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J. Adam, A. A. Solnyshkin, M. Stefanik,
V. M. Tsoupko-Sitnikov, L. Zavorka, V. Kumar*,
Chitra Bhatia*, Aruna Bharti*, S. K. Gupta*

A FEASIBILITY STUDY OF NONELASTIC REACTIONS IN THORIUM AND URANIUM BY THE SPALLATION NEUTRONS

*HENP Laboratory, Department of Physics, University of Rajasthan, Jaipur, India

Адам И. и др. E15-2010-23
Оценка возможного наблюдения неупругих реакций
в тории и уране при облучении их вторичными нейтронами

Поглощение нейтронов и реакции деления играют важную роль в реакторе с тепловыми нейтронами. Когда ядерное горючее облучается нейтронами из реакции глубокого расщепления, открывается ряд неупругих каналов реакций, которые влияют на множественность нейтронов и выход реакций деления.

Расчеты вероятности этих реакций указывают на разное поведение делящихся изотопов ^{238}U и ^{232}Th. Показано, что некоторые неупругие реакции приводят к остаточным ядрам с временами жизни больше нескольких десятков минут, которые могут быть идентифицированы измерением их гамма-спектров.

Чтобы реализовать такие измерения, нужно использовать достаточно большое количество образцов $^{\mathrm{nat}}$U и ^{232}Th для облучения вторичными нейтронами, возникающими при взаимодействии протонов с энергией 660 МэВ с толстой свинцовой мишенью. Предлагается примерная схема облучения и измерений.

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A Feasibility Study of Nonelastic Reactions
in Thorium and Uranium by the Spallation Neutrons

Neutron absorption and fission reactions play an important role in a conventional reactor but when fuel is irradiated by the spallation neutrons then a number of nonelastic reactions will play significant role in both multiplication of neutrons and fission reactions. In theoretical study it has been revealed that the fertile and fissile fuels have a different behavior in this respect. Thus, it is imperative to understand the feasibility of such reactions taking place in the fertile and fissile fuel elements before conducting experiment at Phasotron. In the present study it is found that many nonelastic reactions producing fragments or residuals of half-lives greater than several tens of minutes can be studied using the off-line gamma spectrometry. A large amount of samples can be irradiated with the spallation neutrons produced in 660 MeV proton + Pb collision. A tentative design of the experimental setup is also suggested for the experiment at Phasotron.

The investigation has been performed at the Dzhelepov Laboratory of Nuclear Problems, JINR.

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INTRODUCTION

In a high-energy proton collision with a neutron-rich high mass material like lead spallation neutrons are produced up to the highest beam energy. If a reactor system is driven with the spallation neutrons then the neutronics of such a system will be significantly different compared to a conventional reactor where neutron energy is limited to ~ 10.5 MeV. In theoretical study of the effect of spallation neutrons on the neutron multiplication coefficient k_{eff} of the fertile and fissile fuel systems, recently, it has been shown [1] that a large number of nonelastic reaction channels contribute highly significantly for the k_{eff} compared to that in a conventional reactor where only (n, γ) and (n, f) reactions are highly dominant. It has been revealed in case of irradiation of fertile and fissile fuels by spallation neutron spectrum from interaction of 1 GeV protons with lead target that the sum of nonabsorption, nonfission and $(n, > 2n)$ kinds of reactions is ~ 82 mb compared to ~ 256 mb of the sum $\sigma(n,\gamma) + \sigma(n,2n) + \sigma(n,f)$ in case of ^{232}Th and it is ~ 45 mb compared to $\sigma(n,\gamma) + \sigma(n,2n) + \sigma(n,f) = 2280$ mb in case of fissile ^{233}U. This implies that a fertile fuel produces more neutrons by the $(n, > 2n)$ reactions and the fissile element produces more by the fission reactions. This needs experimental validation. It is worth mentioning that in the theoretical calculations [1] spectrum average cross sections are calculated taking simulated neutron spectrum and the point cross sections from the TALYS 1.0 code. Secondly, on inclusion of all nonelastic reactions in calculation of k_{eff} it is revealed that in fertile ^{232}Th fuel one can obtain $k_{\text{eff}} > 1$ which cannot be obtained in a thermal or a fast reactor. This investigation sets up a big goal before the experimentalists to measure cross sections or the reaction rates for the reactions which normally cannot occur in a conventional reactor but they have high probability of occurrence in the environment of spallation neutrons. In partial support of the aforesaid theoretical work we can point out one observation from our recently published experimental work [2] where very small amounts of ~ 93.1 mg of ^{232}Th and 172.3 mg $^{\text{nat}}$U were used, that in the environment of spallation neutrons rate of $(n, 2n)$ is about three times higher in ^{232}Th than in $^{\text{nat}}$U showing that nonelastic reaction channels might exist with higher reaction rate in ^{232}Th than in $^{\text{nat}}$U which carries within it a small proportion of the fissile ^{235}U. To obtain conclusive results of the reaction rates of nonelastic and fission reactions in fertile and fissile elements one needs to use either very high beam current or large amount of the two fissionable elements. Thus, it is imperative that a series of experiments is required to measure the nonelastic reactions of both fissile and fertile fuels for obtaining a new fuel cycle for an ADS reactor.

In the present work a feasibility study of the fuel elements by considering (n, xn) and (n, pxn) kinds of reactions is done for the samples of ^{232}Th and ^{238}U irradiated by the spallation neutrons produced in 660 MeV proton colliding with a thick Pb target. Towards the last, we have suggested a plan of the future experiment wherein a much larger number of fuel elements with much larger amount of their masses are planned for the irradiation.

1. SIMULATION OF SPALLATION NEUTRON SPECTRUM

In simulation of our experiment with 660 MeV protons, we used the lead cylinder with $2R \times L = 8 \times 20$ cm size, where R is the radius and L is the length of the cylinder and its total area is 602.88 cm^2. Assuming that the proton beam of 660 MeV and 1 μA current equivalently corresponds to 6.241×10^{12} protons/s. Thus, for the 660 MeV proton colliding at the axial centre of the Pb block of the given dimensions we have simulated the neutron flux using the CASCADE code [3] at all axial points separated by 1 cm and an overall average comes out to be 13.5 neutron/proton. In Fig. 1 distribution of n/p has been given as a function of neutron energy E_n (MeV). This shows that 8.424×10^{13} neutrons/s may be produced and this corresponds to a neutron flux N_n to be 1.397×10^{11} neutrons/cm^2/s.

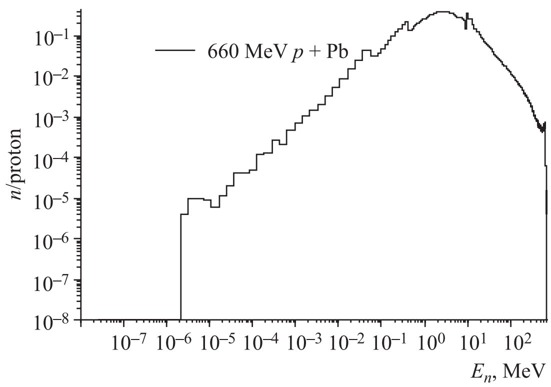


Fig. 1. Simulated number of neutrons $\phi(E_n)$ per beam proton (n/p) plotted as a function of neutron energy E_n (MeV)

2. IDENTIFICATION OF PRODUCED NUCLIDES AND THEIR PRODUCTION RATE

If we irradiate ^{238}U and ^{232}Th having mass $m = 0.15$ g with surface area $S = 1$ cm^2, then the number of atoms at the surface [atom; cm^{-2}], $N_s(= N_{\rm av}m/AS)$ are 3.894×10^{20} and 3.795×10^{20} for ^{232}Th and ^{238}U, respectively. The values of spectrum average cross section are determined as

$$\langle\sigma\rangle = \frac{\int\limits_{E_{\rm thr}}^{E_n(\max)} \sigma(E_n)\phi(E_n)d(E_n)}{\int\limits_{E_{\rm thr}}^{E_n(\max)} \phi(E_n)d(E_n)} \qquad (1)$$

and are calculated using the ENDF/B VII.0 library upto 20 MeV for $(n, 2n)$, $(n, 3n)$ and $(n, 4n)$ reactions, from TALYS 1.0 [4] upto 250 MeV and CASCADE [3] upto 660 MeV. A comparison of (n, xn) cross section calculated using TALYS 1.0 and CASCADE are shown in Fig. 2 and it is clear that these calculations differ significantly. In principle, production rate $Q(A_r, Z_r)$ of radioactive nuclide of mass A_r and atomic number Z_r may be calculated using the relation

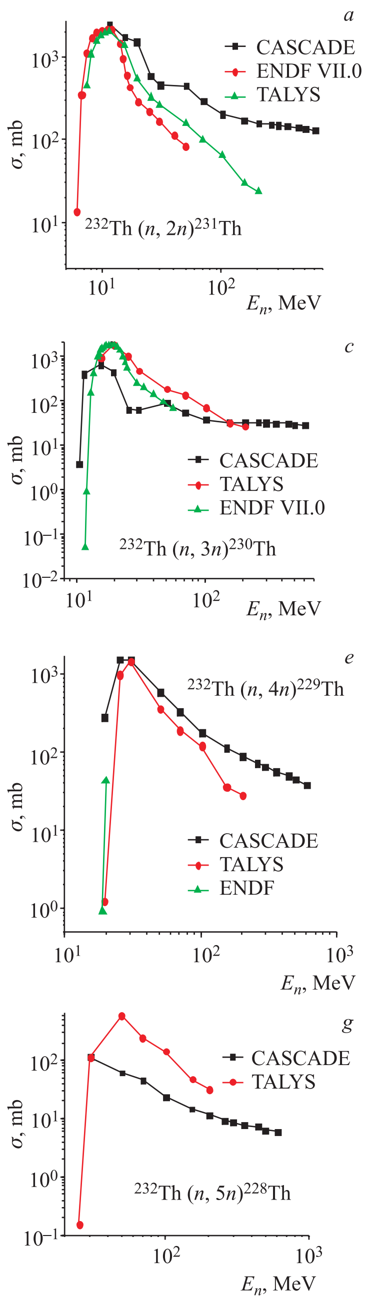
$$Q(A_r, Z_r) = \langle\sigma(A_r, Z_r)\rangle N_s N_n f, \qquad (2)$$

where f is fraction of the simulated neutron flux $\phi(E_n)$ above the threshold energy $(E_{\rm thr})$ of the considered reaction. In terms of various details of the gamma peak (j) there are: the intensity per decay $I_\gamma(j)$, the decay constant (λ), detector efficiency $\varepsilon_\gamma^{\rm abs}(j)$, irradiation (t_1), waiting (t_2) and measurement times $(t_{\rm live}, t_{\rm real})$. The coefficient $\eta_A(Z_t, j)$ accounts for the self-absorption of gamma ray in the sample with Z_t. Then area under the peak $S_\gamma(j)$ is given by [5]

$$S_\gamma(j) = \frac{Q(A_r, Z_r)\varepsilon_\gamma^{\rm abs}(j)I_\gamma(j)(1 - {\rm e}^{-\lambda t_1})(1 - {\rm e}^{-\lambda t_{\rm real}})}{\eta_A(Z_t, j)\lambda{\rm e}^{\lambda t_2}}\frac{t_{\rm live}}{t_{\rm real}}. \qquad (3)$$

Practically, we understand that by analyzing the gamma-ray spectra of the activated nuclides in a sample it is possible to identify the product nuclide (A_r, Z_r) and this depends effectively on the determination of decay constant λ, intensity per decay I_γ and the energy of emitted gamma E_γ from the nuclide. Also, detector efficiency $\varepsilon_\gamma^{\rm abs}$ and the correction factors corresponding to the self-absorption are important for concluding the result of the area under the gamma peak and finally for the estimation of the production rate.

In Table 1 are given basic details such as $T_{1/2}$, E_γ (keV) and I_γ (%) of the product nuclides that may be produced in irradiation of the sample of ^{232}Th by the spallation neutrons, detector efficiency $\varepsilon_\gamma^{\rm abs}$ for the HPGe detector from the



a
CASCADE
ENDF VII.0
TALYS
232Th (n, 2n)231Th
σ, mb
En, MeV
b
CASCADE
TALYS
ENDF VII.0
238U(n, 2n)237U
σ, mb
En, MeV
c
CASCADE
TALYS
ENDF VII.0
232Th (n, 3n)230Th
σ, mb
En, MeV
d
CASCADE
TALYS
ENDF VII.0
238U(n, 3n)236U
σ, mb
En, MeV
e
232Th (n, 4n)229Th
CASCADE
TALYS
ENDF
σ, mb
En, MeV
f
CASCADE
TALYS
238U(n, 4n)233U
σ, mb
En, MeV
g
CASCADE
TALYS
232Th (n, 5n)228Th
σ, mb
En, MeV
h
CASCADE
TALYS
238U(n, 5n)234U
σ, mb
En, MeV

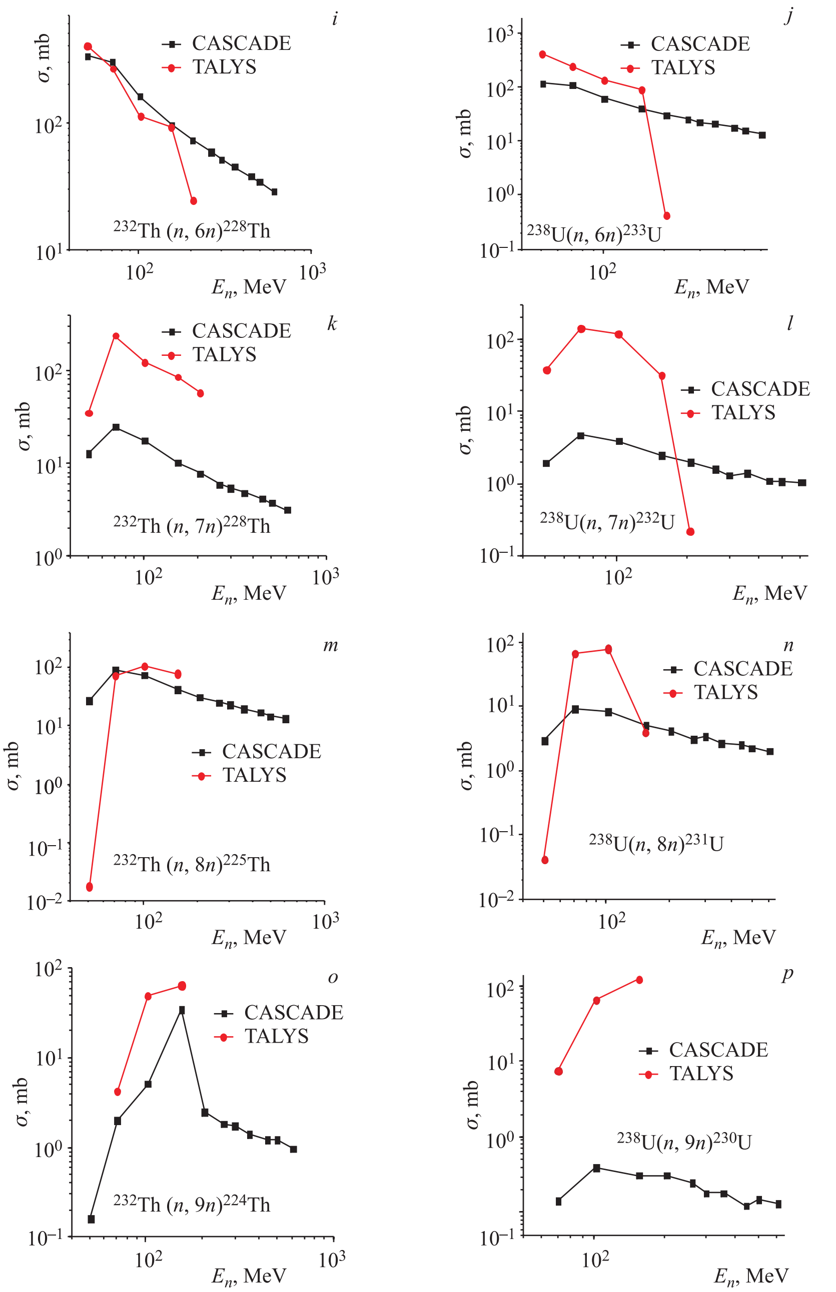


Fig. 2. Comparison of (n, xn) cross section calculated using TALYS 1.0 and CASCADE for the ^{238}U and ^{232}Th samples. The available experimental data for $(n, 2n)$ and $(n, 3n)$ reactions are taken from ENDF/B VII.0 library

ORTEC, self-absorption correction (η_A), product of three time factors $T(t_1,t_2,t_3)$ in Eq. (3), value of $Q(A_r,Z_r)$ calculated from Eq. (2) and the area of the gamma peak S_γ calculated from Eq. (3) corresponding to E_γ in the last column. Similar details are given for the sample ^{238}U in Table 2. The $T(t_1,t_2,t_3)$ factors were calculated for $t_1 = 10$ min, 1 h and 10 h. The time $t_2 = 5$ min was choosen for all combinations. The measurement time $t_{\rm real} = t_{\rm live} = t_3$ was varied from 5 min, 1 h, 19 h, 2 d and 10 d. The different $T(t_1,t_2,t_3)$ factors were calculated to find the optimum conditions for observing the particular residual nuclei. In Tables 1 and 2 the calculations are done with $t_1 = 10$ h, $t_2 = 5$ min and $t_3 = 10$ d.

Table 1. Tentative list of the products of ^{232}Th sample in irradiation by the spallation neutrons

		E_γ, keV	I_γ, %	Efficiency	η_A	$T(t_1,t_2,t_3)$	$Q(A,Z)$	S_γ
^{231}Th	$(n,2n)$	25.646	14.5	0.021	5.453	0.236	1.42E+06	185.22
	25.52 h	84.216	6.6	0.097	1.13	0.236	1.42E+06	1892.31
		89.944	0.94	0.096	1.112	0.236	1.42E+06	271.25
		81.227	0.89	0.097	1.141	0.236	1.42E+06	253.20
^{230}Th	$(n,3n)$	67.67	0.377	0.095	1.224	2.66E-15	3.01E+05	2.34E-13
	7.5E4 y	143.87	0.0488	0.076	1.137	2.66E-15	3.01E+05	2.63E-14
		253.73	0.0111	0.046	1.033	2.66E-15	3.01E+05	3.96E-15
^{229}Th	$(n,4n)$	193.509	4.4	0.060	1.064	2.78E-13	3.46E+05	2.37E-10
	7340 y	210.853	2.8	0.055	1.052	2.78E-13	3.46E+05	1.41E-10
		156.409	1.19	0.072	1.11	2.78E-13	3.46E+05	7.38E-11
		136.99	1.18	0.079	1.156	2.78E-13	3.46E+05	7.75E-11
^{228}Th	$(n,5n)$	84.373	1.22	0.097	1.129	4.07E-06	3.09E+04	1.31E-04
	1.912 y	215.983	0.254	0.054	1.049	4.07E-06	3.09E+04	1.64E-05
		131.613	0.1305	0.081	1.173	4.07E-06	3.09E+04	1.13E-05
		166.41	0.1036	0.068	1.094	4.07E-06	3.09E+04	8.12E-06
^{227}Th	$(n,6n)$	235.971	12.3	0.049	1.039	0.005	1.32E+05	3.67
	18.72 d	256.25	7	0.045	1.032	0.005	1.32E+05	1.93
		50.13	8	0.079	1.55	0.005	1.32E+05	2.55
		79.72	1.89	0.097	1.147	0.005	1.32E+05	1.00
		286.122	1.54	0.041	1.025	0.005	1.32E+05	0.383
^{226}Th	$(n,7n)$	111.12	3.29	0.089	1.269	0.797	7.54E+03	13.89
	30.57 m	131	0.278	0.081	1.175	0.797	7.54E+03	1.16
		242.11	0.866	0.048	1.037	0.797	7.54E+03	2.43
^{225}Th	$(n,8n)$	321.4	23	0.036	1.02	0.452	1.83E+04	66.86
	8.72 m	246	5.06	0.047	1.036	0.452	1.83E+04	19.09
		305.9	4	0.038	1.022	0.452	1.83E+04	12.22
		359	4	0.032	1.016	0.452	1.83E+04	10.38
^{224}Th	$(n,9n)$	178.4	9	0.064	1.079	9.61E-173	2.74E+03	1.41E-171
	1.05 s	413	0.8	0.028	1.012	9.61E-173	2.74E+03	5.73E-173
		234.5	0.4	0.050	1.04	9.61E-173	2.74E+03	5.04E-173
^{230}Ac	$(n,p2n)$	454.95	8	0.025	1.01	—	4.17E+04	0
		508.2	5.15	0.022	1.009	—	4.17E+04	0
^{229}Ac	$(n,p3n)$	146.345	35	0.075	1.131	0.894	4.16E+04	865.97
	62.7(5) m	569.1	91	0.020	1.007	0.894	4.16E+04	657.44

Table 1. Continuation.

		E_γ, keV	I_γ, %	Efficiency	η_A	$T(t_1, t_2, t_3)$	$Q(A, Z)$	S_γ
^{228}Ac	$(n, p4n)$	463.004	4.4	0.024	1.01	0.663	3.26E+04	22.91
	6.15(2) h	794.947	4.25	0.014	1.005	0.663	3.26E+04	12.63
		911.204	25.18	0.012	1.004	0.663	3.26E+04	65.13
^{227}Ac	$(n, p5n)$	160.26	0.0058	0.070	1.104	3.16E-08	3.62E+04	4.21E-09
^{226}Ac	$(n, p6n)$	158.18	17.5	0.071	1.107	0.209	2.17E+04	50.79
	29.37(12)h	230.37	27	0.051	1.042	0.209	2.17E+04	59.33
		253.73	5.7	0.046	1.033	0.209	2.17E+04	11.47
^{225}Ac	$(n, p7n)$	99.91	1.01	0.093	1.085	0.014	1.75E+04	0.21
	10.0(1) d	150.04	0.8	0.074	1.123	0.014	1.75E+04	0.13
		188	0.54	0.061	1.069	0.014	1.75E+04	0.077
^{224}Ac	$(n, p8n)$	131.613	26.9	0.081	1.173	0.88	1.15E+04	188.62
	2.78(17) h	215.983	52.3	0.054	1.049	0.88	1.15E+04	272.50
^{223}Ac	$(n, p9n)$	98.58	0.891	0.093	1.088		1.13E+04	0
		191.3	0.58	0.060	1.066		1.13E+04	0

Table 2. Tentative list of the products of $^{\mathrm{nat}}$U sample in irradiation by the spallation neutrons

		E_γ, keV	I_γ, %	Efficiency	η_A	$T(t_1, t_2, t_3)$	$Q(A, Z)$	S_γ
^{237}U	$(n, 2n)$	59.541	34.5	0.090	1.734	0.027	1.51E+06	721.39
	6.75 d	64.83	1.282	0.093	1.569	0.027	1.51E+06	30.84
		208	21.2	0.056	1.109	0.027	1.51E+06	431.89
		164.61	1.86	0.069	1.199	0.027	1.51E+06	43.14
^{236}U	$(n, 3n)$	49.369	0.078	0.077	2.316	2.84E-20	1.57E+05	1.16E-19
	2.3E7 y	112.75	0.019	0.088	1.128	2.84E-20	1.57E+05	6.63E-20
^{235}U	$(n, 4n)$	185.712	57.2	0.062	1.145	2.82E-23	3.44E+05	2.99E-19
	7.3E8 y	143.764	10.96	0.076	1.284	2.82E-23	3.44E+05	6.30E-20
		163.358	5.08	0.069	1.203	2.82E-23	3.44E+05	2.83E-20
		205.309	5.01	0.056	1.113	2.82E-23	3.44E+05	2.46E-20
^{234}U	$(n, 5n)$	53.2	0.123	0.083	2.04	2.40E-16	1.72E+04	2.06E-16
	2.5E5 y	120.9	0.0342	0.085	1.454	2.40E-16	1.72E+04	8.28E-17
^{233}U	$(n, 6n)$	97.134	0.02	0.094	1.199	5.86E-16	4.88E+04	4.48E-16
^{232}U	$(n, 7n)$	57.766	0.1999	0.088	1.805	3.16E-09	1545.64	4.76E-10
	68.9 y	129.065	0.0682	0.082	1.38	3.16E-09	1545.64	1.98E-10
^{231}U	$(n, 8n)$	25.646	12	0.021	11.32	0.054	2569.20	0.030
	4.2 d	84.216	7	0.097	1.28	0.054	2569.20	0.728
		58.57	0.44	0.089	1.772	0.054	2569.20	0.030
^{230}U	$(n, 9n)$	154.23	0.125	0.072	1.236	0.004	97.43	2.79E-05
	20.8 d	230.37	0.122	0.051	1.085	0.004	97.43	2.17E-05
		72.2	0.6	0.096	1.418	0.004	97.43	1.55E-04
^{236}Pa	$(n, p2n)$	642.35	37	0.017	1.012	0.467	3.47E+04	102.24
	9.1 m	687	9.9	0.016	1.011	0.467	3.47E+04	25.53
^{235}Pa	$(n, p3n)$	380.173		0.030	1.028	—	2.96E+04	0
	24.5 m	645.896		0.017	1.012	—	2.96E+04	0
^{234}Pa	$(n, p4n)$	131.3	18	0.081	1.363	0.634	2.39E+04	162.71
	6.70 h	152.7	6	0.073	1.242	0.634	2.39E+04	53.49
^{233}Pa	$(n, p5n)$	312.17	38.6	0.037	1.042	0.002	1.88E+04	0.623

Table 2. Continuation.

		E_γ, keV	I_γ, %	Efficiency	η_A	$T(t_1,t_2,t_3)$	$Q(A,Z)$	S_γ
	26.967 d	300.34	6.62	0.039	1.046	0.002	1.88E+04	0.111
^{232}Pa	$(n,p6n)$	969.315	41.6	0.011	1.007	0.196	1.38E+04	12.662
	1.31 d	894.351	19.8	0.012	1.008	0.196	1.38E+04	6.536
		453.655	8.61	0.025	1.02	0.196	1.38E+04	5.703
^{230}Pa	$(n,p8n)$	951.95	29.1	0.012	1.007	0.005	4319.52	0.078
	17.4 d	918.48	8.2	0.012	1.008	0.005	4319.52	0.0227
		898.68	5.8	0.012	1.008	0.005	4319.52	0.0164
^{229}Pa	$(n,p9n)$	118.968	0.13	0.086	1.475	0.173	3522.86	0.046
	1.50 d	40.09	0.104	0.059	3.56	0.173	3522.86	0.011
^{228}Pa	$(n,p10n)$	911.204	23	0.012	1.008	0.269	3048.41	2.248
	22 h	968.971	13.9	0.011	1.007	0.269	3048.41	1.278
		964.766	11.4	0.011	1.007	0.269	3048.41	1.052
^{227}Pa	$(n,p11n)$	64.62	5	0.093	1.574	0.834	1778.72	4.394
	38.3 m	110.05	1.24	0.090	1.14	0.834	1778.72	1.445

From the results of calculations presented in Tables 1 and 2 may be pointed out that

1) The reaction product ^{232}Th$(n,2n)^{231}$Th having $T_{1/2} = 25.52$ h can be observed in an experiment being planned at the Phasotron if mass of the sample is large up to $\sim$ 1–2 g and one must take care of the fact that thickness of the irradiated sample is not large for the sake of self-absorption coefficient. At the same time, it will be extremely difficult to observe the products of $(n,3n)$, $(n,4n)$ and $(n,5n)$ reactons because of their half-lives of years. There are week chances of observation of product nuclides corresponding to $(n,6n)$, $(n,7n)$ and $(n,8n)$ reactions and there is completely no chance of observation of product of the $(n,9n)$ reaction. Similarly, amongst the other reaction products, $^{226\text{–}229}$Ac and ^{224}Ac may also be conveniently seen.

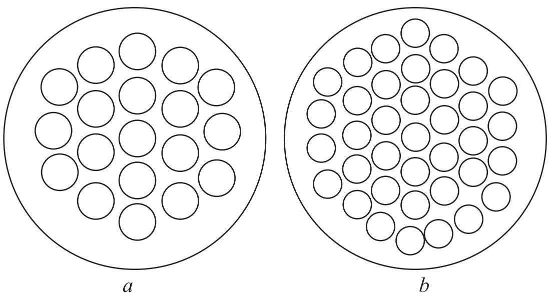


Fig. 3. Design of ^{232}Th (*a*) and $^{\rm nat}$U (*b*) samples each of 8 cm in diameter. Each small block is 11 mm in ^{232}Th and 9 mm in $^{\rm nat}$U

2) The reaction product ^{238}U$(n,2n)^{237}$U is also likely to be observed through both X-ray and gamma-ray detectors and similarly the reaction product ^{238}U$(n,8n)^{231}$U may be observed through the X-ray detector, and there is small chance of observation of the ^{238}U$(n,9n)^{230}$U reaction product through the X-ray detector. In the same way other (n,pxn) kind of reactions may also be observed, e.g., $^{227-235}$Pa, except ^{231}Pa which is having long half-life $\sim 3.38\times10^4$ yrs, may be detected in the planned experiment.

From the results obtained by the above analysis of Tables 1 and 2, we decide to use a big amount of samples and to put for measurements, in such a way so that the self-absorption should not increase and the probability of observing the residual nuclei should enhance. We suggested the irradiation of 19 samples of ^{232}Th and 37 samples of $^{\rm nat}$U and measurement arranged with wide sources as shown in Fig. 3. In such a way the S_γ could be increased by more than one order of magnitude.

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Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/