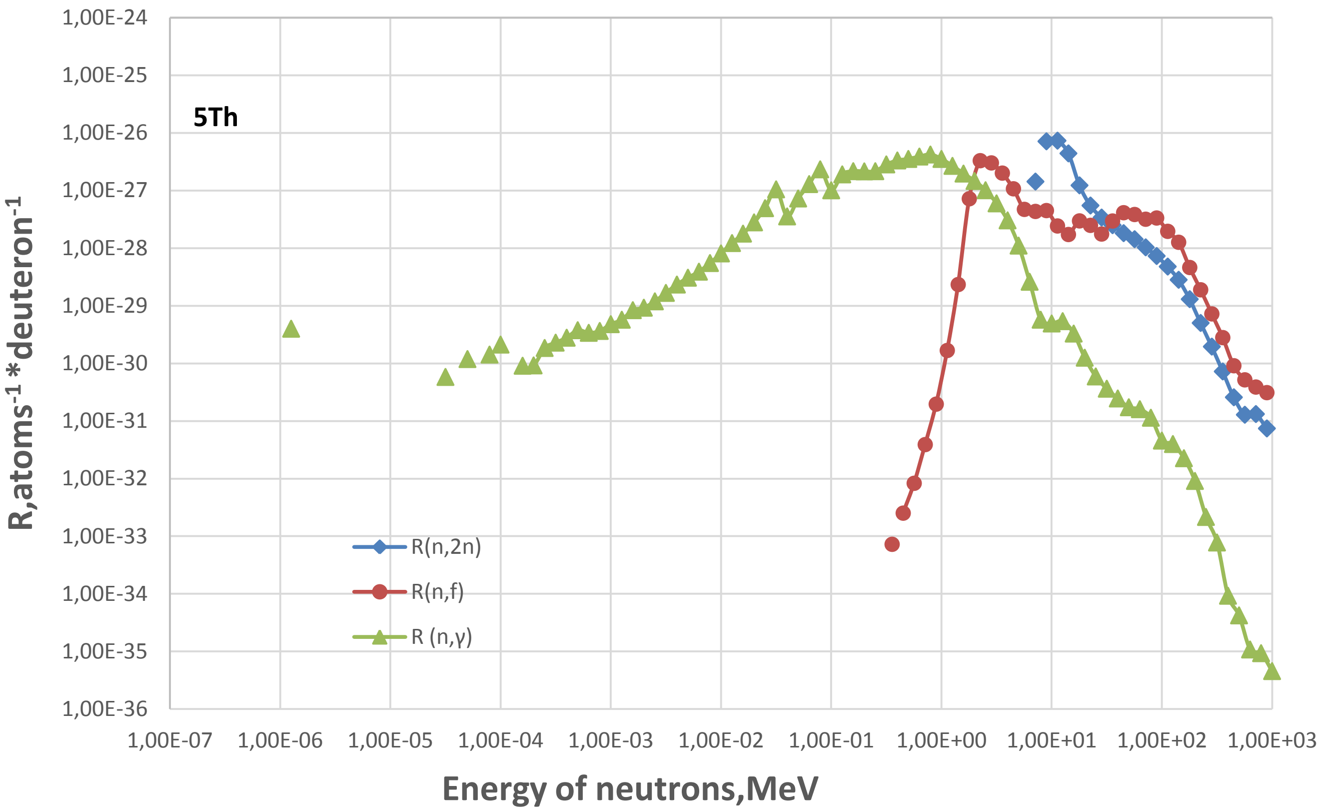


Neutron flux of 5 Th, 7Th, 8 Th .



Cross section, of 232Th(n,γ), 232Th(n,2n), 232Th(n,f)

5Th



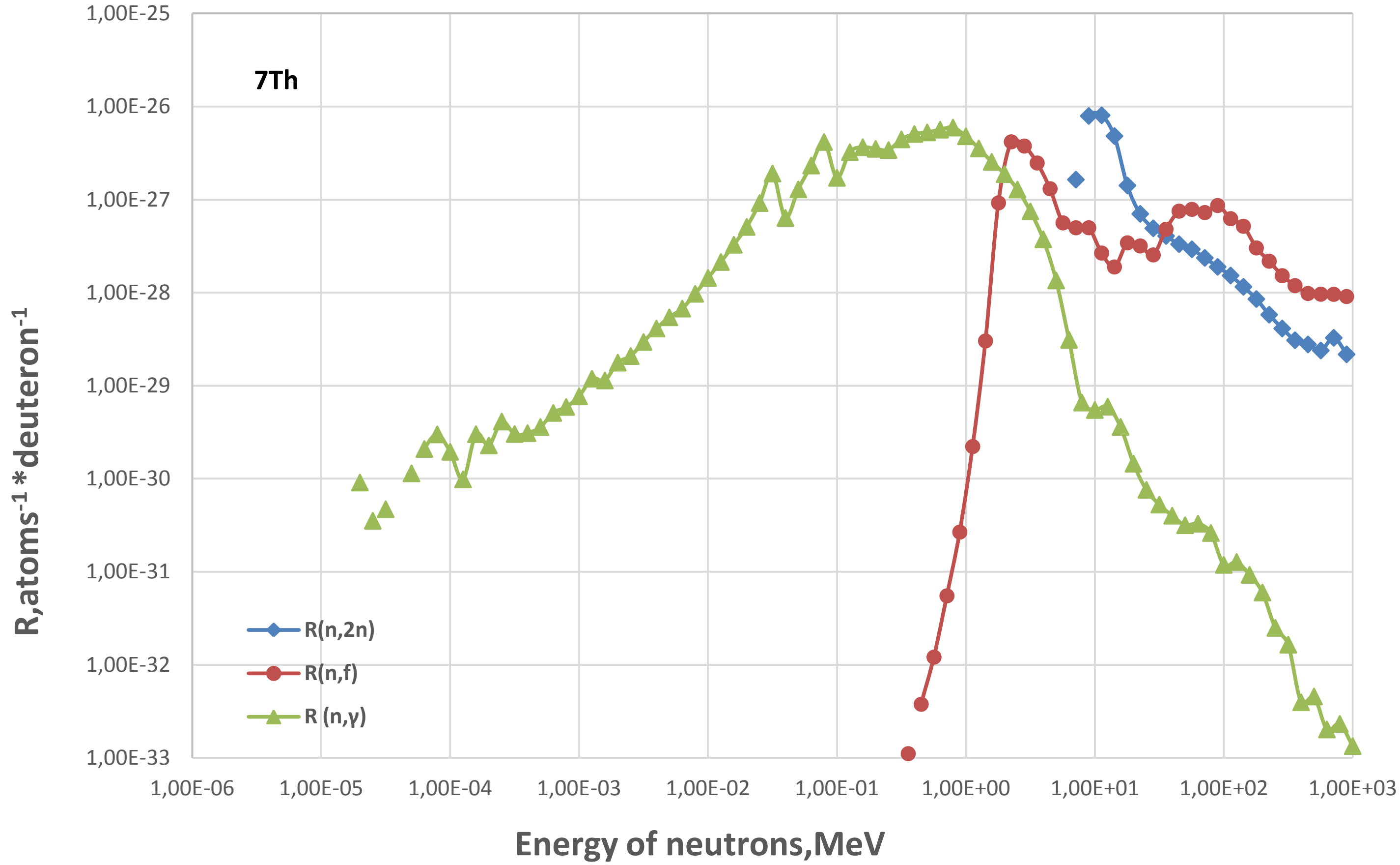
$$R_{calc} = \sum_i \sigma_i(E_n)\Phi_i(E_n)$$

Reaction rate of 232Th(n,2n), 232Th(n,f) and 232Th(n,γ)

σ_i=TALYS1.6

Φ_i=MCNPX 2.7

7Th



$$R_{calc} = \sum_{i} \sigma_i(E_n)\Phi_i(E_n)$$

Reaction rate of 232Th(n, 2n), 232Th(n,f) and 232Th(n,γ)

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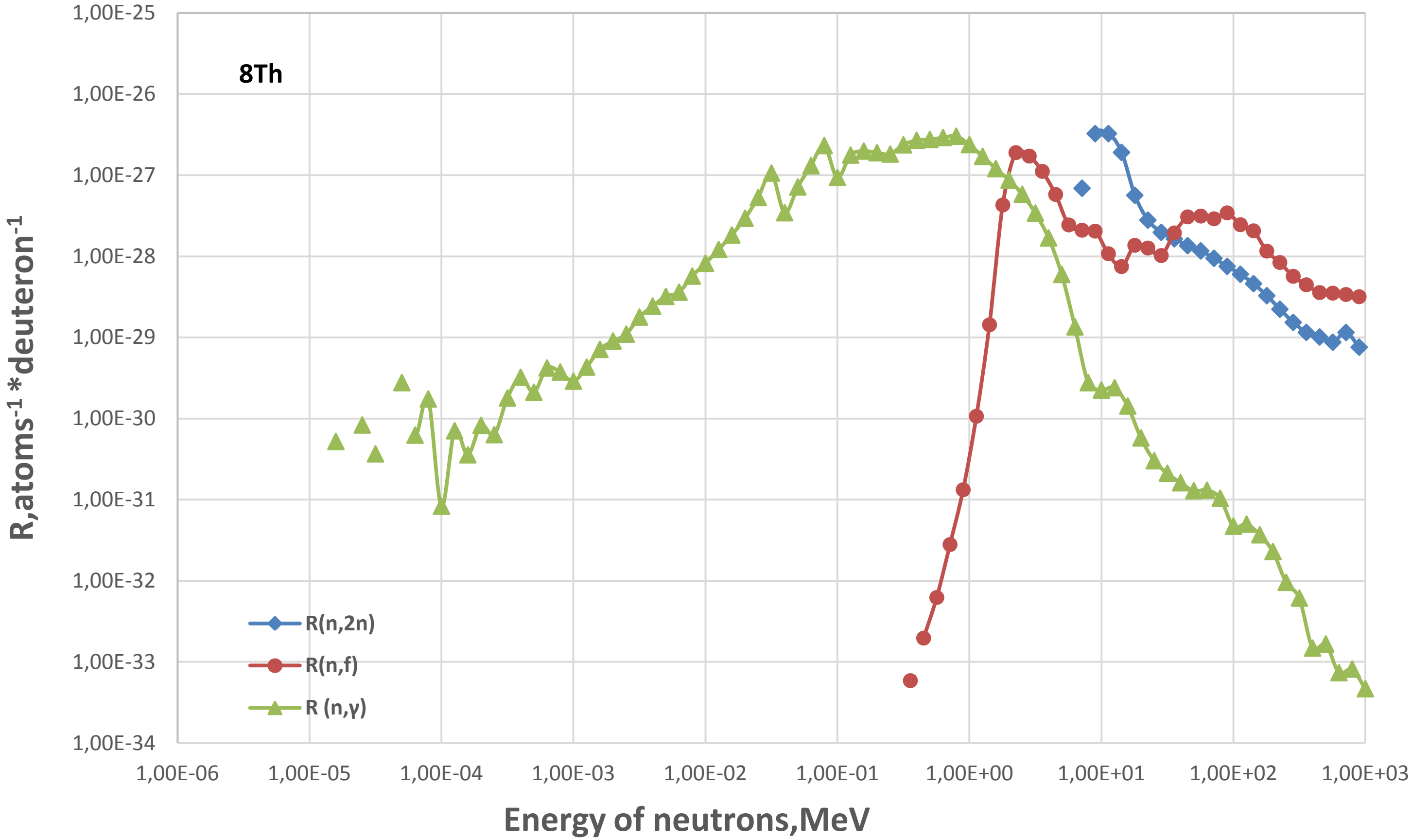
8Th



$$R_{calc} = \sum_{i} \sigma_i(E_n)\Phi_i(E_n)$$

σ_i=TALYS1.6

Φ_i=MCNPX 2.7

Reaction rate of 232Th(n,2n), 232Th(n,f) and 232Th(n,γ)

Comparison of experimentally measured (E) and calculated (C) reaction rates. For the calculations TALYS 1.6 and MCNPX 2.7 by Martin Suchopar.

	7Th		
Reaction	(n,γ)	(n,2n)	(n,f)
R_{expt}(E)	3.58(49)E-26	1.51(13)E-26	6.79(18)E-26
R_{cal}(C)	7.01E-26	2.71E-26	2.15E-26
E/C	0.51(13)	0.56(8)	3.16(27)

	8Th		
Reaction	(n,γ)	(n,2n)	(n,f)
R_{expt}(E)	3.58(49)E-26	1.51(13)E-26	3.38(11)E-26
R_{cal}(C)	3.67E-26	1.10E-26	9.30E-27
E/C	0.98(13)	1.38(8)	3.63(33)

Some important reaction rates in ^{232}Th

5Th

Produced isotope	Reaction	Reaction rate *R*
Pa-233	(n,γ),(d,n)	4.69(82)E-26
Th-231	(d,t),(d,nd),(d,2np),(n,2n)	2.35(63)E-26
Kr-88	(n,f),(d,spallation)	1.22(46)E-27
Sr-91	-//-	1.83(14)E-27
Mo-99	-//-	1.61(60)E-27
Rh-105	-//-	1.37(93)E-27
Ru-105	-//-	1.03(53)E-27
In-110	-//-	2.03(33)E-28
Cd-115	-//-	8.06(45)E-28
I-135	-//-	1.07(10)E-27
Ce-143	-//-	1.22(51)E-27

8Th

Isotope Energy[keV]	I_g [%]	$T_{1/2}$(Library) $T_{1/2}$(Exper.)	<R> R	After beam correction,
K-43		**22.3(1)h**	**5.14(55)E-28**	**4.81(51)E-28**
617.490	79.2	12(9)h	5.14(55)E-28	
Mo-99		**65.94(1)h**	**8.79(18)E-28**	**8.60(17)E-28**
140.511	89.43	2.4(3)d	8.63(51)E-28	
739.50	12.13		3.00(58)E-27	
Rh-105		**35.36(6)h**	**8.60(24)E-28**	**8.25(23)E-28**
319.14	19	20(7)h	8.22(12)E-28	
Ru-105		**4.44(2)h**	**9.99(76)E-28**	**7.25(55)E-28**
316.44	11.1	4.8(20)h	1.76(43)E-27	
469.37	17.5	6(4)h	9.77(14)E-28	
676.36	15.7	7.53(1)h	9.54(26)E-28	
724.2	47	3.7(10)h	9.78(96)E-28	
Cd-115		**2.23(1) d**	**6.70(95)E-28**	**6.52(92)E-28**
336.240	45.9	2.5(8)d	6.34(10)E-28	
527.900	27.45		9.25(26)E-28	
I-133		**20.8(1)h**	**5.98(46)E-28**	**5.57(42)E-28**
529.872	87	12(4)h	5.98(46)E-28	
Ba-140		**12.752(3)d**	**4.50(11)E-27**	**4.48(10)E-27**
537.261	24.39		4.50(11)E-27	
Th-231		**25.52h**	**1.60(14)E-26**	**1.51(13)E-26**
25.646	14.5	15(4)h	1.77(26)E-26	
81.227	0.89	3.2(4)d	2.82(59)E-26	
82.087	0.40	1.0(4)d	2.29(81)E-26	
84.216	6.6	24(4)h	1.50(13)E-26	
102.268	0.41	10(5)h	1.20(46)E-26	
Pa-233		**26.967(2)d**	**3.59(50)E-26**	**3.58(49)E-26**
300.34	6.62	1.7(5)d	8.49(12)E-26	
312.17	38.6	5.1(24)d	3.45(17)E-26	
340.81	4.47	2.7(7)d	5.22(10)E-26	

7Th

Isotope Energy[keV]	I_g [%]	$T_{1/2}$(Library) $T_{1/2}$(Exper.)	<R> R	After beam correction,
Mo-99		**65.94(1)h**	**4.43(37)E-27**	**4.33(36)E-27**
140.511	89.43	2.4(3)d	1.47(44)E-27	
739.50	12.13		8.52(71)E-27	
Ru-105		**4.44(2)h**	**3.83(54)E-27**	**2.78(39)E-27**
316.44	11.1	4.1(13)h	9.35(66)E-27	
724.2	47	4.1(4)h	3.62(17)E-27	
336.240	45.9	1.6(4)d	1.61(11)E-27	
527.900	27.45	1.71(1)d	4.09(40)E-27	
I-133		**20.8(1)h**	**1.96(81)E-27**	**1.83(75)E-27**
529.872	87	10.2(26)h	1.96(81)E-27	
165.864	23.7		2.19(35)E-27	
Ba-140		**12.752(3)d**	**5.88(15)E-27**	**5.85(14)E-27**
162.660	6.22	1.6(5)d	5.37(98)E-27	
537.261	24.39		1.03(29)E-26	
Dy-155		**9.9(2)h**	**3.25(60)E-28**	**2.79(51)E-28**
226.918	68.4	4.9(25)h	3.25(60)E-28	
Hg-192		**4.85(20)h**	**5.60(44)E-28**	**4.16(32)E-28**
274.8	50.4	4.1(15)h	5.31(42)E-28	
306.5	5.4	1.2(3)d	4.05(13)E-27	
Th-231		**25.52h**	**2.91(14)E-26**	**2.75(13)E-26**
25.646	14.5	1.1(6)d	1.28(17)E-26	
81.227	0.89	3.2(5)d	5.00(23)E-26	
84.216	6.6	15.6(25)h	3.36(14)E-26	
102.268	0.41	1.17(1)d	3.68(17)E-26	
163.105	0.155		4.52(21)E-26	
Pa-233		**26.967(2)d**	**6.79(28)E-26**	**6.77(27)E-26**
300.34	6.62		7.22(13)E-26	
312.17	38.6	5.8(29)d	6.94(23)E-26	
340.81	4.47	1.5(6)d	5.75(59)E-26	

As can be seen, the main channels of the interaction of secondary neutrons with nuclei of thorium are 232Th (n, 2n)-reaction with the formation of 231Th. The next is 233Th - the product of (n, γ)- reaction, which decays with a half -life of 22.3 min. in 233Pa, which we have observed.

Products of the fission reaction (n, f) – for example: 85mKr, 99Mo, 115Cd, 133I, 140Ba, 143Ce have been observed as well. Moreover, the products of spallation reactions in 232Th have been detected, such as 43K, 103Ru, 105Rh, 155,157 Dy, 173Hf, 192Hg.

We also note that for samples 6Th, 7Th and 8Th for virtually all products of reaction rate values regularly decrease with the distance from the plane of entry of the deuteron beam into the target.

Conclusions

Interaction of secondary neutrons with ^{232}Th nuclei has been experimentally investigated.

Secondary neutron field has been generated as a result of irradiation of the massive uranium target with the JINR Nuclotron deuteron beam with energy 4 GeV.

A set of four ^{232}Th samples in different positions of the neutron field has been studied.

The reaction rates with $^{232}Th(n,\gamma)$ $^{233}Th \rightarrow ^{233}Pa$, $^{232}Th(d,n)^{233}Pa$, $^{232}Th[(d,t),(d,nd),(d,2np),(n,2n)]^{231}Th$, $^{232}Th[(n,f),(d,spallation)]$ ^{88}Kr and other were obtained and compared with Monte Carlo simulations.


Thank you for
your attention